International Journal of Pharmaceutical and Bio-Medical Science

ISSN(print): 2767-827X, ISSN(online): 2767-830X

Volume 04 Issue 06 June 2024

Page No: 512-522

DOI: https://doi.org/10.47191/ijpbms/v4-i6-06, Impact Factor: 7.792

Excess Thermodynamic Properties of Binary Liquid System Containing (Cyclic Ether + Octan-1-Ol) At Different Temperature

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ABSTRACT

The 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 temperature T = (298.15, 303.15 and 305.15) K. The experimental data have been used to calculate 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) over the entire composition range. 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 results obtained have been discussed and interpreted in terms of the type and nature of the specific intermolecular interactions between the components.

KEYWORDS: Ultrasonic velocity, adiabatic compressibility, internal pressure, molar volume, intermolecular free length and Redlich-Kister polynomial equation.

ARTICLE DETAILS

Published On:
11 June 2024

Available on:
https://ijpbms.com/

1. INTRODUCTION

Sound velocity investigation of liquid mixtures containing of polar components are of considerable importance in understanding intermolecular interactions between the component molecules and find other industrial and technological processes [1-5]. The study of molecular association in binary liquid mixture having alcohol as one of the component is of particular interest, since alcohols are strongly self-associated liquid having a three dimensional network of hydrogen bonds and can be associated with any other group having same degree of polar attractions. The excess molar volume (V^E), excess adiabatic compressibility (β_{ad}^E), excess intermolecular free length (L_f^E) and excess internal pressure (P_i^E) is a good thermodynamic tool to explore the behaviour of mixtures. Alcohols and ethers represent three technically important classes of compounds. Ethers are industrially important solvents in several chemical reactions. Alcohols are self associated organic liquids and well-known polar solvents used in a wide range of applications. The ultrasonic velocity along with density and viscosity furnish wealthy of information about the interaction between ions, dipoles, hydrogen bonding, multi-polar and dispersive forces. Ultrasonic propagation parameters yield valuable information regarding the behaviour of liquid systems because intermolecular and intramolecular association, complex formation, dipolar interactions and related structural changes affect the compressibility of the system, which in turn produces corresponding variations in ultrasonic velocity [6].

In the present study several parameters such as molar volume (V), adiabatic compressibility (β_{ad}), intermolecular free length (L_f) and internal pressure (P_i) of a binary liquid system 1,4-Dioxane + Octan-1-ol have been reported using the experimental data of density, viscosity and ultrasonic velocity of the binary liquid mixture at temperature 298.15, 303.15 and 305.15) K. 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. The source and purity of the chemical compound are shown in table-1.The substances density, viscosity and ultrasonic velocity is compared with the literature data (Table-2) to ascertain the purity, and a good agreement between the experimental data and literature data [12-20] was observed.

2.2 Apparatus and Procedure: All two binary liquid mixtures were prepared by weighing appropriate amounts of pure liquids on a digital electronic balance (Citizen Scale (I) PVT. LTD. Mumbai, India.) with a precision ± 0.1. 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 (%)	Method Purity analysis method	Purification
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.

Compound	ρ (g.cm^{-3})		u (m.s^{-1})		η (mPa s)	
	This work	literature	This work	literature	This work	literature
1,4-Dioxane	1.0108	1.0229[12]	1348	1344[21]	1.0303	1.0690[28]
		1.0286[13]		1345[22]		1.1944[18]
		1.0305[14]		1343[23]		1.1944[29]
		1.02763[15]		1341[24]		1.1960[24]
		1.02792[16]		1342[25]		
Octan-1-ol	0.8242	0.8187[17]	1327	1330[26]	7.8512	7.6630[30]
		0.8220[18]		1346[18]		7.661[30]
		0.8216[19]		1347[27]		7.663[31]
		0.8217[20]		1347[20]		7.5981[32]

2.3 Measurements:

Density: Densities of pure components and liquid-liquid mixtures were measured with 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^{0}C to 85^{0}C with an accuracy ± 0.1^{0}C. Double distilled water used for the calibration of the specific gravity bottle. At least three times for each composition in experimental were generally repeated and the results were treatment.

