

Molecular Interaction Studies on Ester with Cyclohexane in Alcohol at 303, 308 and 313K

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ABSTRACT

The ultrasonic velocity (U), density (ρ) and viscosity (η) have been measured for the ternary mixtures of Methyl Benzoate and Cyclohexane with Butanol at 303, 308 and 313K. From the experimental data, Adiabatic Compressibility (β), Free Length (L_F), Free Volume (V_F), Internal Pressure (π_i), Relaxation time (τ), Acoustic Impedance (Z), Gibbs Free Energy (ΔG), Classical Absorption Coefficient (d/f^2) and Cohesive Energy (H) have been calculated. In addition to that the excess values of certain above parameters are also evaluated. The excess properties have been used to discuss the presence of significant interactions between the component molecules in the ternary mixtures.

Keywords: Density, Viscosity, Adiabatic Compressibility, Excess values.

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INTRODUCTION

In recent years, the measurement of ultrasonic velocity has been successfully employed in understanding the nature of molecular interactions in pure liquids and liquid mixtures. Ultrasonic velocity measurements are highly sensitive to molecular interactions and can be used to provide qualitative information about the physical nature and strength of molecular interaction in liquid mixtures [4,5,8]. The ultrasonic velocity of a molecular interaction in Ternary mixtures has been studied by many authors [9,14,19].

Variations in ultrasonic velocity and related parameters have shed much light upon the structural changes associated with liquid mixtures of weakly or strongly interacting compounds [3,15]. The study of molecular associations in ternary mixtures having an alcohol as one component is of particular interest, since alcohols are strongly self-associated liquids with a three-dimensional network of hydrogen bonds and can be associated with any other group having some degree of polar attractions [2,18]. A survey of literature has shown that a few attempts have been made to obtain ultrasonic velocity data for ternary liquid mixtures [6,16,17].

Methyl benzoate is an ester, reacts with acids to liberate heat along with alcohols and acids. Methyl benzoate is used as a source of benzoyl radical. Cyclohexane is non - polar, it is a solvent and it is used in the production of nylon. Butanol is a four straight chain alcohol, is a volatile and it is polar used as a direct solvent.

Therefore in order to have a clear understanding of the intermolecular interactions between the component molecules, a thorough study on the liquid mixtures (Methyl Benzoate + Cyclohexane + Butanol) using ultrasonic velocity data has been performed at 303, 313 and 323K.

MATERIALS AND METHODS

All the chemicals used in the present work are Analar grade. The purity of the chemicals was ascertained by comparing their densities, viscosities and ultrasonic velocities at 303, 308 and 313K which agrees with the corresponding literature values. The mixtures of Methyl Benzoate + Cyclohexane + Butanol were prepared by weight. The mole fraction of the second component, Cyclohexane ($x_2=0.4$) was kept constant, while the mole fractions of the remaining two (x_1, x_3) were varied from 0.1 to 0.6. The ultrasonic velocity was measured by a single crystal interferometer with a high degree of accuracy operating at a frequency of 3 MHz (model F-05, with digital micrometer) at 303, 308 and 313K. The viscosity was measured by Ostwald's viscometer. An electronically operated constant temperature water bath is used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature. Densities of the mixtures have been found by relative measurement method.

Theory and Calculations

Intermolecular free length (L_F) is calculated using the standard expression

$$L_F = K \beta^{1/2} \quad (1)$$

where K is a temperature dependent constant known as Jacobson constant and β is the adiabatic compressibility that can be calculated from the speed of sound (U) and the density of the medium (ρ) as-

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

The relation for free volume in terms of ultrasonic velocity (U) and the viscosity (η) of the liquid as--

$$V_F = \left(\frac{M_{eff} U}{\eta K} \right)^{\frac{2}{3}} \quad (3)$$

Expression for the determination of internal pressure (π_i) by the use of free volume (V_F) as-

$$\pi_i = bRT \left(\frac{K\eta}{U} \right)^{\frac{1}{2}} \left(\frac{\rho^{\frac{2}{3}}}{M_{eff}^{\frac{2}{3}}} \right) \quad (4)$$

where b stands for cubic packing which is assumed to be 2 for liquids and K is a dimensionless constant independent of temperature and nature of liquids and its value is 4.281×10^9 , T is the absolute temperature and M_{eff} is the effective molecular weight.

