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Ultrasonic, Density and Viscosity Studies in Binary Mixtures of Propylene Glycol and 1-Pentanol at Room Temperature

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***Abstract*— Speed of sound (U), Density (ρ), and viscosity (η) values for the binary mixture systems of Propylene Glycol with 1-pentanol including those of pure liquids were measured over the entire mole fraction range at temperature 303.15 K. From the experimentally determined values, thermo-acoustic parameters such as excess isentropic compressibility (K_s^E), excess molar volume (V^E) and excess free length (L_f^E) and excess enthalpy (H^E) have been calculated. These results have been explained on the basis of intermolecular interactions between the components in the liquid mixture and correlations among the parameters are discussed.**

Keywords: Speed of sound, density, excess molar volume, isentropic compressibility, free length, Propylene Glycol

1. INTRODUCTION

Speed of sound investigations along with the volumetric and viscometric studies of liquids and liquid mixtures are of considerable importance and they play a significant role in understanding the intermolecular interactions occurring among the component molecules besides finding extensive applications in several industrial and technological processes [1, 2]. Several researchers [3-8] have measured the density, viscosity, and speed of sound for a wide range of binary mixtures containing alcohols as one of the components, and these properties were interpreted in terms of specific or nonspecific interactions. Alcohols are strongly associated in solution because of dipole-dipole interaction and hydrogen bonding. They are of great importance for their relevant role in chemistry, biology and studies on hydrogen bonding in liquid mixtures. Alcohols are also widely used as solvents. The molecules containing –OH group form associative liquids due to hydrogen bonding. The effect shown by the molecules with other functional groups on these molecules plays an important role in understanding the behavior of hydrogen bonding. The investigations regarding the molecular association in liquid mixtures having aromatic group as one of the components is of particular interest, since aromatic group is highly non-polar and can associate with any other group having some degree of polar attractions. Even though considerable work has been reported on alcohols as one of the component in binary and ternary mixtures, the data on binary mixtures of Pentanol with Propylene glycol at room temperature (303 K) variation is scanty.

The study of thermodynamic properties of multi component liquid mixtures and data on the analysis in terms of various models are important for industrial and pharmaceutical applications [9]. The excess thermodynamic functions [10] are sensitively dependent not only on the differences in intermolecular forces, but also on the differences in the size of the molecules. The signs and magnitudes of these excess values can throw light on the strength of interactions. So from the experimentally determined values of speed of sound density and viscosity, various thermo-acoustic parameters like excess isentropic compressibility (K_s^E), excess molar volume (V^E), excess free length (L_f^E) and excess enthalpy (H^E) have been calculated. Here we report the results and discuss excess thermo-acoustic parameters using the data regarding the speed of sound, density and viscosity for the binary liquid mixtures of Propylene glycol with 1-pentanol at temperature 303 K.

II. EXPERIMENTAL DETAILS

a. *Materials*

The chemicals used in the present study are, Propylene glycol and 1-pentanol which are of AR grade obtained from Merck Co. Inc., Germany, with purities of greater than 99%. All the chemicals were further purified by standard methods [11] and only middle fractions were collected.

b. Measurements

All binary mixtures were prepared gravimetrically in air-tight bottles and adequate precautions have been taken to minimize evaporation losses. Before use, the chemicals were stored over 0.4nm molecular sieves approximately for 72 hours to remove water content and then degassed. The mass measurements were performed on a digital electronic balance (Mettler Toledo AB 135, Switzerland) with an uncertainty of $\pm 10^{-8}$ kg. The binary mixtures were prepared just before use. The uncertainty in mole fraction was estimated to be less than ±0.0001.

The viscosities were measured with Ostwald viscometer. The viscometer was calibrated at each temperature using redistilled water. The uncertainty in viscosity measurement is up to 0.001mPa-s. The flow time has been measured after the attainment of bath temperature by each mixture. The flow measurements were made with an electronic stop watch with a precision of 0.01s. For all the pure components and

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mixtures, 3 to 4 readings were taken and the average of these values were used in all the calculations.

The densities of the pure compounds and their mixtures were determined accurately using 10 ml specific gravity bottles. The average uncertainty in the measured density was ±0.001 kg/m^3.

