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Thermodynamical Properties of 1,2 -Dichloroethane with -OH Group Molecules using TDR

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Abstract: *The complex permittivity spectra of 1,2-dichloroetahane with -OH group molecules were determined in the frequency range of 0.1 GHz to 20 GHz using time domain reflectometry (TDR) in the temperature range 15°C to 45°C for 11 different concentrations of the system. The dielectric relaxation in this system can be described by a single relaxation time using the Debye model. The static dielectric constant (ε_0) and relaxation time (τ) have been obtained by fitting experimental data with the Debye equation. Thermodynamical properties are obtained by Eyring equation.*

Keywords: *dichloroetahane*

1. Introduction

Dielectric relaxation of liquid mixture gives information about molecular interactions. Considerable dielectric relaxation study has been done in aqueous solutions [1-3] and non-aqueous solutions such as pyridine-amide [4] and dimethylene chloride-dimethylformamide [5]. In this work, we report the dielectric study of dimethylene chloride-n methylformamide mixture. The 1,2-dichloroethane is a non-associative liquid and methanol is an associative liquid, one with chlorine group and other with C=O group. It is interesting to see the effect of chlorine group molecule in NMF. The objective of the present paper is to report the dielectric relaxation study for DCM and NMF mixtures.

2. EXPERIMENTAL SECTION

DC and NMF (AR grade, Qualigens fine chemicals Pvt. Ltd., Bombay, India) were used without further purification. The solutions were prepared at 11 different volume percentage of ethanol in 1,2-dichloroethane from 0 % to 100 % in steps of 10 %. 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:

1,2-dichloroethane - Density - 1.323 gm.cm^{-3} Mol. Wt.- 84.93
Ethanol - Density - 0.7894 gm.cm.$^{-3}$ Mol. Wt.- 46.07

The complex permittivity spectra were studied using the time domain reflectometry [6-8] 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.

The temperature controller system with water bath and a thermostat has been used to maintain the constant temperature within the accuracy limit of ±1°C. The sample cell is surrounded by a heat insulating container through which the water

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of constant temperature using a temperature controller system is circulated. The temperature at the cell is checked using the electronic thermometer.

3. DATA ANALYSIS

The time dependent data were processed to obtain complex reflection coefficient spectra $\rho^*(\omega)$ over the frequency range (0.1 to 20) GHz using Fourier transformation[6,7] of

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

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 $i=\sqrt{-1}$.

The complex permittivity spectra $\varepsilon^*(\omega)$ were obtained from reflection coefficient spectra $\rho^*(\omega)$ by applying bilinear calibration method[8]. For the calibration process, 1,2-dichloroethane and ethanol were used as calibrating liquids.

The experimental values of $\varepsilon^*(\omega)$ were fitted with the Debye equation[9].

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

with ε_0, ε_∞ and τ are the adjustable parameters. A nonlinear Least-Squares fit method[10] was used to determine the values of dielectric parameters.

with ε_0 and τ as adjustable parameters. A nonlinear Least-Squares fit method[10] was used to determine the values of dielectric parameters. The value of ε_∞ is taken to be fixed as 2.0. The parameter ε_∞ has not been used as a fitting parameter, as the data are not found to be sensitive with respect to this parameter.

4. RESULTS AND DISCUSSION

The static dielectric constant (ε_0), relaxation time (τ) obtained by fitting experimental data with the Debye equation are listed in Table 1.

The behavior of static dielectric constant of 1,2-dichloroethane–ethanol system, as a function of mole fraction of ethanol in 1,2-dichloroethane at different temperatures. The static dielectric constant and relaxation time increase with increasing mole fraction of ethanol in the system.

Table 1. Temperature dependent dielectric relaxation parameters for 1,2-dichloroethane-ethanol mixtures [11][a]

x_2	T= 15°C	T= 25°C	T= 35°C	T= 45°C
ε_0				
0.0000	10.88(0)	10.43(0)	10.10(0)	9.82(0)
0.1795	15.60(6)	14.80(2)	14.10(4)	13.60(7)
0.3298	19.40(3)	18.30(7)	17.30(2)	16.40(9)
0.4576	22.90(0)	21.60(5)	20.30(0)	19.10(9)
0.5676	25.70(2)	24.10(5)	22.80(2)	21.50(4)
0.6631	28.00(4)	26.40(2)	24.90(2)	23.40(1)
0.7470	30.10(3)	28.20(1)	26.80(3)	25.10(2)
0.8212	31.70(1)	29.80(1)	28.30(7)	26.40(2)
0.8873	33.20(2)	31.10(1)	29.60(2)	27.50(6)
0.9466	34.10(2)	32.30(2)	30.40(9)	28.30(7)
1.0000	34.64(0)	32.57(0)	30.90(1)	28.91(0)

