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Sound Velocity, Densities and Excess Parameter Studies on 1, 4-Dioxane with 1-Hexanol at the Temperature T= (298.15 To 318.15) K

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ABSTRACT: Sound velocity, densities of binary mixture of 1,4-dioxane with 1-hexanol has been measured over the entire range of composition using Anton Paar at (T=298.15, 303.15, 308.15, 313.15 & 318.15) K. The Excess parameters viz., deviations in isentropic compressibility (ΔK_s), excess molar volumes (V_m^E), excess free length (L_f^E), excess acoustic impedance (Z^E) , excess sound velocity (u^E) are deduced from experimental values and discussed intermolecular interactions present in the mixture. At the end, all the parameters have been fitted to Redlich-Kister equation and their coefficients are obtained.

KEY WORDS: Sound velocity; Densities; Excess parameters; 1,4-dioxane ; 1-hexanol

I. INTRODUCTION

Day to day knowledge of thermodynamic and transport properties liquid/liquid mixtures are essential for the proper design of any chemical industrial processes. 1,4-dioxane is used in polymerization processes is a cyclic ether, an excellent solvent very often used in the manufacture of special chemicals, pesticides, bulk drug intermediates and 1-hexanol is a substance heavily used in the pharmaceutical industry as a solvent, an extraction agent, in processes that produce medicine flavors and in wastewater treatment [1,2]. According to Lagemann and Dunbar et al [3] pointed out the sound velocity approach for the qualitative estimation of interaction in liquids. A parallel measurement of sound velocity, density of liquid mixtures allows one to obtain information about their excess compressibility, volume, free length, acoustic impedance, internal pressure and changes in their properties. Complex formations, formation of hydrogen bond, Dipole – dipole, dipole- induced dipole interactions in solutions and their effect in physical properties of the mixture have received much attention. Also, Romero et al [4] measured speeds of sound of the binary mixtures of 1,3-dioxolane (or 1,4-dioxane) + cyclopentane (or cyclohexane, or benzene) at 283.15, 298.15, and 313.15 K. The excess isentropic compressibilities were calculated from experimental data and fitted with a Redlich-Kister polynomial function. Results were analyzed taking into account molecular interactions and structural effects in the mixtures.

In this paper, sound velocity, density of binary mixture 1,4-dioxane with hexnaol have been measured at (T=298.15, 303.15, 308.15, 313.15 & 318.15) K over the entire range of composition using Anton Paar DSA 5000M. From the experimental values, deviations in isentropic compressibility (ΔK_s), excess molar volumes (V_m^E), excess free length (L_f^E), excess acoustic impedance (Z^E), and excess sound velocities (u^E) for the binary system are estimated using standard equations that are reported by several authors [5-9]. The results obtained were fitted to Redlich–Kister polynomial the results were discussed in terms of molecular interactions.

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II. EXPERIMENTAL

Materials & Measurements: The measurements were performed using the sound analyzer incorporated in the Anton Paar device, model DSA 5000M, equipped with the machine sampler SP-1m (Anton Paar; carousel with 24 vials, 55 cm^3 each). All controls, adjustments, and checks were done using manufacturer's software system put in within the device. A laptop connected to the U tube densimeter enabled us to read the raw data from the device memory and to perform the ensuing analysis. Mole fractions of these samples were determined by measuring the mass of each component with a precision balance (Sartorius, model CP 225D, +/-0.01mg).

1,4-dioxane and 1-hexanol with mass fraction purities >0.998 were purchased from Aldrich Chemical Co. the chemicals were kept in airtight stopped glass bottles were performed in an isothermal mode; that is, the measurements of all prepared solution were done at the similar explicit temperature, then the temperature was changed and the measurements were perennial. The instrument in calibrated with the double distilled water (as suggested by Anton-Paar) for sound velocity and density at room temperature. The accuracy of density measurements was $5*10^{-6}$ $g\cdot cm^{-3}$, and also the temperature of the equipment was maintained constant to within ± 0.01 K.