The speed of sound was measured with a single-crystal variable path interferometer (Mittal Enterprises, New Delhi, India) operating at a frequency of 2 MHz that had been calibrated with water and benzene. The uncertainty in the speed of sound was found to be ±0.1m/s.

c. Computational Details

The values of experimentally determined density and speed of sound for the binary mixtures of Propylene glycol with 1-Pentanol at 303 K over the entire composition range.

In the present work the authors have calculated the excess values of isentropic compressibility and excess free length values to check the applicability of thermo dynamical ideality (the ideal mixing rules) to the components under study.

The excess values of isentropic compressibility K_s^E were calculated as follows,

$$K_s^E = K_s - K_s^{id} \quad (1)$$

Where K_s represent the calculated value of isentropic compressibility for the mixture

$$K_s = \frac{1}{\rho U^2} \quad (2)$$

K_s^E is its excess value, K_s^{id} is the ideal isentropic compressibility value, ρ is the density and U represents the speed of sound. K_s^{id} for an ideal mixture was calculated from the relation recommended by Benson and Kiyohara [12, 13] and Douheret et al [14].

$$K_s^{id} = \sum \phi_i \left\{ K_{s,i}^o + \frac{T V_i^o (\alpha_i^o)^2}{C_{p,i}^o} \right\} - T \left(\sum x_i V_i^o \right) \left(\frac{\sum \phi_i \alpha_i^{o2}}{\sum x_i C_{p,i}^o} \right) \quad (3)$$

in which $K_{s,i}^o$, V_o^i, α_i^o, $C_{p,i}^o$ are the isentropic compressibility, molar volume, isobaric thermal expansion coefficient and molar isobaric heat capacity of pure component i, T represents absolute temperature, ϕ_i is the volume fraction and x_i represents the mole fraction of i in the mixture.

The density values have been used to calculate the excess volumes, V^E, using the following equation,

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \left(\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right) \quad (4)$$

where ρ is the density of the mixture and x_1, M_1, and ρ_1 and x_2, M_2, and ρ_2 are the mole fraction, molar mass, and density of pure components 1 and 2, respectively.

The excess values of free length (L_f^E), Gibbs free energy (ΔG^{*E}) and enthalpy (H^E) were calculated by using the expressions given in literature [15] as follows,

$$L_f^E = L_f - K_T (K_s^{id})^{1/2} \quad (5)$$

Where L_f represents the calculated value for the mixture and K_T represent a temperature dependent constant whose value is K_T=(91.368+0.3565T)x10^{-8}.

Excess enthalpy H^E was calculated from usual relation.

$$H^E = H - (x_1 H_1 + x_2 H_2) \quad (6)$$

Where H represents the calculated value of enthalpy for the mixture and H_1, H_2 represent enthalpy of pure components 1 and 2, respectively.

III. RESULTS AND DISCUSSION

The values of density, viscosity and speed of sound for the binary liquid mixtures of propylene glycol and pentanol at 303 K temperatures were determined and are given in Table 1.

Density : $\rho_l = \frac{\rho_{wt}}{w_{wt}} wt_l$

Viscosity (ŋ): Viscosity (ŋ): $\eta_l = \frac{\eta_{wt}}{t_w \rho_w} \rho_l t_l$

Ultra sonic velocity (U) = f.λ ms^{-1}

f=frequency of the generator = 2 x 10^6 hertz

λ= wavelength = 2d/n

Where d = average of taking oscillations.

n = no. of oscillations

Table 1:Density, ultrasonic velocity and viscosity for different mole fractions at room temperature.

MoleFraction "x1"	Density ρ(Kg . m^{-3})	UltrasonicVelocity U(m.s^{-1})	Viscosity η (m.Pa.S)
0	0.81350	1253.4	3.012
0.14074	0.82614	1273.6	4.28328
0.26923	0.84806	1296.11	5.58673
0.38705	0.87220	1316.28	7.09618
0.49547	0.89595	1344.5	8.54525
0.59564	0.92006	1370.3	10.28555
0.68848	0.94475	1403.2	12.04361
0.77467	0.96788	1438.67	14.3428
0.8549	0.99073	1485.62	16.73178
0.92986	1.01171	1552.61	19.29251
1	1.03070	1624	22.9899

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From these values, various thermo-acoustic parameters like a isentropic compressibility (K_s), free length (L_f), free volume (V_f), and enthalpy (H) have been determined and excess values like the excess isentropic compressibility (K_s^E), excess molar volume (VE), excess free length (L_f^E), and Excess enthalpy (H^E), have been calculated.