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t (ps)				
0.0000	11.47(0)	10.71(0)	10.12(0)	9.92(0)
0.1795	21.60(3)	18.20(2)	15.90(3)	14.1(3)
0.3298	32.30(1)	25.60(3)	22.30(4)	20.4(2)
0.4576	40.20(1)	32.70(4)	28.70(7)	25.9(4)
0.5676	45.20(2)	37.90(3)	33.40(5)	30.4(2)
0.6631	48.20(6)	41.20(6)	36.70(8)	33.9(1)
0.7470	50.50(2)	43.70(2)	39.40(5)	36.5(4)
0.8212	51.90(3)	45.40(3)	41.60(6)	37.8(3)
0.8873	52.90(3)	46.40(6)	43.10(3)	38.8(6)
0.9466	53.50(9)	46.90(4)	43.80(1)	39.1(2)
1.0000	54.36(0)	47.17(0)	43.92(0)	39.44(0)

[a]x_2 is the mole fraction of ethanol in 1,2-dichloroethane. Numbers in bracket represent error in the corresponding value, e.g. 48.20(6) means 48.20 ± 0.06.

The thermodynamic parameters like molar enthalpy of activation ΔH and molar entropy of activation ΔS are determined from the Eyring rate equation[12] utilizing least square fit method as

$$\tau = \frac{h}{KT}\exp\left(\frac{\Delta G}{RT}\right) \quad (3)$$

where, $\Delta G = \Delta H - T\Delta S$

The excess free energy of activation of 1,2-dichloroethane-ethanol binary mixture is given by the equation.

$$\Delta G^E = \Delta G - \left[\Delta G_A X_A + \Delta G_B X_B\right] \quad (4)$$

where, ΔG_A and ΔG_B represents the activation energies for 1,2-dichloroethane and ethanol, respectively and X_A is the mole fraction of 1,2-dichloroethane. Here $X_B = 1 - X_A$ is the mole fraction of ethanol in the studied 1,2-dichloroethane-ethanol mixture.

Table 1: Values of thermodynamical parameters for the mixture of 1,2-dichloroethane-ethanol system.

x_2	ΔG (KJ/mol)				ΔH (KJ/mol)	ΔS (J/mol)
	T=15°C	T=25°C	T=35°C	T=45°C		
0.0000	10.1101	10.4175	10.7250	11.0324	1.2554	-0.0307
0.1795	11.6258	11.7421	11.8584	11.9747	8.2761	-0.0116
0.3298	12.5341	12.6534	12.7727	12.8920	9.0983	-0.0119
0.4576	13.0799	13.2366	13.3933	13.5500	8.5667	-0.0157
0.5676	13.3758	13.5783	13.7808	13.9833	7.5436	-0.0203
0.6631	13.5326	13.7789	14.0252	14.2715	6.4390	-0.0246
0.7470	13.6478	13.9231	14.1984	14.4737	5.7196	-0.0275
0.8212	13.7254	14.0143	14.3031	14.5919	5.4069	-0.0289
0.8873	13.7737	14.0736	14.3735	14.6734	5.1369	-0.0300
0.9466	13.8029	14.1025	14.4022	14.7018	5.1736	-0.0300
1.0000	13.8322	14.1260	14.4198	14.7136	5.3710	-0.0294

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When two liquids are mixed together, there is a change in the energy of the system. This change in energy can be interpreted in terms of its activation energies such as free energy of activation (ΔG), molar enthalpy of activation (ΔH) and molar entropy of activation (ΔS) are tabulated in Table 1.