Relaxation time (τ) can be calculated using viscosity and adiabatic compressibility-

$$\tau = \frac{4}{3} \eta \beta \quad (5)$$

$$\text{Acoustic Impedance } Z = \rho U \quad (6)$$

where ρ is the density and U is the ultrasonic velocity-

$$\text{Gibb's Free Energy } \Delta G = K_B T \ln \left(\frac{K_B T \tau}{h} \right) \quad (7)$$

where $K_B = 1.23 \times 10^{-23}$ J/K

t is the temperature

τ is the relaxation time

$h = 6.626 \times 10^{-34}$ Js (Planck's Constant)

Classical Absorption coefficient is calculated by the formula-

$$\frac{d}{f^2} = \frac{8\pi^2 \eta}{3U^2 \rho} \quad (8)$$

where η is the viscosity of the liquid and ρ is the density of the liquid-

Cohesive energy (H) is given by-

$$H = \pi_i \left(\frac{M_{eff}}{\rho} \right) \quad (9)$$

where π_i is the Internal Pressure and M_{eff} is effective molecular weight-

In order to study the non-ideality of the liquid mixtures, namely excess parameters (A^E) of all the acoustic parameter were computed by

$$A^E = A_{exp} - A_{id} \quad (10)$$

$$A_{id} = \sum A_i X_i$$

where

$$A_i \text{ is any acoustical parameter and } x_i \text{ is the mole fraction of the liquid components.} \quad (11)$$

RESULTS AND DISCUSSION

The experimentally determined values of density (ρ), Viscosity (η) and Ultrasonic velocity (U) for the three systems at different temperatures are given in the Table-(1). From these observed values various acoustical parameters like adiabatic compressibility (β), free length (L_F), free volume (V_F), internal pressure (π_i), acoustic impedance (Z), relaxation time (τ), have been evaluated and is presented in the Tables (2) and (3). Table(4) represents Gibbs free energy (ΔG), Classical Absorption Coefficient (d/f^2) and Cohesive Energy (H) for the mixtures. Excess values are depicted in., Table (5).

From the Table (1) it was observed that the ultrasonic velocity, density of the ternary liquid mixtures decreases with increasing mole fraction of Butanol while the viscosity increases. However the ultrasonic velocity, density, viscosity decreases in all the cases as temperature increases. The same result was obtained by A.N.Kannappan et al [11]. Methyl Benzoate is an industrial ester, Alcohols are polar liquids strongly associated with hydrogen bonding, with an extent of polymerization that may differ depending on temperature, chain length and position of OH group. When the compounds are mixed, the changes that occur in association equilibrium were evidently due to the rupture of the hydrogen bonds in pure Cyclohexane, Butanol and Methyl Benzoate. It is well known that Butanol forms a variety of a species with different degree of association in the pure state; however, polymeric linear associates are expected to be predominant in the pure state.

Table -1 : Density (ρ), viscosity(η) and velocity (U) of Methyl Benzoate, Cyclohexane and Butanol at 303, 308 and 313K.

Methyl Benzoate + Cyclohexane +Butanol										
x_1	x_3	$\rho \text{ Kg/m}^3$			$\eta \times 10^{-3} \text{ Ns/m}^2$			U (m/s)		
		303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
0.60	0.00	957.90	957.00	953.00	0.93	0.88	0.84	1282.80	1270.00	1246.00
0.50	0.10	901.81	900.77	897.59	0.93	0.87	0.82	1361.64	1338.26	1316.07
0.40	0.20	876.69	874.81	871.63	0.96	0.89	0.83	1295.60	1244.12	1226.14
0.30	0.30	848.81	846.49	841.35	0.96	0.89	0.85	1256.12	1194.90	1184.17
0.20	0.40	822.51	820.92	814.21	1.01	0.93	0.89	1224.34	1193.46	1178.00
0.10	0.50	815.05	813.00	809.00	1.06	1.00	0.93	1187.70	1172.17	1127.80
0.00	0.60	782.46	779.22	774.81	1.20	1.08	0.99	1046.20	1010.89	986.30

Table -2: Adiabatic compressibility (β), free length (L_F) and free volume (V_F) of Methyl Benzoate, Cyclohexane and Butanol at 303, 308and 313K.

x_1	x_3	Adiabatic compressibility			Free length			Free Volume		
		$\beta \times 10^{-10} \text{ m}^2/\text{N}$			$L_F \times 10^{-10} \text{ m}$			$V_F \times 10^{-7} \text{ m}^3 \text{ mol}^{-1}$		
		303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
0.60	0.00	6.34	6.48	6.76	0.50	0.51	0.53	2.25	2.41	2.52
0.50	0.10	5.98	6.20	6.43	0.49	0.50	0.51	2.27	2.46	2.63
0.40	0.20	6.79	7.39	7.63	0.52	0.55	0.56	1.86	1.98	2.12
0.30	0.30	7.47	8.27	8.48	0.55	0.58	0.59	1.60	1.68	1.77
0.20	0.40	8.11	8.55	8.85	0.57	0.59	0.60	1.31	1.41	1.48
0.10	0.50	8.70	8.94	9.72	0.59	0.60	0.63	1.04	1.10	1.17
0.00	0.60	11.68	12.56	13.27	0.68	0.71	0.74	0.64	0.70	0.78

Thus, the additional of an ester Methyl Benzoate to a Butanol may result in the following effects:

- (i) rupturing or disruption of associate structures in alcohols,
- (ii) formation of new species because of interaction between ester and alcohols, and
- (iii) free volume changes upon mixing of components of different sizes [1].

