

Viscometric And Volumetric Analysis In Binary Liquid Mixture Of 1,4-Dioxane With n-Octanol At 298.15, 303.15 And 305.15 K And 3 Mhz.

Dhirendra Kumar Sharma^{1*}, Chandra Pal Prajapati², Suneel Kumar³,
Ravi Kant Mishra⁴

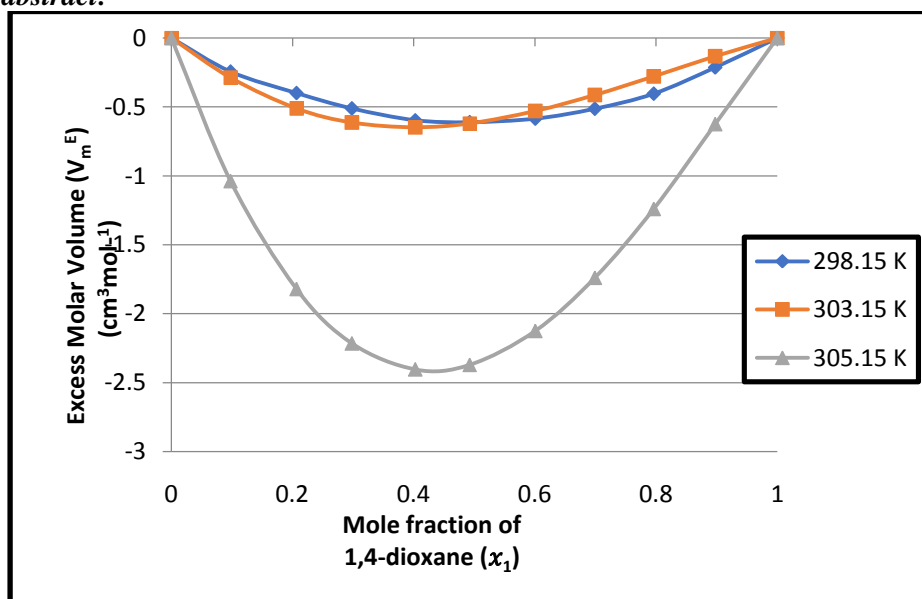
^{1*}Department Of Chemistry, Institute Of Basic Science, Bundelkhand University, Jhansi (U.P), India.

Abstract:

The ultrasonic velocity (u), density (ρ) and viscosity (η) for the binary mixture of cyclic ether (1,4-dioxane (1) + n-octanol (2)) were measured over the whole composition range at different temperature (298.15, 303.15 and 305.15) K and 3 MHz. These experimental values, calculate the excess molar volume (V_m^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). These results have been fitted to the Redlich-Kister polynomial equation. Excess molar volume (V_m^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. A comparison has also been made between observed values and calculated ultrasonic velocities through liquid mixtures.

Keywords: cyclic ether, n-octanol, internal pressure, molar volume, intermolecular free length and Redlich-Kister polynomial equation.

Graphical abstract:



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I. Introduction:

In continuation of our research programme on the thermodynamic and transport properties of binary liquid mixtures at different temperature containing the oxygen (-O-) and hydroxyl functional group (-OH) [1-5]. In this paper we report experimental values of ultrasonic velocity (u), density (ρ) and viscosity (η) for the binary mixture 1,4-dioxane (1) + n-octanol (2) have been measured over the entire composition range and in the different temperature range (298.15, 303.15 and 305.15) K and at the atmospheric pressure. The experimental values of ultrasonic velocity (u), density (ρ) and viscosity (η) are valuable tools to learn about the liquid state [6].

In this work, from the experimental data, we are calculate the excess properties of, molar volume (V_m), adiabatic compressibility (β_{ad}^E), viscosity (η^E), intermolecular free length (l_f^E) and internal pressure (p_i^E). The calculated deviations and excess functions have been explained on the basis of the intermolecular interaction present in this mixture [7-11].

