



Ultrasonic Study of Binary Mixtures of p-xylene with Ethylbenzene as Common Solvent

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Abstract: The experimental density and speed of ultrasound measurements have been measured for pure Ethylbenzene (ETB), p-xylene and their binary mixtures over the whole miscibility range at different temperatures 293, 298, 303, 308 and 313 K. These parameters were used to determine the adiabatic compressibility, intermolecular free length, acoustic impedance and their excess values. The variation of these parameters with composition of mixture indicates the nature and extent of interaction between unlike molecules. The non-ideal behavior of the system studied was explained on the basis of the dipole-induced dipole interactions and hydrogen bonding. The binary data of β^E , Z^E and L_f^E were correlated as a function of mole fraction by using the Redlich-Kister equation.

Keywords: Acoustical properties, Molecular interactions, Adiabatic compressibility, Intermolecular free length Binary mixtures

Introduction:

Ultrasound waves are high frequency mechanical waves [14]. Ultrasonic wave propagation affects the physical properties of the medium and hence can furnish information about molecular interactions of the liquid and liquid mixtures. The measured ultrasonic parameters are being extensively useful to study intermolecular processes in liquid systems [21]. The sign and magnitude of the non-linear deviations from ideal values of velocities and adiabatic compressibility's of liquid mixtures with composition are attributed to the difference in molecular size and strength of interaction between unlike molecules.

Studies of thermodynamic properties of binary mixtures are of considerable interest in the fundamental understanding of the nature of interactions between the unlike molecules. In recent years, the theoretical and experimental investigations of excess and deviation functions are taken as interaction parameters to improve the results [1-4]. The thermodynamic properties of a binary mixture such as viscosity and density are important from practical and theoretical points of view to understand liquid theory. Accurate knowledge of thermodynamic properties of organic liquid mixtures has relevance in understanding the molecular interactions between the components of the mixture [14]. Binary liquid mixtures due to their unusual behavior have attracted considerable attention [19]. In chemical process industries materials are normally handled in fluid form and as a consequence, the physical, chemical, and transport properties of fluids assume importance. Thus data on some of the properties associated with the liquids and liquid mixtures like Density and viscosity find extensive application in solution theory and molecular dynamics[9][16][5]. The present work deals with the qualitative and quantitative study on the binary mixture of ethylbenzene (ETB) with p-xylene (PX). The aromatic hydrocarbon ethylbenzene is





non-polar compounds with no measurable dipole moment. Thus it involves weak intermolecular interactions. The choice of this solvent was done because of its opposite nature of polarity and their wide range of applicability [13]. Ethyl benzene has been used as a solvent in paints. Ethyl benzene is important in petrochemical industry in the production of styrene which is in turn used for making polystyrene [6]. Para anisaldehyde and ethyl benzene mixture is used to prepare allyl anisole analog repellent pesticides [15]. P-xylene is polar molecule and is important components in synthetic chemistry (produce latex systems), in medicine and biological processes (fungicides, drugs, flavouring extracts, and antiseptics) and is widely used in preparing industrial solvents. Therefore, a better understanding of the physicochemical properties of mixed solvent System (Ethylbenzene & p-xylene) is necessary for interpretation of data obtained from thermo chemical, electrochemical, biochemical and kinetic studies [9][18]. In view of their industrial importance, the present study reports the experimental values of densities (ρ), viscosities (η) and ultrasonic speeds (u) of pure ETB, PX and their binary mixture over the entire composition range ($0 < X_1 < 1$) and temp. 293, 298, 303, 308 and 313K. The above experimental data were used to evaluate the excess intermolecular free length (L^E), excess adiabatic compressibility (β^E) and excess acoustic impedance (Z^E) at each temperature. The excess values were correlated using the Redlich-Kister polynomial equation (6) to obtain their coefficients and standard deviations [9]. The study of molecular interactions in the liquid mixture is therefore important in elucidation of the structural properties of the molecules.