Sound velocity:

The speed of sound (u) were measured at a frequency 3 MHz in these solutions using the interferometric method with a (Model F-80D, Mittal Enterprise, New Delhi, India) at 298.15, 303.15 and 305.15) K. 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%. The measured values of ultrasonic velocities of pure 1,4-dioxane and octan-1-ol compare well with the corresponding literature values.

Viscosity:

The viscosities of pure liquids and their binary mixtures were measured by using a Ostawald's viscometer. The viscometer was calibrated with doubly distilled water and benzene, liquid was allowed to stand for about 30 minutes in a thermostatic water bath so that the thermal fluctuations in viscometer were minimized. The accuracy in viscosity data was ± 0.0005 mPa.s. 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 measured values of viscosities of pure 1,4-Dioxane and octan-1-ol compare well with the corresponding literature values.

Theoretical:

The experimentally measured ultrasonic velocity (u), density (ρ) and viscosity (η) are used to evaluate derived parameters like molar volume (V), adiabatic compressibility (β_{ad}), intermolecular free length (L_f) and internal pressure (P_i) using well established relations.

The molar volume (V) of binary liquid mixtures were calculated by using a following equation:

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

The adiabatic compressibility(β_{ad}) has been calculated from the ultrasonic velocity (u) and density (ρ)of the medium using the equation as

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

Intermolecular free length (L_f) has been determined using the standard relation as:

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

Where K ,is a temperature dependent known as Jacobson's constant.

On the basis of statistical thermodynamics, Suryanarayana [33-34] derived an expression for determination of internal pressure by the use of free volume concept 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 b stands for cubic packing factor, which is assumed to be 2 for liquids. 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 have been calculated by using the following relation

$$A^E = A_{exp.} - (X_1A_1 + X_2A_2) \quad (5)$$

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.

3. RESULT AND DISCUSSION

Assuming that ultrasonic absorption is negligible, using experimental results of the 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 temperature T = (298.15, 303.15 and 305.15) K are given in table-3 and their excess parameters have been 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 TemperaturesT = (298.15, 303.15 and 305.15) K.

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.

Mole fraction (x_1)	Excess Viscosity η^E (m.Pa.s)	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 [35]. 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 [36].

The excess properties were fitted to the Redlich-Kister polynomial equation

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

Where Y^E refers to excess properties and x_1 and x_2 are the mole fraction 1,4-Dioxane (1) and Octan-1-ol (2), and A_k represents the coefficients. Adjustable parameters of A_k were evaluated by least-squares method.

The excess molar volume, inspected in project were all negative over the whole range of 1,4-dioxane at different temperatures. These are shown in figure-1. This may suggest that volume construction take place on to mixing1,4-dioxane with alcohol due to the cross-association between these various molecules [37-38]. And the negative values are attributable mainly to the association between ether and alcohol intermolecular hydrogen bonds between the c-OH groups in alcohols and the oxygen atoms in the ether. The strength of the association arising from interactions between the unlike molecules was stronger than the strength of the association between the molecules. The behaviour is explained by the existence of chemical interaction (hydrogen bonding) between unlike molecules of mixture that makes the contraction of solution volume.

