

Certain electrical properties of LiF-PbO-B₂O₃ glass system

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Dielectric constant ϵ , loss $\tan \delta$ ac conductivity σ , of LiF-PbO-B₂O₃ glasses with increasing concentration of PbO (in the range 0-60 mol % in steps of 20 mol%) are studied in the frequency range 10^2 - 10^7 Hz and in the temperature range 30-300 °C. The dc conductivity and dielectric breakdown strength of these glasses are also measured. The values of ϵ , $\tan \delta$, σ_{ac} and σ_{dc} are found to increase with increase in the concentration of PbO. Activation energy for ac conduction, dc conduction and the value of dielectric breakdown strength are found to decrease with increase in the concentration of PbO. With the help of infrared spectra, increase in the values of ϵ and $\tan \delta$ with frequency and temperature are identified due to space charge polarization. An attempt has been made to explain ac conduction phenomenon on the basis of quantum mechanical tunneling model (QMT)/carrier-barrier hopping model.

1 Introduction

The study of dielectric properties such as dielectric constant ϵ , loss tangent $\tan \delta$, ac conductivity σ_{ac} over a range of frequencies and temperatures, dc conductivity and the dielectric break down strength of the glasses help in assessing their insulating character. Work along these lines was carried out in recent years on a variety of inorganic glasses by a number of researchers yielding valuable information¹⁻⁵.

Lithium fluoroborate glasses are well known due to their variety of applications in phosphors and solar energy converters and in a number of electronic devices. Extensive studies on various physical properties like ESR studies and thermoluminescence properties of lithium fluoroborate glasses doped with different transition and rare earth metal ions are available^{6,7}. Quite recently the authors have reported the results of studies on various elastic and some insulating properties of LiF-B₂O₃ glasses doped with certain rare earth ions^{8,9}. These studies have yielded valuable information regarding the mechanical and insulating strength of these glasses. It is expected that when the modifier PbO enters the glass network, it forms the stable glass due to its dual role, one as modifier (if PbO is ionic) and the other as glass former (if PbO is covalent) and in turn brings interesting changes in the insulating character of these glasses. To have a comprehensive understanding over the influence of the modifier PbO on the insulating character of lithium fluoroborate glasses, the authors have undertaken a systematic investigation on dielectric

constant (ϵ) loss $\tan \delta$ and ac conductivity σ in the frequency range 10^2 - 10^7 Hz and in the temperature range 30- 300 °C, dc conductivity in the temperature range 30-300 °C and also the dielectric breakdown strength in the air medium at room temperature.

2 Experimental Methods

For the present investigation the following compositions of the LiF-PbO-B₂O₃ (all in mol %) are chosen: Glass A : 20 LiF-80 B₂O₃, Glass B : 20 LiF-20 PbO-60 B₂O₃, Glass C : 20 LiF-40 PbO-40 B₂O₃ and Glass D: 20 LiF-60 PbO-20 B₂O₃.

Appropriate amounts of Analar grade reagents of PbO, LiF and H₃BO₃ were thoroughly mixed and melted in a platinum crucible at 900 °C for about 2 hr until a bubble free liquid was formed. The resultant melt was then cast in a brass mould and subsequently annealed at 300 °C. The amorphous state of the glasses was checked by X-ray diffraction spectrum recorded on SIEFERT Diffractometer Model SO DEBYE FLUX- 2002 fitted with copper target and nickel filter operated at 40 kV, 30mA. The samples were then ground and finely polished. The final dimensions of the samples were about 1 cm × 1 cm × 0.2 cm. The density d of the glasses was determined to an accuracy of 0.001 by standard principle of Archimedes using xylene (99.99% pure) as the buoyant liquid. A thin coating of silver paint was applied (to the larger area faces) on either side of the samples to serve as electrodes. The dielectric measurements were made on LCR Meter (Hewlett-Packard Model-4263B)

in the frequency range 10^2 – 10^5 Hz and on Radart Q-Meter in the frequency range 10^5 – 10^7 Hz. The dielectric breakdown strength for all the glasses was determined at room temperature in air medium using a high ac voltage breakdown tester (ITL Model AAH-55) operated with an input voltage of 250 V at a frequency of 50 Hz. It was ensured that all the glasses used for this study were of identical thicknesses. The dc conductivity measurements for these glasses were made on Keithly Electrometer Model 236, which can measure the current up to an accuracy of pico Ampere.

3 Results

From the measured values of the density d and calculated average molecular weight $\bar{M}$ various physical parameters such as Pb^{2+} ion concentration N_i , mean Pb^{2+} ion separation distance R_i , and the field strength F_i which are useful for understanding dielectric properties of these glasses are evaluated and presented in Table 1.