Experimental:

Ethylbenzene (S.D. Fine chem., Pvt. Ltd.) were distilled at atmospheric pressure. Ethylbenzene was dried over phosphorus pentoxide for several days, distilled, stored over 4 Å molecular sieve and used immediately [7]. P-xylene (S.D Fine Chem. Ltd. India) also of A.R grade 99.5% was further purified by the method given in the literature [11]. All the chemicals were stored over 0.4 nm molecular sieves to remove the traces of water, if any, and degassed just before use.

In the binary liquid mixture of ethylbenzene & p-xylene, the various concentrations of the binary liquid mixture were prepared in terms of mole fraction. The mole fractions of the first and second component (X_1 and X_2) were varied from 0 to 1. Purities of these chemicals were checked by density determination at the temp. range 293-313 K which showed an accuracy of 0.0001 gm cm⁻³ as compared to reported values. The density, viscosity and velocity were measured as a function of composition of binary liquid mixture at the temp. range 293- 313 K. The density of sample was measured using digital densitometer (Rudolph) with an accuracy of 0.0001. An Ostwald's viscometer was used for the viscosity measurements. An ultrasonic interferometer having the frequency 2 MHz was used for ultrasonic velocity measurements. An electronically operated constant temperature bath was used to circulate water through measuring cell made up of steel containing experimental solution at the temp. range 293- 313 K temperature. An ultrasonic interferometer (Model: F81) working at a frequency 3MHz with an overall accuracy of ± 2 ms⁻¹ has been used for velocity measurement. An electronically digital constant temperature bath has been used to circulate water through the double walled measuring cell made





up of steel containing experimental mixtures at the desired temperature. The accuracy in the temperature measurement is $\pm 0.1\text{K}$. Reliability of the experimental data and the purity of the solvents were ascertained by calculating their densities, ultrasonic speeds and viscosities at different temperatures with the values reported as shown in Table 1.

Results and discussion:

The experimental densities (ρ), ultrasonic speeds (U) and viscosities (η) of pure ethylbenzene, p-xylene and their binary mixture are used to calculate excess thermodynamic properties of mixtures which correspond to the difference between the actual property and the property if the system behaves ideally and thus are useful in the study of molecular interactions and arrangements in the mixtures. In particular, they reflect the interactions that take place between solute-solute, solute-solvent and solvent-solvent species. The effects which are expected to operate between the component molecules under study are (i) structural effect which is due to the differences in shape and size of the component molecules (ii) reorientation effect between component molecules and (iii) energetic effect, i.e., molecular interaction that can be weakened or destroyed or established during the mixing process [22]. Thus, in the present study various acoustical parameters were calculated from measured data by using following equations

$$\text{Adiabatic compressibility } (\beta) = 1 / v^2 \rho \quad \dots\dots (1)$$

$$\text{Intermolecular Free length } (L_f) = K \sqrt{\beta} \quad \dots\dots (2)$$

Where K is temperature dependant constant, values of K varies from 620×10^{-6} to 642×10^{-6} at 293-313 k.

$$\text{Acoustic impedance } (Z) = U\rho \quad \dots\dots (3)$$

$$\text{Molar Volume } (V_m) = M/\rho \quad \dots (4)$$

Where M is mean molecular weight. It is calculated as

$$M = X_1 M_1 + X_2 M_2$$

X_1 and X_2 are mole fractions and M_1 , M_2 are molecular weights of constituent components of binary liquid mixtures.

The values of excess intermolecular free length (L_f^E), excess acoustic impedance, (Z^E) and excess adiabatic compressibility (β^E) were calculated with the help of the following standard relations:

$$Y^E = Y_{\text{exp.}} - (X_1 Y_1 + X_2 Y_2) \quad \dots\dots\dots (5)$$

Where, $Y_{\text{exp.}}$ = experimental values of mixtures

Y_1 & Y_2 = values of parameters for liquids 1 and 2 respectively.

X_1 & X_2 = mole fractions of liquid 1 (PX) and liquid 2 (ETB).

