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(Research Article)

Molecular interaction studies in binary liquid mixtures containing ethyl acetate with n-octanol at different temperatures

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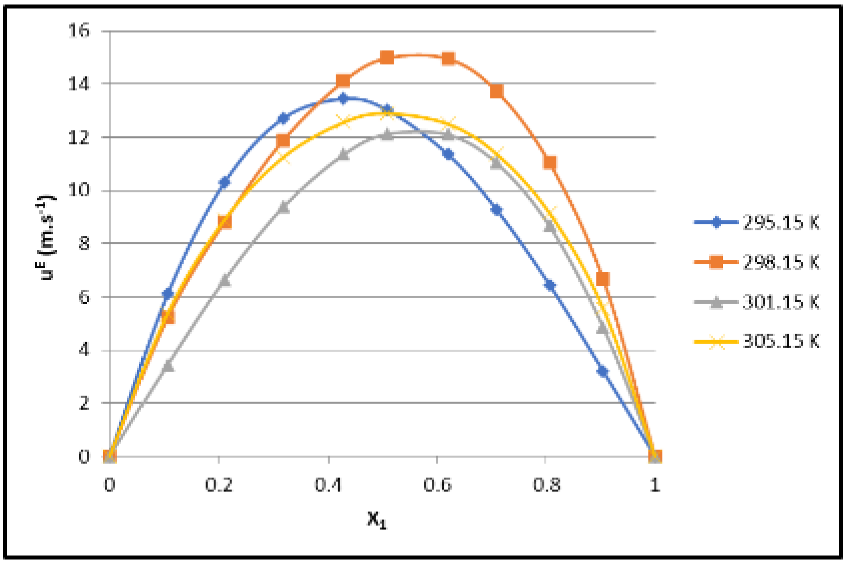
Abstract

Abstract: Experimental values of ultrasonic velocity (u), density (ρ) and viscosity (η) for the binary mixture Ethyl acetate (1) + n-Octanol (2) were measured over the whole composition range at temperature T = (295.15, 298.15,301.15 and 305.15) K and at the atmospheric pressure. From the experimental data excess sound velocity (u^E), excess viscosity (η^E), excess free volume (V_f^E) and excess internal pressure (P_i^E) were calculated. These results have been fitted to the Redlich-Kister polynomial equation. Excess viscosity (η^E), excess free volume (V_f^E) and excess internal pressure (P_i^E) were found to be negative for all temperatures. Excess sound velocity (u^E), excess viscosity (η^E), excess free volume (V_f^E) and excess internal pressure (P_i^E) were plotted against the mole fraction of ethyl acetate over the whole composition range at 295.15, 298.15, 301.15 and 305.15 K. They have been analysed to discuss the nature and strength of intermolecular interactions in these mixtures.

Keywords: Ultrasonic velocity; Density; Viscosity; Binary liquid mixtures; Internal pressure; Free volume; Redlich-Kister polynomial equation; Intermolecular interactions.

Graphical Abstract



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1. Introduction

The present paper forms a part of our ongoing research program of research to measure physical properties of binary liquid mixtures containing ethyl acetate and the prediction of their excess properties [1-5]. In recent years, in the study of molecular interactions in binary liquid mixtures by free volume and internal pressure has gained significant interest.The ultrasonic velocity (u), density (ρ) and viscosity (η) of binary liquid mixture are important from practical and theoretical points of view to understand the liquid theory. Several attempts have been made to calculate the free volume and internal pressure of liquid and liquid mixtures theoretically. Here we made use of empirical methods to estimate the free volume and internal pressure in ethyl acetate (1) + n-Octanol (2) at temperature T = (295.15, 298.15, 301.15 and 305.15) K and at the atmospheric pressure. The ultrasonic velocity (u), density (ρ) and viscosity (η) for the binary mixture Ethyl acetate (1) + n-Octanol (2) were measured over the whole composition range at temperature T = (295.15, 298.15, 301.15 and 305.15) K and at the atmospheric pressure. Ethyl acetate is highly miscible with all organic compounds, such as glycol, ketones, alcohols and esters. It is most commonly used in mixtures with alcohols and a combination with 20% ethanol found application as an excellent solvent.

