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Dielectric Relaxation Study of Liquid Polymers of Ethylene glycol oligomers using Picosecond Time Domain Reflectometry

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ABSTRACT

The dielectric relaxation measurement of polymers in water mixtures have been carried out over entire concentrations, at temperatures 25^{0}C using picoseconds time domain reflectometry technique in the frequency range of 10 MHz to 30 GHz. The complex permittivity spectra of polymer - water mixtures were fitted using Havriliak-Negami equation. The static dielectric constant (ε_0), high frequency permittivity (ε_∞) and relaxation time(τ) for all concentrations have determined using least square fit method. The variations in dielectric parameters with molecular size is studied.

Keywords: Polymers, Dielectric Relaxation, time domain reflectometry, static dielectric constant (ε_0), high frequency permittivity (ε_∞) , relaxation time (τ) etc.

I. INTRODUCTION

Dielectric relaxation study on liquids provides information regarding their molecular behavior and dynamics of the molecules involved at dipolar level. Dielectric studies involve measurements of dielectric permittivity and dielectric loss. The measurement can be used to find dielectric relaxation times and distribution parameters. The relaxation time depends upon the molecular size, shape, intramolecular & intermolecular forces. Time domain reflectometry gives dielectric relaxation study over a wide frequency range [1-7].

The polymers of ethylene glycols such as Ethylene glycol(EG), Diethylene glycol(DEG),Tri ethylene glycol(TEG),Tetra ethylene glycol(TTEG),Penta-ethylene glycol(PEG)& Hexa- ethylene glycol(HEG) are organic liquids and have two -OH groups at the ends of their molecular structure. Due to present of ends -OH groups, molecules of these liquids can enter into intra & intermolecular hydrogen bonding giving rise to several conformations in water mixtures [8].Dielectric relaxation & thermodynamic properties of glycols as a function of temperature and frequency have attracted much attention from industrial and academic researchers due to strong potential in industrial and biological applications.

In this paper we have reported the values of dielectric parameters of polymers of ethylene glycols, which will be useful for the researchers.

II. METHODS AND MATERIAL

2.1 MATERIAL:

Polymers of ethylene glycol i.e. Ethylene glycol (EG) and Diethylene glycol (DEG) were obtained from S.D. Fine-Chem. Limited. Triethylene glycol (TEG), Tetra

ethylene glycol (TTEG), Pentaethylene glycol (PEG) and Hexa ethylene glycol (HEG) were obtained commercially (Aldrich) and used without further purification. The solutions were prepared at different weight fraction of water in EG, DEG, TEG, TTEG, PEG and HEG. The double distilled water is used to prepare mixtures. The dielectric spectra have been obtained by time domain reflectometry (TDR) technique. The Tektronix model no.DSA8200 Digital Serial Analyzer sampling oscilloscope along with sampling module80E08 has been used for the measurement. A repetitive fast rising voltage pulse with 18 picoseconds incident rise time was fed through coaxial line system of impedance of 50Ω. Reflected pulse without sample R_1 (t) and with sample R_x (t) were recorded in time window of 2 nanosecond and digitized in 2000 points. The Fourier transformation of pulses and data analysis were done earlier to determine the complex permittivity spectra $\varepsilon^*(\omega)$ using non linear least squares fit method [9,10].

III. RESULTS AND DISCUSSION

The frequency dependent values of dielectric permittivity (ε′) and dielectric loss (ε″) of polymers of ethylene glycols at 25^{0}C are shown in Fig.1 (a & b).It is observed from the plot that the values of dielectric permittivity (ε′) decreases with increase in the frequency and increase in number of carbon atoms and dielectric loss (ε″) peak shifts from higher to lower frequency.

In general dielectric loss spectrum of the aqueous solutions of polymers of ethylene glycol exhibits an asymmetrical shape and described by the Havriliak-Negami expression. We performed the non linear least square fitting procedure for polymers of ethylene glycol-water mixtures, in order to extract dielectric relaxation parameters with the following equation [11].

$$\varepsilon^*(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{\left[1 + (j\omega\tau)^{1-\alpha}\right]^\beta} \quad (1)$$

Where ε_0 is static dielectric constant,ε_∞ is dielectric constant at high frequency, τ is dielectric relaxation time,α and β are the distribution parameters. The value of α is kept zero and β is varied such that 0≤β≤1. The values of ε_0 and τ for polymers studied here are reported in Table 1.

