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EVALUATION OF A.C. CONDUCTIVITY FOR LEAD SILICATE GLASS FROM DIELECTRIC MEASUREMENTS

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ABSTRACT In the present work, lead silicate glasses has been prepared using powder technology technique. Frequency dependence of dielectric constant, dielectric loss, dissipation losses (tan δ) and a.c conductivity was measured in the frequency range 40 Hz-10MHz and sintering temperature 650 ˚C for 15%, 20%, 25%, and 30 wt% PbO. It was observed that the increases in frequency lead to decreasing in dielectric constant and dielectric loss. Specific frequency introduces suddenly increases in dielectric properties.

Keyword: lead glasses, dielectric constant, loss, green, bulk density.

I. INTRODUCTION

The $PbO–SiO_2$ system, among various lead-oxide-containing glasses, has been found to form glass over a wide composition range and has got many applications in making electron multipliers, TV picture tubes, electrical feedthroughs, and glass-to-metal (GM) seals. Various physical properties like electrical and thermal conductivity, micro hardness, dielectric constant, etc. of the above glass system can be modified by changing the $PbO–SiO_2$ composition [1]. The frequency dependent dielectric and optical properties of binary semiconducting glasses in the system $V_2O_5–TeO_2–PbO$ have been measured as a function of lead content and the effects of composition on refractive index, dielectric constant and optical phonon frequency have been discussed by Memon *et al* (1993)[2].
Polarizing processes in lead monoxide were investigated by V. A. Izvozchikov et al, [3] and the crystallographic structure analysis of the PbO modifications was investigated. They found that some space symmetry groups referred to lead monoxide are characteristic of ferroelectrics semiconductors.
The dielectric constant and a.c. electrical conductivity of the glass systems $60B_2O_3$-(40-x) PbO-$xMC1_2$ and $50B_20_3$ - (50-X)PbO – $xMC1_2$ (M = Pb, Cd) of different compositions (x = 10, 12.5, 17.5, and 20 mole percentage) were studied by [4]. Electric conductivity and dielectric properties of some unconventional lead cuprate glasses have been studied by S. Hazra and A. Ghosh in the temperature range of 80–550 K and in the frequency range 102–106 Hz [5]**.** J C Garg and N C Parakh, measured the electrical conductivity of lead tin monoxide as a function of composition (*x*), temperature and electric field [6].
The object of this work is to evaluate the frequency dependence of lead

silicate glass when it used in any electric device applications

II. FUNDAMENTAL

Dielectric properties are dependent on glass composition, frequency, temperature, and glass microstructures. Polarization of glasses occurs if they are subjected to the action of externally applied electric field. Oxide glasses (e.g., the silicates) are particularly susceptible to such polarization because of the high polarizability of oxygen anions. The polarizability of the cations varies, being least for silica, intermediate for dibasic cations like calcium, and greatest for such monovalent cations as sodium. The total polarization can be indicated by the general equations

$$P = P_e + P_i + P_\circ \quad \cdot\cdot \qquad (1)$$

where P_e relates to displacement of electrons, P_i to that of ions, and P_o to orientation of ionic groups in the lattice substructure (dipole orientation) [7].

The majority of dielectric applications involve alternating fields in which the reversibility of polarization is time dependent (whether there is sufficient time available for electronic, ionic, and dipole displacements to return to their original positions after the field are removed. Dielectric losses in glasses can be qualitative related to the presence of ions and ionic groups in the frequency range where glasses are used as insulators. The complex quantity of dielectric constant content the real and imaginary component K` and K`` relate to the dielectric constant and dielectric absorption. When the glasses are used as insulating materials these absorption result in dielectric losses, usually in the form of heat.

Dielectric losses in glasses consists of conduction ions (movement of ions at low frequency), ionic relaxation losses (depend on mobile and immobile ions in the glass structure at less than 10^6 Hz), and deformation losses (deformation of the glass substrates) [8].

