

Electrical relaxation mechanism in unconventional bismuth cuprate glasses

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The frequency dependent dielectric constant and loss of the unconventional bismuth cuprate glasses have been presented in wide temperature and frequency ranges. Alternating current dielectric loss peak has been observed in the measurable frequency range at higher temperatures, where the measured ac conductivity approaches the dc conductivity. The temperature range where ac loss peak has been observed, varies systematically with glass composition. The dielectric data at these temperature ranges have been analyzed in terms of different theoretical models to find out the possible relaxation mechanism. It has been observed that the dipolar relaxation model with a distribution of relaxation times can best explain the experimental data. The dipolar relaxation occurs due to the hopping of charge carriers between different localized valence states of copper ions within a range of energies near the mobility edge. High value of the dielectric constant observed in these glasses can be attributed to the influence of high polarizability of the Bi^{3+} ions of the unconventional network former Bi_2O_3 to the ac response. The higher distribution of the relaxation times in the present glasses compared to the unconventional lead cuprate glasses indicates less homogeneity of the bismuth cuprate glass system. © 1998 American Institute of Physics. [S0021-8979(98)06213-6]

I. INTRODUCTION

Glasses containing transition metal ions are interesting for their possible applications in threshold and memory switching, etc.^{1,2} The transition metal ion glasses based on the conventional glass network formers such as P_2O_5 , TeO_2 , etc. have been studied extensively.³⁻⁵ Study of ac conductivity of these glasses shows that at low temperatures, ac conductivity is normally proportional to ω^s where ω is the angular frequency and s is the frequency exponent. The value of s is generally less than or equal to one; however, the actual variations of s with temperature and frequency give the information about the conduction mechanism.⁶ On the other hand, at high temperatures, where loss peak is observed, analysis is often made with the dielectric data directly to find out the relaxation mechanism.⁷⁻⁹ Analysis of the dielectric data of the glasses in terms of different models based on Debye or non-Debye types of relaxation mechanisms⁶⁻⁹ give valuable information about the glass system, although there are some controversies in analysis techniques,¹⁰ such as how ac and dc mechanisms coupled, what is the uncertainty in the estimation of peak position, etc.

The glasses based on heavy metal oxide such as Bi_2O_3 and PbO are interesting because they can be used to produce glass ceramics, reflecting windows, layers for optical and optoelectronic devices, etc.¹¹ Recently, the structural studies of the binary cuprate glasses based on Bi_2O_3 and PbO as the unconventional network formers have been reported.¹² These

glasses show unique glass forming ability of the heavy metal ions in presence of transition metal ions. The study of the dc conductivity of bismuth cuprate glasses¹³ reveals that the electronic hopping is the dominant conduction mechanism. The structural studies¹² show that the copper ions occupy the network forming position and thus are unable to diffuse through the host matrix confirming the absence of ionic conduction¹³ in these cuprate glasses. However, the electrical relaxation mechanism of these glasses has not been studied well so far, which is necessary to understand the behavior of the unconventional glass former.

The purpose of the present work is to study the ac relaxation mechanism of the well-characterized unconventional bismuth cuprate glasses in the wide frequency and temperature ranges and the effect of the unconventional network former Bi_2O_3 to the ac response in comparison to the conventional network formers.

II. EXPERIMENTAL PROCEDURE

The preparation of bismuth cuprate glasses has been described in detail elsewhere.¹² In brief, glasses of compositions $x \text{CuO}-(100-x) \text{Bi}_2\text{O}_3$ with $x=27-68$ mol % (Table I) were prepared from the reagent grade chemicals, Bi_2O_3 and CuO . The appropriate mixtures of these chemicals were melted in alumina crucibles in an electrical furnace in the temperature range 1000–1200 °C depending on composition for 1 h. The melts were then quenched by pouring them onto a twin roller. Amorphous nature of the samples was confirmed by x-ray diffraction, while homogeneity was ascertained from scanning electron microscopy. Structural characterization of the samples was made using different techniques.¹²

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TABLE I. Relaxation parameters obtained from dipolar dielectric relaxation (Cole–Cole) model and the experimental values of σ_{dc} and ϵ_{∞} (at 100 kHz and 90 K) for bismuth cuprate glasses.

