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Dielectric study of butanenitrile with 1,2 dichloroethane at 15^{0}C temperature using microwave frequency

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ABSTRACT

The dielectric relaxation study of butanenitrile(BN) with 1,2 Dichloroethane(DCE) mixture has been carried out at temperature 15^{0}C in the frequency range of 10 MHz to 20 GHz using time domain reflectometry (TDR) for 11 different concentrations of the system. The dielectric parameters such as static dielectric constant (ε_0) and relaxation time (τ) have been obtained by fourier transform and the least squares fit method. Excess properties of the mixtures have been determined. In the mixtures excess permittivity is found to be positive in DCE rich region and negative in BN rich region. The excess inverse relaxation time is found to be negative. The investigation shows that there is systematic change in dielectric parameters of the system with change in concentration.

Keywords: Reflectometry, Excss parameters , Relaxation time, nitrile.
PACS: 77.22d

INTRODUCTION

The dielectric relaxation study of solute-solvent mixture at microwave frequencies gives information about formation of monomers and multimers as well as interaction between the molecules of the mixture(1-3). The importance of measuring dielectric constant is that it gives the valuable information about ordering of the molecules in the liquid state. It also provides the information about the charge distribution in a molecular system. Butanenitrile (BN) is less-associative liquid and 1,2 Dichloroethane (DCE) is associative liquid. One with C≡N group and other with chlorine group. It is interesting to see the effect of nitrile group with chlorine-group. The objective of the present paper is to report the detailed study of dielectric relaxation for butanenitrile and 1,2 Dichloroethane mixture using TDR at 15^{0}C temperature at different concentrations for the frequency range of 10MHz to 20GHz range.The dielectric parameters such as dielectric constant, relaxation time and excess dielectric properties for the binary mixtures have also been determined.

MATERIALS AND METHODS

A spectrograde butanenitrile(Fluka cheme Gmbh-9471 Buchs,Steinheim,Swizerland) and AR grade 1,2 Dichloroethane (E-Merck) were used without further purification The solutions were prepared at 11 different volume percentages of BN in DCE from 0 % to 100 % just before the measurements. Using these volume percents the mole fraction is calculated as

$$x_1 = (v_1\rho_{1/}m_1) /[(v_1\rho_{1/}m_1) + (v_2\rho_{2/}m_2)]$$

where m_i, v_i, and ρ_i represent the molecular weight, volume percent, and density of the i^{th} (i=1, 2) liquids, respectively. The density and molecular weight of the liquids are as follows: Butanenitrile- density:0.8329gmcm^{-3}; mol.wt.-67.09 1, 2 Dichloroethane-density:1.256gmcm^{-3}; mol.wt.-98.96 The complex permittivity spectra were studied using the time domain reflectometry [4-6] method. The Hewlett Packard HP 54750 sampling oscilloscope with HP 54754A TDR plug in module has been used. A fast rising step voltage pulse of about 39 ps rise time generated by a pulse generator was propagated through a coaxial line system of characteristic impedance 50 Ohm. Transmission line system under test was placed at the end of coaxial line in the standard military applications (SMA) coaxial connector with 3.5 mm outer diameter and 1.35 mm effective pin length. All measurements were carried out under open load conditions. The change in the pulse after reflection from the sample placed in the cell was monitored by the sampling oscilloscope. In the experiment, time window of 5 ns was used. The reflected pulse without sample $R_l(t)$ and with sample $R_x(t)$ were digitized in 1024 points in the memory of the oscilloscope and transferred to a PC through 1.44 MB floppy diskette drive.

Data Analysis

The time dependent data were processed to obtain complex reflection coefficient spectra $\rho^*(\omega)$ over the frequency range from 10 MHz to 20 GHz using Fourier transformation [7,8] as

$$\rho^*(\omega)=(c/j\omega d)[p(\omega)/q(\omega)] \qquad (1)$$

where $p(\omega)$ and $q(\omega)$ are Fourier transforms of $[R_1(t)-R_x(t)])$ and $[R_1(t)+R_x(t)]$ respectively, c is the velocity of light, ω is angular frequency, d is the effective pin length and $j=\sqrt{-1}$.

The complex permittivity spectra $\varepsilon^*(\omega)$ were obtained from reflection coefficient spectra $\rho^*(\omega)$ by applying bilinear calibration method [4].

