

Dielectric and Electrical Property of the Anhydrous Diglycine Sulfate (DGS) Single Crystals

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Abstract— Anhydrous Diglycine Sulfate (DGS) have been grown from aqueous solution of glycine and sulfate with slow evaporation techniques at constant temperature. The complex relative dielectric function $\epsilon^*(\omega) = \epsilon' - j\epsilon''$ of mixture of Anhydrous Diglycine Sulfate with water in varying concentration have been measured using precision LCR meter in the frequency range 20Hz to 2MHz at room temperature. The dielectric/electrical properties of the liquid samples are represented in terms of intensive quantities namely, complex relative dielectric function $\epsilon^*(\omega)$, conductivity $\sigma^*(\omega)$, electrical modulus $M^*(\omega)$, and extensive quantities, i.e. complex admittance $Y^*(\omega)$ and complex impedance $Z^*(\omega)$. All of these properties are used to explore various processes contributed in the dielectric/electrical presentations of the mixtures.

Index Terms— DGS, Precision LCR meter, Complex relative dielectric function, electrical modulus, Complex admittance, Complex impedance, conductivity.

1 INTRODUCTION

THE most well known glycine sulfate compounds is Triglycine sulfate (TGS) and Anhydrous Diglycine Sulfate (DGS) which have been grown from aqueous solution of glycine and sulfate. DGS exhibits non-ferroelectric property [1], [2], [3], [4], [5], [6], [7]. Whipps et al [3], Wood and Holden [2] have reported that when proportions of amino acid and sulfuric acid are taken in equal molar ratio, they obtained Anhydrous Diglycine Sulfate (DGS) single crystals. It has an orthorhombic structure and exhibits perfect cleavage along the (0 1 0) plane [1]. From the literature survey it has been found that little work has been on the dielectric and morphological properties of the anhydrous diglycine sulfate [5], [6]. Sangwa et al [8] has studied the dielectric measurement, proved to be a powerful method to study of molecular structure, solute-solute and solute-solvent interaction.

The investigation of broadband dielectric spectroscopy (BDS) of the materials covers now a days the extraordinary spectral range from 10^{-6} to 10^{12} Hz [9]. The BDS characterization of the dielectric materials provide the confirmative information on the intra- and intermolecular dynamics, the degree of intermolecular H-bond interactions, cooperation between guest and host molecules and ionic and electrode polarization processes related to the molecular structures [10], [11]. In the past, several attempts have been made to study the dynamics of polar solvents regarding the confirmation of their structural behaviour in microwave dielectric relaxation measurements. However, not much has been done in the frequency range of 20Hz to 2MHz, where the interference between ionic conduc-

tion, electrode polarization and structure relaxation may be the largest, owing to the fact that in complex systems atomic, ionic or molecular units have non-trivial interactions between them. In this paper we study the dielectric spectroscopy over the frequency range 20Hz to 2MHz of binary mixture of DGS crystal and pure water. In view of the present large interest on the molecular interaction in the liquid system, it will be highly informative to make a comparative dielectric study of the liquid samples DGS-water mixtures in the frequency range 20Hz to 2MHz and reporting the result of over accurately measured dielectric constant and other evaluated dielectric parameter at constant temperature. The dielectric and electrical properties of material are represented in terms of intensive quantities i.e. complex relative dielectric function $\epsilon^*(\omega)$, conductivity $\sigma^*(\omega)$, electrical modulus $M^*(\omega)$ and extensive quantities i.e. complex admittance $Y^*(\omega)$, complex impedance $Z^*(\omega)$. Various formulations are used to confirm the effect of ionic and electric polarization processes in the liquid mixtures.