The dielectric constant (ϵ) of pure lithium fluoroborate glasses (20:80 mol % of LiF-B₂O₃) at 30 °C and at 10^6 Hz is found to be 11.26 and is almost independent of frequency; when PbO is added into these glasses, ϵ values at room temperature are found to increase gradually at any frequency with increase in the concentration of PbO and found to decrease with increasing frequency. The dielectric loss ($\tan \delta$) variation with frequency at room temperature for all these glasses has also exhibited a similar behaviour.

The variation of dielectric constant and loss with temperature at different frequencies for all the glasses show that ϵ and $\tan \delta$ values increase considerably at low frequencies and in high temperature region. Fig.1 pre-

sents the variation of dielectric constant with temperature at different frequencies for glass C. The comparison of variation of a dielectric constant with temperature for lithium fluoroborate glasses at 1kHz is shown Fig. 2. It is observed that the value of ϵ (at a fixed temperature) increases with increase in the concentration of PbO. The variation of dielectric loss with temperature has also exhibited a similar behaviour; hence a plot is not presented. Fig. 3 presents the variation of dielectric loss with temperature at different frequencies for glass B. The summary of data on the dielectric constant and dielectric loss for all the glasses is presented in Table 2.

The ac conductivity σ is calculated at different temperatures using the equation

$$\sigma = \omega \epsilon_0 \epsilon \tan \delta \quad \dots(1)$$

(where ϵ_0 is the vacuum dielectric constant) for different frequencies and the plots of $\log \sigma$ against $1/T$ are shown

Table 1 — Various physical properties of LiF-PbO-B₂O₃ glasses

Property	Glass B	Glass C	Glass D
Density d (g/cm ³)	3.676	4.236	4.598
Average molecular weight $\bar{M}$	91.59	122.30	153.02
Pb^{2+} ion concentration N_i (10^{23} ions/cm ³)	4.83	8.34	10.85
Interionic distance of Pb^{2+} ions R_i (Å)	1.30	1.06	0.97
Polaron radius R_p (Å)	0.52	0.56	0.64
Field strength F_i (10^{16} cm ⁻²)	7.31	6.35	4.86

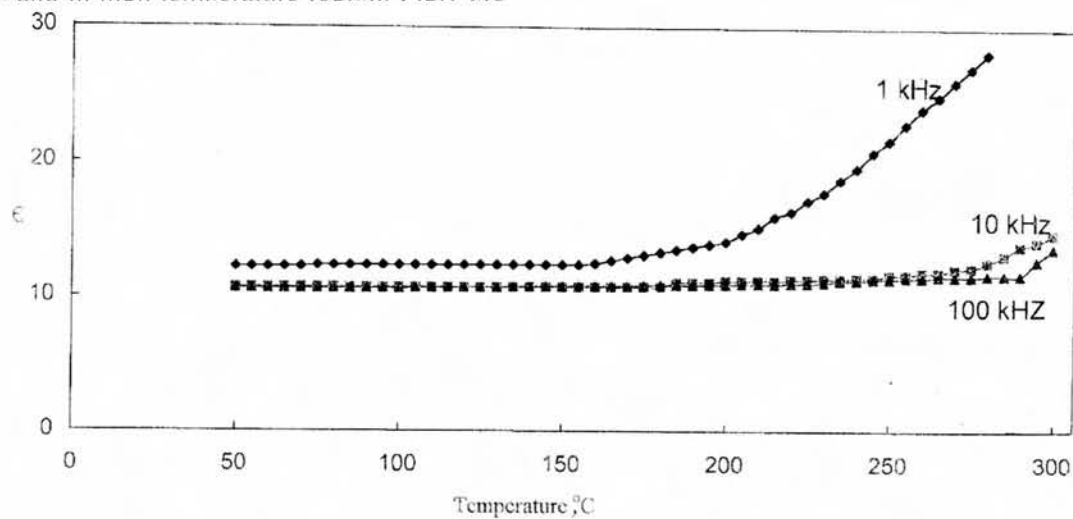


Fig. 1 — Variation of dielectric constant (ϵ) with temperature for glass C at different frequencies

in Fig. 4 for glass B. From these plots the activation energy for ac conduction in the high temperature region over which a near-linear dependence of $\log \sigma$ with $1/T$ could be observed, is calculated and presented in Table 3. This activation energy is found to be highest for pure $\text{LiF-B}_2\text{O}_3$ glasses and decreased with increasing PbO concentration.

The dielectric break down strength for pure glass (glass A) is determined to be 22.5 kV/cm; the value of breakdown strength decreases from glass A to D (Table 3).

The temperature variation of dc conductivity for all the glasses was studied under a dc field 10 V/cm. Fig. 5 shows the variation of dc conductivity with $1/T$ for $\text{LiF-PbO-B}_2\text{O}_3$ glasses. The conductivity is found to be lowest for glass A and highest for glass D. The activation energy for dc conduction too found to be lowest for glass D and highest for glass A (Table 3).