Table 1 Comparison of Experimental sound velocity, density of Pure Liquids with Literature values at (298.15, 303.15, 308.15, 313.15, and 318.15) K

Compound	T (K)	u (ms^{-1})		ρ (kgm^{-3})	
		Observed	Literature	Observed	Literature
1,4-dioxane	298.15	1344.3	1344.8 [10,11]	1026.8	1027.9 [10,12]
	303.15	1322.3		1021.2	1022.5 [13,14]
	308.15	1300.5	1325 [15,16]	1015.5	1016.8 [17]
	313.15	1278.6	1279.8 [10]	1011.8	1011.3 [13,14]
	318.15	1257.3		1004.1	1005.1 [18,19]
1-hexanol	298.15	1303.7	1303.0 [20]	816.1	816.2 [19]
	303.15	1286.7	1285.6 [20]	812.5	811.6 [22,23]
	308.15	1269.8		808.8	808.0 [23]
	313.15	1253.1		805.2	804.6 [24]
	318.15	1236.4		801.4	

III. THEORY

Using the measured values of data, we can calculate the various thermo acoustical parameters such as Isentropic compressibility $K_s = \frac{1}{\rho U^2}$ (1)

Molar volume $\overline{V} = \frac{\overline{M}}{\rho}$ (where $\overline{M} = M_1X_1 + M_2X_2$) (2)

Free length $L_f = K(\beta_{ad})^{1/2}$ (where K is Jacobson's constant) (3)

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Specific acoustic impedance $Z = U \rho$ (4)

The strength of interaction between the component molecules of binary mixtures is well reflected in the deviation of the excess functions from ideality. The excess thermodynamic properties such as ΔK_s, V_m^E, L_f^E, Z^E and u^E have been calculated using the following equation

$$Y^E = Y_{mix} - (x_1 y_1 + x_2 y_2) \quad (5)$$

where x_1 and x_2 are mole fractions of 1,4-dioxane and 1-hexanol respectively.
Further, the excess parameters also fitted to Redlich – Kister polynomial equation to estimate the adjustable parameters.

$$Y^E = x_1 x_2 \sum_{i=0}^{n} a_i (1-2x)^i \quad (6)$$

by the least-squares regression method, the co-efficients a_i were obtained by fitting above equation to the experimental values. The optimum number of coefficients is ascertained from an examination of the variation in standard deviation (σ)

$$\sigma(Y) = \left[\frac{\sum (Y_{\text{expt}} - Y_{calc})^2}{N-n} \right]^{1/2} \quad (7)$$

(N is the number of data points and n is the degree of fitting i.e., number of coefficients)

Table 2(a)
Sound velocity (u), Densities (ρ), deviations in isentropic compressibility (ΔK_s), excess molar volumes (V_m^E), excess free length (L_f^E), excess acoustic impedance (Z^E) and excess sound velocity (u^E) at 298.15 to 318.15 K for 1,4-dioxane (1) +1-hexanol (2) system

