

ELECTRIC CONDUCTIVITY AND DIELECTRIC PROPERTIES OF AgPO_3 GLASS

FATHY SALMAN

Physics departement, Faculty of Science at Banha,
Zagazig University, Egypt.

ABSTRACT

The complex impedance of AgPO_3 glass is measured in the frequency range 50-500KHz from room temperature up to 180°C. The conductivity of AgPO_3 is determined by the impedance method. The frequency dependence of the dielectric constant and dielectric loss tangent are calculated from the impedance data. The temperature dependence of the dielectric constant and dielectric loss tangent are studied.

INTRODUCTION

Recently, glass formation has been reported to be very useful for achieving larger conductivities rather than in crystalline solids. Besides, a fast ion conducting glasses have a wide range of properties : isotropic properties, formation of bulk materials with no grain boundaries, thin - film formation and feasibility of spaoing [1].

Silver ion-containing glasses, which possess high temperature dependent ionic conductivity are of great interest because of their applications as thin films and solid electrolyte[2,3,4]. Silver metaphosphate glass compositions have a well-known structure. X-ray analysis of vitreous sodium metaphosphates [5-6] showed that the structure consists of PO_4 group ideally contains two non-bridging oxygen ions.

In this paper the electric behaviour of AgPO_3 glass is studied by using the complex impedance method over wide ranges of frequencies (50Hz - 500KHz) and temperatures (30°C-180°C).

EXPERIMENTAL

AgPO_3 glass was prepared by melting together stoichiometric amounts of AgPO_3 and $\text{NH}_4\text{H}_2\text{PO}_3$ at about 700°C for three hours until gas evolution ceased. The obtained melt was quenched rapidly at room temperature. The glass samples are shaped in a rectangular form and then polished. Each obtained sample was optically transparent and colourless. Connection was made by silver paste electrodes on both surfaces of the sample.

For the measurements, the sample was placed in a furnace whose temperature was controlled and stabilized within 1 degree. During the measurements the temperature was raised at a rate of $0.5^\circ\text{C}/\text{min}$. The impedance parameters were measured by means of an BM 507 impedancemeter in the frequency range (50Hz-500KHz).

The glass transition temperature T_g of AgPO_3 glass was determined by DTA technique by the Paulik-Paulik method at a rate of $10^\circ\text{C}/\text{min}$.

To ensure that the sample is in a glassy state, X-ray spectrum is recorded.

RESULTS AND DISCUSSIONS

X-ray diffraction pattern obtained for the investigated sample (Fig. 1), shows that it is in the glassy state. DTA thermograms obtained for the studied sample (Fig. 2) show that glass transition temperature is 141.75°C .

The complex impedance Z within the frequency range 50Hz-500KHz was measured in the temperature range (30°C - 180°C) and then was analysed into the real Z' and imaginary Z'' parts on the complex plane[7]. Fig. (3) shows the Z' (Z'') dependence in the complex plane at various temperatures. In this temperature range the impedance diagrams were found to form semicircles in the high frequency limit and only a part of semicircles of bigger diameter in the low frequency limit. Thus the equivalent circuit of the sample consists of a parallel combination of a bulk resistance R_b and a capacity C_b connected in series with a capacity C_e which originates from the electrode

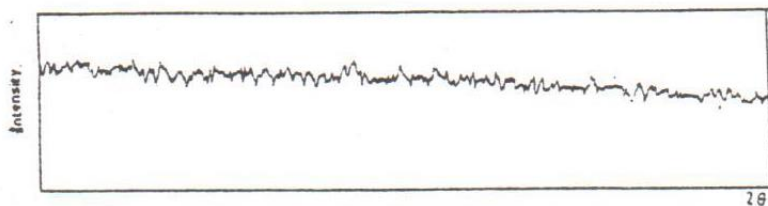


Fig. 1. X - ray diffraction pattern for AgPO_3 sample.