III. EXPERIMENTAL

Lead glasses samples were synthesized using a three-stage heating cycle involving melting, quenching and annealing. The starting material was prepared by mixing and grinding of pre-determined amounts of feldspar, alumina, and lead oxide. The batches were melted to 950 ˚C for a period of time 2h followed by normal quenching of molten liquids onto water kept at room temperature. Regrinding process in a planetary ball mill at room temperature for homogenization has been done. One gram from each batches were weighted and pressed to a disk samples have a diameter 1.5 cm using uniaxial press holder. Prepared samples have sintered at 650°C for 2h. (Precision Impedance Analyzer type Agilen technologies 42942 A made in U.S.A.) was used for dielectric measurements. The dielectric studies of glass samples were carried out by using a dielectric cell. Prepared disk shaped samples were used to find out the dielectric constant. The capacitance and dielectric loss in the frequency range 40 Hz–6MHz were found out. Dielectric constant or relative permittivity were calculated by using the formula

$$\varepsilon_r = \frac{C \times d}{A \times \varepsilon_\circ} \quad \ldots\ldots\ldots\ldots \ (2)$$

where *d* is the thickness of the sample in centimeter, *C* the capacitance, *A* the area of cross section of the sample in cm^2 and ε_o the permittivity of free space (= 8.854×10^{-12} F/m). The relative permittivity ε_r of the material which is a dimensionless quantity. Evaluation of a.c. conductivity for these samples was made using equation (3) from the available measurements of dielectric constant and tan δ at a given frequency.

$$\sigma_{a.c.} = \omega \tan\delta\, \varepsilon_{\circ} \varepsilon_{r} \quad ....(4)$$

where σ is the conductivity, $w = 2\pi f$ is the angular frequency, ε_r is the relative permittivity of the material and ε_o is the permittivity of free space.
With attention that dissipation factor (tan δ) can be introducing using the formula

$$\tan\delta = \frac{\varepsilon_i}{\varepsilon_r} \quad . \qquad (5)$$

IV. RESULTS AND DISCUSSION

The variation of dielectric constant as a function of frequency was shown in figure 1. As seen from the graph that the values of ε′ for all the samples are less than 20. These results were agreeable with most glass silicate samples that reported in [10]. It was notice that the samples that have the higher PbO content have the higher dielectric constant in contrast with that has the lower PbO content. Also, all samples retain the same behaviors at all the frequency ranges. The dielectric constant have its higher value at frequency equal to 200 Hz, this behavior was very conceivable for insulator glass samples because all type of polarizations (electronic, ionic, dipolar, and space charge) can follow the change of frequency at this region, consequently contribute to increases the dielectric constant values. The relatively higher ε' values of these amorphous materials at lower frequencies were explained due to the presence of grain or interphase boundaries. Grain boundaries contain defects such as dangling bonds, vacancies, vacancy clusters, etc. The space charges can move under the application of an external field and when they are trapped by the defects, lots of dipole moments (space charge polarization) are formed. At low frequencies of applied field the dipole moment can easily follow the change of electric field resulting in a high value of polarization and there by a high value of ε' [10]. Sharply decreasing in dielectric constant in the low frequency region between 200 and 650 Hz has been observed. Increase of frequency shows stable state between 1 kHz to 100 kHz for each measured samples with approximately lower dielectric constant ~13 for 15wt% PbO and higher dielectric constant ~16 for 30wt% PbO content. This can be attributed to the limitation of polarizations shared with consequently increasing of applied frequency. The significant phenomenon shown in all prepared samples is the appearance of two pronounceable peaks in two specific frequencies 200 kHz and 6 MHz. This issued can be regarding to the formation of resonance frequency between the presences phases in these samples and the applied field. The increasing in frequency after these regions revealed clearly decreasing in dielectric constant regarded to incapable of material to follow the varying of frequency.