4 Discussion

The properties of glasses depend not only upon its composition, but also to a considerable degree upon its

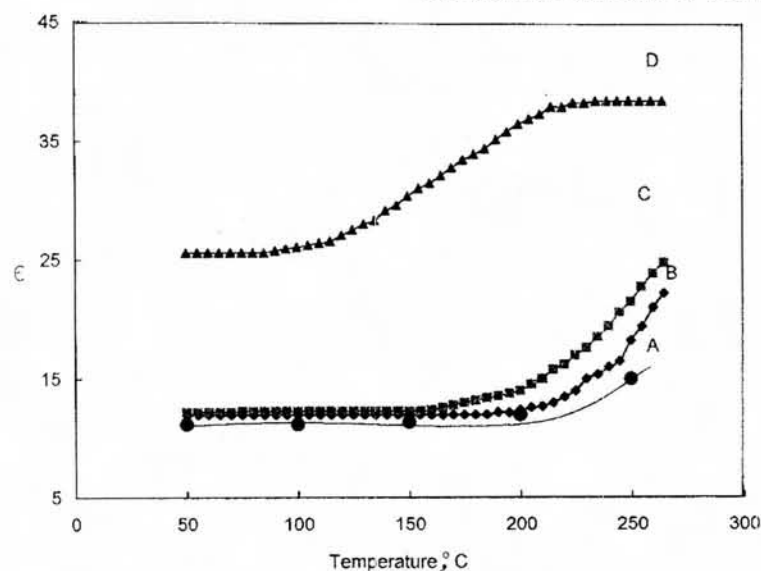


Fig. 2 — Comparison plot of ϵ versus temperature at 1 kHz for $\text{LiF-PbO-B}_2\text{O}_3$ glasses

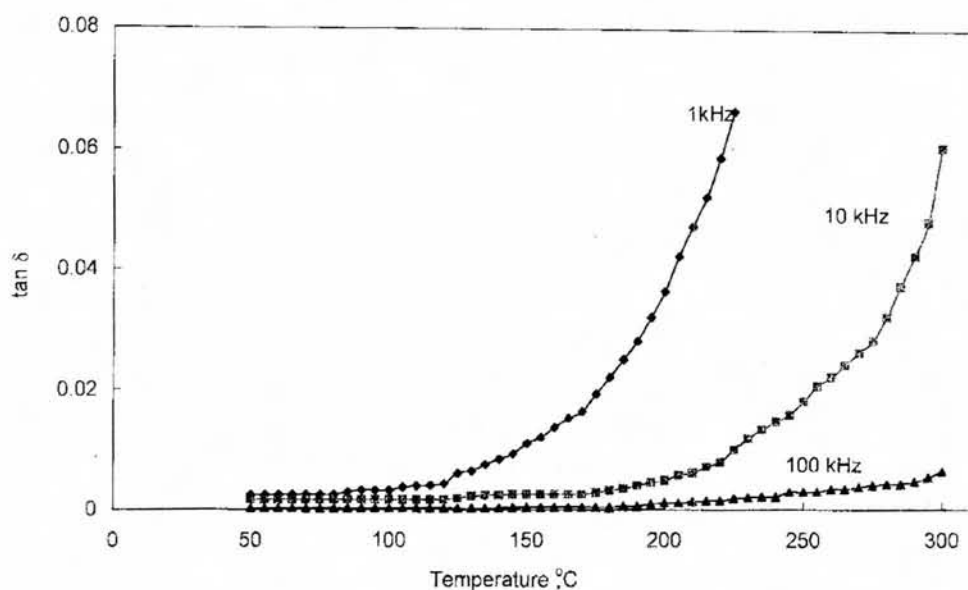


Fig. 3 — Variation of dielectric loss ($\tan \delta$) with temperature for glass B

structure. Deep and important changes in the properties of glass can occur as a result of structural transformation¹⁰. LiF-PbO-B₂O₃ glasses have complex composition and an admixture network formers and modifiers. B₂O₃ is a well-known network former with BO₃ structural units. The presence of BO₄ units is also evident in these glasses. This is confirmed from the study of the IR spectra of these glasses. The infrared spectra of LiF-PbO-B₂O₃ glasses, recorded at room temperature in 200-4000 cm⁻¹ range by an infrared spectro-photometer

(Perkin-Elmer Model-283B) using KBr technique is shown in Fig. 6. In the absence of PbO three groups of bands are observed: (i) in the region 1200-1600 cm⁻¹, due to stretching relaxation of the B-O bond of the trigonal BO₃ units, (ii) in the region 800-1200 cm⁻¹, due to B-O bond stretching of the tetrahedral BO₄ units and (iii) a band at 710 cm⁻¹ due to the bending of B-O-B linkages in the borate network. When PbO is introduced into the glass network, an additional weak but definite band around 550 cm⁻¹ is also observed. This is indicative