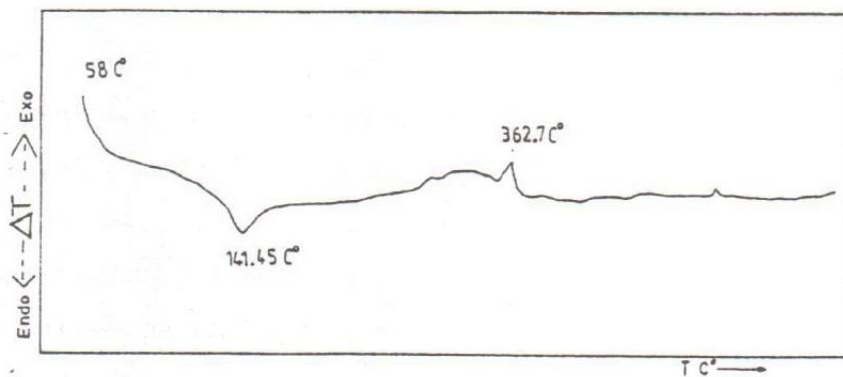


Fig. 2. DTA thermograms for AgPO_3 sample.

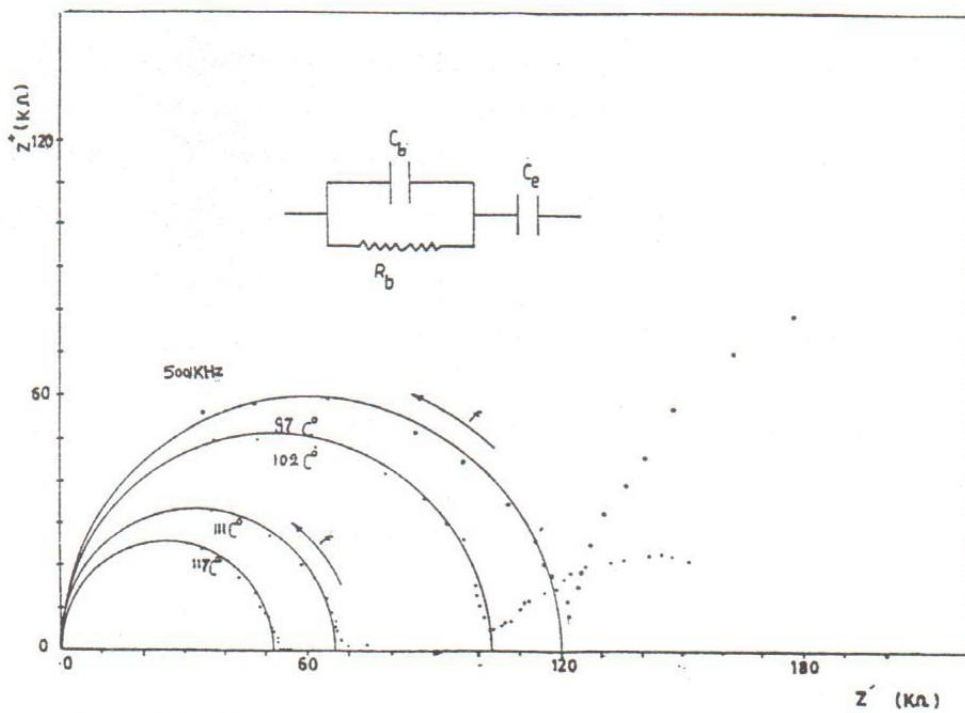


Fig. 3. Z' (Z'') dependence for AgPO_3 at various temperatures.

layer (Fig. 3). At temperatures higher than 120°C the high frequency semicircle disappears (Fig. 4).

The bulk resistance of the glass sample was estimated as the diameter of the semicircle shown in Fig. 3. The conductivity was determined from the bulk resistance of the sample at different temperatures. Above 127°C the resistance of the glass sample is found by the extrapolation of the impedance diagram in the direction of increasing frequencies up to the intercept with the real axis (Fig. 4). Fig. 5 shows the temperature dependence of the obtained bulk conductivity. The conductivity is found to obey Arrhenius law. The value of the activation energy is found to be 0.24 eV. A rapid increase in the conductivity above 127°C . It is related to the glass transition temperature. The observed disagreement between values of T_g obtained from DTA thermograms (141.75°C) in Fig. 2 and that obtained from conductivity measurement 127°C (Fig.5) may be due to the slower rate of increasing temperature during conductivity measurements than that of the DTA thermograms.