2 EXPERIMENTAL PROCEDURE

The compound was synthesized from an aqueous solution of glycine and sulfate in the molar ratio 1:1 by controlled isothermal evaporation at constant temperature. DGS powder samples were mixed with the water in different concentration, which is used for the study of dielectric spectroscopy over the frequency range 20Hz to 2MHz. The complex dielectric function $\epsilon^*(\omega)$ of liquid samples were determined by using precision LCR meter with four terminal liquid dielectric test fixture used for capacitance and resistance measurement in the frequency range 20Hz to 2MHz. The capacitance and parallel resistance of the dielectric liquid test fixture without samples and with samples were measured to compensate for a short. The test fixture correction coefficient was also considered to cancel the effect of stray capacitance during the evaluation of the complex relative dielectric function $\epsilon^*(\omega)$.

The complex relative dielectric function $\epsilon^*(\omega)$ of the liquid samples is determined by using relation (1). [12]

$$\epsilon^*(\omega) = \epsilon' - j\epsilon'' = \alpha \left[\frac{C_p}{C_0} - j \frac{1}{\omega C_0 R_p} \right] \quad (1)$$

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Where C_0 =Capacitance of air, C_p =parallel capacitance with sample, R_p =Parallel resistance with sample, $\omega=2\pi f$ and is the angular frequency, α =correction coefficient of the cell, all measurements were made at room temperature and the temperature was controlled by constant temperature water bath.

3 RESULT AND DISCUSSION

3.1 Dielectric Constant

The complex relative dielectric function contains two terms shown in "(1)" and the dielectric constant is an important property of the dielectric materials. For most materials the value of dielectric constant depends on the frequency of the alternating field, chemical structure imperfection of the material, temperature and pressure [13]. This part is devoted to the discussion of frequency dependence of dielectric constant and dielectric loss of the liquid samples with varying concentration of the DGS sample at room temperature.

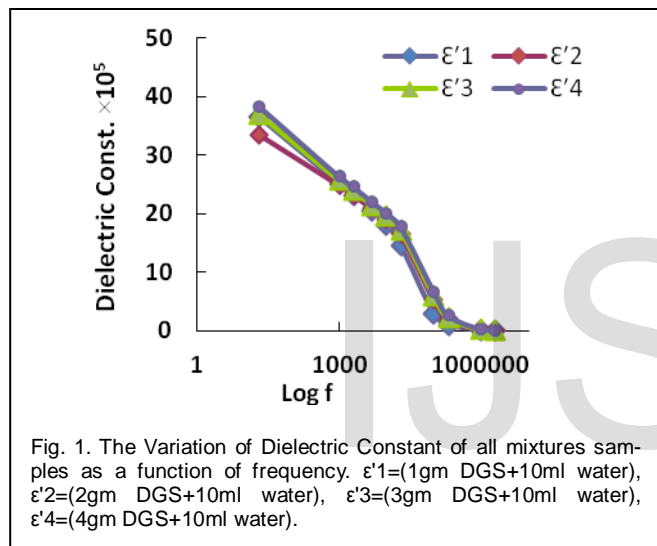


Fig. 1. The Variation of Dielectric Constant of all mixtures samples as a function of frequency. $\epsilon'1$ =(1gm DGS+10ml water), $\epsilon'2$ =(2gm DGS+10ml water), $\epsilon'3$ =(3gm DGS+10ml water), $\epsilon'4$ =(4gm DGS+10ml water).

The $\epsilon^*(\omega)=\epsilon'-j\epsilon''$ is a meaningful quantity under the condition that in the experiment the electric field is an independent variable and the charge is the dependent one. Considering the charge as the independent variable, shows that Fig. 1, the dielectric constant of all mixtures samples decreases as the frequency increasing from 20Hz to 2MHz. it can be seen that dielectric constant of different mixtures of DGS in water over a entire frequency range is high in the low frequency region and decreases gradually with the increases the frequency, its value at 2MHz are 4020, 9120, 10500, 12700 for 1gm, 2gm, 3gm and 4gm in 10ml pure water respectively. This nature of variation governs the various polarizations at low frequency, the value of dielectric constant is high confirming the ionic conduction and electrode polarization and decreases as the concentration of DGS in pure water increases.