Free volume and internal pressure has gained significant interest by chemists, physicists and chemical engineers in past, as it provides a measure of explaining molecular interactions, internal structure, clustering phenomenon and dipolar interactions [6-8]. Free volume and internal pressure has been a subject of active interest among several researchers during recent past. Several attempts have been made by a number of investigators to calculate the internal pressure of liquids and liquid mixtures theoretically. The excess sound velocity (u^E), excess viscosity (η^E), excess free volume (V_f^E) and excess internal pressure (P_i^E) values have been interpreted in terms of the nature of intermolecular interactions between constituent molecules of mixtures [9-11].

2. Experimental Section

2.1. Chemicals

Ethyl acetate 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 T = (295.15, 298.15, 301.15 and 305.15) K were compared with value available in the literature[12-20] and are listed in table-2, were leads to a satisfactory agreement.

2.2. Apparatus and Procedure

Both two mixtures of Ethyl acetate 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 Detailed Description of chemical compounds used

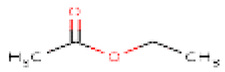
Chemical name	Formula	Structural Formula	Supplier	Mass Fraction Purity	Purification Method
Ethyl acetate	$C_4H_8O_2$		Merck	>99%	none
n-Octanol	$C_8H_{24}O$	CH_3-$(CH_3)_6$-CH_2OH	Merck	>99%	none

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 Atmospheric pressure.

Compound	**(ρ) / (g.cm^{-3})**		**u / (m.s^{-1})**		**η / (mPa s)**	
	expti.	lit.	expti.	lit.	expti.	lit.
Ethyl acetate	0.8820	0.8885[12]	1125	1115[21]	0.4402	0.4000[26]
		0.8941[13]		1138[22]		0.4570[16]
		0.8940[14]		1138.62[23]		0.4233[13]
		0.8943[15]		1144[16]		0.4280[14]
		0.8945[16]		NA		NA
n-Octanol	0.8242	0.8187[17]	1327	1330[24]	7.8512	7.6630[27]
		0.8220[18]		1346[18]		7.661[27]
		0.8216[19]		1347[25]		7.663[28]
		0.8217[20]		1347[20]		7.5981[29]

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^{0}C to 85^{0}C with an accuracy ± 0.1^{0}C.

2.3.2. Sound velocity

The ultrasonic velocity were 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 Ethyl acetate 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 ×10^{-3} m Pas. The measured values of viscosities of pure Ethyl acetate and octanol compare well with the corresponding literature values.

2.3.4. Theoretical

Liquid viscosity has been treated as free volume problem by a number of workers. Suryanarayana et al.[30] derived a formula for the free volume based on one dimensional analysis of the situation. When a ultrasonic wave passes through a liquid medium.

$$V_f = (M\,U / k\,\eta)^{3/2} \qquad (1)$$

Where, M is the molecular weight, u is the ultrasonic velocity, η is the viscosity, V_f , the free volume is in milliliters per mole and K is a constant, independent of temperature and it's value is 4.28 × 10^9 for all liquids.

Suryanarayana and Kuppuswami[31-32] suggested a method for evaluation of internal pressure from the knowledge of ultrasonic velocity, u, density and viscosity, the relation proposed is expressed as

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

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 have been calculated by using the following relation

$$A^E = A_{exp.} - (X_1 A_1 + X_2 A_2) \quad (3)$$

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

The experimental values of ultrasonic velocity (u), density (ρ) and viscosity (η) for the binary mixture Ethyl acetate (1) + n-Octanol (2) at temperature T = (295.15, 298.15, 301.15 and 305.15) K and at the atmospheric pressure, as a function of ethyl acetate mole fraction (x_1) have been reported in table-3 and excess properties are enlisted in table - 4. The results for the density of the mixture show that it increase with temperature and increase with ethyl acetate mole fraction, but the determined free volume increase with temperature and increase with ethyl acetate mole fraction. This indicates the complex formation and intermolecular weak association may be due to hydrogen bond formation[33].

Table 3 Values of density, sound velocity, viscosity, free volume and internal pressure properties for binary liquids mixture of ethyl acetate + n-octanol at different temperature.