Glass composition	Temperature (K)	τ_d (s)	α	ϵ_{∞}	ϵ_0	σ_{dc} (10^{-10}) ($\Omega^{-1} \text{cm}^{-1}$)	ϵ_{∞} (exp)
35 CuO-65 Bi ₂ O ₃	389	2.45×10^{-4}	0.46	32	59	10.0	24.5
	433	3.20×10^{-5}	0.46	32	59	95.0	
	478	5.62×10^{-6}	0.45	33	61	820.0	
46 CuO-54 Bi ₂ O ₃	306	2.55×10^{-4}	0.49	34	58	0.35	25.2
	338	3.85×10^{-5}	0.49	34	60	3.50	
	378	5.50×10^{-6}	0.49	35	63	39.00	
56 CuO-44 Bi ₂ O ₃	287	4.50×10^{-4}	0.59	34	158	1.0	29.5
	317	8.50×10^{-5}	0.58	34	158	7.9	
	346	1.50×10^{-5}	0.56	35	155	40.0	
68 CuO-32 Bi ₂ O ₃	208	3.90×10^{-3}	0.48	33	208	0.03	32.5
	253	8.25×10^{-5}	0.48	34	208	0.79	
	310	3.50×10^{-6}	0.48	34	212	53.0	

Depending on conductivity levels of the well-characterized bismuth cuprate glasses, the ac measurements were carried out in a capacitance bridge (GenRad, model-1615A) in the frequency range 10^2 – 10^5 Hz or in an impedance analyzer (Hewlett Packard, model-4192A LF) in the frequency range 10^2 – 10^6 Hz, using gold as an electrode material. The dc measurements were made in a Keithley electrometer (model-617). All measurements were taken in the temperature range 80–500 K. The sample cell was placed in an electric furnace and in a cryostat for measurements above and below room temperature, respectively.

III. RESULTS AND DISCUSSION

The variation of the measured ac conductivity as a function of reciprocal temperature and the dielectric constant as a function of temperature for the 35 CuO-65 Bi₂O₃ glass composition are shown in Figs. 1 and 2, respectively, at three frequencies. The dc conductivity is also included in Fig. 1 for comparison. Two regions have been observed in both Figs. 1 and 2. In the low temperature range, the measured ac conductivity is substantially higher than the dc conductivity and shows a weak temperature dependence but a strong frequency dependence, while dielectric constant shows both weak temperature and frequency dependence. In the high temperature range, the measured ac conductivity approaches the dc conductivity and shows a strong temperature dependence but an almost frequency independence, while dielectric constant shows both strong temperature and frequency dependence. The temperature and frequency dependence of the ac conductivity and the dielectric constant for the other glass compositions are qualitatively similar. The dielectric constant observed in the present glass system is higher compared to the glasses formed with the conventional glass former. It is worth mentioning that at low frequencies and high temperatures, electrode polarization might have significant contribution to the dielectric constant. However, for the present glass compositions, the dielectric constant is found to be independent of the thickness of the samples and the electrode area for all frequencies and temperatures, indicating

that the bulk effect is dominant and the electrode polarization has little contribution to the dielectric properties. This is also confirmed from the values of the high frequency dielectric constant, ϵ_{∞} , shown in Table I which are also high. All these clearly suggest the influence of the unconventional network former Bi₂O₃ on the dielectric properties due to the higher polarizability of bismuth ions.

At higher temperatures, where the measured ac conductivity approaches the dc conductivity, it makes no sense to determine the exponent, s , which is, however, useful and essential for the analysis of the low temperature conductivity data, where ac loss peak cannot be observed in the measured frequency range. The data at high temperatures are analyzed here in terms of different relaxation models to find out the shape as well as frequency and temperature variation of the loss peaks and dielectric data directly.