x_1	u ms^{-1}	ρ kgm^{-3}	ΔK_s*10^{-10} m^2N^{-1}	$V_m^E*10^{-4}$ $m^3\,mol^{-1}$	L_f^E $*10^{-11}$ m	Z^E*10^6 $kgm^{-2}s^{-}$	u^E ms^{-1}
				298.15 K			
0.0000	1303.7	816.1	0.0000	0.0000	0.0000	0.0000	0.0000
0.1327	1300.3	834.2	0.1214	0.0100	0.0530	-0.0211	-8.8064
0.2630	1298.5	854.6	0.2104	0.0171	0.0929	-0.0377	-15.8759
0.3906	1299.6	876.5	0.2538	0.0211	0.1141	-0.0481	-19.7904
0.4903	1302.8	895.9	0.2617	0.0224	0.1195	-0.0521	-20.9677
0.5947	1307.2	917.7	0.2493	0.0218	0.1158	-0.0524	-20.6385
0.6848	1312.3	938.7	0.2219	0.0197	0.1046	-0.0487	-18.9746
0.7732	1319.4	960.7	0.1783	0.0160	0.0854	-0.0410	-15.8356
0.8541	1327.0	982.9	0.1231	0.0112	0.0601	-0.0298	-11.4212
0.9273	1335.0	1004.5	0.0630	0.0059	0.0315	-0.0163	-6.1503
1.0000	1344.3	1026.8	0.0000	0.0000	0.0000	0.0000	0.0000
				303.15 K			
0.0000	1286.7	812.5	0.0000	0.0000	0.0000	0.0000	0.0000
0.1327	1283.1	830.5	0.1229	0.0100	0.0531	-0.0202	-8.3291
0.2630	1281.0	850.6	0.2133	0.0172	0.0932	-0.0362	-15.0368
0.3906	1281.8	872.2	0.2584	0.0213	0.1148	-0.0462	-18.8090
0.4903	1284.4	891.5	0.2666	0.0225	0.1203	-0.0501	-19.9024
0.5947	1288.3	913.1	0.2533	0.0219	0.1162	-0.0502	-19.4820

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0.6848	1293.3	933.8	0.2245	0.0197	0.1046	-0.0466	-17.8088
0.7732	1299.4	955.7	0.1801	0.0160	0.0852	-0.0392	-14.8252
0.8541	1306.4	977.7	0.1250	0.0112	0.0602	-0.0285	-10.7420
0.9273	1313.8	999.1	0.0649	0.0059	0.0318	-0.0156	-5.8670
1.0000	1322.3	1021.2	0.0000	0.0000	0.0000	0.0000	0.0000
308.15 K							
0.0000	1269.8	808.8	0.0000	0.0000	0.0000	0.0000	0.0000
0.1327	1266.0	826.6	0.1253	0.0101	0.0536	-0.0194	-7.9554
0.2630	1263.5	846.5	0.2173	0.0172	0.0939	-0.0348	-14.3064
0.3906	1263.9	868.0	0.2636	0.0213	0.1157	-0.0444	-17.8884
0.4903	1266.0	887.1	0.2724	0.0226	0.1214	-0.0482	-18.9430
0.5947	1269.4	908.5	0.2593	0.0220	0.1175	-0.0483	-18.5630
0.6848	1273.9	929.0	0.2302	0.0198	0.1058	-0.0449	-16.9819
0.7732	1279.4	950.7	0.1849	0.0161	0.0863	-0.0377	-14.1427
0.8541	1285.8	972.4	0.1285	0.0113	0.0609	-0.0274	-10.2479
0.9273	1292.6	993.7	0.0667	0.0059	0.0322	-0.0150	-5.5961
1.0000	1300.5	1015.5	0.0000	0.0000	0.0000	0.0000	0.0000

Table 2(b)
Sound velocity (u), Densities (ρ), deviations in isentropic compressibility (ΔK_s), excess molar volumes (V_m^E), excess free length (L_f^E), excess acoustic impedance (Z^E) and excess sound velocity (u^E) at 298.15 to 318.15 K for 1,4-dioxane (1) +1-hexanol (2) system