Further the adiabatic compressibility, Free Length shows an inverse behavior compared to the ultrasonic velocity in the mixtures, as shown in Fig (iv, v). It is primarily the compressibility that changes with the structure and this lead to the change in ultrasonic velocity.

The addition of interacting molecules breaks up the molecular clustering of the other releasing several dipoles for the interactions [8]. In view of greater force of interaction between the molecules there will be an

increase in cohesive energy and the occurrence of structural changes takes place due to the existence of electrostatic field.

Thus structural arrangement of molecules results in increasing adiabatic compressibility there by showing intermolecular interactions. Similar results in some liquid mixtures were also reported by others [7,13,18].

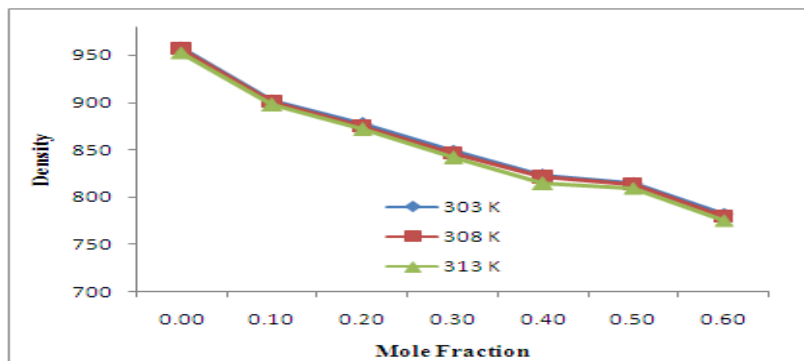


Fig.-1

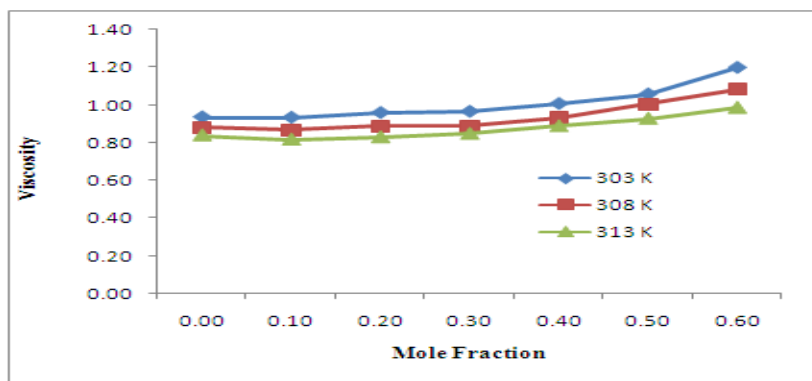


Fig.-2

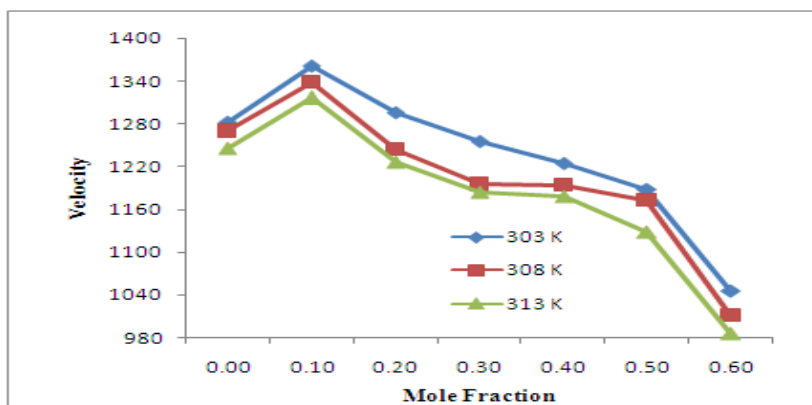


Fig.-3

Fig.(1-3): Variation of Density, Viscosity, Velocity Versus Mole fraction of (Methyl Benzoate + Cyclohexane + Butanol) at 303, 308, 313K.