The conductivity results reported above are due to the ionic type of conduction and to structural features of silver phosphide. According to the previous study (8) on AgPO_3 structure, it is formed of an assembly of elementary units (PO_4 tetrahedral) in which at least one atom of oxygen is shared (Bridging oxygen). Some oxygen atoms that are non-bridging are negatively charged keeping in their vicinity the silver cation which are free to move through the rigid glassy matrix.

FREQUENCY DEPENDENCE OF THE DIELECTRIC CONSTANT ϵ AND DIELECTRIC LOSS TANGENT $\tan \delta$

The frequency dependence of the dielectric constant ϵ was studied at different temperatures for AgPO_3 glass below T_g . The results are illustrated in Fig. 6. These curves show that the highest value of ϵ starts to decrease by a slow rate then by a high rate as the frequency increases. The low frequency region is closely connected to dc conduction, with substantial charge movement in the material. The high frequency region is due to more "dipole-like" localized hopping of charges[9].

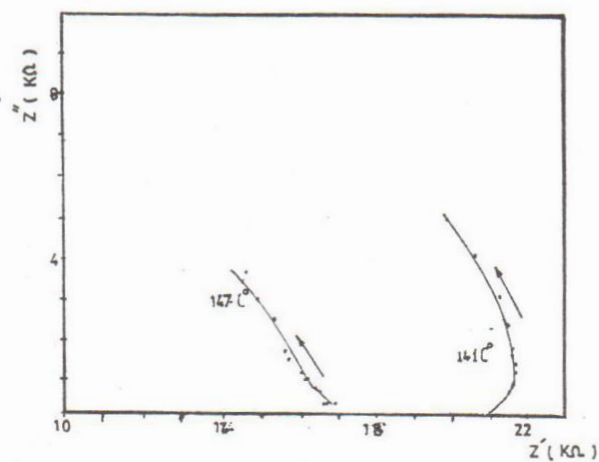


Fig. 4. $Z''(Z')$ dependence for AgPO_3 at various temperatures.

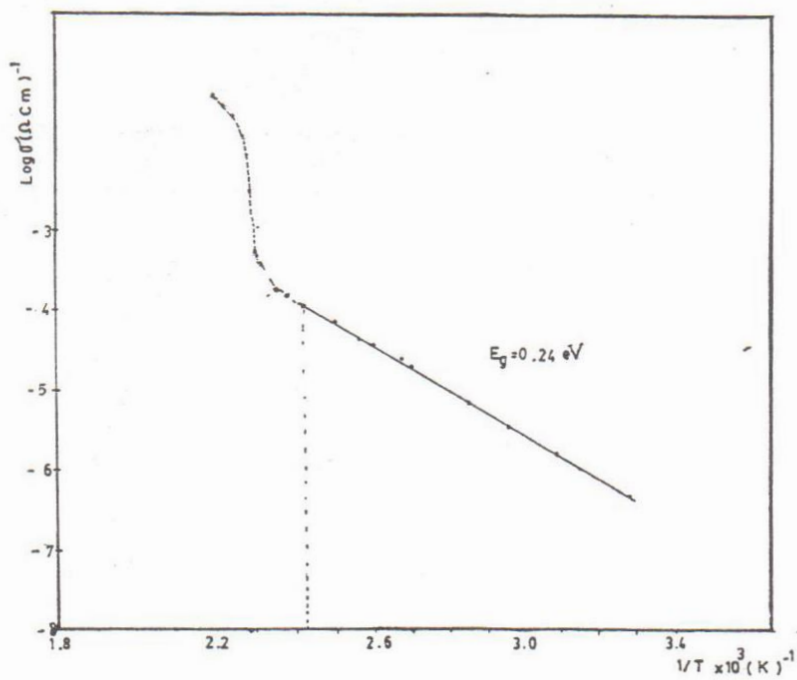


Fig. 5. Temperature dependence of the electric conductivity of AgPO_3 glass.