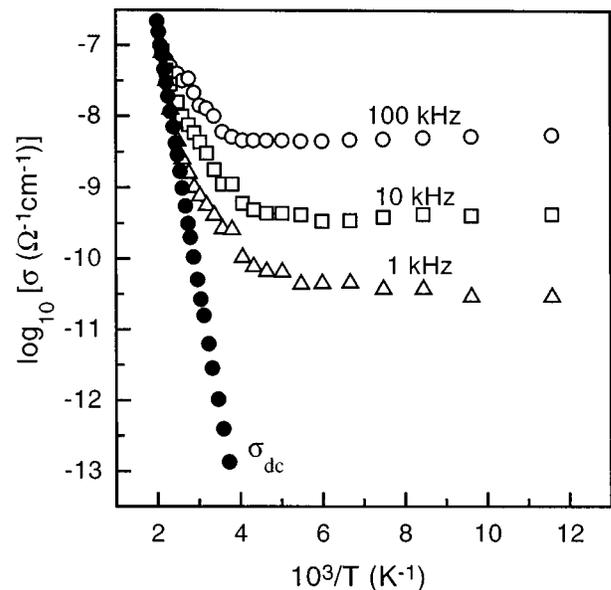


FIG. 1. The temperature dependence of the measured ac conductivity at three different frequencies and dc conductivity for the 35 CuO-65 Bi₂O₃ glass composition.

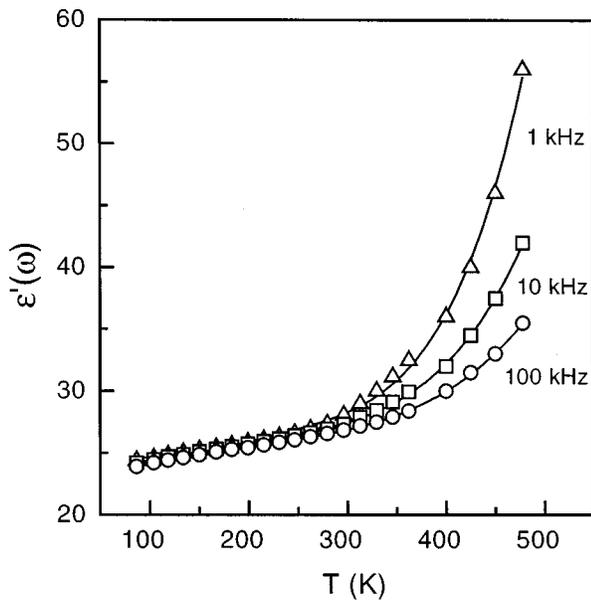


FIG. 2. Variation of dielectric constant, $\epsilon'(\omega)$ with temperature for three different frequencies for the 35 CuO-65 Bi₂O₃ glass composition. Solid curves are to guide the eye.

A. Dipolar relaxation model

Dielectric constant $\epsilon'(\omega)$ and loss $\epsilon''(\omega)$ as a function of frequency at different temperatures for the 46 CuO-54 Bi₂O₃ glass composition are shown in Figs. 3(a) and 3(b), respectively. The dc contribution $\sigma_{dc}/\epsilon_0\omega$ (where ϵ_0 is the free space permittivity) is subtracted from the measured $\epsilon''(\omega)$ in Fig. 3(b) where a broad loss peak is observed. The dielectric constant and loss data of all glass compositions were fitted to the following empirical function proposed by Cole–Cole⁷ for symmetric distribution of relaxation time

$$\epsilon^*(\omega) = \epsilon_\infty + \frac{(\epsilon_0 - \epsilon_\infty)}{1 + j(\omega\tau_d)^{1-\alpha}}, \tag{1}$$