II. Experimental

chemicals: 1,4-dioxane and n-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 [12-20] and are listed in table-2, were leads to a satisfactory agreement.

apparatus: 1,4-dioxane and n-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. Provenance and purity of the materials used.

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
N-octanol	111-87-5	sigma-aldrich	≥ 99	chromatography by the supplier	none

Table 2. Physical properties of pure liquids at 298.15k and 3mhz.

Compound	$\rho / (g.cm^{-3})$		$U / (m.s^{-1})$		$H / (mpa.s)$	
	Expti.	Lit.	Expti.	Lit.	Expti.	Lit.
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		
N-octanol	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

Measurements:

Density: densities of pure liquids and their binary mixtures were determined by using a 30-ml specific gravity bottle by relative measurement method with an accuracy of $\pm 0.01 \text{ kg.m}^{-3}$. The specific gravity bottle with the experimental mixture was immersed in the temperature controlled water bath (msigoyal scientific, meerut, u.P. India.), the precision of the density measurements was estimated to be $\pm 0.0002 \text{ g cm}^{-3}$. The observed values of densities of pure 1,4-dioxane and n-octanol compare well with corresponding literature values of respectively.

Sound velocity: the ultrasonic velocities were measured using a multifrequency ultrasonic interferometer (model f-80d, mittal enterprise, new delhi, india) working at 3 m.hz. The meter was calibrated with water and benzene at 298.15k. The measured values of ultrasonic velocities of pure 1,4-dioxane and n-octanol compare well with the corresponding literature values.

Viscosity:

The viscosity of pure liquids and their binary mixture were measured using suspended ostwald's 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 time of pure liquids and liquid mixtures and it was calibrated with triply distilled water, methanol and benzene at 298.15 k. The efflux time was measured with an electronic stop watch (racer) with a time resolution (± 0.015), and an average of at least four 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 viscosities have reproducibility within $\pm 0.002 \text{ m.pa.s}$. The measured values of viscosities of pure 1,4-dioxane and n-octanol compare well with the corresponding literature values.

Theoretical:

The molar volume V_m calculated from the measured values of density (ρ), molar volume (V_m) was calculated using the relation

$$V_m = \frac{(X_1M_1 + X_2M_2)}{\rho} \tag{1}$$

Where X_1, X_2 and M_1, M_2 are the mole fraction and molecular weight of the component 1 and 2 respectively

The adiabatic compressibility(β_{ad}), calculated from the measured values of density (ρ) and sound velocity (u) was calculated using the relation

$$\beta_{ad} = u^{-2}\rho^{-1} \tag{2}$$

Where ρ , is the density of liquid.

I in the year 1952, jacobson, suggested an empirical relation for calculating the free length (L_f) of liquids. Intermolecular free length (L_f), can be calculated from the isentropic compressibility(β_{ad}), by the relation given below

$$L_f = k \beta_{ad}^{1/2} \tag{3}$$

Suryanarayana and kuppuswami [33-34]derived a formula for evaluation of internal pressure from the ultrasonic velocity, u , density, ρ and viscosity, η , the relation proposed is expressed as

$$P_i = b \left(\frac{k\eta}{u} \right)^2 \frac{\rho^{2/3}}{M_{eff}^{7/6}} \tag{4}$$

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 A^E of these thermodynamic parameters have been obtained by subtracting the ideal value from the experimental value

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

Where a represents the parameter such as intermolecular free length, molar volume, available volume, free volume and isentropic compressibility and X_1 and X_2 are the mole fractions of components whose parameters.

III. Result And Discussion:

The experimental values of density, viscosity and ultrasonic velocity at different temperature over the entire composition range are presented in table 3. The excess function, excess intermolecular free length (L_f^E), excess viscosity (η^E), excess molar volume (V_m^E), excess adiabatic compressibility (β_{ad}^E) and excess internal pressure (P_i^E) for binary liquid mixtures evaluated using equation (5) are summarized in table 4.