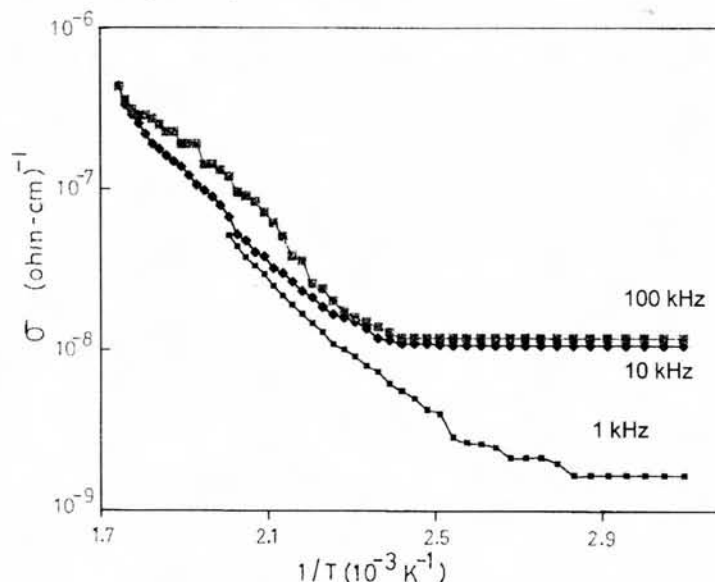


Fig. 4 — Variation of ac conductivity (σ_{ac}) with $1/T$ at different frequencies for glass B

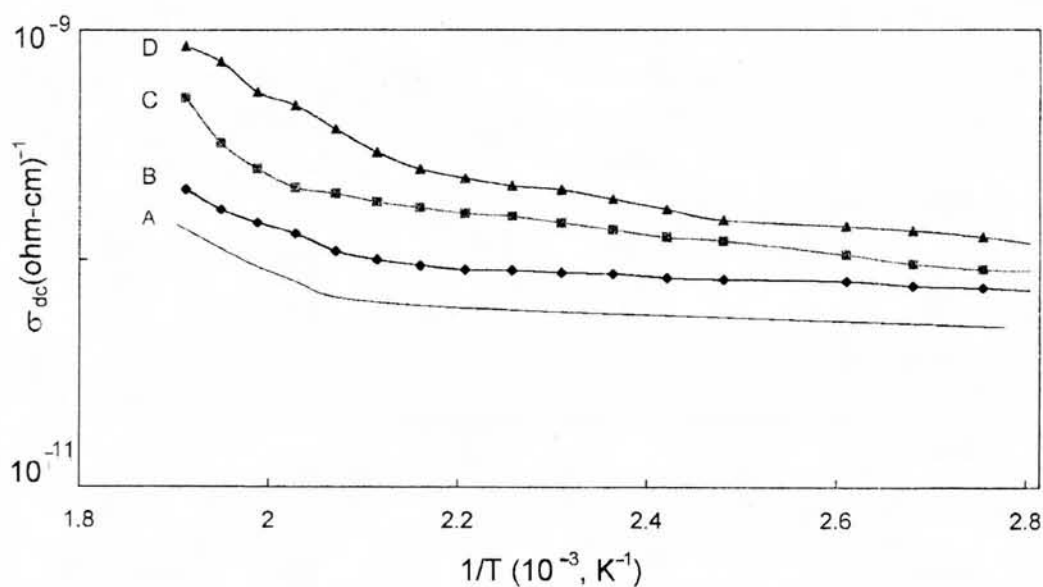


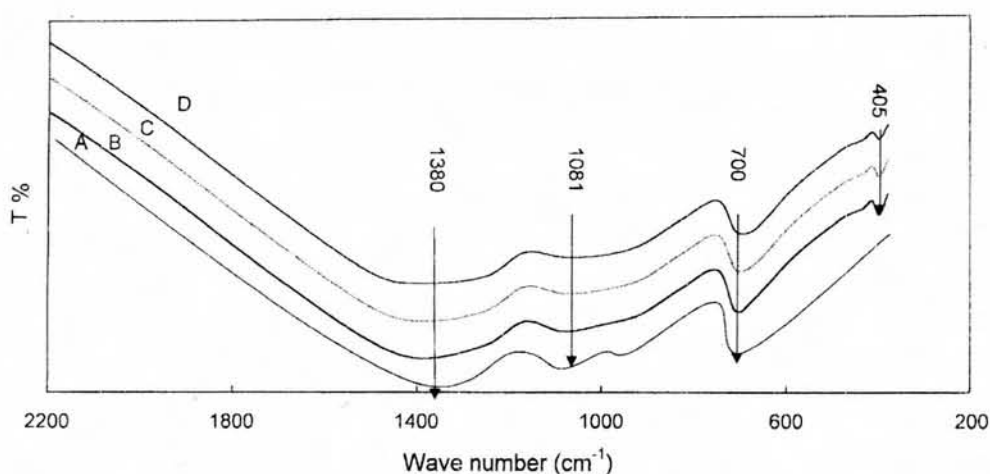
Fig. 5 — Variation of $\log \sigma_{dc}$ with $1/T$ for all glasses

