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Research Article

INTERNAL PRESSURE, ULTRASONIC VELOCITY AND VISCOSITY OF BINARY LIQUID MIXTURE CONTAINING (1, 4-DIOXANE + 1-OCTANOL) AT DIFFERENT TEMPERATURE AND UNDER ATMOSPHERIC PRESSURE

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Abstract

Experimental measurements of density, viscosity, and sound speed at 298.15 K and atmospheric pressure are reported for the binary liquid mixture of 1,4-dioxane (1) + 1-octanol (2). Based on these experimental data, various thermodynamic excess and deviation properties were calculated and fitted using the Redlich-Kister polynomial equation. The variation of these properties with composition has been analyzed in terms of molecular interactions between the mixture components and structural effects. From the experimental data, excess molar volume (V^E), excess adiabatic compressibility (β_{ad}^E), excess viscosity (η^E), excess intermolecular free length (L_f^E), and excess internal pressure (P_i^E) were determined. These properties were found to be negative at all measured temperatures. The results have been interpreted in terms of the nature of molecular interactions between the components of the mixture.

Keywords: Redlich-Kister polynomials, Thermodynamic properties, Binary mixtures; Refractive index, Sound of sound, binary liquid mixtures, internal pressure, molar volume, intermolecular free length.

INTRODUCTION

This study is part of a systematic research program focused on the measurement and mathematical modelling of various thermodynamic properties of binary and ternary liquid mixtures containing important compounds [1–5]. The investigation of thermodynamic properties in binary mixtures of alkanols with cyclic ethers is significant from both practical and theoretical perspectives. In particular, knowledge of excess adiabatic compressibility provides insights into molecular orientation and helps assess the extent of intermolecular interactions between the components of a liquid mixture[6]. In this work, ultrasonic velocity (u), density (ρ), and viscosity (η) were measured for the binary mixture of 1,4-dioxane (1) + 1-octanol (2) over the entire composition range at three different temperatures T = (298.15, 303.15 and 305.15) K and at the atmospheric pressure. From the experimental data, excess molar volume (V^E), excess adiabatic compressibility (β_{ad}^E), excess viscosity (η^E), excess intermolecular free length (L_f^E) and excess internal pressure (P_i^E) were calculated across all compositions and temperatures. These excess properties were correlated using the Redlich-Kister polynomial equation. The observed deviations and excess functions were analyzed in terms of intermolecular interactions present in the mixture. This work extends our ongoing research on the thermodynamic, transport, and optical properties of liquid-liquid mixtures[7–11]. The studied thermodynamic properties were fitted using Redlich-Kister polynomials, one of the most widely used mathematical representations of excess thermodynamic properties in binary liquid mixtures. Additionally, various theoretical and semi-empirical models were employed to predict the sound speeds of the investigated liquid mixtures.

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The predictive accuracy of each model was evaluated using the mean absolute percentage deviation between experimental and calculated data.

Experimental Procedure

Chemicals

1,4-Dioxane and 1-octanol were obtained from Merck Chem. Ltd India with mass purity >99%. Both liquids were used without further purification. The experimental values of ultrasonic velocity (u), density (ρ) and viscosity (η) of pure liquids at temperature 298.15K were compared with value available in the literature and are listed in table-1, were leads to a satisfactory agreement.

Apparatus and Procedure

Both two mixtures of 1,4-Dioxane and 1-octanol have been prepared by mixing known masses of the pure components. The mass is performed by using a digital electronic balance (Citizen Scale (I) PVT. LTD. Mumbai, India.),with a resolution of 10^{-5}g. The experimental uncertainty in mole fractions did not exceed ± 0.0005. All the solutions were prepared by mass ratios and stored in the air-tight stopper measuring flasks.

