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Article

Dielectric Properties of Ternary $(ZnO)_{30}(MgO)_x(P_2O_5)_{70-x}$ (x = 5, 8, 13) Glasses

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ABSTRACT

$(ZnO)_{30}(MgO)_x(P_2O_5)_{70-x}$ glasses of the composition x = 5, 8 and 13 mol % have been prepared by melt quenching technique. The dielectric permittivity (ε′) and loss factor (ε″) were measured in the frequency range from 0.01 Hz to 1 MHz and in the temperature range 303 to 573 K. From the results there are evidence of dipolar relaxation occurring between 10^3 – 10^6 Hz while at low frequency the spectrum is dominated by dc conduction which manifested by the 1/ω slope of loss factor plot. Value of the relaxing frequency (ω_p) plotted against 1/T shows one electrical transportation mechanism. The empirical data was sufficiently fitted by using Harviliak-Negami equation.

| Dielectric relaxation | Complex permittivity | Phosphate glasses |

1. Introduction

The study on dielectric behavior of phosphate glasses over a wide range of frequency and temperature is expected to reveal comprehensive idea about the nature and origin of the loss occurring in these materials as well as conduction mechanism [1, 2]. The experimental data in present glass system were fitted by the Havriliak-Negami (HN) dielectric relaxation function, superimposed by a conductivity term as shown below:

$$\varepsilon^* = \varepsilon' - i\varepsilon''$$

$$= \sum_{k=1}^{m}\left\{\frac{\Delta\varepsilon_k}{\left[1+(i\omega\tau_k)^{\alpha_k}\right]^{\beta_k}} + \varepsilon_{\infty_k}\right\} - i\left(\frac{\sigma_\circ}{\varepsilon_\circ\omega}\right)^n \qquad (1)$$

where $\sigma_\circ$ is dc conductivity, $\Delta\varepsilon$ is dielectric strength, τ is relaxation time, α is width parameter, β is asymmetry parameter, ε_∞ is infinite permittivity and n is exponential factor.

2. Materials and Methods

The starting chemical powders, phosphorus (V) oxide (99.99%), magnesium oxide (98%) and zinc oxide (99.7%) were weighed and mixed together in an alumina crucible in appropriate quantities to constitute a 15-20 g batch. The crucible was covered and heated in an electric furnace for about 1 hour at a temperature of 400°C. The crucible was then transferred to another electric furnace and kept at 1200°C for 2 hours. The melt was stirred occasionally every 20 minutes to ensure homogeneity and proper mixing. Each melt was cast by pouring it into a preheated stainless steel cylindrical two-split mould to form glass rods about 20 mm long and 10 mm in diameter.

After casting each glass was immediately transferred to an annealing furnace, and held at temperature of 400°C for 1 hour and slowly cooled to room temperature. The samples were prepared by cutting the glasses into disc-form and both faces of the samples were polished using silica carbide in order to get a smooth and parallel sample. Pure aluminum (99.999% Al) was evaporated onto both sides of the samples as electrode by using Edwards Auto 306 Vacuum Coating. The samples were then stored in a desiccators until the dielectric relaxation measurements were performed by using Novocontrol Alpha High-Resolution Dielectric Analyzer.