3.1 STATIC DIELECTRIC CONSTANT (ε_0):

The value of static dielectric constant depends upon number of dipoles per unit volume of given liquid system. As number of carbon atoms increases from EG to HEG, the number of dipoles per unit volume of decreases. Hence the value of static dielectric constant decreases from EG to HEG.

3.2 DIELECTRIC RELAXATION TIME (τ):

The dielectric relaxation time depends upon volume of molecule and hydrogen bonding between the molecules. As number of carbon, hydrogen and oxygen atoms increases, molecular weight increases. Hence relaxation time increases. Therefore relaxation time for DEG is more than EG. But relaxation time from TEG to HEG decreases. Because as molecular size of polymer increases, instead of whole molecule individual segments rotates fastly and relaxation time decreases.

IV. CONCLUSION

The temperature dependent complex permittivity spectra of Diethylene glycol in aqueous solution have been studied using time domain reflectometry technique in the frequency range 10 MHz to 30 GHz. The variation in dielectric constant and relaxation time with increase in molecular weight of polymers indicates the structural dynamics of polymers.

V. ACKNOWLEDGEMENTS

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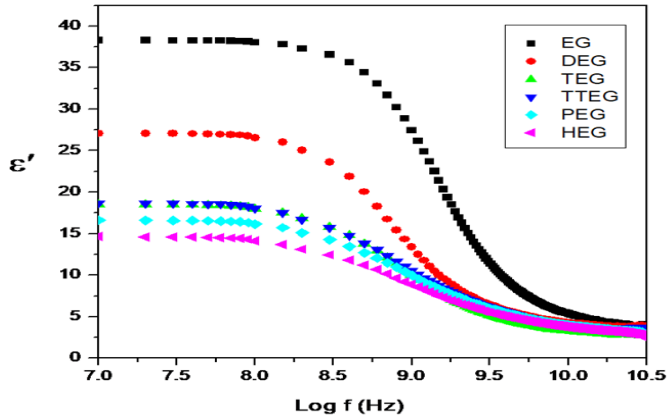

Figure 1 (a) Frequency dependent dielectric permittivity (ε′) for pure polymers of EG at 25⁰C.

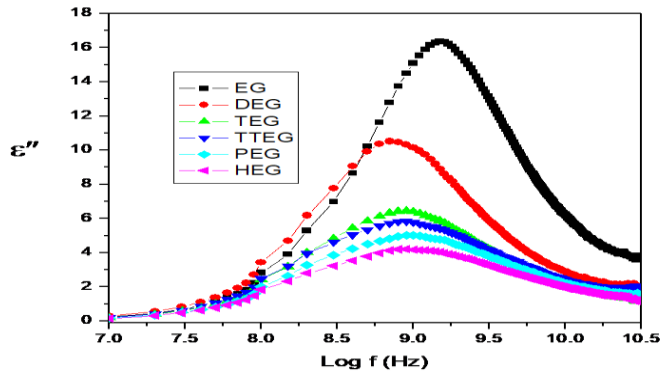

Figure 1(b) Frequency dependent dielectric loss (ε″) for pure polymers of EG

Table 1: Dielectric parameters obtained from Havriliak-Negami expression.

Sample	Static dielectric constant (ε_0)		Relaxation Time (τ) ps	
	Experimental Value	Literature Value	Experimental Value	Literature Value
EG	41.94	40.7[a]	104.42	121[a]
DEG	29.50	30.7[a]	216.82	237[a]
TEG	21.87	23.0[a]	221.36	205[a]
TTEG	20.02	20.90[b]	181.76	----------
PEG	18.82	17.80*	156.63	----------
HEG	15.12	16.60*	152.50	----------

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* Data from Japan group.

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