Table 3. Experimental results of binary liquid mixture of 1,4-dioxane (1) + n-octanol (2)) at different temperatures (298.15, 303.15 and 305.15) k and 3mhz.

Mole fraction of (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. Excess properties of molar volume, viscosity, adiabatic compressibility, free length and internal pressure of binary liquid mixture of 1,4-dioxane (1) + n-octanol (2) at different temperatures (298.15, 303.15 and 305.15) k and 3mhz.

Mole fraction (x_1)	Excess viscosity (η^E) M.pa.	Excess molar volume (V_m^E) ($\text{cm}^3\text{mol}^{-1}$)	Excess adiabatic compressibility $B_{ad}^E \times 10^{-7}$	Excess internal pressure $P_1^E \times 10^4$	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 excess parameters, excess molar volume (V_m^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 plotted against mole fraction of 1,4-dioxane in figure 1,2,3,4,5.

It is observed that sound velocity (u), density (ρ), viscosity (η) and excess molar volume (V_m^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_m^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_m^E), at each studied temperature obtained from equation-6, have been correlated the following type of redlich-kister polynomial equation at each temperature [36].

$$Y^e = x_1 x_2 \sum_{k=1}^K a_k (x_1 - x_2)^k \quad (6)$$

$K=1$

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.

A perusal of table 4 reveal that the value of excess molar volume (V_m^E) are negative in all binary liquid system over the entire range of composition at different temperatures. The value of V_m^E are plotted against the mole fraction of 1,4-dioxane (X_1) and are shown in figure 1. In the present investigation the negative V_m^E values for binary mixtures of 1,4-dioxane with n-octanol may be attributed to hydrogen bond formation through dipole-dipole interaction between n- alkanol and 1,4-dioxane molecule or to structure contribution arising from geometrical fitting of one component (n-alkanol) in to the other (1,4-dioxane) due to differences in the molar volumes between components. Excess molar volume value of all binary liquid systems increases with an increases of concentration of 1,4-dioxane(X_1). This is attributed to decreased ether-ether and alkanol-alkanol contacts with an increase the concentration of 1,4-dioxane(X_1)[37-38]. The behaviour is explained by the existence of chemical interaction (hydrogen bonding) between unlike molecules of mixture that makes the contraction of solution volume.