From tables-2 and 3, it was observed that as the concentration of primary alcohol increases, free volume decreases. Internal pressure increases with increasing concentration of alcohol. However, with rise in temperature increase in free volume and decrease in internal pressure are noticed. This suggests that there is a closed packing of molecules inside the shield. Such an increase in internal pressure generally indicates association through hydrogen bonding and hence supports the present investigation.

In the Methyl Benzoate structure there are a lot voids available for O-H to penetrate and enter into complexion, and also provides information regarding the hydrogen bond between interacting components[1].In the present investigation, from the table it is observed that these acoustic impedance (Z) values decreases with increasing concentration of alcohols. Such decreasing trends of acoustic impedance further support the possibility of molecular interaction due to H-bonding between the Methyl Benzoate and alcohol molecules.

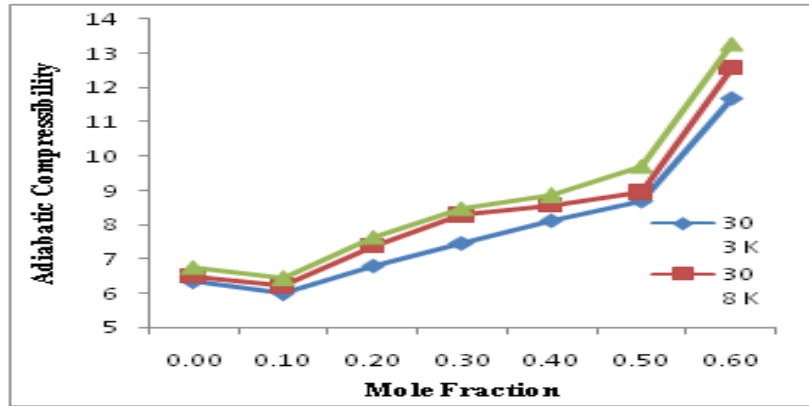


Fig.-4

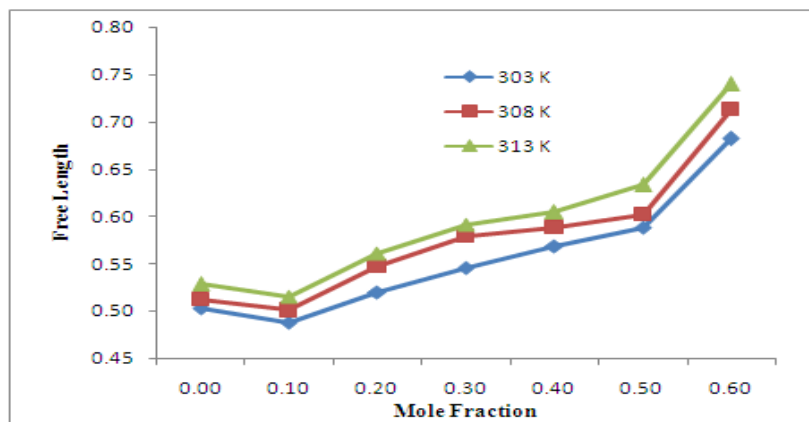


Fig.-5

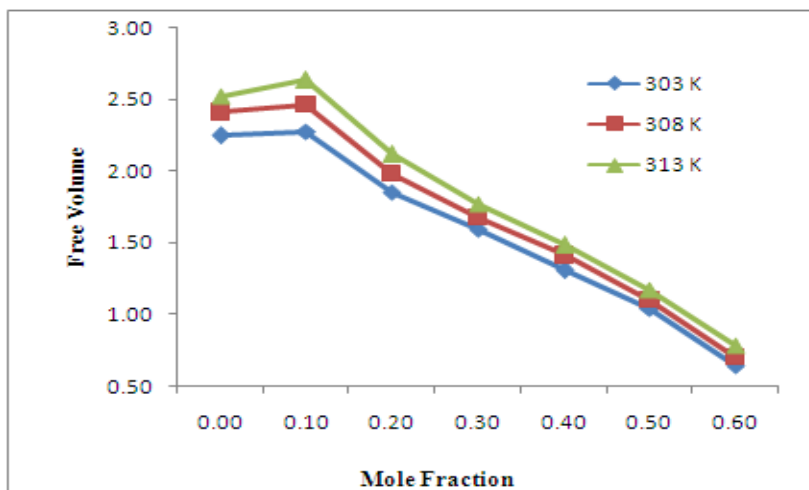


Fig.-6

Fig. (4-6): Variation of Adiabatic Compressibility, Free Length ,Free Volume Versus Mole fraction of (Methyl Benzoate + Cyclohexane + Butanol) at 303,308,313K.

