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Thermo-Acoustical Molecular Interaction Studies In Binary Liquid Mixture Containing (1, 4-Dioxane + Octan-1-Ol) At Different Temperature And 3 Mhz.

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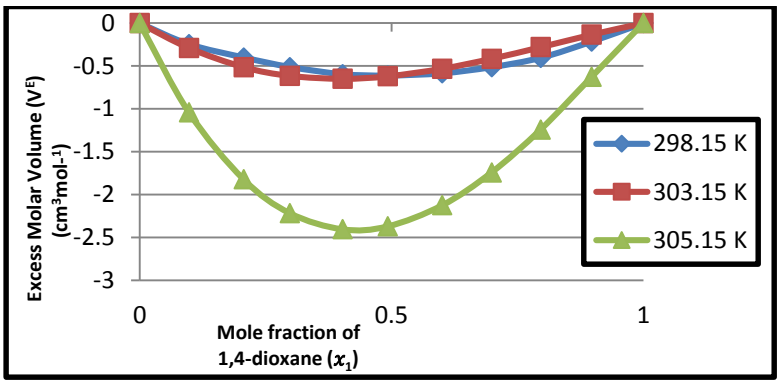
Abstract

Density (ρ), speed of sound (u), and viscosity (η) for the binary mixture 1, 4-Dioxane (1) + Octan-1-ol (2) were measured over the whole composition srange at temperatureT = (298.15, 303.15 and 305.15) K and 3 MHz. 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. These results have been fitted to the Redlich-Kister polynomial equation. 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 found to be negative for all temperatures. The thermodynamic properties have been discussed in term of nature of molecular interactions between the components of the mixture.

Keywords: Speed of sound, density, viscosity, binary liquid mixtures, internal pressure, molar volume, intermolecular free length and Redlich-Kister polynomial equation

Graphical Abstract



1. INTRODUCTION

Cyclic ether (1, 4-Dioxane) and octanol are important organic solvents that can be used in medical, petrochemical and pharmaceutical industries to a great extent [1-5]. The ultrasonic measurements have been widely used to study the molecular structure moreover, they provide useful information on molecular interactions required for optimizing thermodynamic model development.

The knowledge of excess adiabatic compressibility helps in understanding the molecular orientation and to study the extent of intermolecular interaction between the component molecules of liquid mixture [6]. In this work, the ultrasonic velocity (u), density (ρ) and viscosity (η) for the binary mixture1, 4-Dioxane (1) + Octanol (2) have been measured over the entire composition range and in the temperaturerange 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) have been calculated over the entire composition range and each temperature. 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) data have been correlated using the Redlich-Kister polynomial equation. The calculated deviations and excess functions have been explained on the basis of the intermolecular interaction present in this mixture. This work is a continuation of our research work studies on thermodynamic, transport and optical properties of liquid-liquid mixtures [7-11].

2. Experimental Procedure

2.1 Chemicals.

1, 4-Dioxane and octanol were obtained from Merck Chem. Ltd India with mass purity >99%. Both liquids were used without further purification as indicated in table-

1. 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-2, were leads to a satisfactory agreement.

2.2 Apparatus and Procedure

Both two mixtures of 1, 4-Dioxane and 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.

Table 1: CAS Registry Number, Mass Fraction Purity of the chemicals

Component	CAS Reg. No.	Supplier	Mass Fraction Purity (%)	Purity analysis method	Purification Method
1,4-Dioxane	17647-74-4	Sigma-Aldrich	≥99	Chromatography by the supplier	None
Octan-1-ol	111-87-5	Sigma-Aldrich	≥ 99	Chromatography by the supplier	None

Table 2: 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
		1.0305		1343		1.1944
		1.02763		1341		1.1960
		1.02792		1342		
Octan-1-ol	0.8242	0.8187	1327	1330	7.8512	7.6630
		0.8220		1346		7.661
		0.8216		1347		7.663
		0.8217		1347		7.5981

2.3 Measurements

2.3.1 Density

The densities of the pure liquid and its mixture were measured using a 25-ml specific gravity bottle by relative measurement method with an accuracy of ± 0.01 $kg.m^{-3}$. The specific gravity bottle with the experimental mixture was immersed in the temperature controlled water bath (MSI Goyal scientific, Meerut, U.P. India.), operating in the temperature range of -10^0C to 85^0C with an accuracy ± 0.1^0C.