Measurements

Density

The densities of the pure liquid and its mixtures were measured using a 25-mL specific gravity bottle by the relative measurement method, with an accuracy of ±0.01 kg·m^{-3}. The specific gravity bottle containing the experimental mixture was immersed in a temperature-controlled water bath

Table 1. Comparison of Experimental and Literature density (ρ), sound velocity (u) and viscosity (η) of pure Components with Available Literature Values at T = 298.15K and 3MHz

Compound	(ρ) / (g.cm^{-3})		u / (m.s^{-1})		η / (mPa s)	
	expti.	lit.	expti.	lit.	expti.	lit.
1,4-Dioxane	1.0108	1.0229	1348	1344	1.0303	1.0690
		1.0286		1345		1.1944
		1.02763		1341		1.1960
1-Octanol	0.8242	0.8187	1327	1330	7.8512	7.6630
		0.8220		1346		7.661

(MSI Goyal Scientific, Meerut, U.P., India) to maintain a stable measurement environment. Operating in the temperature range of -10°C to 85°C with an accuracy of ±0.1°C.

Sound velocity

The ultrasonic velocity was measured using a multi-frequency ultrasonic interferometer (Model F-80D, Mittal Enterprise, New Delhi, India) operating at 3 MHz. The instrument was calibrated using water and benzene. The measurement of sound velocity through the medium was based on the precise determination of the wavelength of ultrasonic waves of known frequency, generated by a quartz crystal in the measuring cell. The interferometer cell was filled with the test liquid, and a water bath was used to circulate water around the measuring cell to maintain temperature stability. The uncertainty in the measurements was estimated to be ±0.1 m·s^{-1}. The measured values of ultrasonic velocities for pure 1,4-dioxane and 1-octanol were found to be in good agreement with the corresponding literature values

Viscosity The viscosity of pure liquids and liquid mixtures was measured using an Ostwald viscometer with a capacity of approximately 15 mL, a capillary length of about 90 mm, and an internal diameter of 0.5 mm. The viscometer was calibrated using doubly distilled water and benzene. The flow times of pure liquids and liquid mixtures were recorded five times to ensure accuracy. The efflux time was measured using an electronic stopwatch (Racer) with a time resolution of ±0.015 s, and the average of at least five readings was taken. A glass stopper was placed at the opening of the viscometer to prevent evaporation losses during measurements. The uncertainty in viscosity measurements was estimated to be ±0.005 × 10^{-3} mPa·s. The measured viscosity values for pure 1,4-dioxane and 1-octanol were found to be in good agreement with corresponding literature values.

Theoretical

The ultrasonic velocity (u), density (ρ), and viscosity (η) of pure liquids and liquid mixtures at various concentrations were measured at 298.15 K, 303.15 K, and 305.15 K at a frequency of 3 MHz. Thermodynamic and acoustical parameters, including molar volume (V), adiabatic compressibility (β_{ad}), intermolecular free length (L_f), and internal pressure (P_i), were determined using the experimentally observed values of sound velocity, density, and viscosity, applying standard relations as given below.

The molar volume (V) of binary liquid mixtures at a given mole fraction is calculated using the following equation:

$$V = \frac{(X_1M_1 + X_2M_2)}{\rho} \quad (1)$$

Where:

- X_1 and X_2 are the mole fractions of components 1 and 2,
- M_1 and M_2 are the molar masses of components 1 and 2, respectively,
- ρ is the density of the mixture.

The adiabatic compressibility (β_{ad}) is determined using the experimentally measured ultrasonic velocity (u) and density (ρ) with the following relation:

$$\beta_{ad} = \frac{1}{u^2\rho} \quad (2)$$

where:

- β_{ad} is the adiabatic compressibility (in mPas),
- u is the ultrasonic velocity (in m/s),
- ρ is the density of the liquid or mixture (in g.cm^{-3}).

The intermolecular free length (L_f) is calculated using the standard expression:

$$L_f = K\,\beta_{ad}^{1/2} \quad (3)$$

where:

- L_f is the intermolecular free length (in meters),
- K is the temperature-dependent Jacobson's constant,
- β_{ad} is the adiabatic compressibility (in mPa.s).

This equation is derived based on the assumption that sound waves propagate through the liquid medium by intermolecular free paths, and it helps in understanding molecular interactions in binary liquid mixtures.