where $\epsilon^*(\omega) = \epsilon'(\omega) + j\epsilon''(\omega)$, and $\epsilon_0, \epsilon_\infty$ are the low- and high-frequency dielectric constants, respectively, τ_d is the dielectric relaxation time, and α is the Cole–Cole distribution parameter having values between 0 and 1. The value of $\alpha = 0$ gives the single Debye type relaxation, while the value of α away from 0 indicates higher distribution of relaxation times. The parameters $\epsilon_\infty, \epsilon_0, \tau_d$, and α were varied to get best fit at different temperatures and frequencies. Such best fits are shown in Figs. 3(a) and 3(b) for 46 CuO-54 Bi₂O₃ glass composition. The agreement between theoretical and experimental values is good. However, the fitting of the loss data below the peak frequency in Fig. 3(b) is not so good. This is because the uncertainties in the low ac loss value which is estimated by subtracting high dc loss value from measured total loss. Similar fits have been observed for other glass compositions. Attempts were also made to fit the dielectric data to the Davidson–Cole function,⁸ where the distribution of relaxation times is asymmetric, but the fittings of the Davidson–Cole function to the measured data at different temperatures and frequencies are not good and thus are not

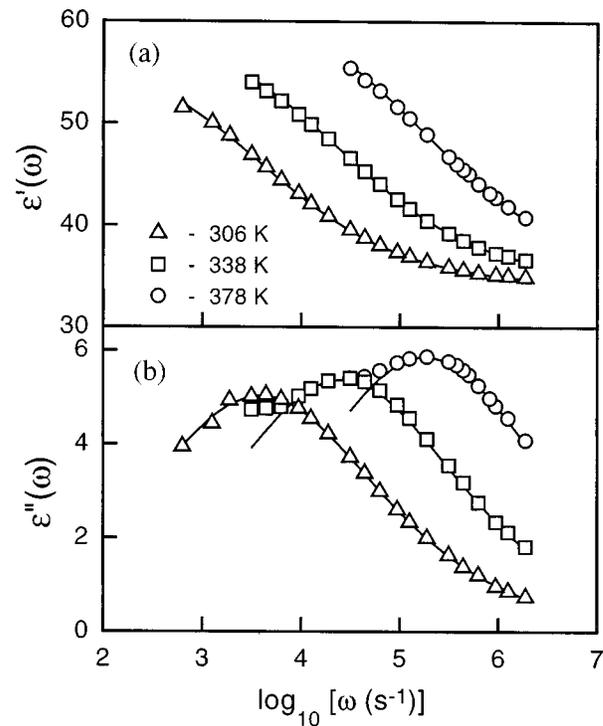


FIG. 3. (a) and (b). The frequency dependencies of $\epsilon'(\omega)$ and $\epsilon''(\omega)$, respectively, at three different temperatures for the 46 CuO-54 Bi₂O₃ glass composition. The dc contribution ($\sigma_{dc}/\epsilon_0\omega$) has been subtracted from the measured $\epsilon''(\omega)$. Solid curves are the best fits to the dipolar relaxation model.

shown here. The parameters obtained from the fits of the Cole–Cole function at different temperatures are shown in Table I for the different glass compositions. Table I indicates that the estimated values of α, ϵ_∞ , and ϵ_0 for all compositions are almost independent of temperature. It may be noted that the estimated values of α and ϵ_∞ do not vary significantly with composition. The values of ϵ_∞ estimated from the model are close to that obtained from experiment. The values of ϵ_∞ and ϵ_0 are much higher than those of the transition metal ion glasses formed with conventional network formers, such as P₂O₅.³ These results clearly suggest higher influence of the unconventional network former Bi₂O₃ similar to PbO (Ref. 14) on the dielectric properties than the conventional formers.³ The value of α for the present glass system is less than that for glasses with conventional former but it is higher compared to the lead cuprate glass system.¹⁴ This is the indication of less homogeneity of the bismuth cuprate glasses compared to lead cuprate glasses. The higher value of α observed for the 56 CuO-44 Bi₂O₃ glass composition compared to other compositions may be due to the clustering effect of the glass.