The values of thermo-acoustical parameters of intermolecular free length (L_f), adiabatic compressibility (β),free volume (V_f), and enthalpy (H) at temperatures T=303 K are given in Table 2

Table 2: The values of thermo-acoustical parameters such as intermolecular free length (L_f), adiabatic compressibility (K_s),free volume (V_f), internal pressure (π), enthalpy (H)in binary liquid mixtures containing propylene glycol and pentanol at temperatures T = 303 K

MoleFraction "x1"	Isentropic compressibility(K_s)	Free length (L_f)	Free volume (V_f)	Enthalpy (H)
0	7.82462 E-10	5.57744E-11	6.36449E-4	26589.11438
0.14074	7.46243 E-10	5.44683E-11	3.97333E-4	31179.24195
0.26923	7.01922 E-10	5.2826E-11	2.82055E-4	34876.60505
0.38705	6.61736 E-10	5.12915E-11	2.07085E-4	38527.16384
0.49547	6.1744E -10	4.95451E-11	1.65717E-4	41348.60072
0.594	5.7883-10	4.79711E-11	1.3198E-4	44430.76897
0.68848	5.37581 E-10	4.62302E-11	1.10119E-4	46989.08418
0.77467	4.99176 E-10	4.45482E-11	8.9594E-5	50133.4254
0.8549	4.57325 E-10	4.26399E-11	7.58871E-5	52773.44256
0.92986	4.10033 E-10	4.0375E-11	6.65123E-5	54951.04174
1	3.67871 E-10	3.82429E-11	5.54934E-5	58196.7742

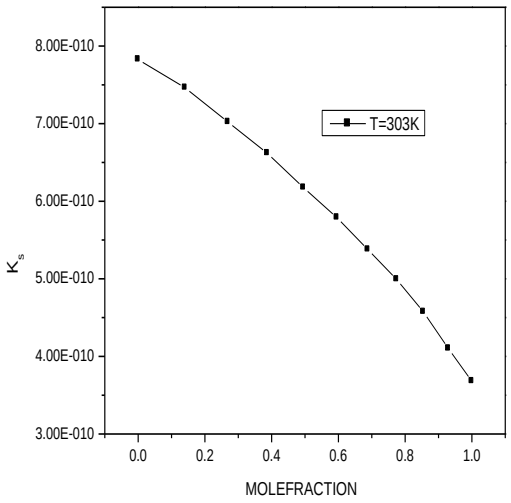

Fig.1. Mole fraction with K_S

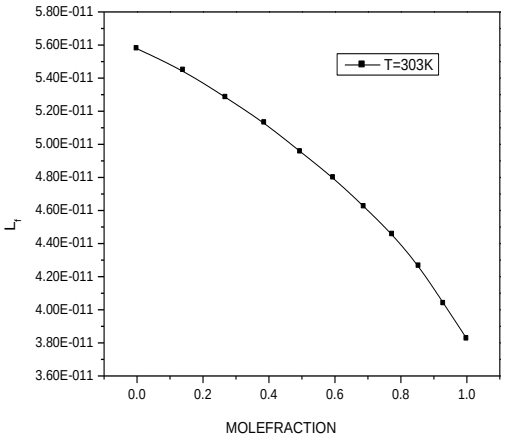

Fig.2. Mole fraction with L_f

and the values of excess thermo-acoustical parameters such as excess intermolecular free length (L_f^E), excess adiabatic compressibility (K_s^E), excess free volume (V_f^E), excess enthalpy (H^E) and at temperatures T=303K are given in Table 3 . The variations of the above excess thermo-acoustical parameters at temperatures T=303 K with the mole fractions of propylene glycol are represented in the figures from Fig-1to Fig-9

MoleFraction "x1"	Excess Adiabatic compressibility K_S^E ($m^2.N^{-1}$)	Excess Intermolecular free length $L_f^E*10^{-13}$/m	Excess Free volume V^E($m^3.mol^{-1}$)	Excess Enthalpy H^E ($J.mol^{-1}$)
0	0	0	0	0
0.14074	22.1305	11.61243	-9.33185	-22569.0505
0.26923	31.08067	17.71627	-5.50393	-14810.40503
0.38705	39.7411	23.02696	-2.39783	-7435.81878
0.49547	40.39519	24.57029	0.34902	-1187.46747
0.59564	43.31699	26.39159	2.71898	5060.85109
0.68848	40.55666	25.25842	4.74302	10553.63167
0.77467	37.88533	23.54939	6.65703	16422.24659
0.8549	29.29733	18.53169	8.367	21598.15513
0.92986	13.08271	9.02469	10.03936	26145.07275
1	0	0	0	0