Suryanarayana and Kuppuswami[12-13] suggested a method for evaluating internal pressure (Pi) using the knowledge of ultrasonic velocity (u), density (ρ), and viscosity (η). The proposed relation is expressed as:

$$p_i = bRT\left(\frac{k\eta}{u}\right)^{\frac{1}{2}}\frac{\rho^{\frac{2}{3}}}{M_{eff}^{\frac{7}{6}}} \quad (4)$$

where:

- Pi ,is the internal pressure,
- b, is a constant, which is assumed to be 2 for all liquids and solution.
- ρ ,is the density of the liquid,
- u, is the ultrasonic velocity,
- η ,is the viscosity of the liquid.

Table 2. Values of excess viscosity (η^E),molar volume (V^E), adiabatic compressibility ($\beta_{ad}{}^E$), internal pressure ($P_i{}^E$) and free length ($L_f{}^E$)for Various 1,4-Dioxane Mole Fractions x_1 of the Binary Mixture (1,4-Dioxane (1) + 1-Octanol (2)) at Temperatures T = (298.15, 303.15 and 305.15) K

Mole fraction (5_1)	Density (ρ) g.cm^{-3}	Sound velocity (u) m/s	Viscosity (η) m.Pa.s	Excess Viscosity (η□) m.Pa.	Excess Molar volume(V^E) (cm^3mol^{-1})	Excess adiabatic compressibility $\beta_{ad}{}^E\times10^{-7}$	Excess internal pressure $p_i{}^E\times10^4$	Excess free length $L_f{}^E\times10^{-10}$
At 298.15 K								
0.00000	0.8312	1330.0	7.9215	0.0000	0.00000	0.00000	0.00000	0.00000
0.09780	0.8582	1332.0	6.2565	-0.1825	-0.24561	-0.27232	-0.15262	-0.10256
0.20653	0.8763	1334.0	5.2685	-0.2854	-0.40125	-0.44362	-0.28325	-0.21561
0.29810	0.8952	1336.0	4.4523	-0.4128	-0.51234	-0.53245	-0.36328	-0.32125
0.40275	0.9152	1339.0	35698	-0.5321	-0.59652	0.60251	-0.52456	-0.55851
0.49229	0.9315	1341.0	2.6525	-0.6215	-0.61245	-0.64258	-0.62354	-0.60851
0.60068	0.9456	1346.0	2.6325	-0.6859	-0.58632	-0.53236	-0.71235	-0.85451
0.69888	0.9623	1351.0	1.8956	-0.4525	-0.51456	-0.42582	-0.56540	-0.72542
0.79610	0.9836	1356.0	1.4685	-0.3287	-0.40562	-0.35652	-0.64892	-0.59875
0.89749	0.9946	1360.0	1.0525	-0.2003	0.21564	-0.22481	-0.15684	-0.20568
1.00000	1.0215	1367.0	1.0652	0.0000	0.00000	0.00000	0.00000	0.00000
At 303.15 K								
0.00000	0.8242	1327.0	7.8512	0.0000	0.00000	0.00000	0.00000	0.00000
0.09780	0.8284	1329.0	5.1466	-0.2012	-1.15434	-0.08590	-0.08222	-0.12088
0.20653	0.8370	1330.0	4.6513	-0.3633	-1.80656	-0.16204	-0.05674	-0.22926
0.29810	0.8529	1332.0	3.2294	-0.4856	-1.97558	-0.14911	-0.10819	-0.22024
0.40275	0.8595	1334.0	2.5625	-0.5567	-2.29511	-0.22970	-0.11758	-0.33295
0.49229	0.8852	1336.0	2.3806	-0.6540	-2.54289	-0.15022	-0.08558	-0.23132
0.60068	0.9030	1338.0	1.8916	-0.5241	-2.73983	-0.16391	-0.08626	-0.25166
0.69888	0.9266	1339.0	1.4950	-0.4018	-1.27332	-0.13934	-0.08901	-0.21745
0.79610	0.9564	1341.0	1.3490	-0.3126	-1.11434	-0.07472	-0.05664	-0.12426
0.89749	0.9859	1345.0	1.1845	-0.2121	-0.68611	- 0.01397	-0.02768	-0.03111
1.00000	1.0108	1348.0	1.0303	0.0000	0.00000	0.00000	0.00000	0.00000
At 305.15 K								
0.00000	0.8153	1324.0	7.1025	0.0000	0.00000	0.00000	0.00000	0.00000
0.09780	0.8326	1328.0	5.0123	-0.2254	-0.28451	-0.23541	-0.10253	-0.18562
0.20653	0.8523	1334.0	4.2513	-0.3689	-0.45682	-0.42362	-0.18962	-0.24562
0.29810	0.8845	1338.0	3.1202	-0.4525	-0.59452	-0.55632	-0.29632	-0.33652
0.40275	0.9021	1340.0	2.2256	-0.5485	-0.65785	-0.60258	-0.35245	-0.65231
0.49229	0.9263	1342.0	2.1251	-0.7523	-0.69892	-0.65284	-0.56242	-0.86542
0.60068	0.9512	1345.0	1.5641	-0.6218	-0.58452	-0.56452	-0.66547	-0.95241
0.69888	0.9725	1348.0	1.3025	-0.4287	-0.35241	-0.41258	-0.45632	-0.70125
0.79610	0.9901	1350.0	1.2351	-0.3258	-0.20145	-0.31745	-0.28564	0.54282
0.89749	1.0095	1352.0	1.625	-0.312	-0.18563	-0.20513	-0.12325	-0.32542
1.00000	1.0102	1355.0	1.1032	0.0000	0.00000	0.00000	0.00000	0.00000