Mole fraction of ethyl acetate (X_1)	Density (ρ) g.cm^{-3}	Sound velocity (u) m.s^{-1}	Viscosity (η) m.Pa.s	Free volume (V_f) m^{3}mol^{-1}	Internal pressure (p_i×10^4) N m^{-2}
At 295.15 K					
0.0000	0.8242	1327	7.8512	0.01165	0.66872
0.1056	0.8259	1312	4.7776	0.02292	0.55668
0.2095	0.8300	1294	3.2258	0.03838	0.49024
0.3174	0.8318	1275	2.2206	0.06206	0.43728
0.4286	0.8387	1239	1.5414	0.09634	0.37509
0.5083	0.8400	1225	1.2853	0.11928	0.38467
0.6196	0.8444	1214	0.9417	0.17562	0.35719
0.7090	0.8586	1192	0.8858	0.17723	0.37519
0.8064	0.8651	1164	0.6239	0.27175	0.34396
0.9044	0.8716	1148	0.5565	0.29587	0.35364
At 298.15 K					
0.0000	0.8242	1327	7.8512	0.01165	0.66872
0.1056	0.8259	1312	4.7776	0.02292	0.55668

0.2095	0.8300	1294	3.2258	0.03838	0.49024
0.3174	0.8318	1275	2.2206	0.06206	0.43728
0.4286	0.8387	1239	1.5414	0.09634	0.37509
0.5083	0.8400	1225	1.2853	0.11928	0.38467
0.6196	0.8444	1214	0.9417	0.17562	0.35719
0.7090	0.8586	1192	0.8858	0.17723	0.37519
0.8064	0.8651	1164	0.6239	0.27175	0.34396
0.9044	0.8716	1148	0.5565	0.29587	0.35364
1.0000	0.8820	1125	0.4402	0.38161	0.34494
0.0000	0.8225	1310	7.8502	0.01132	0.66572
0.1056	0.8232	1308	4.7745	0.02185	0.55452
0.2095	0.8316	1282	3.2212	0.03785	0.49002
0.3174	0.8386	1265	2.2156	0.06179	0.43652
0.4286	0.8452	1224	1.5315	0.09529	0.37450
0.5083	0.8462	1216	1.2752	0.17356	0.38356
0.6196	0.8536	1182	0.9325	0.17652	0.35625
0.7090	0.8623	1145	0.8832	0.27150	0.37425
0.8064	0.8726	1130	0.6186	0.29456	0.34285
0.9044	0.8821	1122	0.5521	0.29125	0.35225
1.0000	0.8895	1115	0.4830	0.38056	0.34332
At 301.15 K					
0.0000	0.8185	1308	7.8500	0.02132	0.66021
0.1056	0.8192	1306	4.5625	0.05412	0.60213
0.2095	0.8205	1290	3.2145	0.06524	0.54214
0.3174	0.8245	1250	2.2130	0.10231	0.41235
0.4286	0.8336	1225	1.5021	0.12561	0.40021
0.5083	0.8350	1201	1.2785	0.13524	0.38561
0.6196	0.8362	1189	0.9421	0.14286	0.37452
0.7090	0.8486	1145	0.8898	0.16542	0.36021
0.8064	0.8564	1110	0.6215	0.17451	0.35854
0.9044	0.8615	1105	0.5565	0.28523	0.35121
1.0000	0.8890	1103	0.4845	0.38098	0.34856
At 305.15 K					
0.0000	0.8172	1308	7.7721	0.02325	0.65215
0.1056	0.8200	1302	4.4445	0.06123	0.64214
0.2095	0.8236	1295	3.1952	0.06852	0.63541
0.3174	0.8257	1265	2.1956	0.11421	0.62589
0.4286	0.8324	1245	1.4852	0.12854	0.60213

0.5083	0.8421	1236	1.2045	0.13452	0.59457
0.6196	0.8498	1190	0.9410	0.14872	0.57461
0.7090	0.8542	1175	0.9345	0.15871	0.48521
0.8064	0.8612	1160	0.8562	0.16214	0.40125
0.9044	0.8745	1124	0.5210	0.24521	0.38785
1.0000	0.8856	1100	0.4810	0.38102	0.34125

Table 4 Excess values of sound velocity (u^E), viscosity (η^E), free volume (V_f^E) and internal pressure (P_i^E) properties for binary liquids mixture of ethyl acetate + n-octanol at different temperature.