The values of excess adiabatic compressibility (β^E), free length (L_f^E) and acoustic impedance (Z^E) for the mixture have been least-squares fitted to Redlich-Kister type polynomial equation given in literature [10] by taking the limits $n=0$ to i .

$$F(X) = X_1 \cdot (1-X_1) \sum A_i \cdot (1-2 \cdot X_1)^i \quad \dots\dots (6)$$

Where $F(x)$ refers to β^E , Z^E or L_f^E . The coefficient A_i is the polynomial coefficient tabulated by using the least square method computed by the MAPLE software has been used. The values of the standard deviation (σ) were obtained from the expression

$$\sigma = \{ \sum (F(x)_{\text{exp}} - F(x)_{\text{cal}})^2 / (k/n) \}^{1/2} \quad \dots\dots (7)$$





Where k is the number of experimental points excluding the end points and n is order of polynomial equation.

The values of $F(X)$ cal are obtained from Eq. (6) by using the best fit values of A_i coefficients. The coefficients A_0 , A_1 , A_2 , A_3 and A_4 along with standard deviations σ of fit for all the mixtures are listed in Table 2 and it has been observed that standard deviations are very low.

Fig.1. shows the variation of ultrasonic velocity with the entire miscibility range of ethylbenzene in p-xylene at different temperatures. Obtained values of ultrasonic velocity for pure ETB and PX are in agreement with the reported values. In all the measurement temperatures and concentrations, the plot comprises two linear portions with different slopes. These slopes show a break at the mole fraction of 0.6 of ETB/PX. This nonlinear behavior of velocity with concentration indicates the deviation from ideal mixing behavior, similar to the previous reports of the water/ether system [23, 24]. Even though the ultrasonic velocity decreases with 'x', there are no maxima or minima. The linear behavior with decrease in velocity with concentration indicates the interactions between unlike molecules [25- 27]. Also it is noted that the velocity of ultrasonic wave decreases with temperature. At 293 K, the velocity of ETB is 1328.27 m/s and it is decreased to 1252 m/s when the temperature is increased to 313 K. As the temperature is increased, available thermal energy facilitates the breaking of the bonds between the associated molecules into their monomers. Moreover, increase of thermal energy weakens the molecular forces which tend to decrease the ultrasonic velocity as expected [8, 28].

The experimental results of density measurements of binary mixtures of ETB with p-xylene over the whole composition range expressed in mole fraction x_1 of p-xylene ($0 < x < 1$) at different temperatures is shown in Fig.2. decrease in densities with increase in temperature is seen at higher values of mole fractions. However decrease in density gradually decreases on increasing the mole fractions. This indicates the dominant character of ethylbenzene over p-xylene, that binds the molecules together and therefore the density variation with temperature is less at higher values of ' x_1 '.

Fig.3. illustrates the variation of the adiabatic compressibility for various mole fraction of ETB in forming solution with p-xylene. The nature of variation indicates the possible interaction among molecules. The adiabatic compressibility gradually increases with increasing the concentration of ETB in the mixture. The reverse behavior of adiabatic compressibility occurs because of structural changes present in the mixture that brings the molecules to a closer packing. Also, it reveals negative deviation with temperature, when compared with ultrasonic velocity. These deviations can be explained in terms of the dispersion forces that increase the intermolecular path lengths as described by Jacobson [29, 30]. Decrease of the intermolecular path lengths on the other hand leads to a positive deviation in sound velocity and negative deviation in the compressibility. In our observation it seems that the negative deviation in compressibility at higher mole fractions of PX is due to the presence of dipole-induced dipole interactions. ETB being nonpolar molecules induces dipole moment in p-xylene, causing dipole-induced dipole interactions to operate. These findings are in accordance with the previously reported results of Yadava and Yadav [31].