- K,is a constant, independent of temperature and its value is 4.28×10^9 for all liquids,
- R ,is universal gas constant and
- T, is absolute temperature.

This equation provides insights into the molecular interactions and cohesive forces present in the liquid mixture. Let me know if you need further elaboration!

The excess values of ultrasonic-related parameters, such as excess adiabatic compressibility ($\beta_{ad}{}^E$), excess intermolecular free length ($L_f{}^E$), and excess internal pressure ($p_i{}^E$), can be calculated using the general relation:

$$A^E = A_{exp.} - (X_1 A_1 + X_2 A_2) \qquad (5)$$

where:

- A^E represents the excess property (e.g., excess adiabatic compressibility, excess intermolecular free length, excess internal pressure).
- $A_{mix.}$ is the experimentally determined value of the property for the binary mixture.
- X_1 and X_2 are the mole fractions of components 1 and 2, respectively.
- $A_1$and A_2 are the values of the property for the pure components.

This equation helps in understanding molecular interactions, structural effects, and deviations from ideal mixing behavior in binary liquid mixtures.

RESULT AND DISCUSSION

The experimentally determined values of ultrasonic velocity (u), density (ρ), and viscosity (η) for the binary mixture 1,4-Dioxane (1) + 1-Octanol (2) across the entire composition range at temperatures T= (298.15, 303.15, and 305.15 K) and a frequency of 3 MHz are presented in Table-2. Additionally, the derived excess properties excess molar volume (V^E), excess adiabatic compressibility ($\beta_{ad}{}^E$), excess viscosity (η^E), excess intermolecular free length ($L_f{}^E$), and excess internal pressure ($P_i{}^E$)are also reported in Table-2. These excess thermodynamic and acoustic parameters help analyze the nature of molecular interactions, structural effects, and deviations from ideal behavior in the binary system. The values of excess molar

volume (V^E), excess adiabatic compressibility ($\beta_{ad}{}^E$), excess viscosity (η^E), excess intermolecular free length ($L_f{}^E$), and excess internal pressure (Pi^E) are plotted against the mole fraction of 1,4-dioxane at different temperatures, as shown in Figures 1–5.