The experimental values of ε^* are fitted with the Debye equation [9]

$$\varepsilon^*(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + j\omega\tau} \qquad (2)$$

with ε_0, ε_∞, and τ as fitting parameters. A nonlinear least-squares fit method [10] was used to determine the values of dielectric parameters. In Eq.(2), ε_0 is the static dielectric constant, ε_∞ is the limiting high-frequency dielectric constant and τ is the relaxation time.

RESULTS AND DISCUSSION

The static dielectric constant (ε_0) and relaxation time (τ) obtained by fitting experimental data with the Debye equation are listed in Table 1. The values of static dielectric constant (ε_0)increases and relaxation time (τ) values increases with the increase of BN into DCE.

Figure 1, shows behavior of excess static dielectric constant for the system as a function of volume concentration of BN in DCE at 15^{0}C temperature

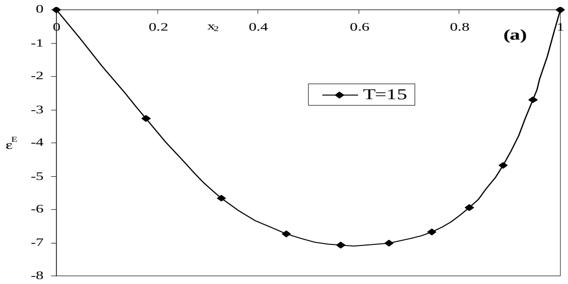

FIGURE 1. Static dielectric constant (ε_0) versus volume percentage of BN into DCE .

The information related to liquids 1 and 2 interaction may be obtained by excess properties[11] related to the permittivity and relaxation times in the mixture The excess permittivity ε^E is defined as

$$\varepsilon^E=(\varepsilon_0-\varepsilon_\infty)_m-[(\varepsilon_0-\varepsilon_\infty)_1\ x_1+(\varepsilon_0-\varepsilon_\infty)_2\ x_2] \qquad (3)$$

where x- mole fraction and suffices m, 1, 2 represents mixture, liquid 1 (BN) and liquid 2 (DCE) respectively. The excess permittivity may provide qualitative information about multimers formation in the mixture.

Similarly, the excess inverse relaxation time is defined as

$$(1/\tau)^E = (1/\tau)_m-[(1/\tau)_1\ x_1+(1/\tau)_2\ x_2\ ] \qquad (4)$$

where $(1/\tau)^E$ is excess inverse relaxation time which represents the average broadening of dielectric spectra. The inverse relaxation time analogy is taken from spectral line broadening (which is inverse of the relaxation time) in the resonant spectroscopy [12].

The experimental values of both the excess parameters were fitted to the Redlich-Kister equation[13, 14]

$$A^E = (x_1\ x_2)\sum_n B_n\left(x_1-x_2\right)^n$$

where A is either ε^E or $(1/\tau)^E$. By using these B_n values, A^E values were calculated.

The excess permittivity (ε^E) increases sharply towards the peak values then decreases slowly up to pure PN. The excess permittivity values are positive from 0 to 0.5947 concentration and are negatives from 0.6953 to pure BN concentrations. The positive values represents that the dipoles of the system are aligned in parallel direction. The total effective dipole of the system increases. There may be the formations of monomeric or polymeric structures of the molecules. In BN rich region there is decrease in dipoles and antiparallel alignment of molecules.

The excess inverse relaxation time values are negatives for all concentrations. The negative values of inverse relaxation time indicates that there is a opposing field in the molecules and there is slower effective dipole rotation of the molecules in the system.

Table1: Static dielectric constant (ε_0) and relaxation time (τ) for 15^0C temperature.

Vol. % of BN	ε_0	τ (ps)
0	10.95(0)	12.47(0)
10	13.59(4)	15.20(24)
20	15.45(2)	14.80(13)
30	16.62(1)	14.03(6)
40	16.74(2)	13.73(14)
50	17.63(1)	12.95(5)
60	18.54(1)	12.21(4)
70	19.64(2)	11.79(5)
80	20.67(1)	10.88(8)
90	21.74(1)	10.22(3)
100	23.70(4)	9.28(2)

Number in bracket represent error in the corresponding value, e.g. means 13.42(23) means 13.42 ± 0.23.

CONCLUSION

The dielectric constant ,relaxation parameters, excess parameters are reported for Butanenitrile and 1,2 Dichloroethane system at 15^0C temperature at 11 different concentrations. These data provide information regarding solute-solvent interaction. From the present study we can conclude that there is a strong molecular interaction between solute and solvent.

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