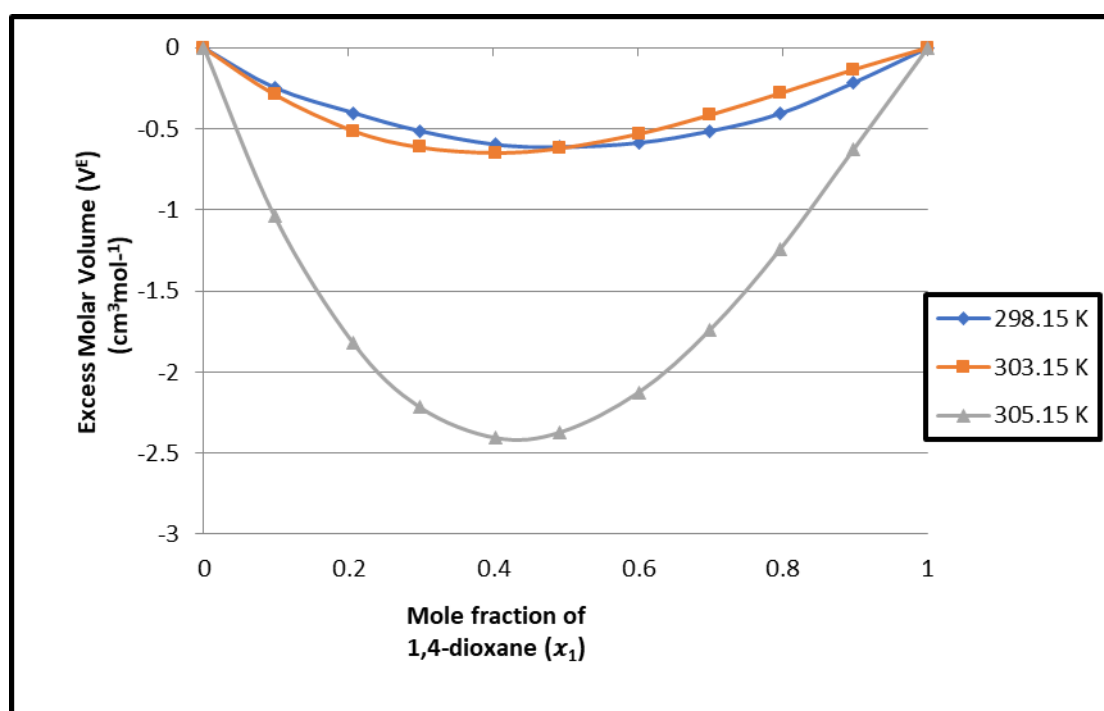


Figure 1. Comparison between excess molar volume V_m^E , against the mole fraction of 1,4-dioxane x_1 , for the binary mixture (1,4-dioxane (1) +n-octanol(2)) at different temperatures (blue \blacklozenge , 298.15 k orange \blacksquare ,303.15 k and gray \blacktriangle , 305.15 k).

The excess adiabatic compressibility (β_{ad}^E) for the binary liquid mixture 1,4-dioxane (1) + n-octanol (2) at the different temperatures from (298.15,303.15 and 305.15) k as a function of 1,4-dioxane mole fraction have been reported in table-4. The excess adiabatic compressibility (β_{ad}^E) is plotted in figure 2 for the binary systems of 1,4-dioxane (1) + n-octanol (2) at the different temperatures from (298.15,303.15 and 305.15) k. The excess

adiabatic compressibility (β_{ad}^E) values are negative over the entire composition range at the different temperature which are under the investigation and this may be attributed due to the relative strength of effect which influence the free space between component molecules as described in the literature[39-41]. Fort and moore suggested that the liquid having different molecular size and shape mix well there by reducing the volume which causes the values of (β_{ad}^E) to be negative. It is also suggested that liquids are less compressible when compared to their ideal mixtures signifying the chemical effects including charge transfer forces, formation of hydrogen bond and other complex forming interactions. It can also be said that the molecular interaction are strong in these binary liquid mixtures and that the medium is highly packed.

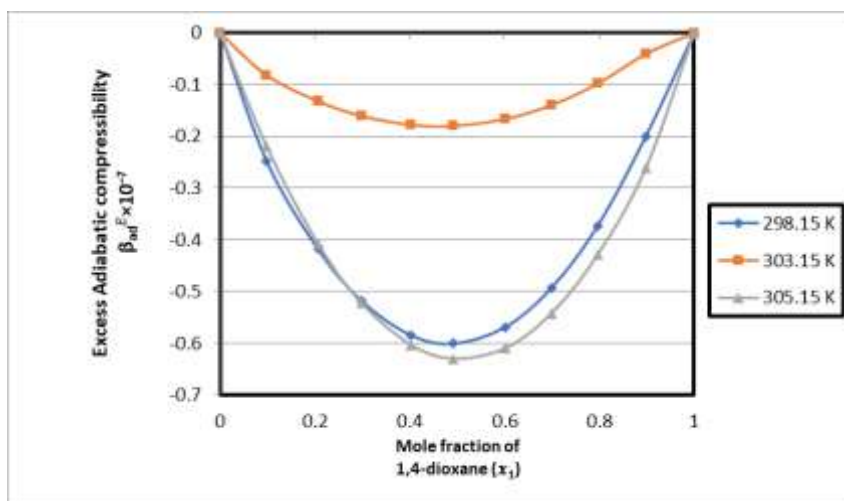


Figure 2. Comparison between excess adiabatic compressibility β_{ad}^E , against the mole fraction of 1,4-dioxane x_1 , for the binary mixture (1,4-dioxane (1) +n- octanol(2)) at diff erent temperatures (blue \blacklozenge , 298.15 k orange \blacksquare ,303.15 k and gray \blacktriangle , 305.15 k).