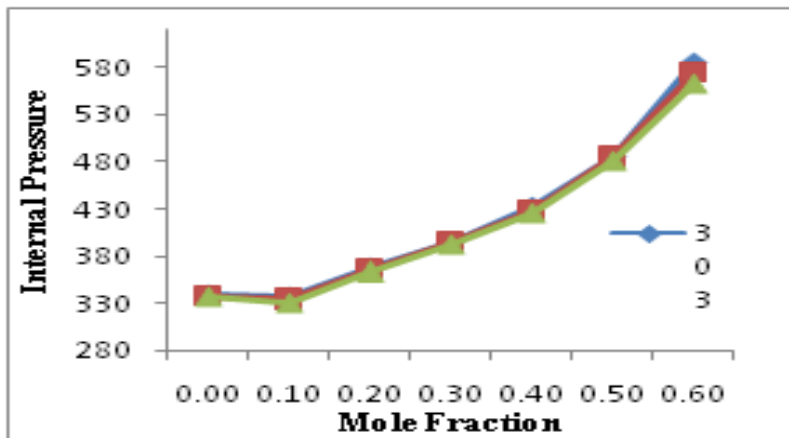


Fig.-7

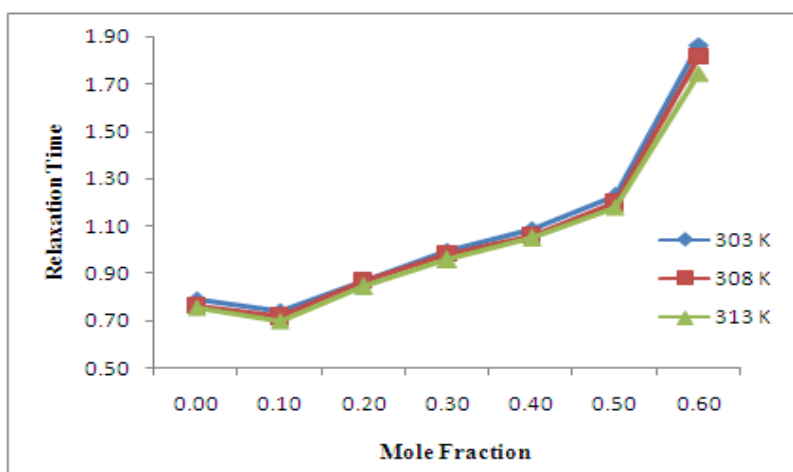


Fig.-8

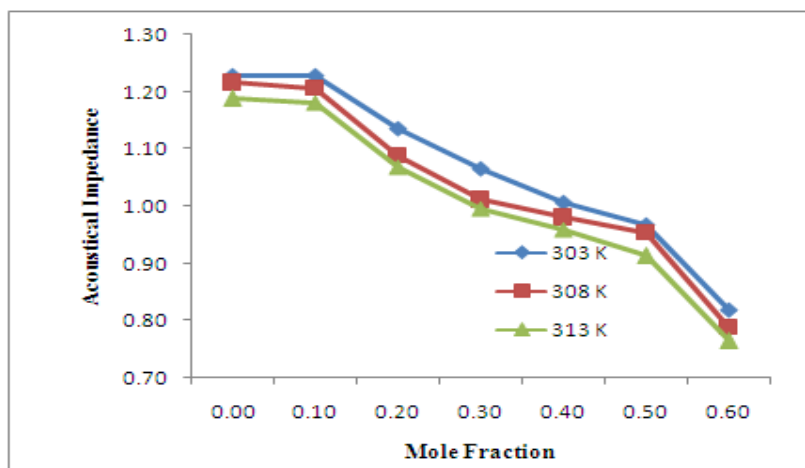


Fig.-9

Fig. (7-9):Variation of Internal Pressure,Relaxation Time,Acoustical Impedance Versus Mole fraction of (Methyl Benzoate + Cyclohexane + Butanol) at 303,308,313K.