Intermolecular free length is an important parameter that has association with adiabatic compressibility. Fig.4. shows the variation of free length with mole fractions. It is clear that the intermolecular free length shows a similar behaviour as reflected by ' β '. The increased compressibility brings the molecules to a closer packing resulting an increase in intermolecular free length. Moreover, free length is a predominant factor in determining the variation of ultrasonic velocity in solutions. The inter dependence of ' L_f ' and ' U ' has been evolved from a model for sound propagation proposed by Eyring and Kincaid [32]. According to the proposed theory, the increase in the value of ' β ' and ' L_f ' with decrease in ultrasonic velocity further strengthens the process of complex formation between the solute molecules due to which structural arrangement is considerably altered. In the present study also, there is a possibility of complex formation due to interactions as revealed by the nonlinear variation of ultrasonic velocity and their related parameters due to strong interaction of forces [33, 34].

When an acoustic wave travels in a medium, there is a variation of pressure from particle to particle. When a plane ultrasonic wave is set up in a liquid, then the pressure and hence density and refractive index of the liquid show a periodic variation with distance from the source along the direction of propagation. If there is stationary ultrasonic pattern in the liquid, the density will be greater in the nodal plane. The acoustic impedance is the parameter related to the elastic properties of the medium. Therefore, it is important to examine specific acoustic impedance in relation to concentration and temperature. Fig.5.shows the variation of acoustic impedance with mole fraction and temperature. Acoustic impedance exhibits a nonlinear variation with concentration at $x = 0.4-0.6$. The nonlinear behaviour further supports the possibility of molecular interactions due to the H bonding [35, 36].

In addition to above observations, the nature and strength of molecular associations in the present system (ETB+PX) have been studied from excess acoustic parameters such as excess adiabatic compressibility (β^E), excess acoustic impedance (Z^E) and excess free length (L_f^E) as shown in Table 2. It is expected that the dispersion forces should make positive contributions to excess values while dipole-dipole, dipole-induced dipole, charge-transfer interaction between unlike components should make negative contributions [33].

The variation of adiabatic compressibility (β^E) and excess free length (L_f^E) with mole fraction of PX in the binary solution (ETB+PX) is represented in Fig. 7 & Fig. 8 respectively. The value of excess adiabatic compressibility is found to be varying over the mole fraction of 0-1 ETB indicating the presence of interactions between the molecules. The change from negative to positive L_f^E values indicating greater be the dissociation between the components and may be quantitatively interpreted in terms of detachment of unlike molecules leading to increase in compressibility and volume. The β^E and L_f^E minima occur at the same concentrations further strengthens the occurrence of molecular associations. The positive contribution is mainly to the dispersion forces and this type of variation of L_f^E has also been reported for binary mixtures [8, 40, 41]. This may be qualitatively interpreted in terms of closer approach of unlike molecules leading to reductions in compressibility and volume [31, 33]. This type of interactions for the binary mixture DMF-toluene [37], DMF-water [38] has been already reported. The pronounced compressibility minimum at $x = 0.35$ may be





attributed to strong molecular interaction which eventually results in complexation [38]. The excess properties are found to decrease with increasing temperature which is in agreement with the previously reported result [39].

The excess acoustic impedance variation is shown in Fig.6. Over the mole fraction range 0–1 of the investigated system, the interaction between unlike molecules are quite obvious. In the higher compositions $x = 0.2-0.8$, excess impedance value is in the negative as well as positive side and this indicates the association or dissociation reaction between the molecules at different temp. as revealed in the excess adiabatic compressibility and free length variations [20]. The change from negative to positive values in Z^E over the entire mole fraction of the system investigated again support our view that the molecular interactions between unlike molecules are quite obvious [12]. Negative excess values are due to closely packed molecules which accounts for the existence of strong molecular interactions, whereas positive excess values reflect weak interactions between unlike molecules [17].

Conclusion:

The concentration dependencies of ultrasonic velocity and density of ethylbenzene–p-xylene binary system have been measured at different temperatures. The nonlinear variation of the related parameters such as ultrasonic velocity, density, adiabatic compressibility, intermolecular free length and acoustic impedance were elaborated to understand the molecular interactions that leads to the process of complex formation between the solute molecules through dipole-induced dipole interactions. The positive and negative variations of the excess values with concentration and temperature of the same acoustic parameters supported the presence of interaction between unlike molecules.