B. Conductivity relaxation model

Dielectric modulus [$M^*(\omega) = M'(\omega) + jM''(\omega)$] of a system defined by $M^*(\omega) = 1/\epsilon^*(\omega)$, can be used to get information about the relaxation mechanism in absence of a well-defined dielectric loss peak. Figures 4(a) and 4(b) show the variation of $M'(\omega)$ and $M''(\omega)$ with frequency at three different temperatures for 68 CuO-32 Bi₂O₃ glass composi-

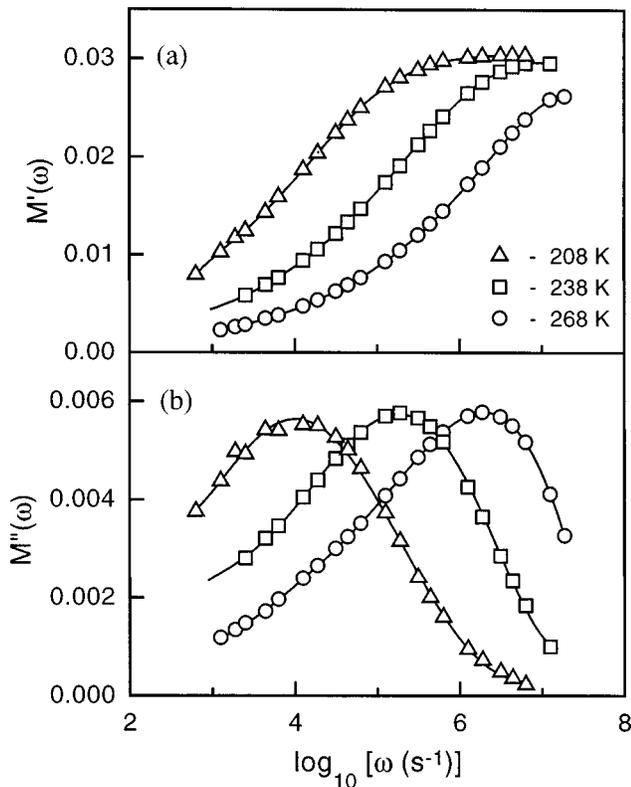


FIG. 4. (a) and (b). The frequency variations of $M'(\omega)$ and $M''(\omega)$, respectively, at three different temperatures for the 68 CuO-32 Bi₂O₃ glass composition. Solid curves are the best fits to the conductivity relaxation model.

tion. It can be noted that $M'(\omega)$ increases to a maximum asymptotic value $M_\infty (=1/\epsilon_\infty)$ with the increase of frequency, while the spectra of $M''(\omega)$ show an asymmetric peak approximately centered in the dispersion region of $M'(\omega)$. The peak shifts to higher frequencies with the increase of temperature. The frequency ω_c , at which the maximum of $M''(\omega)$ (M''_{\max}) occurs, defines the conductivity relaxation time τ_c by $\omega_c \tau_c = 1$. The temperature and frequency dependence of $M'(\omega)$ and $M''(\omega)$ for the other glass com-

positions are similar, except their values are different. The dielectric modulus in the frequency domain can be written as⁹

$$M^*(\omega) = M_\infty [1 - N^*(\omega)], \quad (2)$$

where $N^*(\omega)$ is the one-sided Fourier transform of $-d\phi(t)/dt$ and is given by

$$N^*(\omega) = \int_0^\infty dt \exp(-j\omega t) \left[-\frac{d\phi(t)}{dt} \right]. \quad (3)$$

The distribution of the relaxation times can be predicted from the Kohlraush-Williams-Watts (KWW) function,¹⁵ $\phi(t) = \exp[-(t/\tau_c)^\beta]$, where β is a stretching exponent tending to unity for single Debye type relaxation. Moynihan *et al.*⁹ developed a procedure by taking the following equivalent form of the KWW function