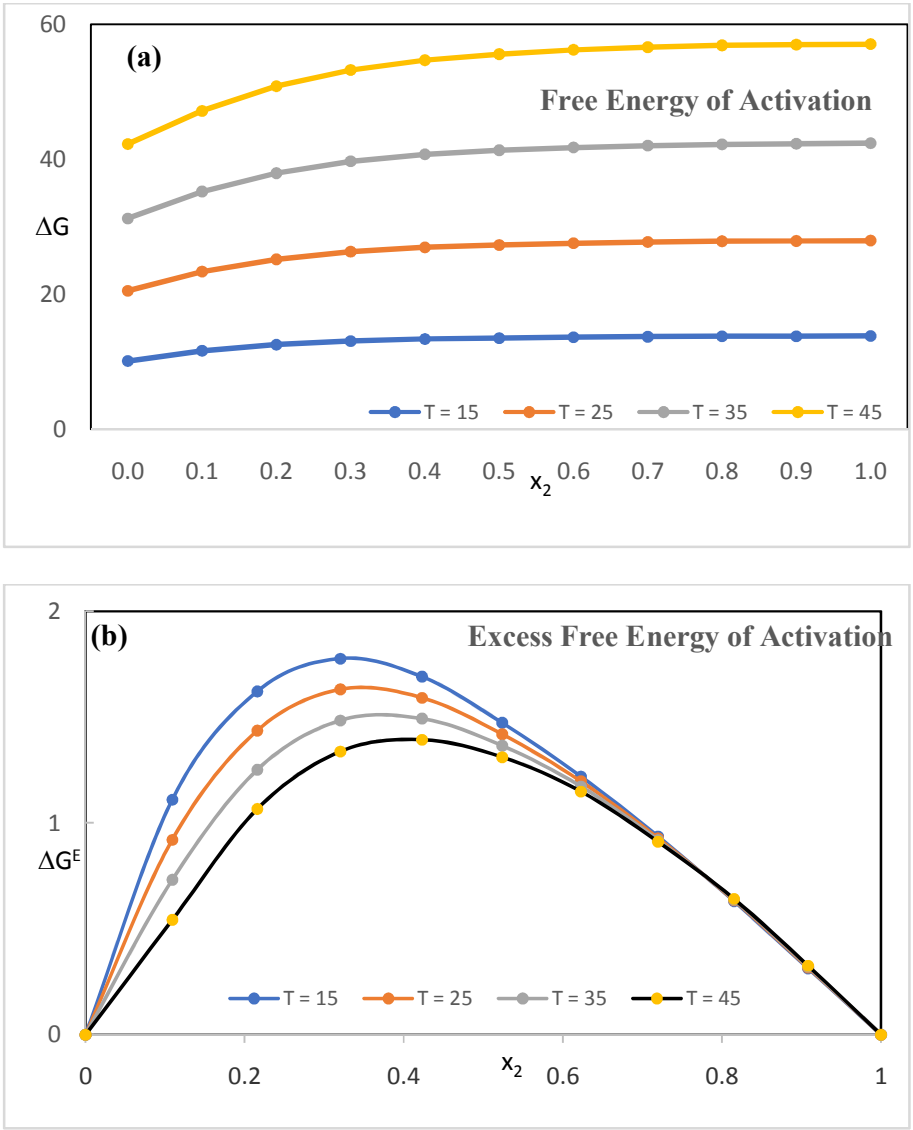

Figure 2(a): Free energy of activation (ΔG) and **(b):** Excess free energy of activation (ΔG^E) versus mole fraction (x_2) of ethanol in 1,2-dichloroethane at different temperatures.

The value of free energy of activation (ΔG) is least for 1,2-dichloroethanel and increases with increase in concentration of ethanol at all temperatures as shown in Figure 2 (a). This indicated that the ethanol molecules in the mixture dominate over 1,2-dichloroethanel molecules. This dominance of ethanol over 1,2-dichloroethanel is primarily due to its large dipole moment values. From the Figure 2 (b), the excess activation free energy (ΔG^E) values are positive for all concentrations. These positive values confirms that there is an increase in hindrance to the reorientation for the molecules of 1,2-dichloroethanel, ethanol and in their binary mixtures.

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The values of molar enthalpy of activation (ΔH) are positive for all concentrations of the system. The positive value shows endothermic interaction between 1,2-dichloroethanel-ethanol mixture of the system. The values of molar entropy of activation (ΔS) of the system are a measure of the orderly nature of the molecule. In this system, these values are negative for all concentrations, it means the environment of the system is non-cooperative resulting in the activated state, which is more disordered than the normal state.

5. CONCLUSION

Dielectric relaxation parameters are reported in the paper for binary mixture of 1,2-dichloroethane and -OH molecule (ethanol) for various temperature and concentrations. The dielectric constant and relaxation time provide information regarding solute-solvent interaction in liquids. This data provides information regarding solute-solvent interaction in liquids.

Free energy of activation increases with increasing the mole fraction of ethanol in 1,2-dichloroethane of the system and its excess values are positive for all concentrations.

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