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Symbols and abbreviations:

| | |
|-------------|----------------------------------|
| ρ - | Density |
| η - | Viscosity |
| U- | Ultrasonic Velocity |
| β - | Adiabatic Compressibility |
| L_f - | Intermolecular free length |
| Z - | Acoustic Impedance |
| β^E - | Excess Adiabatic compressibility |





| | |
|------------|-----------------------------------|
| L_f^E - | Excess Intermolecular Free Length |
| Z^E - | Excess acoustic Impedance |
| σ - | Standard deviation |
| X_1 - | Mole fraction for p-xylene |
| X_2 - | Mole fractions for ethylbenzene |
| M_1 - | Molecular weight of p-xylene |
| M_2 - | Molecular weight of ethylbenzene |
| K- | Kelvin |
| ETB- | Ethylbenzene |
| PX- | P-xylene |

Table 1: Values of density (ρ), viscosity (η), ultrasonic velocity (U), adiabatic compressibility (β), free length (L_f), acoustic impedance (Z) of pure liquids at 313 K.

| Component | Temp. T/K | ρ Kg m ³ | η x10 ³ Nsm ² | U ms ⁻¹ | β x 10 ¹⁰ m ² N ⁻¹ | L_f x 10 ¹⁰ m | Z x10 ⁻⁶ Kg m ⁻² s ⁻¹ |
|--------------|--------------|-----------------------------|--|-----------------------|---|-------------------------------|---|
| Ethylbenzene | 293K | 867.30 | 0.0670 | 1328.27 | 6.5352 | 158.50 | 1.1520 |
| | 298K | 862.80 | 0.0638 | 1289.73 | 6.9677 | 164.98 | 1.1128 |
| | 303K | 858.20 | 0.0623 | 1271.87 | 7.2032 | 169.35 | 1.0915 |
| | 308K | 853.50 | 0.0586 | 1265.60 | 7.3148 | 172.01 | 1.0802 |
| | 313K | 848.60 | 0.0569 | 1252.00 | 7.5178 | 176.03 | 1.0624 |
| p-xylene | 293K | 861.1 | 0.6781 | 1316.93 | 6.696 | 160.43 | 1.134 |
| | 298K | 856.6 | 0.6441 | 1298.8 | 6.9205 | 164.42 | 1.1125 |
| | 303K | 852.2 | 0.6154 | 1281.73 | 7.1427 | 168.64 | 1.0923 |
| | 308K | 847.5 | 0.5351 | 992.67 | 11.9744 | 220.08 | 0.8413 |
| | 313K | 842.6 | 0.5074 | 969.2 | 12.6343 | 228.2 | 0.8166 |

Table 2. Values of A_i parameters of Eq. (6) along with standard errors, σ (Y^E) for the binary liquid mixtures at different temperatures.