2.3.2 Sound velocity

The ultrasonic velocity was measured using a multi-frequency ultrasonic interferometer (Model F-80D, Mittal Enterprise, New Delhi, India) working at 3 M.Hz. The meter was calibrated with water and benzene. Measurement of sound velocity through medium was based on the accurate determination of the wavelength of ultrasonic waves of known frequency produced by quartz crystal in the measuring cell. The interferometer cell was filled with the test liquid, and water was circulated around the measuring cell from a water bath. The uncertainty was estimated to be 0.1 ms^{-1}.The measured values of ultrasonic velocities of pure 1,4-dioxane and octanol compare well with the corresponding literature values.

2.3.3 Viscosity

The viscosity of the pure liquids and liquid mixtures are measured using an Ostwald's viscometer. This viscometer having a capacity of about 15 ml and the capillary having a length of about 90 mm and 0.5 mm internal diameter has been used to measure the flow times of pure liquids and liquid mixtures and it was calibrated with doubly distilled water and benzene. The flow time of pure liquids and liquid mixtures were repeated for five times. The efflux Time was measured with an electronic stopwatch (Racer) with a time resolution (± 0.015), and an average of at least five flow time readings was taken. Glass stopper was placed at the opening of the viscometer to prevent the loss due to evaporation during measurements. The uncertainty of viscosity was ± 0.005 $\times 10^{-3}$ m Pas. The measured values of viscosities of pure 1,4-Dioxaneand octanol compare well with the corresponding literature values.

3. Theoretical

The ultrasonic velocity (u), density (ρ) and viscosity (η) in pure liquids and liquid mixtures of various concentrations have been measured at 298.15, 303.15 and 305.15 K and 3 MHz.

Thermodynamics and acoustical parameters such as molar volume (V), adiabatic compressibility (β_{ad}), intermolecular free length (L_f) and internal pressure (P_i) were determined using the observed values of sound velocity, density and viscosity using the standard relations given below.

The molar volume (V) of binary liquid mixtures at a given mole fraction is given by:

$$V = \frac{(X_1 M_1 + X_2 M_2)}{\rho} \quad \textbf{(1)}$$

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

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

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

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

Where K is Jacobson's constant which is temperature dependent parameter.

Suryanarayana and Kuppuswami [12-13] suggested a method for evaluation of internal pressure from the knowledge of ultrasonic velocity, u, density and viscosity, the relation proposed is expressed as

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

Where b is packing factor, which is assumed to be 2 for all liquids and solution. 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.

The excess value of ultrasonic related parameters has been calculated by using the following relation

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

Where A represents the parameter such as intermolecular free length, molar volume, isentropic compressibility, viscosity and internal pressure and $X_1 \, and X_2$ is the mole fractions of components whose parameters.

4. Result and Discussion

The experimental values of ultrasonic velocity (u), density (ρ) and viscosity (η) for the binary mixture 1,4-Dioxane (1) + Octan-1-ol (2) were measured over the whole composition range at temperatureT = (298.15, 303.15 and 305.15) K and 3 MHz. are given in table-3 and calculated values of 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) are given in table-4.

Table 3: Values of density (ρ), sound velocity (u) and viscosity (η) for Various 1, 4-Dioxane Mole Fractions x_1 of the Binary Mixture (1,4-Dioxane (1) + Octan-1-ol (2)) at Temperatures T = (298.15, 303.15 and 305.15) K and 3MHz.

Mole fraction of 1,4-dioxane (x_1)	**Density (ρ) g.cm^{-3}**	**Sound velocity (u) m.s^{-1}**	**Viscosity (η) m.Pa.s**
T = 298.15 K			
0.00000	0.8312	1330.0	7.9215
0.09780	0.8582	1332.0	6.2565
0.20653	0.8763	1334.0	5.2685
0.29810	0.8952	1336.0	4.4523
0.40275	0.9152	1339.0	3.5698
0.49229	0.9315	1341.0	2.6525
0.60068	0.9456	1346.0	2.6325
0.69888	0.9623	1351.0	1.8956
0.79610	0.9836	1356.0	1.4685
0.89749	0.9946	1360.0	1.0525
1.00000	1.0215	1367.0	1.0652
T = 303.15 K.			
0.00000	0.8242	1327.0	7.8512
0.09780	0.8284	1329.0	5.1466
0.20653	0.8370	1330.0	4.6513
0.29810	0.8529	1332.0	3.2294
0.40275	0.8595	1334.0	2.5625
0.49229	0.8852	1336.0	2.3806
0.60068	0.9030	1338.0	1.8916
0.69888	0.9266	1339.0	1.4950
0.79610	0.9564	1341.0	1.3490
0.89749	0.9859	1345.0	1.1845
1.00000	1.0108	1348.0	1.0303
T = 305.15 K.			
0.00000	0.8153	1324.0	7.1025
0.09780	0.8326	1328.0	5.0123
0.20653	0.8523	1334.0	4.2513
0.29810	0.8845	1338.0	3.1202
0.40275	0.9021	1340.0	2.2256
0.49229	0.9263	1342.0	2.1251
0.60068	0.9512	1345.0	1.5641
0.69888	0.9725	1348.0	1.3025
0.79610	0.9901	1350.0	1.2351
0.89749	1.0095	1352.0	1.1625
1.00000	1.0102	1355.0	1.1032