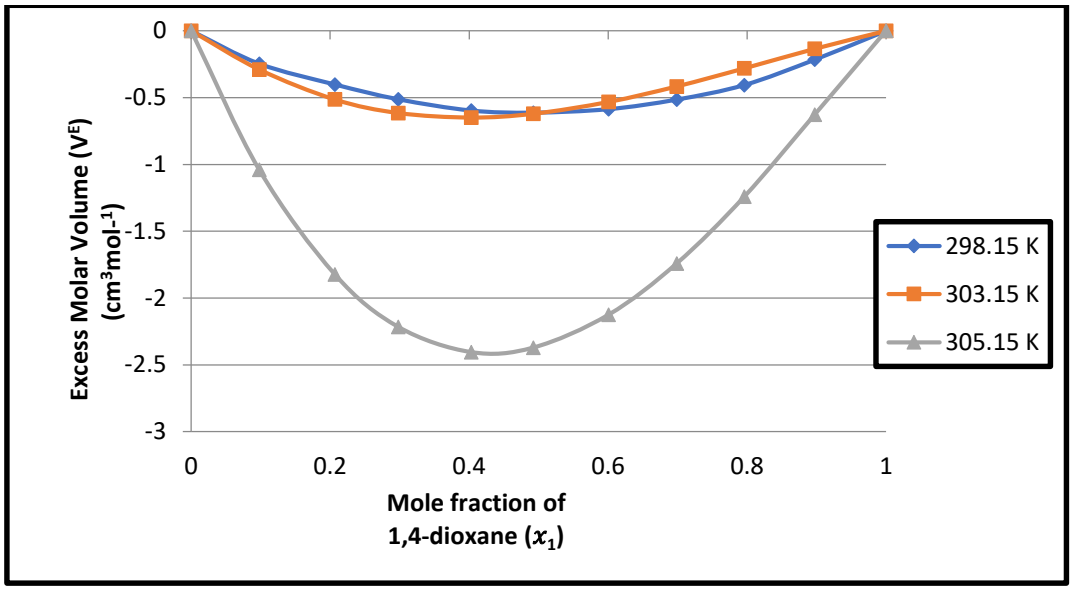

Figure 1. Curves of excess molar volume V^E against the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister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

The excess adiabatic compressibility (β_{ad}^E) data of the mixture 1,4-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. The excess adiabatic compressibility (β_{ad}^E), values were ascribed the negative excess values have been due to the closely packed molecules which account for existence of strong molecular interaction. As mentioned in the literature [39 - 41] 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 [42- 43]. 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.

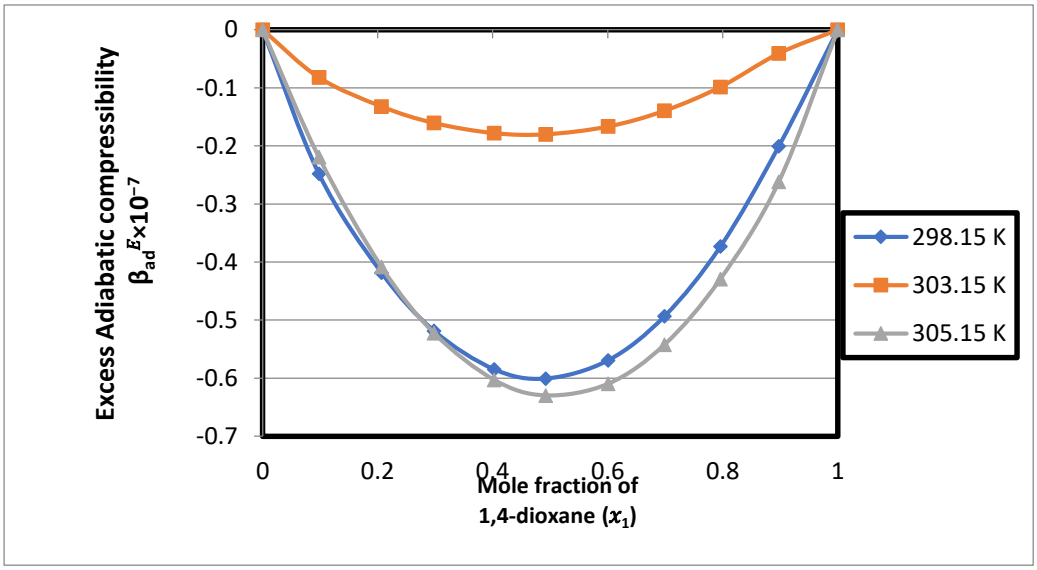

Figure 2. Curves of excess adiabatic compressibility β_{ad}^E against the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from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