The measurement of viscosity in binary liquid mixture gives some reliable information in the study of intermolecular interaction. The molecules of one or more components forming the temarise are either polar, associating or accordingly show non-ideal behaviors in mixtures. Negative values of η^E in most of the cases are the consequence of lower viscosity contributions of similar non-specific interaction and hydrogen bonding effect of molecular species in real mixtures rather than those in the corresponding ideal mixtures. In the present study, it is observed that, for the binary system 1,4-dioxane (1) + n-octanol (2), the η^E values gradually decrease up to the mole fraction around 0.5 and then begins to increase figure 3 more over it is observed that the η^E values decrease as the concentration of x_1 increase. The negative values imply the presence of dispersion forces between the mixing components in the mixtures [42-43].

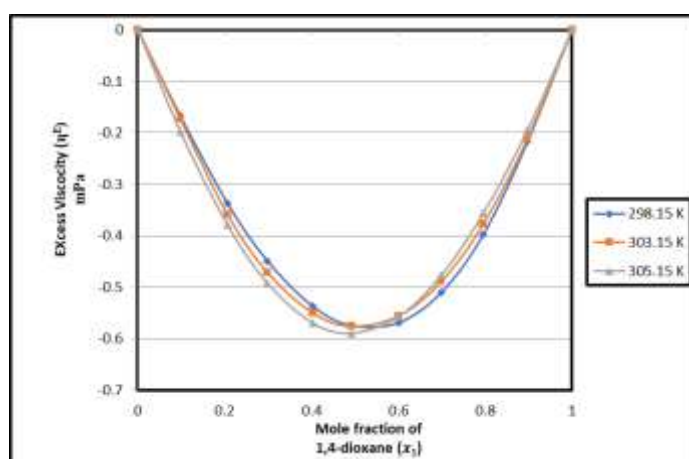


Figure 3. Comparison between excess viscosity η^E , against the mole fraction of 1,4-dioxane x_1 , for the binary mixture (1,4-dioxane (1) +n- octanol(2)) at different temperatures (blue \blacklozenge , 298.15 k orange \blacksquare ,303.15 k and gray \blacktriangle , 305.15 k).

The excess intermolecular free length (l_f^E) for the binary mixture 1,4-dioxane (1) + n-octanol (2) at different temperatures (298.15, 303.15 and 305.15) k as a function of 1,4-dioxane mole fraction have been

reported in table-4. The negative values of excess intermolecular free length (L_f^E) play a very important role in description of molecular interaction in liquid mixtures through dipole-dipole interaction and hydrogen bonding. Due to polar nature of 1,4-dioxane and alkanol, the dipole-dipole interactions prevail in these mixtures. When the compounds are mixed the changes that occur in association equilibria are evidently rupture of the hydrogen bonds in pure 1,4-dioxane and alkanol, dipole-dipole interactions and the formation of o-h ---o hydrogen bonds between the components. This suggests the existence of strong interaction between the components in all the binary systems. The values of L_f^E suggest that strong specific interaction like the formation of h- bond association through weaker physical forces of attraction