$$\phi(t) = \sum_i g_i \exp(-t/\lambda_i \tau_c), \quad (4)$$

where g_i is the weight factor of the i th peak having relaxation time $\lambda_i \tau_c$ and $\sum g_i = 1$. This KWW function has been used earlier to describe the relaxation behavior of many ionic and electronic glasses and polymers.¹⁶ The data for $M'(\omega)$ and $M''(\omega)$ presented in Fig. 4 have been fitted simultaneously to the theoretical values predicted by Eq. (2) together with Eqs. (3) and (4). Best fits are shown in Fig. 4 for 68 CuO-32 Bi₂O₃ glass composition. The other glass compositions also showed similar fits. The values of ϵ_∞ , τ_c , and β obtained from different glass compositions are shown in Table II. The low frequency dielectric constant (ϵ_0) and the dc conductivity (σ_c) were also estimated from the modulus analysis⁹ and are shown in Table II. Although the fitting of experimental data with the theory is good it can be seen from Table II that the values of ϵ_0 and σ_c estimated from conductivity relaxation model do not agree with the experimental values. Thus the model can predict the data only qualitatively. However, the low values of the β indicate the higher distribution of the relaxation times for the present glass system similar to that predicted from dielectric relaxation model.

TABLE II. Relaxation parameters obtained from conductivity relaxation model for bismuth cuprate glasses.

Glass composition (CuO mol %)	Temperature (K)	τ_c (s)	β	$\epsilon_\infty (=1/M_\infty)$	ϵ_0	σ_c (10^{-8}) ($\Omega^{-1} \text{ cm}^{-1}$)
35	389	6.92×10^{-5}	0.30	33.0	498	0.5
	433	8.30×10^{-6}				3.8
	478	1.38×10^{-6}				22.8
46	306	7.94×10^{-5}	0.20	34.5	4347	0.03
	338	1.26×10^{-5}				0.20
	378	1.78×10^{-6}				1.43
56	252	1.26×10^{-4}	0.25	35.0	1225	0.1
	287	1.00×10^{-5}				1.3
	317	1.58×10^{-6}				8.2
68	208	7.94×10^{-5}	0.35	32.5	271	0.7
	253	1.82×10^{-6}				31
	310	7.08×10^{-8}				810

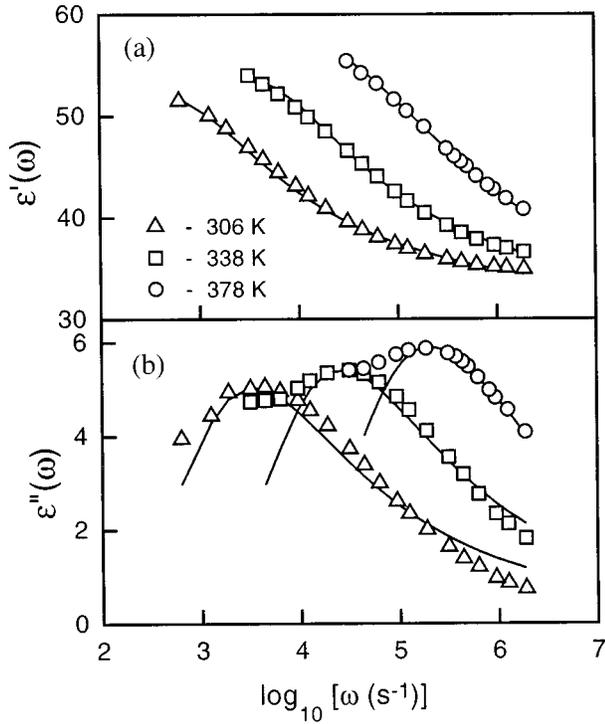


FIG. 5. (a) and (b). The frequency dependencies of $\epsilon'(\omega)$ and $\epsilon''(\omega)$, respectively, same as in Fig. 3. Solid curves are the best fits to the random-free-energy-barrier model.

C. Random-free-energy-barrier model

Based on continuous time random walk approximation,¹⁷ Dyre proposed following random-free-energy-barrier model¹⁰ in which the ac and dc conductivities arise from the same hopping mechanism

$$\epsilon^*(\omega) = \frac{\sigma_R}{\epsilon_0} \left[\frac{\tau_R}{\ln(1 + j\omega\tau_R)} + \frac{j}{\omega} \right], \quad (5)$$