Excess viscosity ($\Delta\eta$), is found to be negative for the binary mixture over the entire composition range at all the three temperatures (Figure-3). From experimental results suggest that the negative values of ($\Delta\eta$) may be attributed to the formation of hydrogen bonding (O-H…….O) resulting in the formation of complexes between the component molecules and negative values suggest that the rupture f hydrogen bonded chain of the dipolar interaction between solute and alcohol exceed the intermolecular interaction through dipole-dipole and hydrogen bonding between solute and alcohol. This lead to less negative values of $\Delta\eta$ as temperature is raised as observed in the present binary mixture. Many workers [44-45], have reported similar behaviour where negative value of $\Delta\eta$ indicates dispersive interaction.

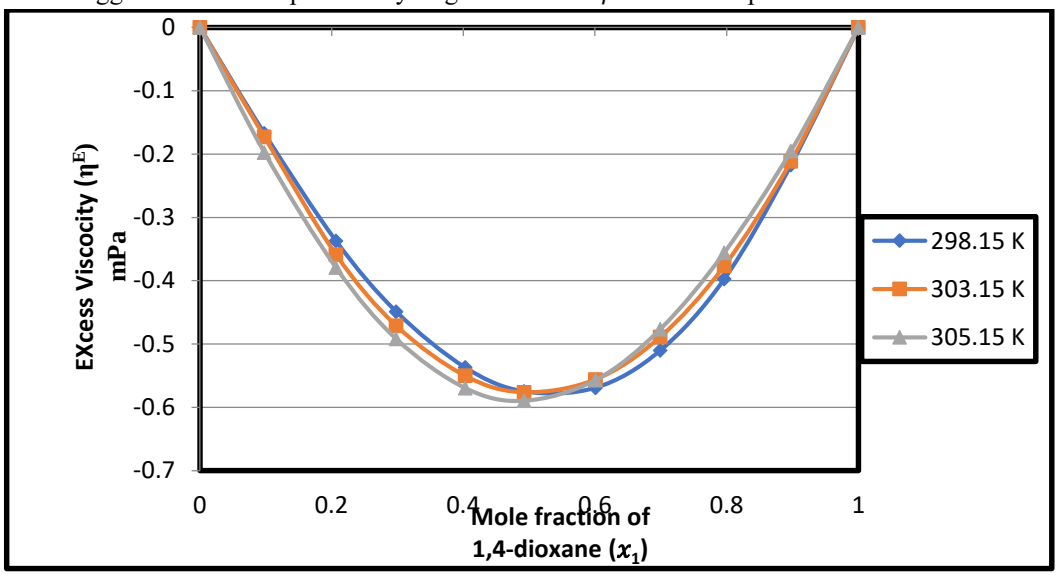

Figure 3.Curves of excess viscosityη^Eagainst the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister equation.

The excess 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 temperature (Figure-4), In the present paper, negative values of excess free length for 1,4-Dioxane (1) + Octan-1-ol (2) mixture can be attributed to formation of molecular complexes through dipole-dipole interaction. 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.