Table 4: Values of excess viscosity (η^E),molar volume (V^E), adiabatic compressibility (β_{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) + Octan-1-ol (2)) at Temperatures T = (298.15, 303.15 and 305.15) K and 3MHz.

Mole fraction (x_1)	Excess Viscosity (η^E) m.Pa.	Excess Molar volume (V^E) (cm^3mol^{-1})	Excess adiabatic compressibility $\beta_{ad}^E \times 10^{-7}$	Excess internal pressure $p_i^E \times 10^4$	Excess free length $L_f^E \times 10^{-10}$
At 298.15 K					
0.00000	0.0000	0.00000	0.00000	0.00000	0.00000
0.09780	-0.1825	-0.24561	-0.27232	-0.15262	-0.10256
0.20653	-0.2854	-0.40125	-0.44362	-0.28325	-0.21561
0.29810	-0.4128	-0.51234	-0.53245	-0.36328	-0.32125
0.40275	-0.5321	-0.59652	0.60251	-0.52456	-0.55851
0.49229	-0.6215	-0.61245	-0.64258	-0.62354	-0.60851
0.60068	-0.6859	-0.58632	-0.53236	-0.71235	-0.85451
0.69888	-0.4525	-0.51456	-0.42582	-0.56540	-0.72542
0.79610	-0.3287	-0.40562	-0.35652	-0.64892	-0.59875
0.89749	-0.2003	0.21564	-0.22481	-0.15684	-0.20568
1.00000	0.0000	0.00000	0.00000	0.00000	0.00000
At 303.15 K					
0.00000	0.0000	0.00000	0.00000	0.00000	0.00000
0.09780	-0.2012	-1.15434	-0.08590	-0.08222	-0.12088
0.20653	-0.3633	-1.80656	-0.16204	-0.05674	-0.22926
0.29810	-0.4856	-1.97558	-0.14911	-0.10819	-0.22024
0.40275	-0.5567	-2.29511	-0.22970	-0.11758	-0.33295
0.49229	-0.6540	-2.54289	-0.15022	-0.08558	-0.23132
0.60068	-0.5241	-2.73983	-0.16391	-0.08626	-0.25166
0.69888	-0.4018	-1.27332	-0.13934	-0.08901	-0.21745
0.79610	-0.3126	-1.11434	-0.07472	-0.05664	-0.12426
0.89749	-0.2121	-0.68611	- 0.01397	-0.02768	-0.03111
1.00000	0.0000	0.00000	0.00000	0.00000	0.00000
At 305.15 K					
0.00000	0.0000	0.00000	0.00000	0.00000	0.00000
0.09780	-0.2254	-0.28451	-0.23541	-0.10253	-0.18562
0.20653	-0.3689	-0.45682	-0.42362	-0.18962	-0.24562
0.29810	-0.4525	-0.59452	-0.55632	-0.29632	-0.33652
0.40275	-0.5485	-0.65785	-0.60258	-0.35245	-0.65231
0.49229	-0.7523	-0.69892	-0.65284	-0.56242	-0.86542
0.60068	-0.6218	-0.58452	-0.56452	-0.66547	-0.95241
0.69888	-0.4287	-0.35241	-0.41258	-0.45632	-0.70125
0.79610	-0.3258	-0.20145	-0.31745	-0.28564	0.54282
0.89749	-0.312	-0.18563	-0.20513	-0.12325	-0.32542
1.00000	0.0000	0.00000	0.00000	0.00000	0.00000

The value of 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) are plotted against the mole fraction of 1,4-dioxane at different temperature are shown in Figure1-5 respectively. It is observed that ultrasonic velocity (u), density (ρ) and viscosity (η) and 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) parameter shows nonlinear increasing variation with increase in molar concentration. This indicates the complex formation and intermolecular weak association may be due to hydrogen bond formation [14]. This behaviour is the result of structural changes occurring in the mixture.