where σ_R is the dc conductivity and τ_R is the relaxation time predicted by this model and are related by $\epsilon_0\Delta\epsilon = \sigma_R\tau_R/2$, where $\Delta\epsilon = \epsilon_0 - \epsilon_\infty$. The data of the present glass compositions have been analyzed in the light of the random-free-energy-barrier model. The experimental data for $\epsilon'(\omega)$ and $\epsilon''(\omega)$ at different temperatures have been fitted simultaneously to the random-free-energy-barrier model, using σ_R , ϵ_∞ , and τ_R as variable parameters. Best fits to the dielectric constant $\epsilon'(\omega)$ and loss $\epsilon''(\omega)$ are shown in Figs. 5(a) and 5(b), respectively, for a glass composition at three temperatures. Reasonable fits were observed at all temperatures and frequencies. The values for the estimated parameters σ_R , ϵ_∞ , and τ_R for all glass compositions are shown in Table III. The estimated values of ϵ_0 and ϵ_∞ are independent of temperatures. However, the values of ϵ_∞ are lower than the experimental values for the glass compositions with higher CuO content and the values of ϵ_0 are lower than the values obtained from dielectric and conductivity relaxation models. The estimated values of σ_R also differ considerably from that of the experimental values. Thus this model also can predict the dielectric data for the present glass system only qualitatively.

TABLE III. Relaxation parameters obtained from random-free-energy-barrier model for bismuth cuprate glasses.

Glass composition (CuO mol %)	Temperature (K)	τ_R (s)	ϵ_0	ϵ_∞	σ_R ($10^{-8} \Omega^{-1} \text{cm}^{-1}$)
35	398	7.80×10^{-4}	53.5	30.5	0.5
	433	1.05×10^{-4}	54.0	30.4	3.9
	478	1.90×10^{-5}	54.0	30.2	21.5
46	306	1.40×10^{-3}	53.0	28.4	0.3
	338	1.80×10^{-4}	54.5	28.0	2.5
	378	2.30×10^{-5}	56.5	27.8	20.0
56	287	2.00×10^{-3}	118	16.0	0.9
	317	3.00×10^{-4}	121	16.0	6.1
	346	6.20×10^{-5}	122	15.0	29.5
68	208	1.50×10^{-2}	180	10.0	0.2
	253	4.00×10^{-4}	180	10.0	7.5
	210	1.20×10^{-5}	182	10.0	155.0

D. Conduction and relaxation mechanism

The inverse relaxation times (i.e., relaxation frequencies) predicted from different models are plotted as a function of inverse temperature in Fig. 6 for 46 CuO-54 Bi₂O₃ glass composition. The experimental dc conductivity in the same temperature range is also included in Fig. 6 for comparison. The other glass compositions also showed similar temperature dependence. The dc conductivity in this limited temperature range can be fitted to the Arrhenius equation: $\sigma_{dc} = \sigma_0 \exp(-W/kT)$, where W is the activation energy for the dc conductivity. The activation energy calculated using the Arrhenius equation is shown in Table IV for all glass compositions. It may be noted from Fig. 1 that at low temperatures dc conductivity is not Arrhenius. The activation energy decreases with the decrease of temperature which can be

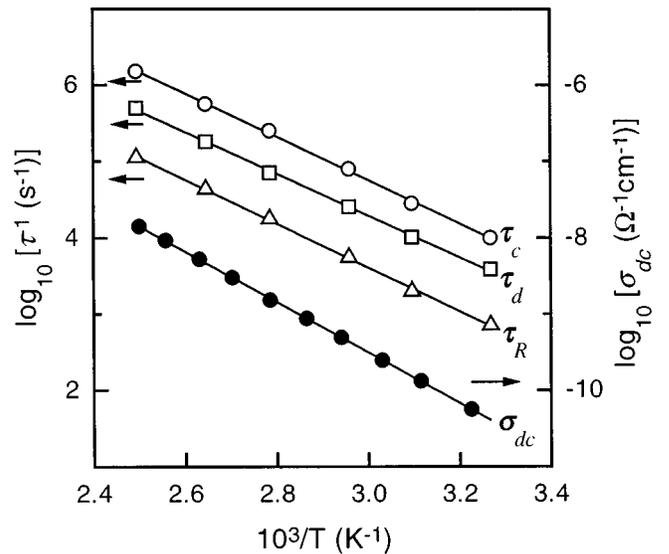


FIG. 6. Temperature dependence of the dc conductivity (σ_{dc}) and the relaxation times predicted by the dipolar relaxation (τ_d), conductivity relaxation (τ_c), and random-free-energy-barrier (τ_R) models for the 46 CuO-54 Bi₂O₃ glass composition. Solid lines are the least-square straight line fits to the data.