3.2 Dielectric Loss

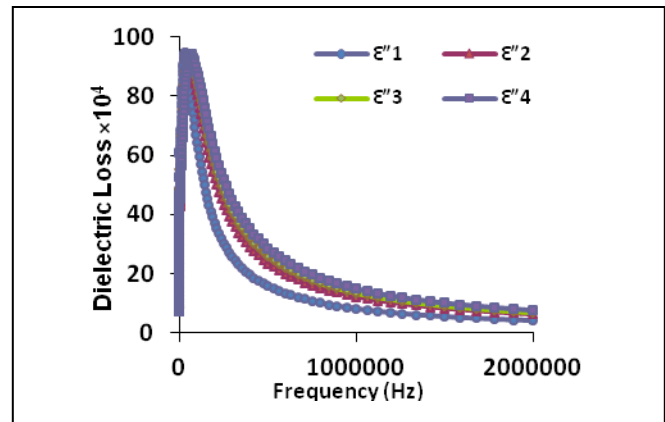


Fig. 2. The variation of Dielectric Loss of all mixtures samples as a function of frequency with peak values. $\epsilon''1$ =(1gm DGS+10ml water), $\epsilon''2$ =(2gm DGS+10ml water), $\epsilon''3$ =(3gm DGS+10ml water), $\epsilon''4$ =(4gm DGS+10ml water).

Fig. 2 shows the dielectric loss of the mixtures DGS 1gm+10ml water, 2gm DGS + 10ml water, 3gm DGS + 10ml water, 4gm DGS + 10ml water. In general the peak in dielectric loss spectra which corresponds to electrode polarization (EP), relaxation frequency (f_{EP}) is used to separate the bulk material property [14] and shows the peaks at lower frequency for all mixture of DGS in pure water in various four concentrations and after that it decreases as the frequency increases. The τ_{EP} involves charging and discharging time of Electric Double Layer (EDL) capacitance, which is associated with the overall dynamics of the adsorbed ions on the electrode surfaces in the alternating electric field [14, 15].

3.3 Conductivity

The frequency dependent real part σ' and the imaginary part σ'' of the alternating current (ac) complex conductivity $\sigma^*(\omega)$ of the liquid samples were obtained from the (2).

$$\sigma = \sigma' - j\sigma'' = \omega\epsilon_0\epsilon'' - j\omega\epsilon_0\epsilon' \quad (2)$$

Where ϵ_0 (8.854×10^{-12} F m⁻¹) is free space dielectric constant.

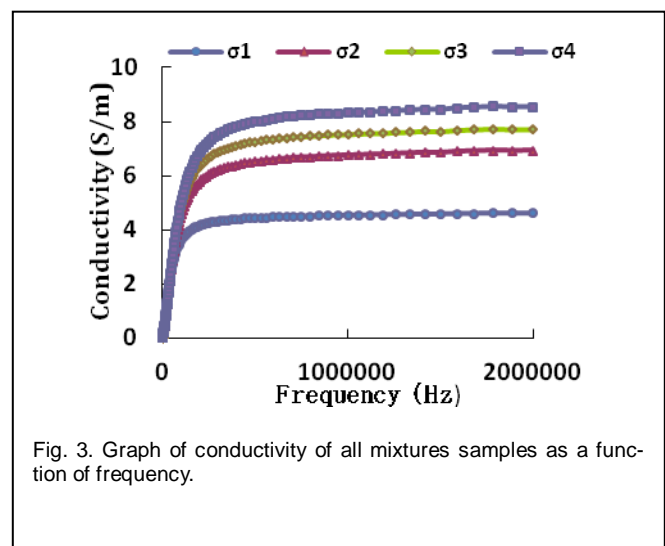


Fig. 3. Graph of conductivity of all mixtures samples as a function of frequency.