Mole fraction of ethyl acetate (X_1)	**Excess sound velocity (u^E)**	**Excess Viscosity (η^E) N Sm^{-2}**	**Excess Free volume (V_f^E) m^{3}mol^{-1}**	**Excess internal pressure ($p_i^E \times 10^4$) N m^{-2}**
At 295.15 K				
0.0000	0.00	0.0000	0.0000	0.0000
0.1056	+6.36	-2.2921	-0.0278	-0.0778
0.2095	+9.36	-3.0735	-0.0508	-0.1106
0.3174	+12.14	-3.2790	-0.0670	-0.1286
0.4286	+13.42	-3.4681	-0.0738	-0.1548
0.5083	+14.73	-4.2162	-0.0804	-0.1694
0.6196	+12.18	-2.7995	-0.0967	-0.1194
0.7090	+8.25	-2.3178	-0.0652	-0.1109
0.8064	+5.07	-1.7108	-0.0503	-0.0632
0.9044	+3.72	-1.1250	0.0315	-0.0222
1.0000	0.00	0.0000	0.0000	0.0000
At 298.15 K				
0.0000	0.000	0.0000	0.0000	0.0000
0.1056	+6.38	-2.8542	-0.0352	-0.0845
0.2095	+10.45	-3.2145	-0.0452	-0.1002
0.3174	+11.54	-4.7852	-0.0640	-0.1258
0.4286	+12.84	-5.2142	-0.0854	-0.1387
0.5083	+13.45	-5.8989	-0.0974	-0.1542
0.6196	+15.87	-4.8745	-0.0810	-0.1701
0.7090	+13.54	-3.4578	-0.0745	-0.1542
0.8064	+12.45	-2.9856	0.0578	-0.1365
0.9044	+6.85	-1.8745	-0.0381	-0.1025
1.0000	0.00	0.0000	0.0000	0.000
At 301.15 K				
0.0000	0.000	0.0000	0.0000	0.0000
0.1056	+6.12	-2.8745	-0.0385	-0.0945
0.2095	+9.78	-3.4217	-0.0487	-0.1065
0.3174	+10.25	-4.9874	-0.0578	-0.1285
0.4286	+11.85	-5.023	-0.0687	-0.1475

0.5083	+13.45	-4.5235	-0.0789	-0.1700
0.6196	+14.45	-3.7412	-0.0954	-0.1523
0.7090	12.85	-3.1245	-.07998	-0.1278
0.8064	+10.45	-2.4521	-0.0612	-0.1085
0.9044	+0.747	-1.2584	-0.0421	-0.8524
1.0000	0.00	0.000	0.0000	0.0000
At 305.15 K				
0.0000	0.00	0.0000	0.0000	0.0000
0.1056	+6.05	-2.9745	-0.0452	-0.8798
0.2095	+8.65	-3.5689	-0.0542	-0.1085
0.3174	+10.32	-4.8752	-0.0651	-0.1278
0.4286	+12.45	-59874	-0.0796	-0.1387
0.5083	+14.75	-6.4521	-0.0985	-0.1545
0.6196	12.02	-4.2153	-0.0845	-0.1725
0.7090	+10.75	-3.4541	-0.0641	-0.1463
0.8064	+0.8421	-2.4215	-0.0521	-0.1287
0.9077	+0.6887	-1.8542	-0.0410	-0.0978
1.0000	0.00	0.000	0.0000	0.0000

For the binary liquid mixture Ethyl acetate (1) + n- Octanol (2), the obtained excess sound velocity (u^E) values are positive over the whole composition range at the different temperature T = (295.15, 298.15, 301.15 and 305.15)K as depicted in figure-1.