x_1	u ms^{-1}	ρ kgm^{-3}	ΔK_s*10^{-10} m^2N^{-1}	$V_m^E*10^{-4}$ $m^3\ mol^{-1}$	L_f^E $*10^{-11}$ m	Z^E*10^6 $kgm^{-2}s^{-1}$	u^E ms^{-1}
313.15 K							
0.0000	1253.1	805.2	0.0000	0.0000	0.0000	0.0000	0.0000
0.1327	1248.9	822.8	0.1272	0.0101	0.0537	-0.0187	-7.5149
0.2630	1246.1	842.5	0.2223	0.0173	0.0949	-0.0335	-13.6688
0.3906	1246.0	863.7	0.2689	0.0214	0.1166	-0.0427	-16.9782
0.4903	1247.7	882.6	0.2775	0.0226	0.1220	-0.0462	-17.9118
0.5947	1250.6	903.8	0.2648	0.0221	0.1183	-0.0463	-17.5746
0.6848	1254.6	924.2	0.2359	0.0199	0.1068	-0.0430	-16.1273
0.7732	1259.2	945.6	0.1896	0.0162	0.0871	-0.0362	-13.4210
0.8541	1265.3	967.2	0.1308	0.0113	0.0611	-0.0262	-9.6208
0.9273	1271.6	988.2	0.0666	0.0059	0.0318	-0.0142	-5.1214
1.0000	1278.6	1009.8	0.0000	0.0000	0.0000	0.0000	0.0000
318.15 K							
0.0000	1236.4	801.4	0.0000	0.0000	0.0000	0.0000	0.0000
0.1327	1231.9	818.9	0.1308	0.0101	0.0546	-0.0180	-7.2538
0.2630	1228.8	838.4	0.2278	0.0174	0.0961	-0.0323	-13.0915
0.3906	1228.2	859.4	0.2773	0.0215	0.1186	-0.0412	-16.3815
0.4903	1229.5	878.2	0.2870	0.0227	0.1244	-0.0446	-17.3200
0.5947	1231.8	899.1	0.2734	0.0222	0.1203	-0.0447	-16.9275
0.6848	1235.3	919.3	0.2431	0.0200	0.1084	-0.0415	-15.4619

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0.7732	1239.7	940.5	0.1957	0.0162	0.0886	-0.0349	-12.8908
0.8541	1244.9	961.9	0.1367	0.0113	0.0627	-0.0254	-9.3880
0.9273	1250.6	982.7	0.0716	0.0059	0.0334	-0.0139	-5.1758
1.0000	1257.3	1004.1	0.0000	0.0000	0.0000	0.0000	0.0000

Table 3
Coefficients (a_i) and standard deviation (σ) for 1,4-dioxane (1) + 1-hexanol (2)

Property	T (K)	a_0	a_1	a_2	a_3	a_4	σ (10^2)
	298.15	1.0456	0.0778	0.1007	-0.0508	-0.2810	0.2007
	303.15	1.0651	0.0875	0.0556	-0.0735	-0.2143	0.1619
ΔK_s (10^{-10} m^2N^{-1})	308.15	1.0884	0.0785	0.0588	-0.0650	-0.2158	0.1425
	313.15	1.1091	0.0768	0.1099	-0.0619	-0.3264	0.2188
	318.15	1.1468	0.0769	0.0531	-0.0870	-0.2099	0.1671
	298.15	0.0896	-0.0047	0.0028	0.0054	-0.0092	0.0173
	303.15	0.0900	-0.0041	0.0020	0.0043	-0.0090	0.0172
V_m^E (m^3 mol^{-1})	308.15	0.0903	-0.0044	0.0019	0.0057	-0.0084	0.0186
	313.15	0.0906	-0.0048	0.0036	0.0057	-0.0112	0.0162
	318.15	0.0910	-0.0045	0.0033	0.0054	-0.0121	0.0146
	298.15	0.4784	-0.0033	0.0452	-0.0191	-0.1108	0.0798
	303.15	0.4812	0.0014	0.0291	-0.0285	-0.0879	0.0670
L_f^E (10^{-11} m)	308.15	0.4858	-0.0023	0.0298	-0.0230	-0.0859	0.0556
	313.15	0.4884	-0.0027	0.0499	-0.0216	-0.1299	0.0883
	318.15	0.4979	-0.0016	0.0282	-0.0331	-0.0843	0.0650
	298.15	-0.2093	0.0377	-0.0217	0.0015	0.0315	0.0238
	303.15	-0.2010	0.0350	-0.0173	0.0030	0.0263	0.0230
Z^E (10^6 $kgm^{-2}s^{-1}$)	308.15	-0.1934	0.0342	-0.0166	0.0020	0.0257	0.0211
	313.15	-0.1853	0.0316	-0.0213	0.0023	0.0348	0.0274
	318.15	-0.1792	0.0306	-0.0146	0.0031	0.0229	0.0214
	298.15	-84.0147	6.0595	-12.2046	7.9790	22.2153	17.8591
	303.15	-79.7178	4.1994	-7.4132	9.9613	14.8167	9.0017
u^E (ms^{-1})	308.15	-75.8826	4.3843	-7.2660	8.6973	13.7777	8.2718
	313.15	-71.7440	3.8471	-11.6603	7.8407	23.3737	13.2685
	318.15	-69.3653	3.1780	-5.7993	10.0204	10.8547	7.6972