Table 2 — Summary of data on dielectric constant and loss

Glass	ϵ at 1 kHz			100 kHz			$\tan \delta$ at 10 kHz ($\times 10^{-3}$)		
	R.T	100 °C	250 °C	R.T	100 °C	250 °C	R.T	100 °C	250 °C
Glass A	11.20	11.30	15.30	11.10	11.15	11.20	1.1	1.3	3.0
Glass B	12.00	12.10	18.20	11.52	11.50	12.10	1.6	1.7	5.8
Glass C	12.20	12.30	21.50	11.50	11.50	12.20	4.4	4.6	6.3
Glass D	25.30	26.10	38.20	22.50	22.70	25.10	4.6	4.7	7.0

Table 3 — Data on ac conductivity of LiF-PbO-B₂O₃ glasses

Glass	$N(E_F)$ in (10^{20} , $\text{eV}^{-1}/\text{cm}^3$)				A.E (eV) from		Breakdown strength (kV/cm)	A.E (eV) from
	Austin & Mott	Butcher & Hyd	Polak	S_{exp}	σ_{ac}	$\Delta\epsilon$		
A	—	—	—	—	0.38	0.37	22.50	0.45
B	3.60	1.50	3.65	0.91	0.34	0.33	21.62	0.49
C	5.65	2.36	5.74	0.87	0.32	0.31	18.54	0.48
D	6.48	2.70	6.58	0.85	0.31	0.30	15.10	0.46

Fig. 6 — Infrared transmission spectra of LiF-PbO-B₂O₃ glasses

of the presence of PbO₄-units in the LiF-PbO-B₂O₃ glass network¹¹⁻¹³. With the increase of PbO into the network, the intensity of IInd group of bands slowly diminished and shifted towards slightly higher frequencies. Intensity of the band due to PbO₄ units also reduced with increasing PbO concentration (Fig. 6) and peak position of this band is also shifted towards slightly lower frequencies.

Further, lithium can also occupy both tetrahedral sites (substitutional or network formers) and octahedral sites (interstitial or network modifiers) in these glasses. The modifier, PbO enters the glass network by breaking up tetrahedral bonds (the oxygen of PbO break the local symmetry while Pb²⁺ ion occupy interstitial position) of BO₄ units and introduces coordinated defects known as dangling bonds in these glasses. This is evidenced from IR spectra as discussed above.

The dielectric constant of a material is due to electronic, ionic, dipolar and space charge polarizations. Among these, space charge contribution depends on the purity and perfection of the glasses. Recollecting data, the slight increase in ϵ and $\tan \delta$ observed at room temperature for these glasses may be ascribed to the defects produced in the glass lattice, which contribute to the space charge polarization¹⁴.

Variation of ϵ and $\tan \delta$ with temperature is highest for glass D, in which the modifier PbO is present in larger quantities. This indicates an increase in the lattice distortion in LiF-B₂O₃ glasses with increase in PbO concentration, thus resulting in the enhancement of the space charge polarization, which ultimately causes larger increase of ϵ and $\tan \delta$ values as observed in the present measurements.

It is interesting to find that the nature of variation of ϵ and $\tan \delta$ with temperature is similar. The variation of ϵ with temperature can be connected to frequency through modified Debye equation as reported earlier⁵. A plot of $\log(\Delta\epsilon)$ against $1/T$ (where $\Delta\epsilon$ is the difference between dielectric constant at any temperature T and that at room temperature) at different frequencies for (glass D) is shown in Fig. 7. The plots are straight lines having about the same slope for all the frequencies. The computed activation energy is 0.30 eV. This value is practically the same as the activation energy for ac conduction in the same temperature region in these glasses. This seems to suggest that the charge carriers responsible for change in ϵ and σ with temperature in this temperature range are the same. A similar analysis was also carried out on the

other glasses and found to yield similar results. Further, this activation energy too is found to decrease from glass A to D (Table 3).

In general the ac conductivity of the amorphous material, where charge carriers experience an approximately random potential energy on diffusing, is found to obey the equation¹⁵:

$$\sigma(\omega) = A\omega^s \quad \dots(2)$$

with $s < 1$ up to the frequency of 1 MHz. Though various models have been developed to interpret the ac conduction mechanism in the glasses, the authors have first attempted to explain ac conduction in the present LiF-PbO-B₂O₃ glasses on the basis of quantum mechanical tunneling (QMT) model.

According to QMT model, only those pairs of carriers separated by hopping distance R_i given by:

$$R_i = (1/2\alpha) \ln \left(\frac{V_{ph}}{\omega} \right) \quad \dots(3)$$

contribute significantly to conduction. With this, the equation for ac conductivity due to quantum mechanical tunneling is given by¹⁵:

$$\sigma(\omega) = \frac{\eta e^2 K T}{3} \left[N(E_F) \right]^2 \alpha^{-5} \omega \left[\ln \frac{V_{ph}}{\omega} \right]^4 \quad \dots(4)$$

where $N(E_F)$ is the density of the energy states near the Fermi level, α is the electronic wave function decay constant, v_{ph} is phonon frequency and η a constant and its value is given by: $\eta = \pi/3$ (Ref. 16), $= 3.66 \pi^2/6$ (Ref. 17), $= \pi^4/96$ (Ref. 18).