3. Results and Discussion

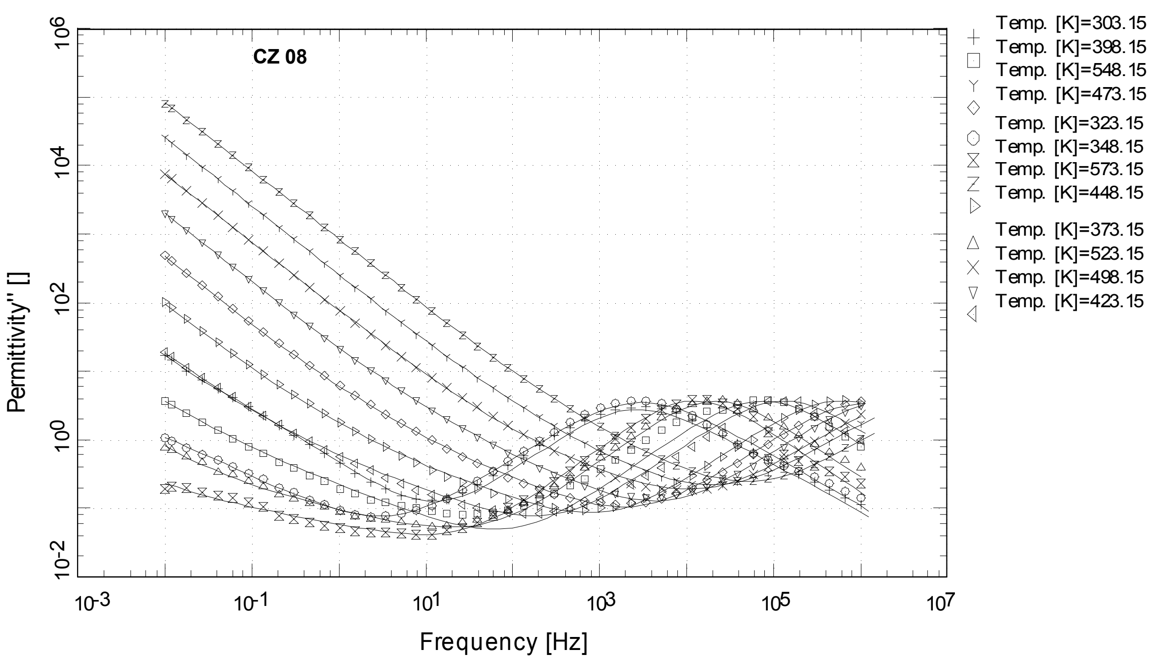

Figure 1: Dielectric permittivity in the frequency domain for $(ZnO)_{30}(MgO)_x(P_2O_5)_{70-x}$ glasses at $x = 8$. Solid line indicates fitting by the Havriliak-Negami equation superimposed with a conductivity term by using Winfit.

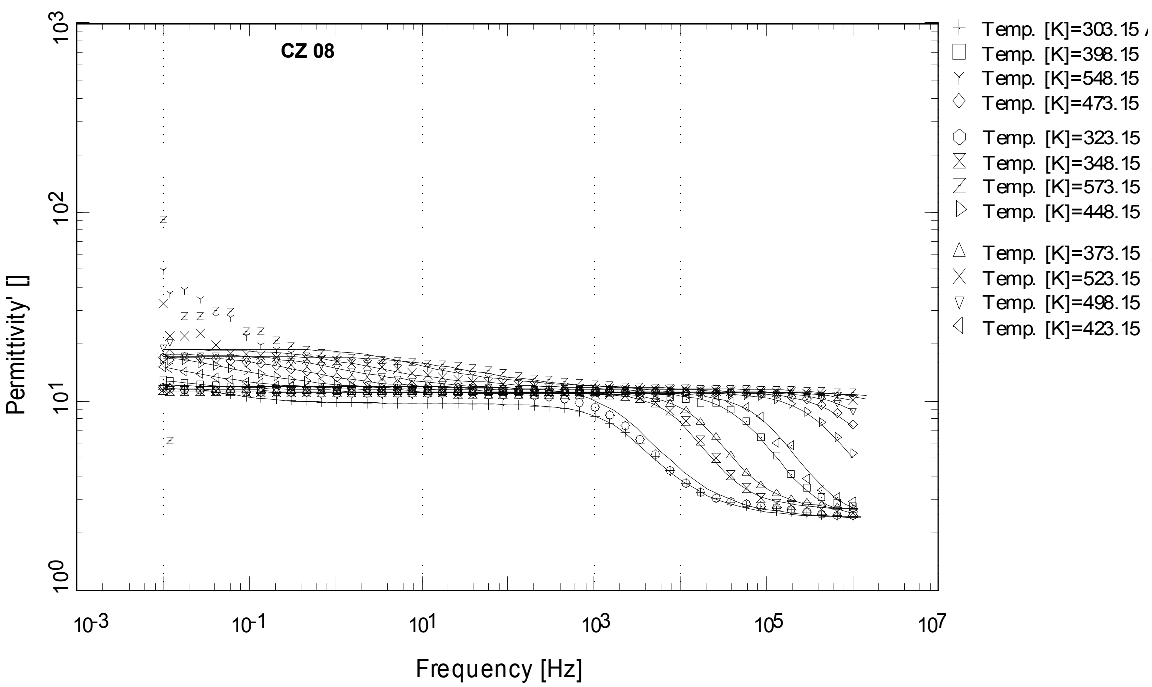

Figure 2: Dielectric loss factor in the frequency domain for $(ZnO)_{30}(MgO)_x(P_2O_5)_{70-x}$ glasses at $x = 8$. Solid line indicates fitting by the Havriliak-Negami equation superimposed with a conductivity term by using Winfit.

There are four main types of polarization mechanisms, namely electronic, atomic, orientation and interfacial or space charge polarization [3]. Figure 1 and 2 show the dielectric response of $(ZnO)_{30}(MgO)_x(P_2O_5)_{70-x}$ for $x = 8$ which is a representative of the dielectric characteristic behaviour for all glasses studied. Figure 1 shows that in the frequency limit $\omega < \omega_p$, ε' is a plateau until it reached to ω_p where dispersion takes place and ε' decreases to a constant value, ε_∞. As observed, the steplike decrease shifts uniformly towards high frequency between the ranges 10^3 - 10^6 Hz.