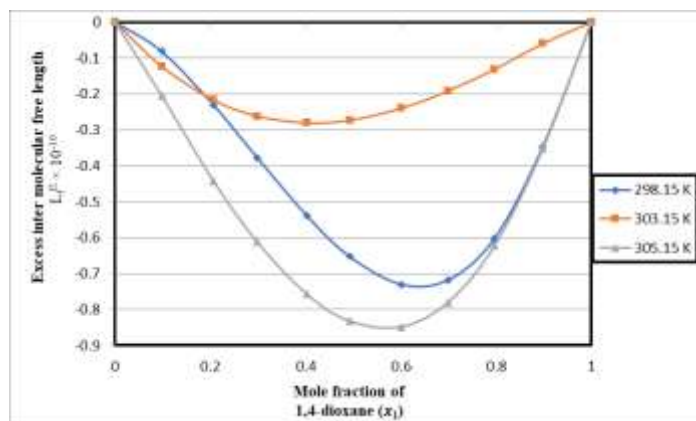


Figure 4. Comparison between excess intermolecular free length (L_f^E), against the mole fraction of 1,4-dioxane x_1 , for the binary mixture (1,4-dioxane (1) + n-octanol(2)) at different temperatures (blue \blacklozenge , 298.15 k orange \blacksquare , 303.15 k and gray \blacktriangle , 305.15 k).

For the binary mixture 1,4-dioxane (1) + n-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. The excess internal pressure (p_i^E) is another important parameter through which molecular interactions can be explained. In the present investigation for the binary system 1,4-dioxane (1) + n-octanol (2), it is observed that, as the mole fraction of 1,4-dioxane increase, the p_i^E values decrease. The values of p_i^E are almost negative and gradually decrease and move towards the positive values by the increase of mole fraction of ethyl acetate. More over the p_i^E decrease with increase in x_1 . This situation is observed for binary system 1,4-dioxane + n-octanol under study and can be viewed from plots figure 5.

The negative values of excess internal pressure (p_i^E) indicate the presence of strong molecular interaction. We may conclude that alkanol, which is a self – associating polar organic liquid has a tendency to form complexes with 1,4-dioxane and the increase in its dilution causes disruption of aromatic c – h bond stretching as the self – association of alkanol is disrupted. It is also concluded that suryanarayana approach for estimating internal pressure of binary liquid mixtures, based on dimensional analysis using thermodynamic consideration is very well applicable in the present case.

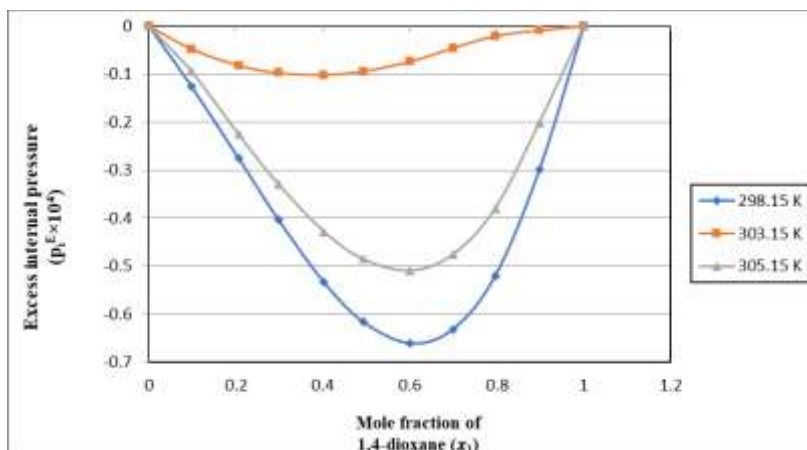


Figure 5. Comparison between excess internal pressure (p_i^E), against the mole fraction of 1,4-dioxane x_1 , for the binary mixture (1,4-dioxane (1) + n-octanol(2)) at different temperatures (blue \blacklozenge , 298.15 k orange \blacksquare , 303.15 k and gray \blacktriangle , 305.15 k).

IV. Conclusion:

The ultrasonic velocity (u), density (ρ) and viscosity (η) have been measured over the whole composition range at different temperature range (298.15, 303.15 and 305.15) K for the binary liquid mixture 1,4-dioxane (1) + n-octanol (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. The results assure what has been formerly announced that in the solvents system investigated, the negative values are attributable to stronger hydrogen bond formations between unlike molecules.

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