Observations:

- **Nonlinear Variation:** The ultrasonic velocity (u), density (ρ), and viscosity (η), along with the excess parameters (V^E, $\beta_{ad}{}^E$, η^E, $L_f{}^E$, and $P_i{}^E$), exhibit a nonlinear increasing trend with an increase in molar concentration.
- **Intermolecular Interactions:** The observed variations suggest complex formation and weak intermolecular associations, likely due to **hydrogen bond formation** between 1,4-dioxane and 1-octanol.
- **Structural Effects:** The behavior can be attributed to **structural rearrangements** in the mixture, which influence the observed excess parameters.

The values of **excess molar volume (V^E)** at each temperature (298.15 K, 303.15 K, and 305.15 K) are recorded in Table-2. The experimental values of V^E at each studied temperature, obtained from Equation-5, have been correlated using a Redlich-Kister polynomial equation**,** a well-established model for fitting excess properties in binary mixtures [15].

$$Y^E = x_1 x_2 \sum_{k=1} A_k (x_1 - x_2)^k \qquad (6)$$

Where Y^E represent an excess or deviation property, subscripts 1 and 2 represent the pure components, k is the number of fitted parameter and A_k represents the coefficients. Adjustable parameters of A_k were evaluated by least-squares method. The excess molar volume (V^E) values for the 1,4-Dioxane (1) + 1-Octanol (2) binary mixture are negative over the entire composition range at the studied temperatures. This trend is depicted in Figure-1 and suggests that contractive effects dominate over expansive effects in the mixture.

Key Observations:

- **Negative V^E Values:** The negative sign of V^E indicates that volume contraction occurs upon mixing, which can be attributed to specific molecular interactions between the components[16-17].
- **Temperature Dependency:** The magnitude of negative V^E increases with temperature**,** suggesting stronger interactions or changes in molecular packing at higher temperatures.
- **Minima at X_1=0.5:** The excess molar volume shows a minimum at mole fraction X_1=0.5, indicating optimal interaction between 1,4-dioxane and 1-octanol at this composition.
- **Hydrogen Bonding Contribution:** The observed contraction in volume can be explained by hydrogen bonding between the oxygen atoms in 1,4-dioxane (which act as hydrogen bond acceptors) and the hydroxyl groups (-OH) in 1-octanol (which act as hydrogen bond donors). This strong interaction leads to closer packing of molecules in the solution.

The excess adiabatic compressibility (β_{ad}^E) for the mixture1,4-Dioxane (1) + 1-Octanol (2) at temperature from (298.15,303.15 and 305.15) K as a function of 1,4-dioxane mole fraction have been reported in table-2.

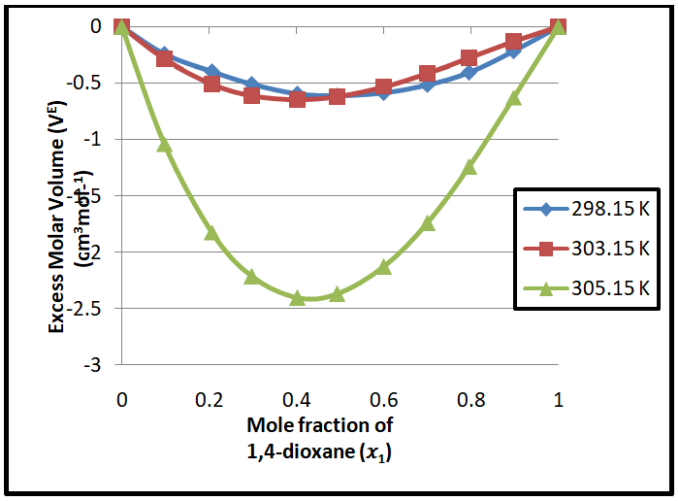

Figure 1. Curves of excess molar volume V^E against the mole fraction of 1, 4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + 1-Octanol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister equation

As depicted in figure-2, for the studied binary system, the excess adiabatic compressibility (β_{ad}^E) values over the entire composition range are negative. As mentioned in the literature[18] the negative values of excess adiabatic compressibility (β_{ad}^E) suggest the presence of the dispersion forces or weak interactions between the component molecules in the mixture. Strong molecular interaction occur through charge transfer, dipole-induced dipole and dipole-dipole interactions, interstitial accommodation, and oriental ordering and all lead to a more compact structure, which makes excess adiabatic compressibility (β_{ad}^E) negative [19-20]. In the present studied binary system the negative excess adiabatic compressibility (β_{ad}^E) values may be indicate clustering of octanol molecules in the presence of 1,4-dioxane.