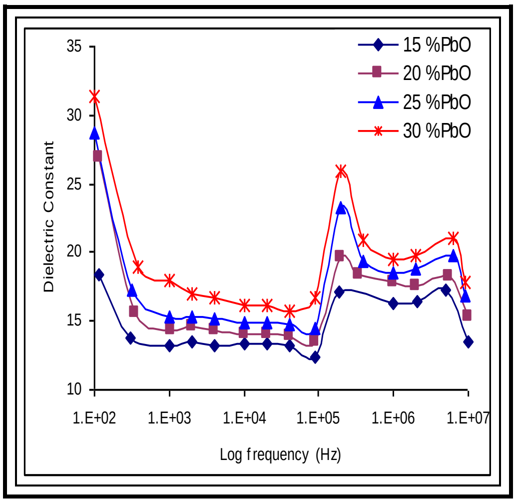

Figure 1: Dielectric constant ($\mathcal{E}$r) as a function of frequency at room temperature for different lead glasses.

Figure 2, shows the dielectric losses ε_i versus the applied field for prepared glass samples. The low dielectric loss value was shown in all samples. This result was obtained in most silicate glasses dependent from the strong bonding between atoms [11]. It can see clearly that the dielectric loss decreasing slightly at lower frequencies at distinguished regions between 200Hz to 1 kHz. Increasing of applied frequency leads to produce pronounced peaks at 200 kHz and suddenly high increasing in dielectric losses at 10 MHz. This behavior can explain in which that, the mobile ions content were slightly low and the prepared samples has small deformation. Thus, as the frequency increases, the contribution of ionic losses and deformation losses were inhibiting by the strong bonds between the glass components which blocking the ions movements then leads to decreases in dielectric losses [11]. The suddenly increase in 200 kHz can be attributed to the resonance state of constituents ions. The highly increase in dielectric losses at 10MHz can be related to contribution of all types of losses (electronic, ionic, deformation, ionic groups, and vibration losses). This can be also explained by Maxwell- Wagner-Sillars theory, which is to account for the dielectric loss due to the interfacial polarization of heterogeneous materials having the volume fraction of conductive phases lower than the percolation threshold [12].

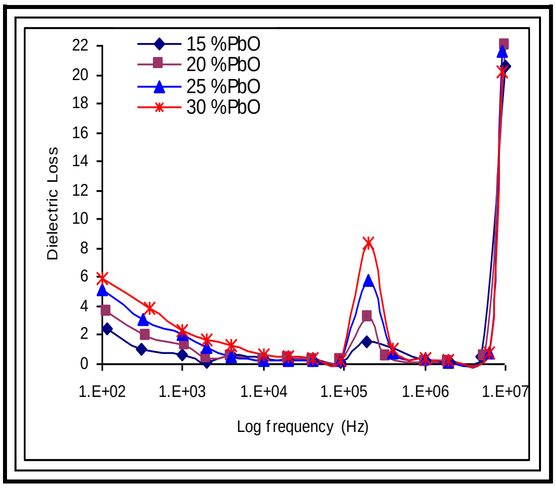

Figure 2: Dielectric losses ($\mathcal{E}_i$) as a function of frequency at room temperature for different glasses.

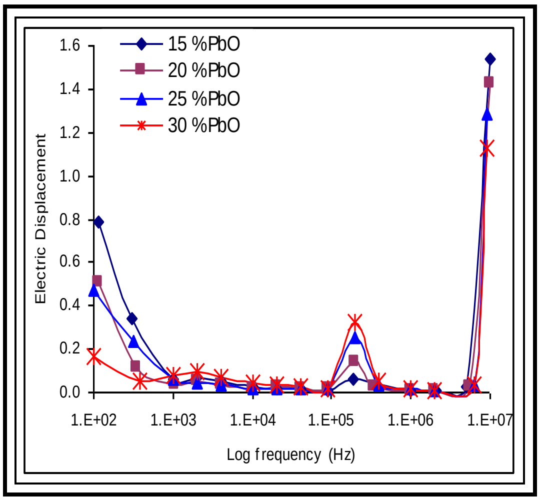

Figure 3: the electric displacement (tan δ) as a function of frequency for different lead glasses.

The dependence of electric displacement on the applied field was shown in figure 3. This property shows slightly value for all prepared glass samples at all the range. The maximum value it reaches for these samples at low frequency are between ~0.2 and

0.8 dependence on glass composition. At higher frequency (10 MHz) the dielectric retained its topping value ~ 1.12 to 1.5. This result has explained as the same as the dielectric losses because of the relation between tan δ and dielectric losses eq.5.