| Property | T/K | A0 | A1 | A2 | A3 | A4 | σ (YE) |
|---|------|---|---------|---------|---------|----------|---------------|
| ETB+PX | | | | | | | |
| $10^{10}L_f^E$ /(m) | 293K | 0.8626 | -0.3745 | 0.6047 | -0.5676 | -0.0041 | 0.007900 |
| | | $10^{-6}Z^E$ /(Kgm ⁻² S ⁻¹) | -0.0060 | 0.0027 | -0.0030 | 0.0038 | -0.0021 |
| $10^{10}\beta^E$ /(m ² N ⁻¹) | 298K | 0.0629 | -0.0318 | 0.0502 | -0.0471 | -0.0001 | 0.000659 |
| | | $10^{10}L_f^E$ /(m) | -0.3527 | 0.3335 | -0.4690 | 0.1612 | 0.3122 |
| $10^{-6}Z^E$ /(Kgm ⁻² S ⁻¹) | 298K | 0.0030 | -0.0029 | 0.0045 | -0.0023 | -0.0033 | 0.000052 |
| | | $10^{10}\beta^E$ /(m ² N ⁻¹) | -0.0310 | 0.0280 | -0.0384 | 0.0136 | 0.0251 |
| $10^{10}L_f^E$ /(m) | 303K | 1.2391 | -1.2098 | 1.8661 | -0.2054 | -1.3955 | 0.020833 |
| | | $10^{-6}Z^E$ /(Kgm ⁻² S ⁻¹) | -0.0091 | 0.0099 | -0.0144 | 0.0004 | 0.0105 |
| $10^{10}\beta^E$ /(m ² N ⁻¹) | 303K | 0.1039 | -0.1025 | 0.1570 | -0.0176 | -0.1164 | 0.001772 |
| | | $10^{10}L_f^E$ /(m) | 91.8226 | 72.7004 | 58.9980 | 189.1650 | 195.6206 |
| $10^{-6}Z^E$ /(Kgm ⁻² S ⁻¹) | 308K | 0.4432 | -0.3634 | 0.3200 | -0.8253 | 0.7941 | 0.002747 |
| | | $10^{10}\beta^E$ /(m ² N ⁻¹) | 11.7018 | 6.1920 | -5.6956 | 19.1882 | -20.4638 |





| | | | | | | | |
|--|------|---------|---------|---------|----------|---------|----------|
| $10^{10}L_f^E$ (m) | 313K | 49.2435 | 68.5620 | 65.4416 | -33.2330 | 87.2299 | 1.508423 |
| $10^{-6}Z^E$ (Kgm ⁻² S ⁻¹) | | 0.1800 | -0.2942 | 0.2441 | 0.1900 | -0.3930 | 0.006755 |
| $10^{10}\beta^E$ (m ² N ⁻¹) | | -8.3190 | 6.1509 | -7.3418 | -2.8773 | 8.6067 | 0.150467 |

Fig. 1. Ultrasound velocity (U) with mole fraction (x₁) in binary mixture of ETB. mixture of ETB and PX at different temperatures.

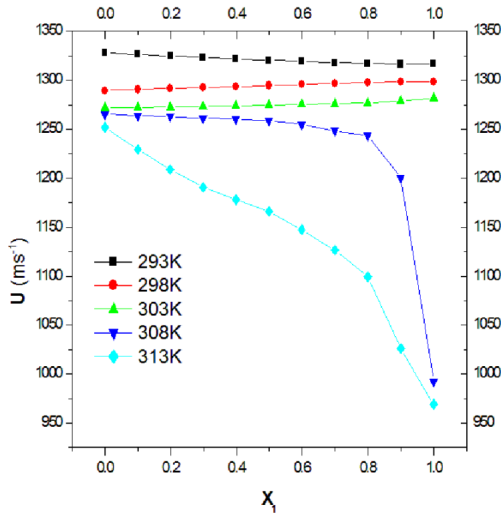


Fig. 2. Density (ρ) with mole fraction (x₁) in binary mixture of ETB and PX at different temperatures.

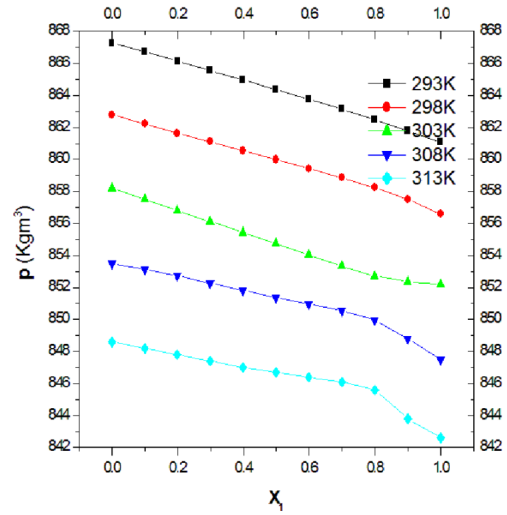


Fig. 3. Adiabatic compressibility (β) with mole fraction (x₁) in binary mixture of ETB. (x₁) in binary and PX at different temperatures.