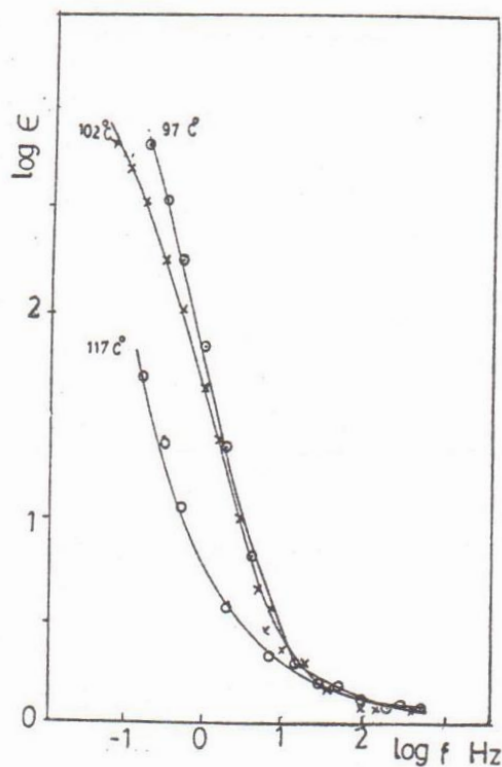


Fig. 6. Frequency dependence of dielectric constant ϵ at various temperatures for AgPO_3 .

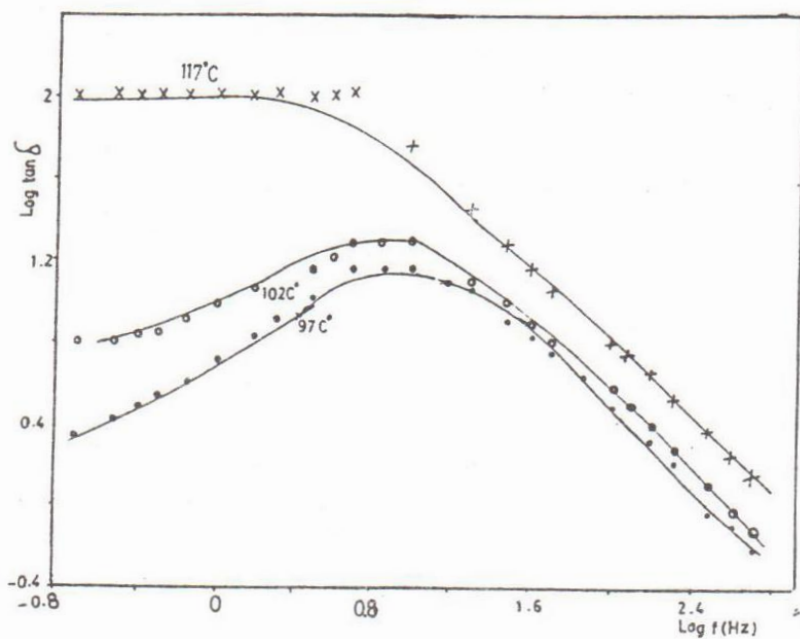


Fig. 7. Frequency dependence of dielectric loss tangent δ .