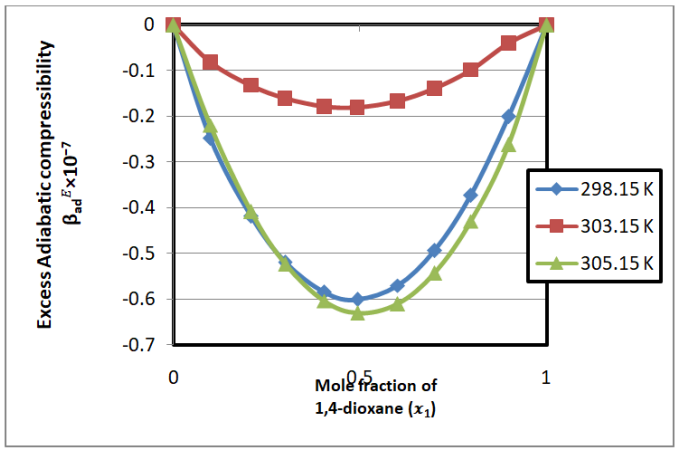

Figure 2.Curves of excess adiabatic compressibility β_{ad}^E against the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) +1- Octanol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister equation.

The viscosities of binary mixture increase linearly with increase in mole fraction of 1,4-dioxane.Viscosity deviation ($\Delta\eta$), is found to be negative for the binary mixture over the entire composition range at all the three temperatures (Figure-3), which suggest the presence of weak intermolecular interaction. It can be seen from figure-3 that in the mixture,

absolute value of $\Delta\eta$ decrease at temperature in raised. An increment of temperature diminishes the self association of the pure component and also the hetro association between unlike molecule, because of the increase of the thermal energy. This lead to less negative values of $\Delta\eta$ as temperature is raised as observed in the present binary mixture. Many workers have reported similar behaviour where negative value of $\Delta\eta$ indicates dispersive interaction.

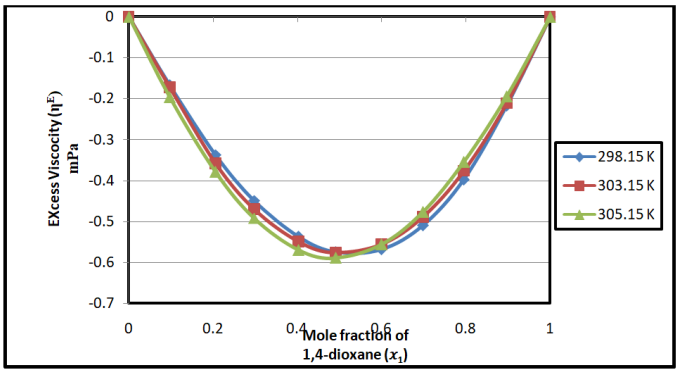

Figure 3. Curves of excess viscosityη^Eagainst the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + 1-Octanol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister equation

As the simplest molecular property is the free length (L_f) between the surfaces of the molecules. It seems interaction to find out the variation of intermolecular free length with concentration and temperature. The excess intermolecular free length (L_f^E)for the binary mixture1,4-Dioxane (1) + 1-Octanol (2) at temperature from (298.15,303.15 and 305.15) K as a function of 1,4-dioxane mole fraction have been reported in table-2. Excess intermolecular free length (L_f^E) is found to be negative for the binary mixture over the entire composition range at all three temperatures (Figure-4), The negative excess intermolecular free length (L_f^E) has been found to be negative for the binary mixture 1,4-Dioxane (1) + 1-Octanol (2). Which suggest that the sound wave needs to cover a large distance. This again supports the possibility of interaction due to hydrogen bonding between unlike molecules.