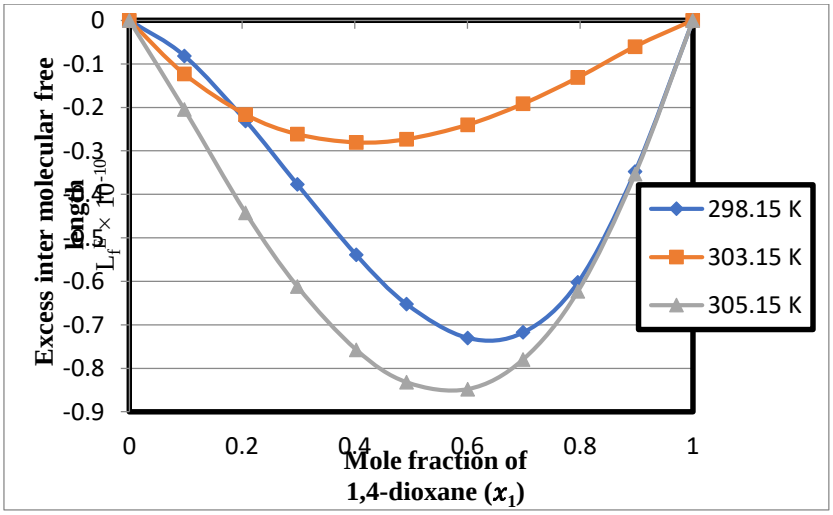

Figure 4. Curves of excess intermolecular free length (L_f^E)against the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from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

The internal pressure increases with increase in molar concentration indicate the association through hydrogen bonding. It show the increasing magnitude of interaction between the 1,4-Dioxane (1) and Octan-1-ol (2).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.

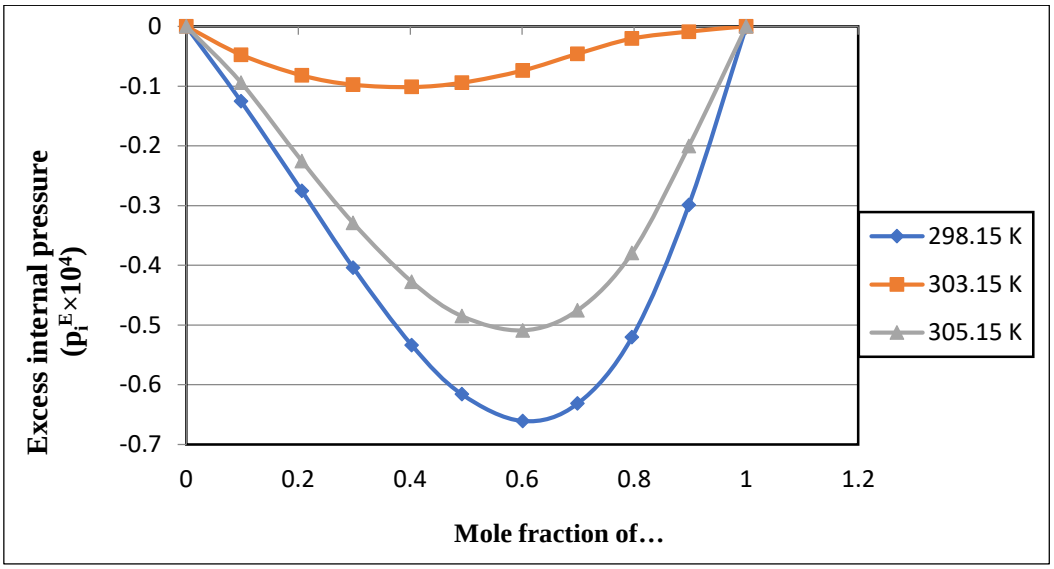

Figure 5. Curves ofexcess internal pressure (P_i^E) against the mole fraction of 1,4-dioxane x_1, for the binary mixture (1,4-dioxane (1) + Octan-1-ol (2)) at different temperatures (blue ♦, 298.15 K orange ■,303.15 K and gray ▲, 305.15 K). The solid lines represent the values calculated from the Redlich−Kister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

The 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).

4. CONCLUSION

In this paper the sound 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, excess adiabatic compressibility, excess 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 octan-1-ol.

ACKNOWLEDGEMENT

The authors thank the University authorities for providing the necessary facilities to carry out the work.

DECLARATIONS CONFLICT OF INTEREST

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

FUNDING

The author(s) reported there is no funding associated with the work featured in this article.

DATA AVAILABILITY STATEMENT

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

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