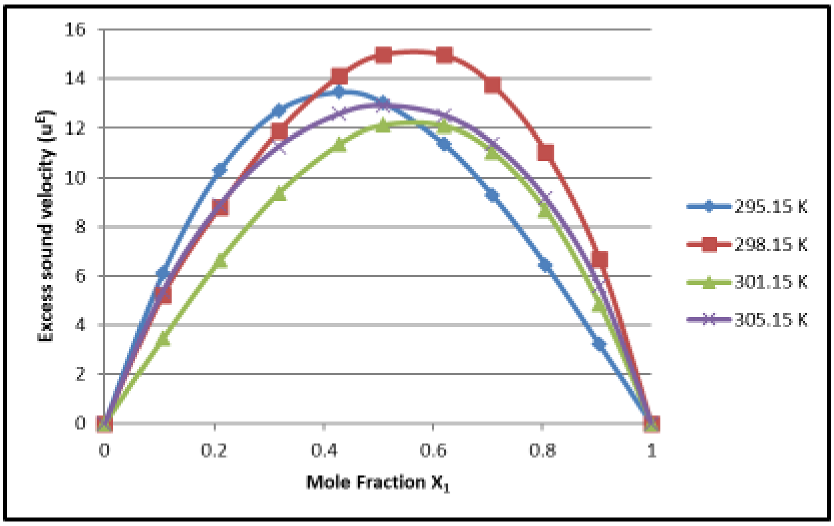


Figure 1 Curves of excess sound velocity u^E against the mole fraction of Ethyl acetate x_1, for the binary mixture (Ethyl acetate (1) + n- Octanol (2)) at different temperatures (blue ⧫, 295 K orange ■, 298.15 K, gray▲, 301.15 K and ×, 305.15K). The solid lines represent the values calculated from the Redlich–Kister equation.

The ultrasonic velocity in a mixture is mainly influenced by its molecular property. The results for the excess sound velocity (u^E) plotted in figure-1 are positive for all the four temperature studied. The observed positive trends in excess sound velocity indicate that the effect due to the breaking up of self-associated structure of the components of the mixtures is dominant over the effect of hydrogen bonding and dipole-dipole interaction between unlike molecule. The positive values of excess sound velocity (u^E) increase with the increase in temperature which indicates the increase in strength of interaction with all four temperatures in the mixture. The higher positive values of excess sound velocity (u^E) are observed at 298.15K.The positive excess sound velocity (u^E) clearly suggests that there exist strong molecular interaction between the molecules of all the four temperatures.

The viscosities of binary liquid mixture Ethyl acetate (1) + n- Octanol (2) at temperature T = (295.15, 298.15, 301.15 and 305.15) K decrease linearly with increase in mole fraction of ethyl acetate. Excess viscosity (η^E) values are negative for the all four temperature T = (295.15, 298.15, 301.15 and 305.15) K over the whole mole fraction range (Figure-2).

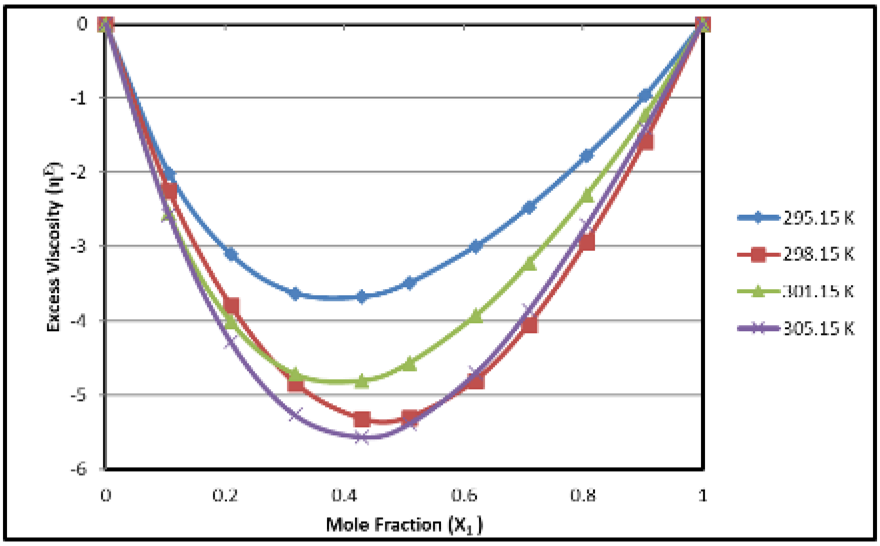


Figure 2. Curves of excess viscosity η^E against the mole fraction of Ethyl acetate x_1, for the binary mixture (Ethyl acetate (1) + n-Octanol (2)) at different temperatures (blue ♦, 295 K orange ■, 298.15 K, gray▲, 301.15 K and ×, 305.15K). The solid lines represent the values calculated from the Redlich–Kister equation.