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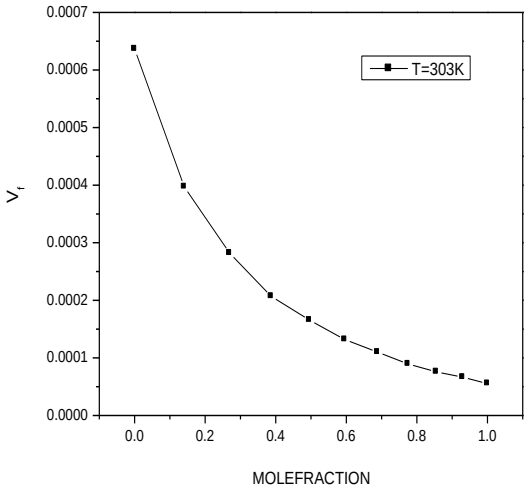

Fig.3. Mole fraction with V_f

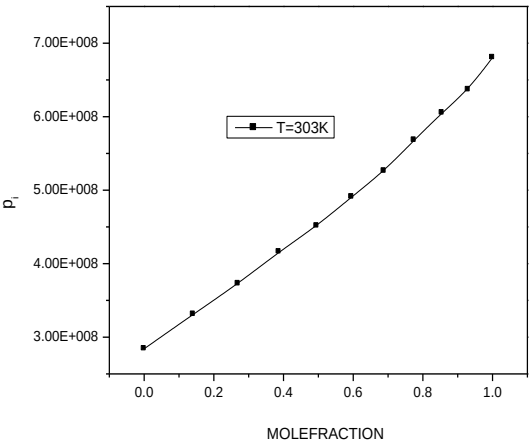

Fig.4. Mole fraction with p_i

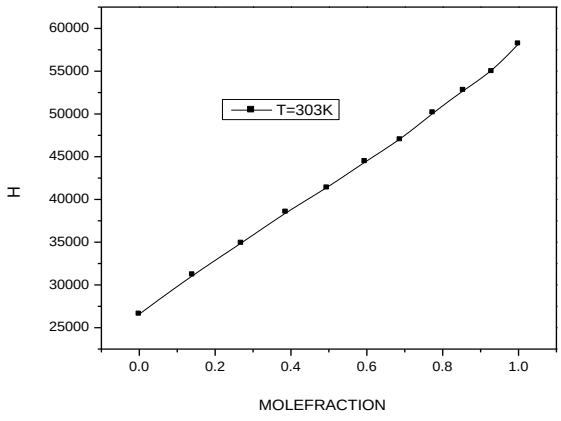

Fig.5. Mole fraction with H

Table 3 : The values of excess thermo acoustical parameters such as excess intermolecular free length (L_f^E), excess adiabatic compressibility (K_S^E), excess free volume (V_f^E),) and excess enthalpy (H^E) in a binary liquid mixture containing propylene glycol and pentanol at temperatures T=303 K are given below in Table 3 and in figures 6-9.

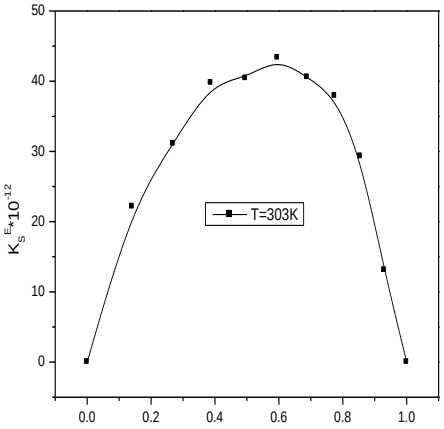

Fig.6. Mole fraction with K_S^E

From Fig-6 represents the variations of excess adiabatic compressibility (K_S^E) in binary liquid mixtures containing propylene glycol and pentanol over the entire mole fraction range of propylene glycol at temperature T= 303 K.