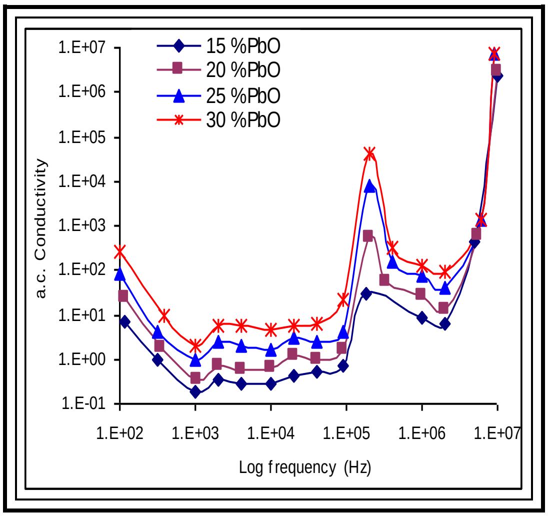

Figure 4: Variation of a.c. conductivity as a function of frequency for different lead glasses contents.

Frequency dependence of conductivity for measured prepared glass samples at room temperature was shown in figure 4. Higher PbO wt% content has the higher a.c. conductivity in contrast with the lower lead glass samples at all the frequency range. This means that the frequency dependence of samples conductivity was generated by the addition of lead oxide. At low frequencies, the decreasing in a.c. conductivity was revealed. This can be regarded to the fact that, at low frequency, ions make random jumps over a barrier separating two sites then leads to decreasing conductivity. A frequency independent conductivity is reported between 1 kHz and 100 kHz which is attributed to the resistive conduction through the bulk glass. The presence and rising of applied field leads to ions migrate preferentially in the direction of an applied field, which effectively lowers the energy barriers in the forward direction .

PbO in general is a glass modifier and inters the glass network by breaking up the Si-O, Si-O-Al bonds (normally the oxygen's of PbO break the local symmetry while Pb^{+2} ions occupy interstitial positions) and introduces coordinate defects known as dangling bonds along with non-bridging oxygen ions. In this case Pb^{+2} are octahedral coordinated. However, PbO may also participate in the glass network with PbO_4 structural units when lead ion is linked to four oxygen in a covalence bond configuration [13].

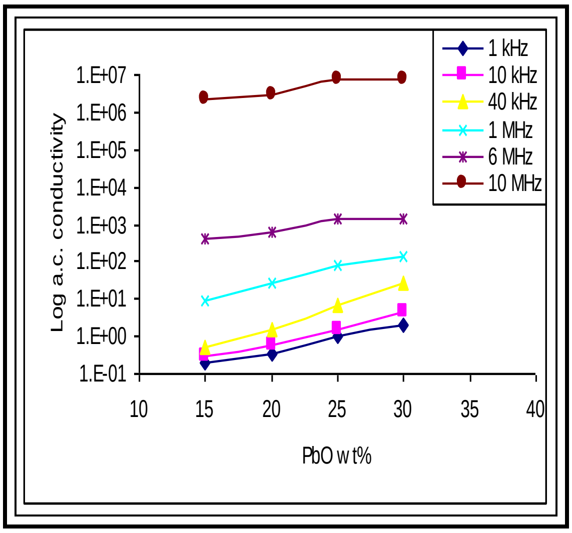

Figure 5: Log a.c. conductivity of different lead glass content at some selected frequencies.

The dependence of conductivity and lead oxide content at specific frequencies was shown in figure 5. It can see that the conductivity were increases with increasing of both lead oxide content and applied field. This result was derived from the presence of Pb ions and the energy absorbed from the field.

V. CONCLUSION

The dielectric constant of prepared samples increases with increasing of lead oxide content and decreasing with applied field with appearance of significant peaks at specific frequencies. Small dielectric losses for all samples and obtained its high losses at 10 MHz. All samples reveled low dissipation factor and the a.c. conductivity increases with increasing of PbO content and applied field.

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