The frequency component s is defined as:

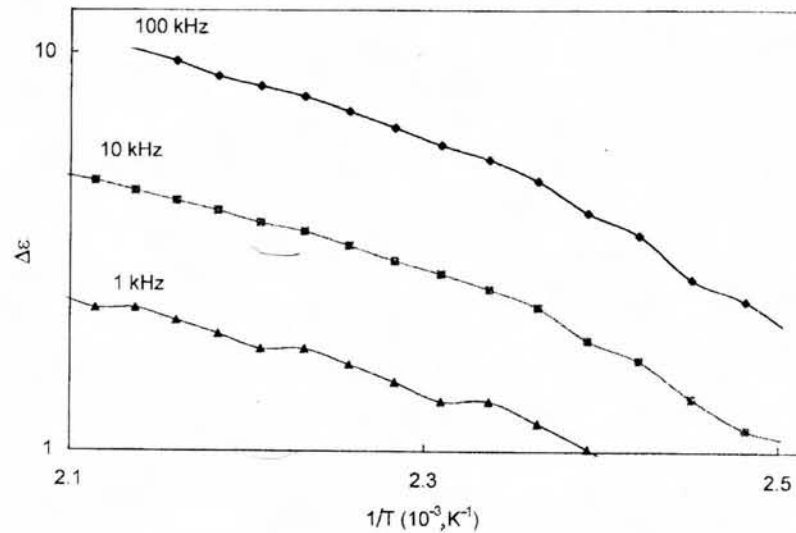


Fig. 7 — Variation of $\Delta\epsilon$ with $1/T$ for glass D at different frequencies

$$s = \frac{\partial (\ln \sigma)}{\partial (\ln \omega)} \quad \dots(5)$$

Substitution of Eq. (4) into Eq. (5) results

$$s = 1 - \frac{4}{\ln (1/\omega \tau_0)} \quad \dots(6)$$

Thus the quantum mechanical tunnelling of a carrier through the potential barrier between the sites separated by a distance R_i demands that s should be temperature independent.

The plot of $\log \sigma (\omega)$ versus $\log \omega$ for (glass C) at a temperature of 373 K is shown in Fig. 8. The graph obtained is a straight line. Plots drawn for the other glasses have also yielded straight lines. These results indicate that the exponent s is independent of frequency, hence it is not fully consistent with Eq. (6). Perhaps, the more accurate model for ac conduction in which s is less dependent on frequency (as observed) for ac conduction in LiF-PbO-B₂O₃ glasses is the correlated barrier hopping (CBH) model¹⁸, in which the height of the potential barrier is correlated with inter-site separation. According to this model:

$$s = 1 - \frac{6 K T}{E_0 - K T \ln (1/\omega \tau_0)} \quad \dots(7)$$

where E_0 is the optical band gap. According to Eq. (6), the value of s changes by 0.07 for a frequency change of 10^2 – 10^6 Hz, whereas this change is only 0.003 as per Eq. (7) for the same change in the frequency for a fixed optical band gap. Hence Eq. (7) can be assumed to give more accurate value of s than Eq. (6) for the present

glasses and the CBH model can be assumed to be more appropriate than the QMT model for ac conduction in these glasses. However, further analysis on ac conduction for these glasses is carried out on the basis of QMT model for the lack of accurate value of optical band gap E_0 at different temperatures. The experimental values of s obtained for these glasses are presented in Table. 3 along with the other important data on ac conduction.

Considering

$$\sigma \propto \epsilon^{-2\alpha} P_i \quad \dots(8)$$

as valid at least for low frequencies, the value of α is determined by plotting $\log \sigma$ against the hopping distance; the average value of the α obtained is $1.5 \text{ \AA}^{-1}$.

Among various mechanisms for conduction (like band conduction, conduction in extended states, conduction in localized states near the band edge and conduction in the localized states near the Fermi level) in the amorphous materials, the conduction in the localized states near the Fermi level occurs when ac. conductivity is nearly temperature independent and varies linearly with frequency. The conduction in present LiF-PbO-B₂O₃ glasses in the low temperature region (up to 400 K) can safely be attributed to take place by this mechanism. The value of $N(E_F)$, i.e. the density of energy states near Fermi level, for a frequency of 10^4 Hz and at 373 K, taking α equal to $1.5 \text{ \AA}^{-1}$ and $\nu_{ph} \sim 5 \times 10^{12}$ Hz is calculated using Eq. (4), with value of numerical constant η suggested by different investigators and presented in Table 3. The value of $N(E_F)$ is found to increase