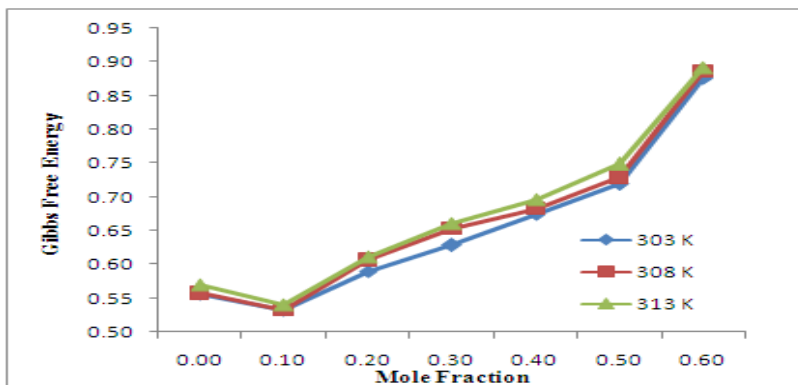


Fig.-10

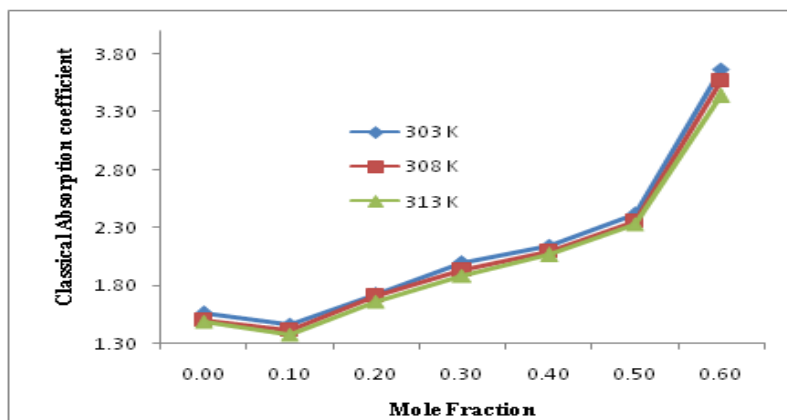


Fig.-11

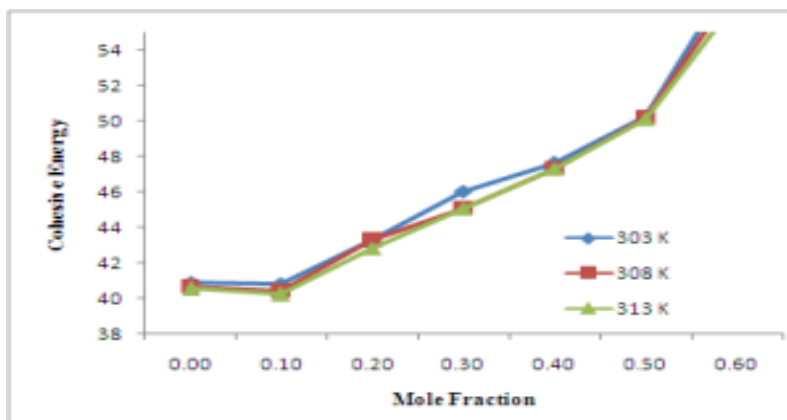


Fig.-12

Fig. (10-12): Variation of Gibbs Free Energy, Classical Absorption Coefficient, Cohesive Energy Versus Mole fraction of (Methyl Benzoate + Cyclohexane + Butanol) at 303,308,313K.

The relaxation time (τ) increases with increases in mole fraction of alcohol, which shows the presence of molecular interaction. The relaxation time (τ) shows continuous increase with chain length of alcohols and offer hindrance to the rotation of the molecule. At high concentration of alcohol in the mixture, there are a large number of alcohol molecules surrounding the ester molecules. At low concentration of alcohol in the mixtures, there are only a small number of alcohol molecules to enable dipole-dipole interaction through hydrogen bonding with the non associative ester molecules. The associative alcohol molecules act as proton donor enabling hydrogen bonding with Methyl Benzoate molecules. From the table (4) Gibbs free energy, Classical Absorption Coefficient and Cohesive Energy

are increases with the increase value of mole fraction. This shows that there is a strong interaction between the mixture.

In order to understand more about the nature of the interaction between the components of liquid mixture, it is necessary to discuss the same in terms of excess parameters rather than the actual values.

They can yield an idea about the non linearity of the system as association or other type of interactions [18]. The variation of excess parameters versus mole fraction of alcohol have been depicted form the Figures-13 and 16.

Table - 3: Internal Pressure (π_i), Relaxation time (τ) and acoustical impedance (Z) of Methyl Benzoate , Cyclohexane and Butanol at 303K, 308K and 313K

x_1	x_3	Internal Pressure			Relaxation time			Acoustical Impedance		
		$\pi_i \times 10^6 \text{ N/m}^2$			$\tau \times 10^{-12} \text{ Sec}$			$Z \times 10^{-6} \text{ Kg m}^2/\text{sec}$		
		303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
0.60	0.00	339.93	337.30	336.73	0.79	0.76	0.74	1.23	1.22	1.19
0.50	0.10	337.45	333.71	330.83	0.74	0.72	0.70	1.23	1.21	1.18
0.40	0.20	368.33	365.99	362.70	0.87	0.87	0.84	1.14	1.09	1.07
0.30	0.30	394.99	394.50	391.83	1.00	0.98	0.96	1.07	1.01	1.00
0.20	0.40	432.26	427.86	425.22	1.09	1.06	1.05	1.01	0.98	0.96
0.10	0.50	485.81	484.23	480.52	1.22	1.20	1.18	0.97	0.95	0.91
0.00	0.60	585.99	574.97	562.58	1.86	1.81	1.75	0.82	0.79	0.76