It is observed that the excess adiabatic compressibility (K_S^E) values are negative. This indicates that the attractive forces between the molecules of components are stronger [16] than the intra-molecular attractions in each component. According to Fort and Moore [17] a negative excess compressibility is an indication of strong hetero-molecular interaction in the liquid mixtures which is attributable to charge transfer, dipole-dipole, dipole-induced dipole interactions, and hydrogen bonding between unlike components. In the present study, the excess compressibility is negative and it suggests the existence of strong intermolecular interactions in the binary liquid mixture.

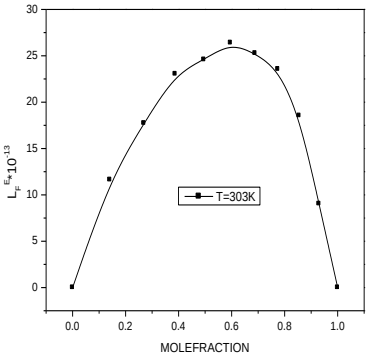

Fig. 7. Mole fraction with L_f^E

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From Fig-7, it is observed that the excess intermolecular freelength values are negative for the entire mole fraction range. The negative values of excess intermolecular free length suggest that there exist strong interactions between the components of liquid mixture[16]

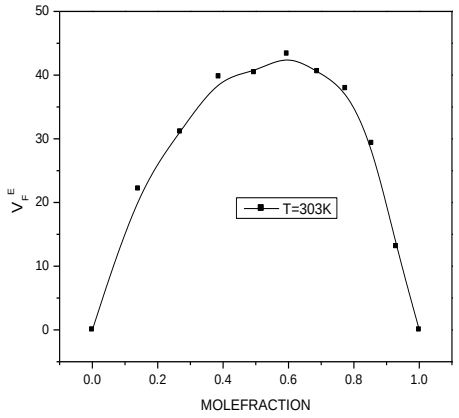

Fig.8. Mole fraction with V_f^E

The variations of excess free volume (V_f^E) with the mole fraction of propylene glycol is represented in Fig-8. It is observed from Fig-8 values are negative over the entire composition range. This suggests that the component molecules are more close together in the liquid mixture than in pure liquids forming the mixture, indicating that strong attractive interactions[18].

The variation of excess enthalpy in the present binary system is as shown in Fig-9 it is observed that H^E values are negative over the entire composition range of propylene glycol. The negative values of H^E suggest strong interactions [19].

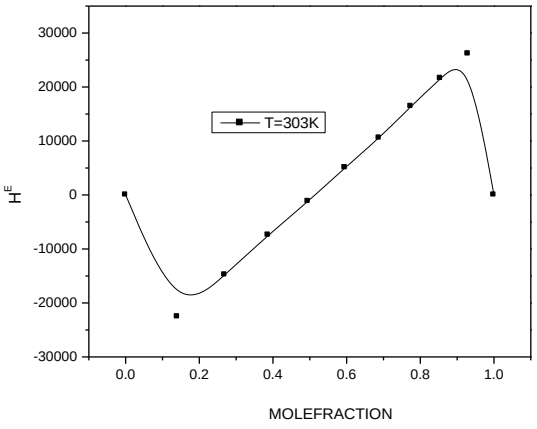

Fig. 9. Mole fraction with H^E

the variations of adiabatic compressibility(K_S) with respect to the mole fraction at temperatures T= 303 K. It is observed table from table 2. the value of adiabatic compressibility decreases with increase in mole fraction. According to Ali *et al* , these types of variations indicate that there exist strong molecular interactions between the component molecules of liquid mixture and also interactions become weaker with increase of temperature. The variations of intermolecular free length (L_f) with mole fraction at temperatures T=303 K is represented and it is observed that, the value of intermolecular free length decreases with increase in mole fraction. The decrease in intermolecular free length with mole fraction indicates strong intermolecular interactions between the component molecules of the liquid mixture.

The variations of excess intermolecular free length (L_f^E) with the mole fraction of propylene glycol ranging from 0 to 1 at temperatures T=303 K in the binary liquid mixtures containing propylene glycol and pentanol is at 303 K is as shown in negative. Therefore the molecular interactions are observed and may due to formation hydrogen bond between the constituents' molecules.

IV. CONCLUSIONS

The excess parameters like K_S^E , V_f^E, L_f^E, and H^E are calculated from the experimentally determined speed of sound, density and viscosity values. The formation of hydrogen bond between the mixtures is identified by studying the variations in these parameters through molecular interactions between the two moieties.

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