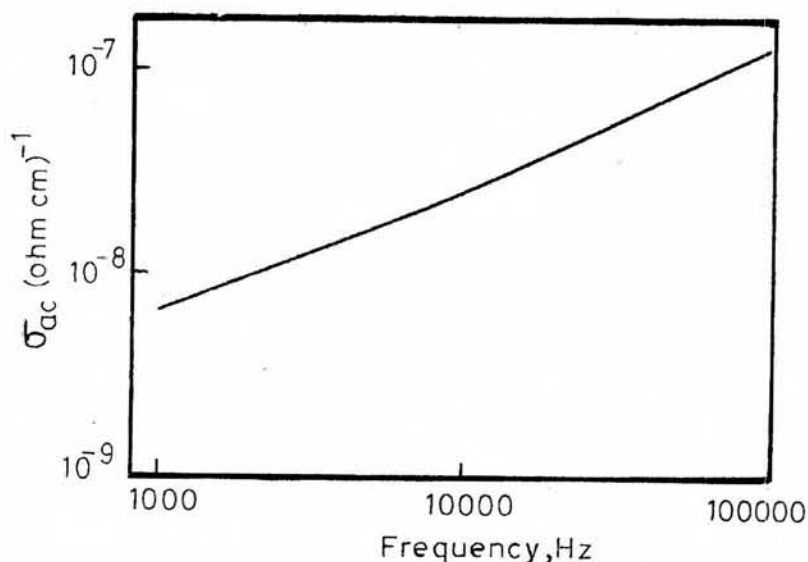


Fig. 8 — Variation of σ_{ac} with frequency for glass C

from glass B to D. Further, the range of $N(E_F)$ values obtained $\approx 10^{20}$ eV⁻¹/cm³. Such values of $N(E_F)$ suggest the localized states near the Fermi level¹⁸. The increase in the conductivity from glass B to D can be understood due to the increase in the density of these energy states.

When the dielectric is placed in the electric field, the heat of dielectric loss is liberated. If the applied field is an alternating field, the specific dielectric loss, i.e. the loss per unit volume of the dielectric is given by¹⁹:

$$\rho_1 = E^2 \omega \epsilon \epsilon_0 \tan \delta \text{ W/m}^3 \quad \dots(9)$$

This equation indicates that the higher the values of $\epsilon \tan \delta$ of the glass at a given frequency, the higher are the losses. In a dielectric across which the voltage is applied, heat is liberated, the temperature of the dielectric then rises and the loss increases still further. The dielectric breakdown strength is in fact inversely proportional to the specific dielectric loss represented by Eq. (9).

Recollecting the data on breakdown strengths of LiF-PbO-B₂O₃ glasses, as seen earlier, the rate of increase of $\epsilon \tan \delta$ with temperature is highest for glass D and minimum for pure glass (glass A). Though the breakdown strengths are actually determined at room temperature, the heat liberated during the breakdown raises the temperature of the glass and hence raises the $\epsilon \tan \delta$ value. Since the rate of increase of $\epsilon \tan \delta$ with temperature is highest for glass D, the breakdown strength is lower when compared with the other glasses. Thus the experiments on the dielectric breakdown strength of LiF-PbO-B₂O₃ glasses reveal that there is an increase in the concentration of defects (such as dangling bonds and point defects) in the glasses with increase in the concentration of Pb²⁺ ions. It may be noted here that though there is a possibility for Pb²⁺ to take part in the network forming positions and as a result there should have been an increase in the activation energies for ac conduction, which is not observed in this study.

The characteristic variations of dc conductivity with temperature for LiF-PbO-B₂O₃ glasses follows the typical semi-conducting behaviour. Several factors such as coordination number of modifier ions, character of the neighbouring oxygen ligands or the state of the polarisability of oxygen anions, whether the modifier metal ion behaves as a network former or a network modifier and valence state of modifier metal ion as governed by oxidation reduction nature of glass and the melting conditions etc., are considered to affect the conductivity of LiF-PbO-B₂O₃ glasses. As mentioned before, the modifier PbO enters the glass lattice network by break-

ing up the random network thus introducing coordinated defects in the network which are nothing but simple dangling (broken) bonds. These dangling bonds may be under coordinated or sometimes may be over coordinated or floating bonds. These bonding defects give rise to electron states within the band gap of the material and these will therefore be localized in that region.

The variation of dc current with temperature can be attributed to ionic drift current, due to electron hopping and current due to electrons in the conduction band. Current due to ionic drift is known to decrease the current with temperature, whereas the current due to electron hopping, and electrons in the conduction band results in an increase of current with temperature. Recollecting the nature of variation of dc current with temperature for these glasses, it appears that there is no contribution to the current due to ionic drift and hence the conduction in these glasses is mainly due to the last two mechanisms. As has been mentioned earlier, there is a localization of structural defects in the band gap of the material; these localized states cannot carry the current by themselves. The conduction involving the localized states can only take place by means of transition of electrons from the filled states to neighbouring empty states with phonon assistance. This type of transfer of electrons between the sites localized at different positions is known as the current due to hopping, which is mainly responsible for increase in the conduction with temperature in these glasses.