Table - 4: Gibbs Free Energy (ΔG), Classical Absorption Coefficient (d/f^2) and Cohesive Energy (H) of Methyl

x_1	x_3	Gibbs Free Energy			Classical Absorption Coefficient			Cohesive Energy (H)		
		$(\Delta G) \times 10^{-20} \text{ KJ/mol}$			$(d/f^2) \times 10^{-11}$			$\times 10^3$		
		303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
0.60	0.00	0.56	0.56	0.57	1.56	1.50	1.49	40.91	40.64	40.54
0.50	0.10	0.53	0.53	0.54	1.47	1.42	1.38	40.84	40.44	40.23
0.40	0.20	0.59	0.61	0.61	1.72	1.71	1.66	43.25	43.25	42.84
0.30	0.30	0.63	0.65	0.66	1.99	1.93	1.89	46.01	45.08	45.05
0.20	0.40	0.67	0.68	0.70	2.14	2.09	2.07	47.58	47.28	47.28
0.10	0.50	0.72	0.73	0.75	2.42	2.36	2.33	50.26	50.17	50.08
0.00	0.60	0.88	0.89	0.89	3.67	3.58	3.44	58.51	57.65	56.73

Rai et al suggested that the negative excess compressibility has been due to closed packed molecules and the positive excess values are due to weak interaction between the unlike molecules[17,12]. Similar conclusions were also arrived by Debey et al [6]. From the table (5) the excess properties were found to decrease with increase in temperature. The value of excess inter molecular free length follows the same trend as that of excess adiabatic compressibility. The value of excess inter molecular free length are negative.

The negative deviations of excess free volume are an indication of the existence of the strong interaction between the components [10,11]. The negative excess internal pressure over the entire range of mole fraction of the system also supports the presence of interaction.

CONCLUSIONS

The results obtained for the present study indicate that the thermodynamic parameters are sensitive to the molecular interactive present in liquid mixtures.

From Ultrasonic velocity and related acoustical parameters for ternary mixtures it is observed that at high concentration of alcohol in the mixture, there are a large number of alcohol molecules surrounding the ester molecules.

At low concentration of alcohol in the mixtures, there are only a small number of alcohol molecules to enable dipole-dipole interaction through hydrogen bonding with the non associative ester molecules.

Table - 5 : Excess Adiabatic compressibility (β^E), Excess Free length (L_f^E), Excess Free Volume (V_f^E) and Excess Internal Pressure (π_i^E) of Methyl Benzoate, Cyclohexane and Butanol at 303, 308 and 313K

x_1	x_3	Excess Adiabatic compressibility			Excess Free length			Excess Free Volume			Excess Internal Pressure		
		$\beta^E \times 10^{-10} \text{ m}^2/\text{N}$			$L_f^E \times 10^{-10} \text{ m}$			$V_f^E \times 10^{-7} \text{ m}^3 \text{ mol}^{-1}$			$\pi_i^E \times 10^6 \text{ N/ m}^2$		
		303K	308K	313K	303K	308K	313K	303K	308K	313K	303K	308K	313K
0.6	0	-0.16	-0.32	-0.34	0.00	-0.01	-0.01	0.72	0.74	0.67	-49.12	-45.81	-38.25
0.5	0.1	-0.83	-1.06	-0.98	-0.03	-0.03	-0.03	0.86	0.90	0.90	-92.15	-85.99	-78.05
0.4	0.2	-0.33	-0.15	-0.09	-0.01	0.01	0.01	0.55	0.53	0.51	-101.85	-90.33	-80.12
0.3	0.3	0.03	0.46	0.45	0.01	0.02	0.02	0.41	0.34	0.28	-115.78	-98.44	-84.93
0.2	0.4	0.37	0.45	0.52	0.02	0.02	0.02	0.23	0.19	0.12	-119.13	-101.73	-85.51
0.1	0.5	0.65	0.57	1.07	0.02	0.02	0.04	0.08	0.00	-0.07	-106.08	-81.91	-64.08
0	0.6	3.31	3.90	4.31	0.10	0.12	0.13	-0.21	-0.29	-0.34	-46.53	-27.83	-16.00