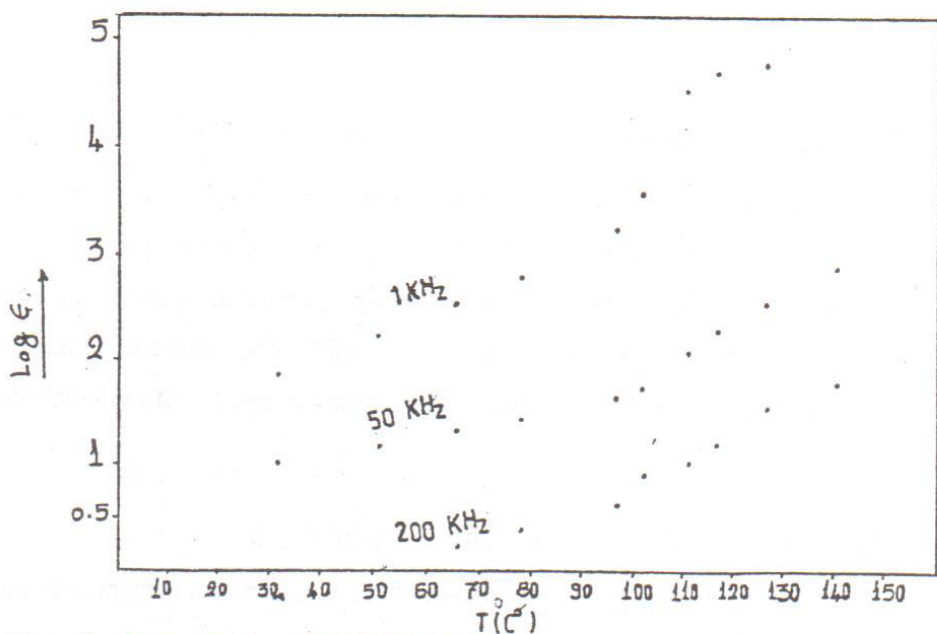


Fig. 8. Temperature dependence of dielectric constant ϵ at various frequencies for AgPO_3 .

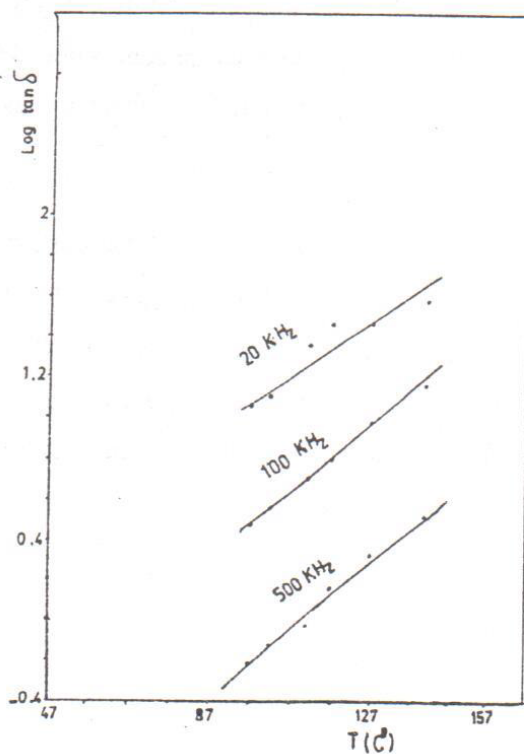


Fig. 9. Temperature dependence of dielectric loss tangent δ at various frequencies for AgPO_3 .

The frequency dependence of the dielectric loss $\tan \delta$ for AgPO_3 is shown in Fig. 7. It is clear that the dielectric loss tangent undergoes dielectric loss peak. The peak amplitude increases steadily with decreasing temperature while the peak frequency decreases. This is the behaviour of many dipolar materials at moderate temperature[9]. In principle, one would therefore expect these dipoles to be almost free to move in order to behave like ideal Debye systems except for the interactions between the neighbouring dipoles.

TEMPERATURE DEPENDENCE OF ϵ AND $\tan \delta$:

Fig. 8 shows the temperature dependence of the dielectric loss tangent $\tan \delta$ at different frequencies. It can be observed that as the temperature increases the dielectric loss tangent increases. This is explained by Stevels[10] who divided the dielectric losses into three parts : conduction losses, dipole relaxation losses and deformational losses. The conduction loss increases with temperature since $(1/\sigma)$ decreases. At higher temperatures conduction loss, dipole loss and vibration loss all contribute to the dielectric loss.

Fig.9 shows the temperature dependence of the dielectric constant ϵ at different frequencies. It can be observed that the dielectric constant increases as temperature increases. In the low temperature range the increase of ϵ is with low rate. While at high temperature range the rate of increasing becomes more pronounced. This behaviour can be explained as follows : at high temperature ranges, multicomponents contribute to the polarization of the sample while at low temperatures this contribution was only due to the space charge.

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