The values of excess molar volume (V^E), at each temperature from (298.15, 303.15 and 305.15) K are listed in table-4. The values of excess molar volume (V^E) at each studied temperature obtained from equation-5, have been correlated the following type of Redlich-Kister polynomial equation at each temperature [15].

$$Y^E = x_1 x_2 \sum_{k=1} A_k (x_1 - x_2)^k \quad \textbf{(5)}$$

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.

For the binary mixture 1, 4-Dioxane (1) + Octan-1-ol (2), the obtained excess molar volume (V^E) values are negative over the whole composition range at the studied temperature are depicted in figure-1. The sign of excess molar volume (V^E) depends upon the relative magnitude of contractive and expansive effects that arise on mixing of the components [16-17]. The studied system show negative excess molar volume (V^E) values that increase in magnitude with temperature and with minima displayed at the composition X_1 = 0.5, as observed in figure-1. The behaviour is explained by the existence of chemical interaction (hydrogen bonding) between unlike molecules of mixture that makes the contraction of solution volume.

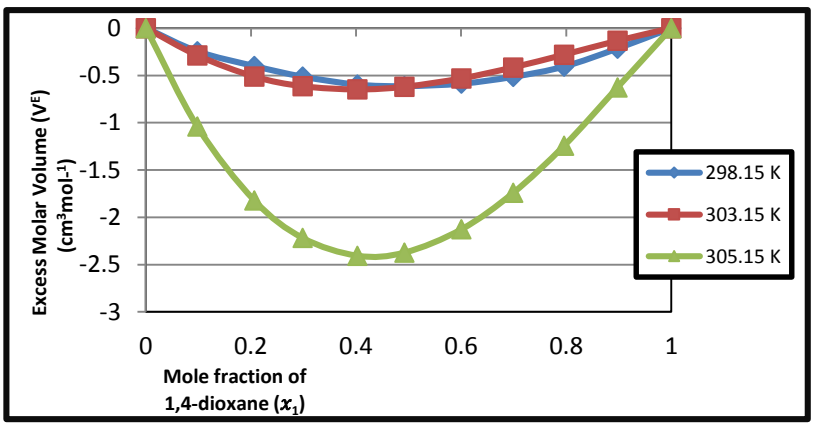


Figure 1. Curves of excess molar volume V^E against the mole fraction of 1, 4-dioxane x_1, for the binary mixture (1, 4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■, 303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister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

The excess adiabatic compressibility (β_{ad}^{E}) for the mixture1, 4-Dioxane (1) + Octan-1-ol (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-4. 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.

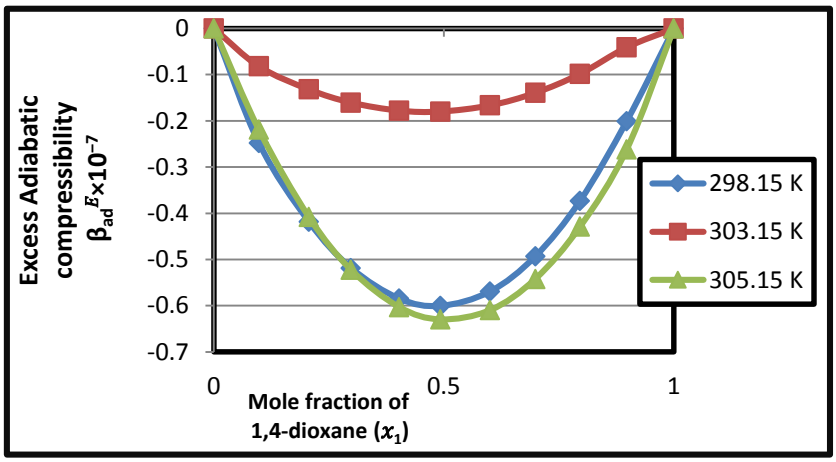


Figure 2. Curves of excess adiabatic compressibility β_{ad}^{E} against the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated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