The excess viscosity values, which represent the deviation from rectilinear dependence of η_{exp} of binary mixture on mole fraction, have been used to explain the mixture component's intermolecular interaction. The relatively large η^E values at the initial stages indicated intermolecular hydrogen bonding among the binary mixture components. The negative magnitude of η^E values also confirmed the non-existence of charge transfer or the absence of robust hydrogen bond interactions that would result in complexation between the binary mixture components. The negative η^E values might indicate that the average degree of cross-association between alkan-1-ol and ethyl acetate gradually decreased as the chain length of alkan-1-ol increased. Thus, the larger negative deviation for the system containing longer chain alkan-1-ol confirmed strong dispersion forces in this system. It can be seen from figure-2 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[34,35], have reported similar behaviour where negative value of $\Delta\eta$ indicates dispersive interaction. The negative values of excess viscosity (η^E) observed in Ethyl acetate (1) + n-Octanol (2) mixture indicate the presence of strong inter molecular interaction amongst the mixing components. The values of excess viscosity (η^E) for all the four temperatures studied are indicative of the predominance of dispersion forces.

The definition of free volume given by Eyring and Hirschfelder reveals that the deviation in free volume from ideal behaviour is indicative of molecular interaction between molecules of the components. The thermodynamic properties of mixtures have been interpreted in many ways. These are mainly hydrogen bonding, dipole-dipole moment, charge transfer, molecular association etc. which make a negative contribution to the excess value of various thermodynamic parameters. These negative values indicate strong interaction between the components of the mixture. Dispersion forces and dilution effects make a positive contribution to the excess value which is indicative of weaker interaction. More than one type of interaction may be concerned in any given system. Fort and Moore, however, explained the behaviour of liquid mixtures on the basis of the excess compressibility for the systems he studied. In the present work, the excess free volume is used. The results have been presented in the form of graphs in Figure 3, where excess free volumes for the systems have been plotted against the mole fraction of one of the components of the mixture.

The results of excess free volume (V_f^E) verses mole fraction (x_1) exhibit negative deviations over the entire composition range of ethyl acetate and all four temperature T = (295.15, 298.15, 301.15 and 305.15) K as shown in figure-3. Negative excess free volume (V_f^E) leads to reduction in volume. This may be due to the formation of new bonds. However negative excess free volume (V_f^E) values for binary mixture of Ethyl acetate (1) + n- Octanol (2) cannot be assigned to molecular complexation. These negative value are accounted on the fact that with increasing alkanol size, interstial accommodation becomes increasingly important and hence excess free volume (V_f^E) become negative for longer n-alkanol[36]. The values of excess free volume (V_f^E) indicates the contributions made by the strong dipole-dipole interaction between the unlike molecule of the component[37].

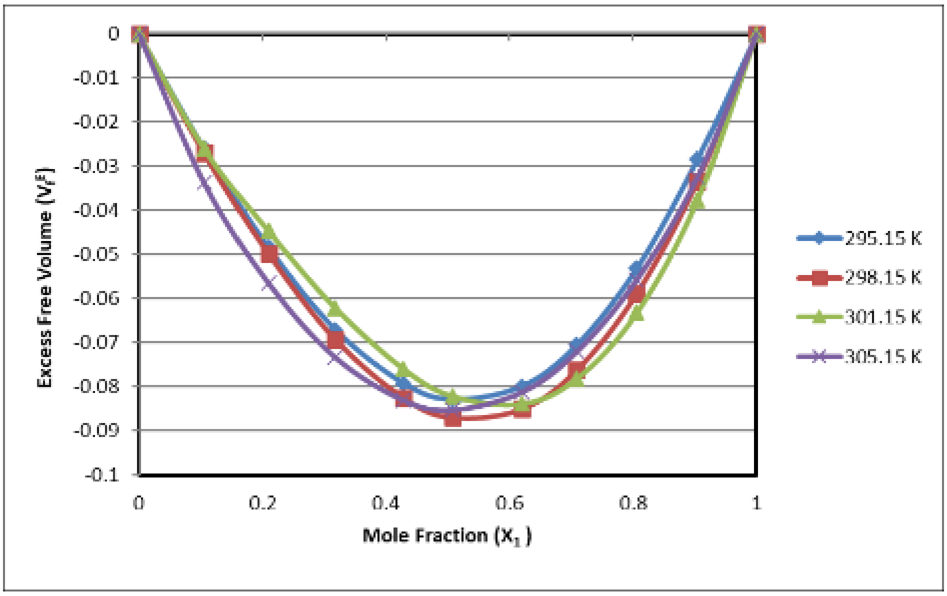


Figure 3 Curves of excess free volume (V_f^E) against the mole fraction of Ethyl acetate x_1, for the binary mixture (Ethyl acetate (1) + n- Octanol (2)) at different temperatures (blue ⧫, 295 K orange ■, 298.15 K, gray▲, 301.15 K and ×, 305.15K). The solid lines represent the values calculated from the Redlich–Kister equation.