Fig. 3 shows the σ' spectra of the DGS-water mixture system. Usually, increase of ionic conductivity of a mixture is due to the increment of the number of mobile charge carriers pro-

duced in the liquid system with the change in concentration of the constituent. The real part of ac conductivity $\sigma(ac)$ of the mixture increases with the increase in the DGS crystal concentration and its values are 4.6, 6.9, 7.7, 8.54 for 1gm, 2gm, 3gm and 4gm DGS in pure water respectively. which suggests the higher conductivity of DGS crystal and the conductivity of DGS - water mixture system increases linearly in lower frequency region then after it will become almost saturation.

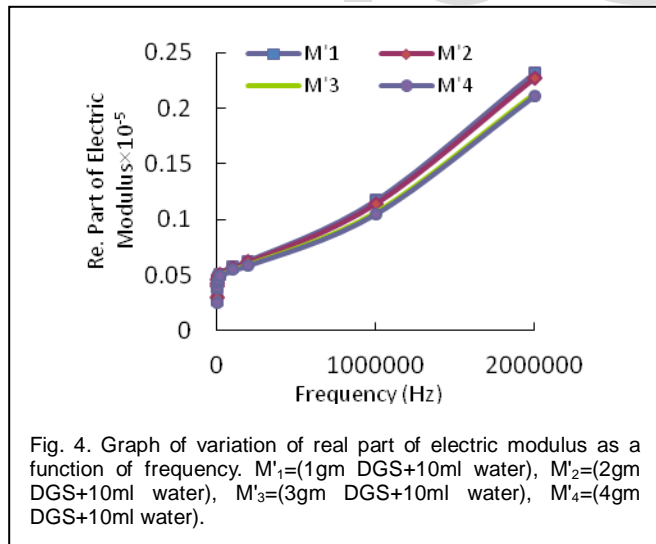
3.4 Electric Modulus

In Electric modulus study, the ionic conduction in these polar liquids and their binary mixtures may be due to electrolyte dissociation of impurities, thermal dissociation, photo ionization etc. similar effect is observed by Sengwa et al in case of pure water and alcohol [8]. Considering the charges as the independent variable, conductivity relaxation effects can be suitably analyzed within the modulus formalism in terms of a dimensionless quantity electric modulus.

The frequency dependent value of real and imaginary part of electric modulus is obtained using (3). [16], [17], [18], [19], [20], [21].

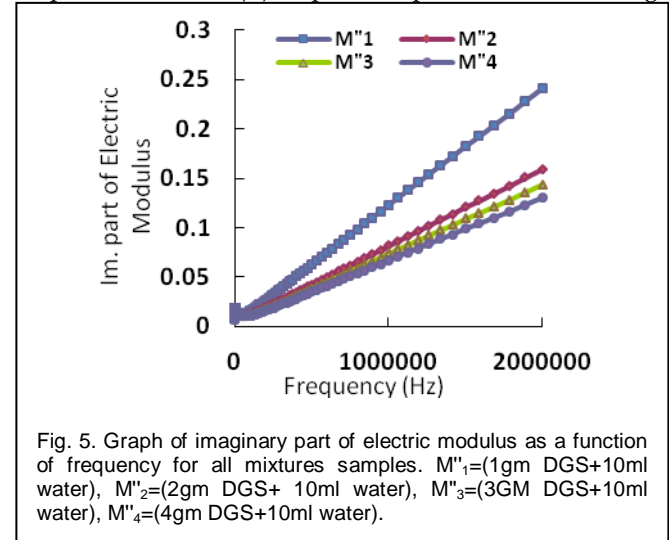
$$M^*(\omega) = M' - jM'' = \left[\frac{\epsilon'}{\epsilon'^2 + \epsilon''^2} - j \frac{\epsilon''}{\epsilon'^2 + \epsilon''^2} \right] \quad (3)$$

Interpretation of relaxation phenomena via electric modulus formalism offers some advantages upon permittivity and conductivity relaxation treatments since large variation in the ϵ' and ϵ'' values at low frequencies and high temperature are minimized. Further, difficulties occurring in the analysis of dielectric spectra from the electrode nature, the electrode-specimen electrical contact and the injection of space charges and absorbed impurities can be neglected in the electric modulus spectra.