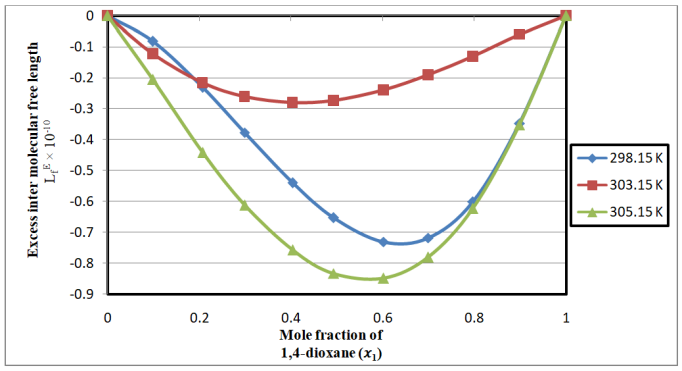

Figure 4.Curves of excess intermolecular free length (L_f^E)against the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + 1-Octanol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister equation

The internal pressure is a cohesive force, which is the result of attractive and repulsive forces between the molecules. The attractive forces mainly consist of hydrogen bonding, dipole-dipole, and dispersion interactions. Repulsive forces, acting over very small intermolecular distances, play a minor role in the cohesion process under normal circumstances. For the binary mixture1,4-Dioxane (1) + 1-Octanol (2), the obtained excess internal pressure (P_i^E) values are negative over the whole composition range at the studied temperatures as depicted in figure-5.

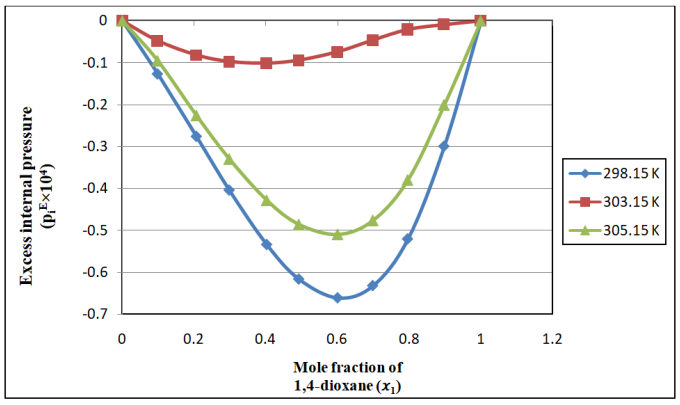

Figure 5. Curves of excess internal pressure (P_i^E) against the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + 1-Octanol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister equation

The excess internal pressure is often discussed in terms of molecular interactions in liquid mixtures. The variation of excess internal pressure is entirely negative similar to the deviation in excess molar volume, excess intermolecular free length excess adiabatic compressibility, and excess viscosity at all temperatures for1,4-Dioxane (1) + 1-Octanol (2) at temperature from (298.15,303.15 and 305.15) K. The less magnitude of these values suggests that weak interactions present in the system. The excess internal pressure decreasing with the increase in mole-fraction of1,4-Dioxane up to the mole-fraction (0.6) and then increases with increase in mole-fraction. This negative trend in (P_i^E) indicates that the only dispersion and dipolar forces operating with complete absence of specific interaction. It show the increasing magnitude of interaction between the 1,4-Dioxane (1) + 1-Octanol (2).

Conclusion

In this paper the ultrasonic velocity (u), density (ρ) and viscosity (η) have been measure over the whole composition range at temperature T = (298.15, 303.15 and 305.15) K for the binary mixture1,4-Dioxane (1) + Octan-1-ol (2). Excess molar volume, adiabatic compressibility, deviations in viscosity, excess intermolecular free length and excess internal pressure for binary mixtures have been calculated and fitted to a Redlich–Kister equation. This measured and calculated value of various thermo-acoustic parameters suggest the occurrence of complexation and through hetero molecular H-bonding between 1,4-Dioxane (1) + Octan-1-ol (2) in the binary liquid mixture. Hence it is concluded that the association in the mixture is result of hydrogen bonding between the molecule and octanol.

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