TABLE IV. Activation energies (W and W_r) for the dc conductivity (σ_{dc}) and relaxation times (τ), and pre-exponential factor (τ_0) predicted by different models for different bismuth cuprate glass compositions.

Glass composition (CuO mol %)	Temperature range (K)	τ_d		τ_c		τ_R		σ_{dc} W (eV)
		W_d (eV)	τ_0 (10^{-13} s)	W_c (eV)	τ_0 (10^{-14} s)	W_R (eV)	τ_0 (10^{-13} s)	
35	389–478	0.68	12.6	0.70	7.50	0.66	15.0	0.71
46	306–401	0.54	3.45	0.56	5.45	0.57	6.0	0.60
56	252–346	0.48	16.5	0.62	3.80	0.50	25.0	0.49
68	208–310	0.38	22.4	0.39	1.58	0.38	27.5	0.42

accounted for the polaron hopping¹⁸ and we have discussed in details elsewhere.¹³ The relaxation times predicted by each model also show similar activated behavior: $\tau = \tau_0 \exp(W_r/kT)$, where W_r is the activation energy of the relaxation time and τ_0 is the high temperature limit of the relaxation time. Note W_r and τ_0 for each models are shown in Table IV for all glass compositions. The activation energies of the relaxation times in the three cases are nearly same and are very close to the activation energy of the dc conductivity. The values of the pre-exponential factor, τ_0 , on the other hand, are different for the three models.

The different parameters obtained from different models by best fitting procedure are not all physical. The values of dc conductivity predicted from conductivity relaxation and random-free-energy-barrier models vary significantly from the experimental values. Similarly, the values of ϵ_0 predicted from conductivity relaxation model are too high and some of the values of ϵ_∞ predicted from random-free-energy-barrier model are too low. On the other hand, the fitting of the data to the dipolar relaxation model is good and the parameters obtained from it are also physical. In transition metal ion glasses the states are localized and distributed at random within the tail of an energy band associated with the transition metal ions, where the density of states may be higher than that in more conventional semiconductors. In these glasses the localization is enhanced by polaron formation.¹⁹ At low temperatures, hopping occurs between localized states near the Fermi level (within kT). But at higher temperatures in the dispersion regions, hopping occurs between localized states within an appropriate range of energies and site separations lying below and closer to the mobility edges. The dipole is formed between two valence states of copper ions which are localized. The behavior of these dipoles in the mobility edge controls the relaxation mechanism. The values of α and β on the other hand, reflect the degree of coupling of these dipoles. The values of these parameters obtained for the present glass system indicate the presence of different dipoles having different relaxation times due to the inhomogeneity or strong interaction of these dipoles with the glass network.

IV. CONCLUSIONS

Alternating current conductivity and dielectric properties of the unconventional bismuth cuprate glasses have been

measured over the frequency range 10^2 – 10^6 Hz and temperature range 80–500 K. Analysis of the data at high temperatures, where the ac conductivity approaches the dc conductivity has been made in terms of different empirical relaxation models of which the dipolar dielectric relaxation with a distribution of relaxation times, provides a better description of the electrical relaxation mechanism. The distribution parameters, α or β obtained for the present glass system indicate the higher distribution of relaxation times (i.e., less homogeneity) compared to the lead cuprate glass system. The high value of the dielectric constant obtained for the bismuth cuprate glasses similar to the lead cuprate glass system can be attributed to the influence of the unconventional network former (i.e., high polarizability of bismuth ions) to the relaxing dipoles.

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