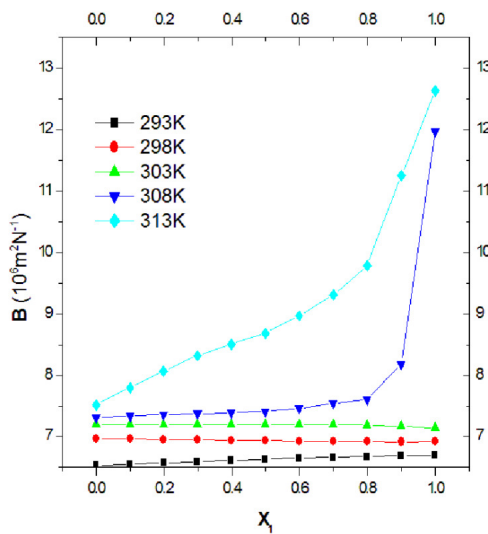


Fig. 4. Intermolecular free length (L_f) with mole fraction (x₁) in binary mixture of ETB and PX at different temperatures.

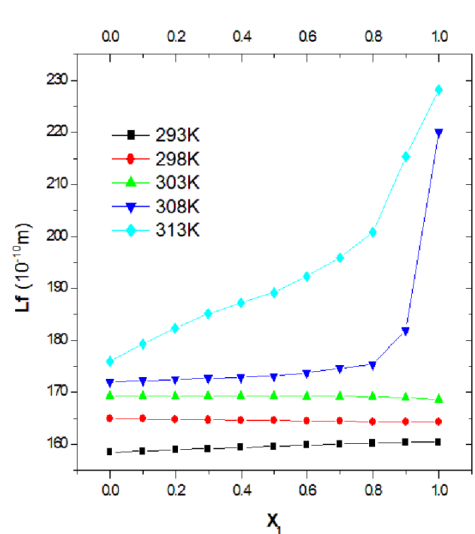


Fig.5. Acoustic impedance (Z) with mole fraction (x₁) in binary mixture of ETB.

Fig.6. Excess acoustic impedance (Z^E) with mole fraction (x₁) in binary





and PX at different temperatures.

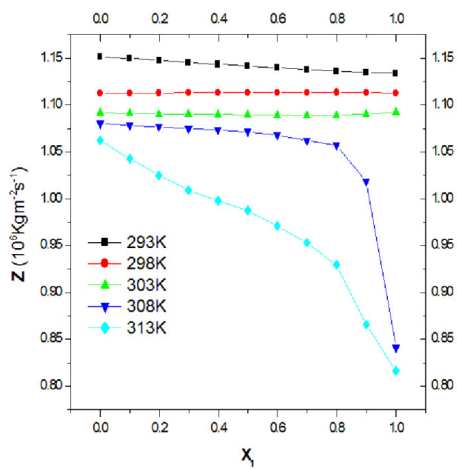


Fig. 7. Excess adiabatic compressibility (β) with mole fraction (x_1) in binary mixture of ETB and PX at different temperatures.

mixture of ETB and PX at different temperatures.

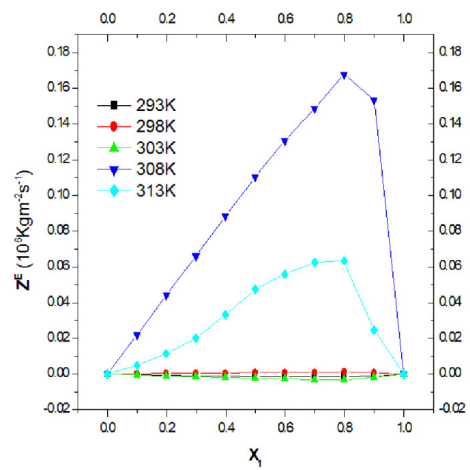


Fig. 8. Excess intermolecular free length (L) with mole fraction (x_1) in binary mixture of ETB and PX at different temp.