At lower frequency and at temperatures above 448 K, an unexpected additional relaxation process appears and its corresponding loss peak cannot be seen in the measured ε'' data on account of the dominant influenced of conduction loses. While, the strong increase of ε' at lower frequencies and at temperatures above 498 K is attributed to electrode polarization effect [4]. At low frequency the ε'' spectrum is dominated by dc conduction which manifested by the $1/\omega$ slope while dielectric loss peak at higher frequency range is observed. In the loss factor plot shown in Figure 2, the $1/\omega$ slope of ε'' at low frequency is found to increase with increasing temperature while the position of the loss peaks gradually shifted toward higher frequencies with increasing temperature.

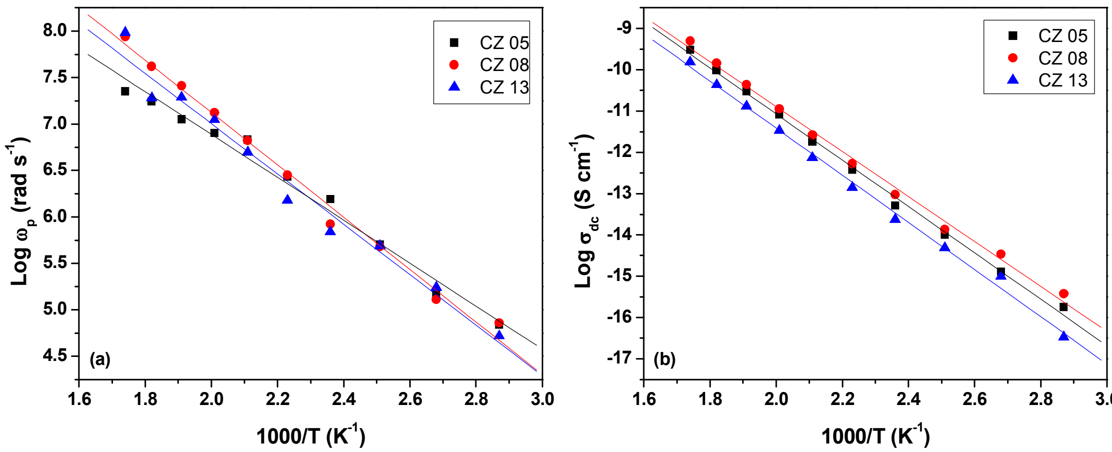

Figure 3: (a) Temperature dependence of hopping rate and (b) dc conductivity for $(ZnO)_{30}(MgO)_x(P_2O_5)_{70-x}$ glasses with different MgO concentrations.

The value of relaxation frequency, ω_p can be expressed by the usual relation: $\omega_p = \omega_\circ \exp(-E_\omega/kT)$, where E_ω is the activation energy of hopping rate, k is the Boltzmann constant and $\omega_\circ$ is the pre-exponential factor. From Figure 3(a), value ω_p plotted against 1/T shows linear dependency. Figure 3(b), the electrical conduction in glass also obeying Arrhenius relation: $\sigma = \sigma_\circ \exp(-E_\sigma/kT)$, where E_σ is activation energy of conduction. The values of E_ω and E_σ for different concentrations of MgO have been calculated and tabulated in Table 1. The value of E_σ is found to be larger than E_ω indicating that both the conduction and relaxation processes are due to different hopping conduction mechanism.

Table 1: Activation energies, E_ω and E_σ for $(ZnO)_{30}(MgO)_x(P_2O_5)_{70-x}$.

Sample Code	**E_ω (eV)**	**E_σ (eV)**
CZ 05	0.56	1.11
CZ 08	0.46	1.08
CZ 13	0.54	1.13

4. Conclusion

The dielectric permittivity of $(ZnO)_{30}(MgO)_x(P_2O_5)_{70-x}$ with different MgO concentrations is frequency and temperature dependent. From the thorough analysis of the experimental data, dc conduction loss dominates at low frequencies while dipolar type relaxation occurs at higher frequencies as observed in dielectric loss factor plot. An unexpected additional relaxation process appears at relatively lower frequency region in ε′ plots. Spectral curve fitting for the studied samples, using two Havriliak-Negami relaxation processes superimposed with a conductivity term, the estimation of the parameters $\sigma_\circ$ and ω_p is well suited and applicable for monitoring its natural characteristics of these phosphate glasses systems.

5. Conclusion

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