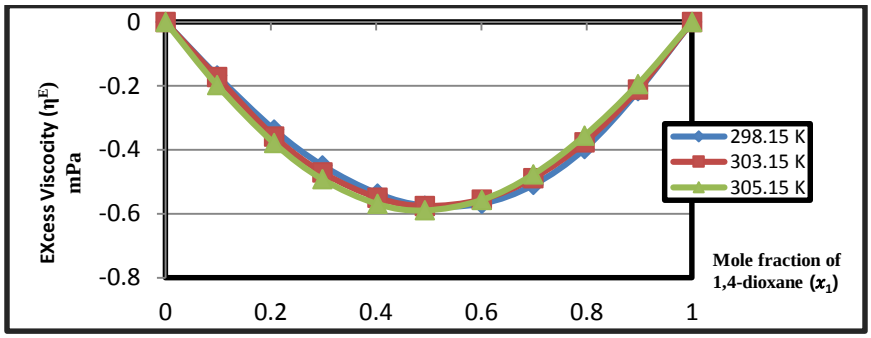


Figure 3. Curves of excess viscosityη^Eagainst the mole fraction of 1, 4-dioxane x_1, for the binary mixture (1, 4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■, 303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister 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.Theexcess intermolecular free length (L_f^E)for the binary mixture1, 4-Dioxane (1) + Octan-1-ol (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-4. 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) + Octan-1-ol (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.

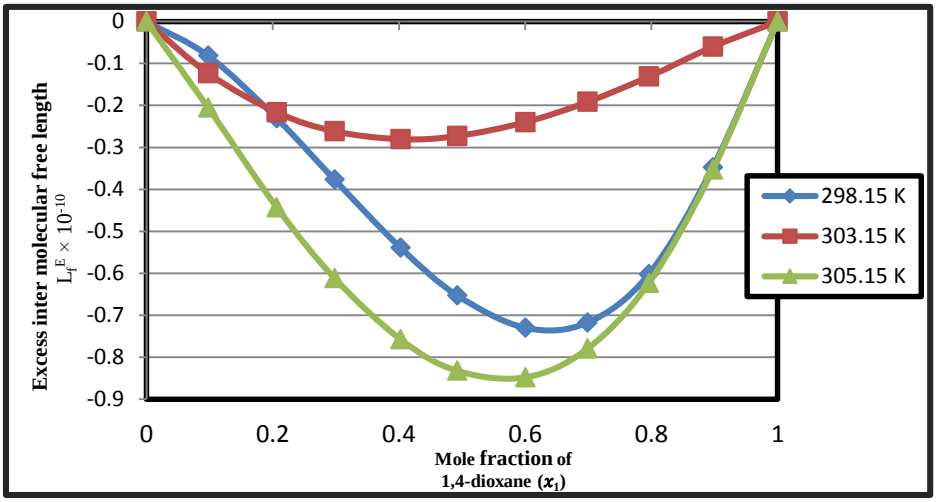


Figure 4. Curves of excess intermolecular free length (L_f^E)against the mole fraction of 1, 4-dioxane x_1, for the binary mixture (1, 4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■, 303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated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

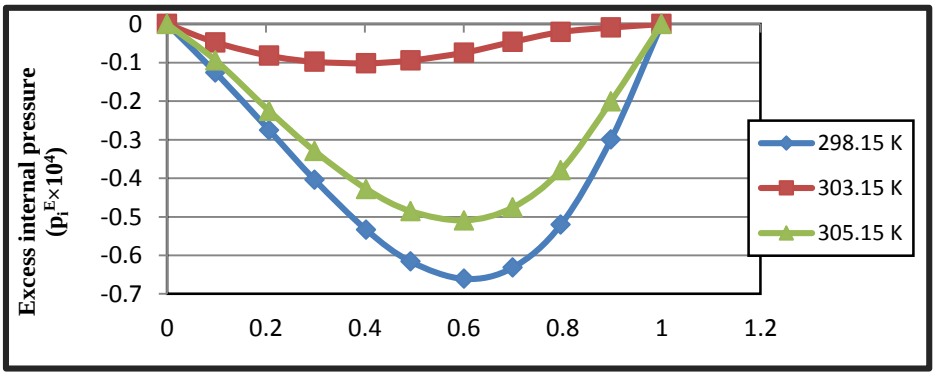


Figure 5. Curves of excess internal pressure (P_i^E) against the mole fraction of 1, 4-dioxane x_1, for the binary mixture (1, 4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■, 303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from the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

The authors have no competing interests to declare that are relevant to the content of this article.

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Data Availability Statement

All data generated or analyzed during this study are included in this published article.

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