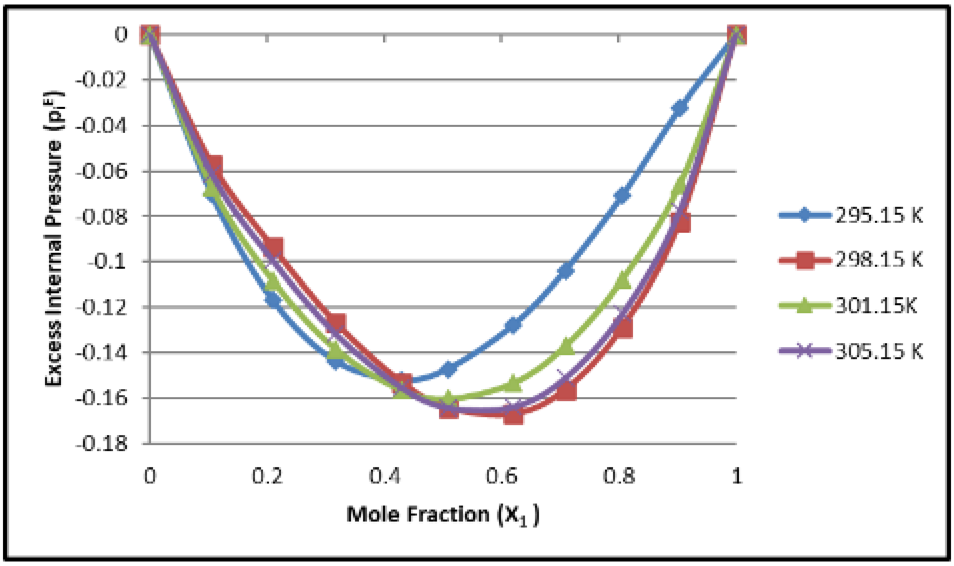


Figure 4 Curves of excess internal pressure (p_i^E) against the mole fraction of Ethyl acetate x_1, for the binary mixture (Ethyl acetate (1) + n- Octanol (2)) at different temperatures (blue ⧫, 295 K orange ■, 298.15 K, gray▲, 301.15 K and purple ×, 305.15K). The solid lines represent the values calculated 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. The excess internal pressure (P_i^E) of a liquid mixture from linearly, reflects changes of structure and cohesive forces during the mixing process. Excess values of internal pressure (P_i^E) are negative over the entire composition range and for all four temperature T = (295.15, 298.15, 301.15 and 305.15) K. The results have been presented in the form of graphs in figure 4, where excess internal pressure for the systems has been plotted against the mole fraction of one of the components of the mixture. The results of excess internal pressure (P_i^E) verses mole fraction (x_1) exhibit negative deviations over the entire composition range of ethyl acetate and all four temperature T = (295.15, 298.15, 301.15 and 305.15) K as shown in figure-4.

Figure 4, indicates weak interaction between the components of the mixture. In the Ethyl acetate (1) + Octanol (2) mixture internal pressure decrease with increase in mole fraction of ethyl acetate (x_1) which indicate reduction in the intermolecular interaction. This negative trend in (P_i^E) indicates that the only dispersion and dipolar forces operating with complete absence of specific interaction. It shows the increasing magnitude of interaction between the Ethyl acetate (1) + n-Octanol (2).

4. Conclusion

In this paper the ultrasonic velocity (u), density (ρ) and viscosity (η) have been measure over the whole composition range at temperature T = (295.15, 298.15, 301.15 and 305.15) K for the binary mixture Ethyl acetate (1) + n-Octanol (2). Excess sound velocity, deviations in viscosity, excess free volume and excess internal pressure for binary mixtures have been calculated and fitted to a Redlich–Kister equation. It is obvious that, there exist a molecular interaction between the components of the mixture. In specific weak molecular interaction like dipole-dipole, dipole-induced dipole and dispersive forces are found to exist between the components of the individual mixtures.

Compliance with ethical standards

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