From figure 4, it is observed that the real part of electric modulus $M'(\omega)$ of all liquid samples increases with the increase of frequency at lower frequencies of the $M'(\omega)$ spectra, which confirms that the measured dielectric/electric properties are free from the contribution of EP effect. Generally, the dielectric materials, having the contribution of EP effect in their measured dielectric properties, shows a plateau at lower frequencies and an abrupt increase in $M'(\omega)$ spectra at higher

frequencies. The $M''(\omega)$ dispersion spectra is shown in Fig. 5.



The main advantages of this formalism is that the space charge effect often do not mask the features of the spectra, owing to the suppression of high capacitance phenomena in $M''(\omega)$ spectra.

3.5 Complex Impedance Behaviour

The frequency dependent values of real parts z' and reactive part z'' of the complex impedance $z^*(\omega)$ of the liquid samples is evaluated by (4).

$$Z^*(\omega) = Z' - jZ'' = \left[\frac{R_p}{1 + (\omega C_p R_p)^2} - j \frac{\omega C_p R_p^2}{1 + (\omega C_p R_p)^2} \right] \quad (4)$$

The complex impedance plan plots (z'' vs z') are commonly used to separate the bulk material and the electrode polarization formula. A common feature of dielectrics with dc conductivity is a discontinuity at electrode/dielectric interface, which has different polarization properties than the bulk of dielectric material.

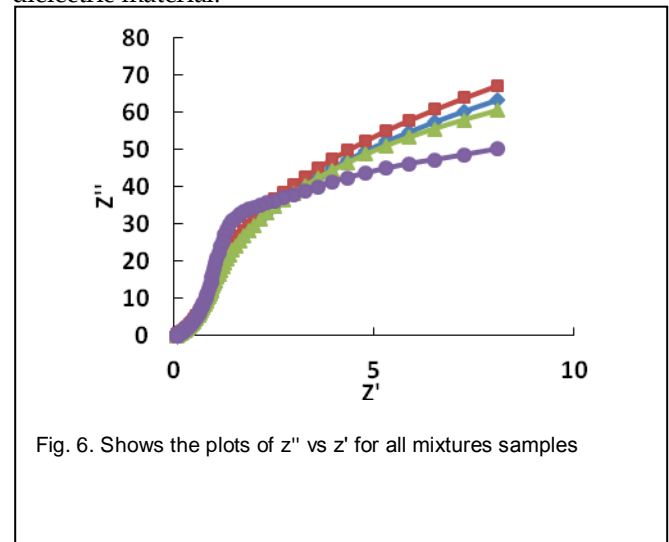


Figure 6 shows the complex impedance plan plots for all samples of DGS-pure water, usually the frequency values corresponding to z'' minimum values in the z'' vs. z' plots separates the crystal effect and surface polarization phenomena, in present study it is found to vary with concentration of DGS in

pure water and Complex admittance $Y^*(\omega)$ can be found from the equation (5). [13]

$$Z^*(\omega) = \frac{1}{Y^*(\omega)} = \frac{1}{\frac{1}{R_P} + j\omega C_P} \quad (5)$$

Where R_P and C_P is the resistance and capacitance with samples.

4 CONCLUSION

This paper reports experimental data for complex relative dielectric function, conductivity, Electric Modulus, Complex impedance of DGS and pure water mixtures with different concentration. Dielectric constant and Dielectric Loss decreases as the frequency and as DGS concentration increases, Dielectric loss shows the peak at low frequency and then decreases as the frequency increases. Conductivity also increases as the frequency increases and in Electric modulus study the value of real and imaginary part increases linearly as the frequency increases and decreases as the concentration of DGS increases. This shows that electric polarization is absent. The comparative analysis of the various dielectric and electric quantity spectra confirms that the behaviour is governed by the concentration of the DGS mixtures and plays important role in the finding of the insight of the intermolecular of the liquids mixtures.

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