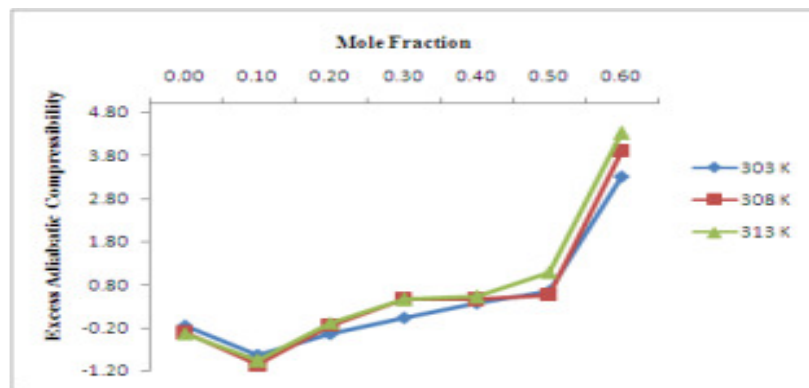


Fig.-13

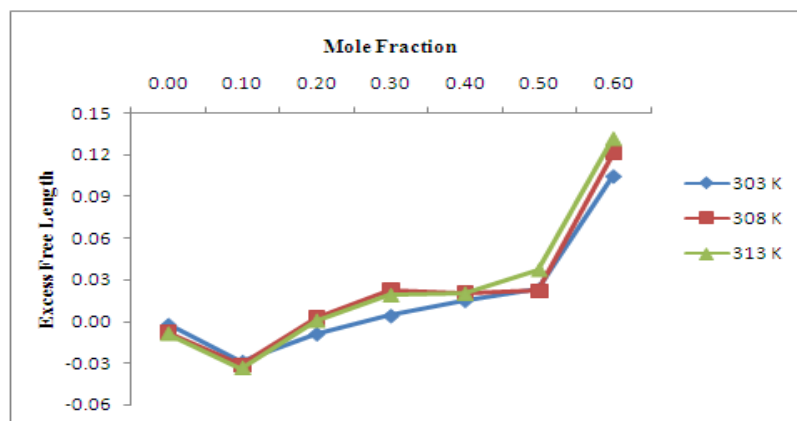


Fig.-14

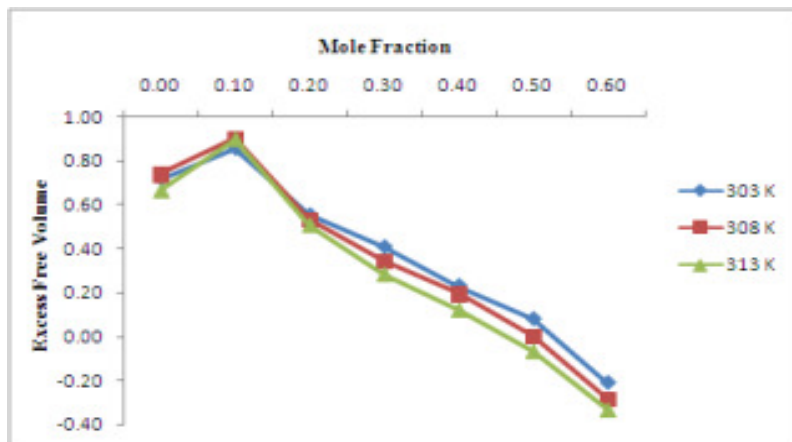


Fig.-15

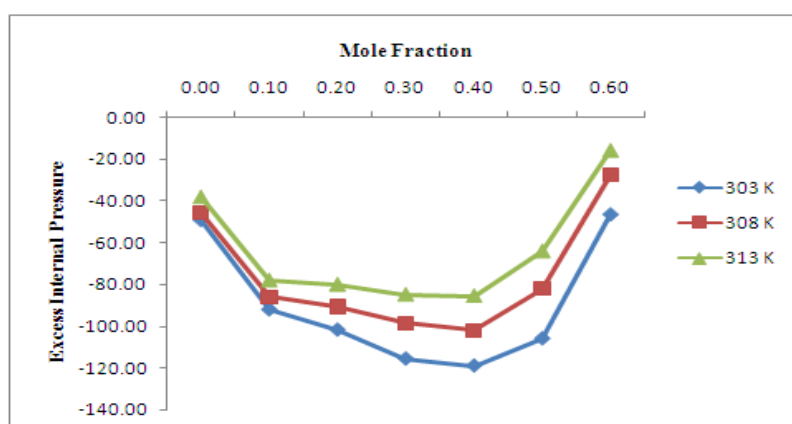


Fig.-16

Fig.(12-16): Variation of Excess Adiabatic Compressibility, Excess Free Length, Excess Free Volume, Excess Internal Pressure Versus Mole fraction of (Methyl Benzoate + Cyclohexane + Butanol) at 303,308 and 313K.

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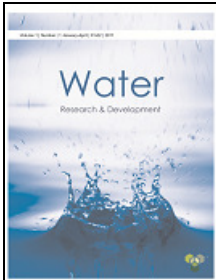
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