The dc conductivity of the present glasses for the hopping of the polarons in non-adiabatic regime is given by²⁰:

$$\sigma_{dc} = \frac{v_{ph} N_i e^2 R^2}{KT} C (1 - C) \exp(-2\alpha R_i) \exp(-W/KT) \quad \dots(10)$$

where N_i is the concentration of Pb²⁺ ions, v_{ph} the phonon frequency, C the mole fraction of the sites occupied by electron and α the electron wave function decay constant.

The term $\exp(-\alpha R_i)$ represents electron overlap integral. The activation energy W can be expressed as:

$$\begin{aligned} W &= W_H + (1/2) W_D & \text{for } T > \theta_D/2 \\ &= W_D & \text{for } T < \theta_D/4 \end{aligned} \quad \dots(11)$$

where W_H is the polaron hopping energy, W_D the disorder energy and θ_D the Debye temperature.

When $\exp(-\alpha R_i)$ approaches unity the hopping conduction is adiabatic and mainly controlled by activation energy. Then Eq. (10) reduces to:

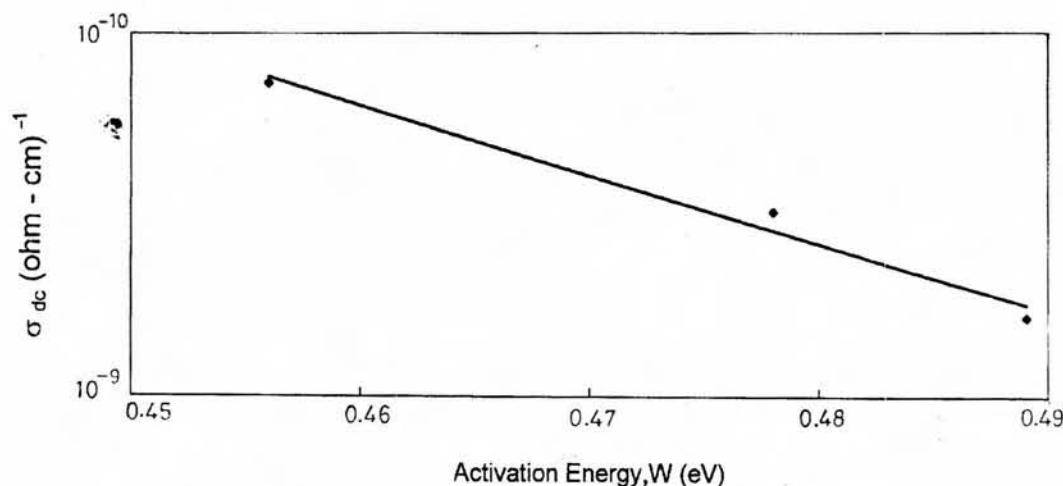


Fig. 9 — $\log \sigma_{dc}$ versus activation energy at 510 K for different compositions of PbO

$$\sigma_{dc} = \frac{v N_i e^2 R^2}{kT} C (1 - C) \exp(-W/kT) \dots (12)$$

Plot of $\log \sigma_{dc}$ versus $1/T$ for LiF-PbO-B₂O₃ glasses shows a linear nature (Fig. 5), indicating that σ_{dc} increases with increase in temperature T and also with increase in the concentration of Pb²⁺ ions. The activation energy W is independent of temperature and depends on Pb²⁺ ion concentration. The activation energy W values are found to decrease with increase in the concentration of Pb²⁺ ions (Table 3). The linear nature of the curves also indicates that the conduction is electronic.

To explore the nature of the hopping conduction in LiF-PbO-B₂O₃ glasses a graph between $\log \sigma_{dc}$ and W is plotted at fixed temperature (510 K) in Fig. 9; the graph obtained is a straight line. This indicates that Eq. (12) is valid. From the slope of this curve the value of $1/kT$ is obtained and the temperature T is estimated at which the graph is drawn. The value of T is found to be ~ 450 K which is closed to the actual temperature. This suggests that the hopping is adiabatic and the conduction is mainly controlled by the activation energy only as represented³ by Eq. (12).

5 Conclusions

From the results of various electrical measurements on LiF-PbO-B₂O₃ glasses, it is observed that with increasing the concentration of the modifier PbO in LiF-B₂O₃ glass network; there is an increase in the rates of increase of $\epsilon \tan \delta$, σ_{ac} and σ_{dc} with temperature and decrease in the values of dielectric breakdown strength and activation energy for ac/dc conduction. The carrier-barrier hopping model seems to be more appropriate for

ac conduction and dc conduction is due to adiabatic hopping.

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