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IV. RESULTS AND DISCUSSION

The values of sound velocities and densities for pure liquids are experimentally measured and are compared with the literature values and they are good agreement with each other as given in the table-1. The experimental data related to ΔK_s, V_m^E, L_f^E, Z^E and u^E for the binary liquid mixture at different temperatures are given in tables-2 (a), 2 (b). Redlich-Kister coefficients of the various parameters at different temperatures with respective standard deviations are reported in table- 3.

Deviation in Isentropic Compressibility (ΔK_s)

The values of deviation in isentropic compressibility for 1, 4-dioxane + 1-hexanol binary mixture studied at five different temperatures are given in tables 2 (a), 2 (b).The corresponding plots of deviation in isentropic compressibility against mole fraction of 1, 4-dioxane are plotted in Fig 1. The ΔK_s plots are more positive at intermediate composition x_1 =0.5. The positive values of ΔKs suggest that the mixture is more compressible than the corresponding ideal mixture. This leads decrease of strength of the interactions between component molecules in the mixture.

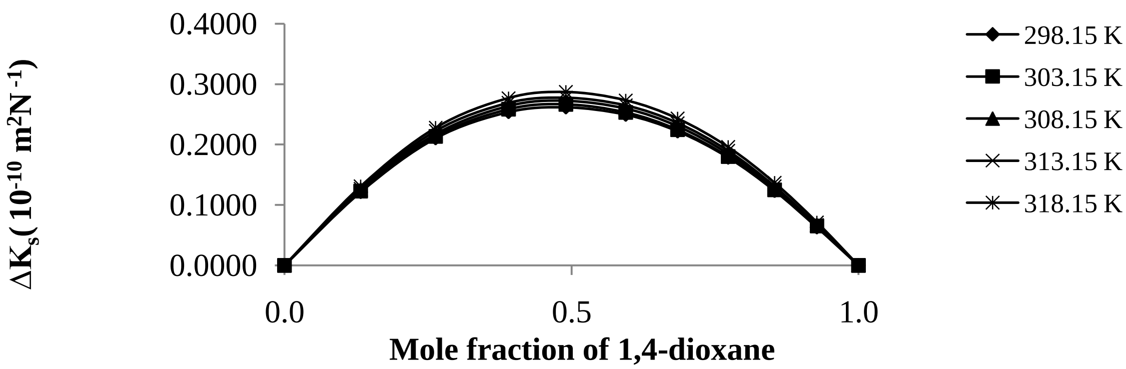

Fig. 1 - Variation of deviations in isentropic compressibility ΔK_s with mole fraction of 1,4-dioxane + pentanol system at different temperatures

Excess molar volume (V_m^E)

Fig.2 shows the excess molar volumes V_m^E in the case of 1, 4-dioxane +1-hexanol are found to be more positive at x_1 = 0.5 at all temperatures studied. The positive values of excess molar volumes suggest that the mixture prefers to have loose structure than compact structure in the solutions [25] i.e. weak intermolecular interactions are present in the mixtures.

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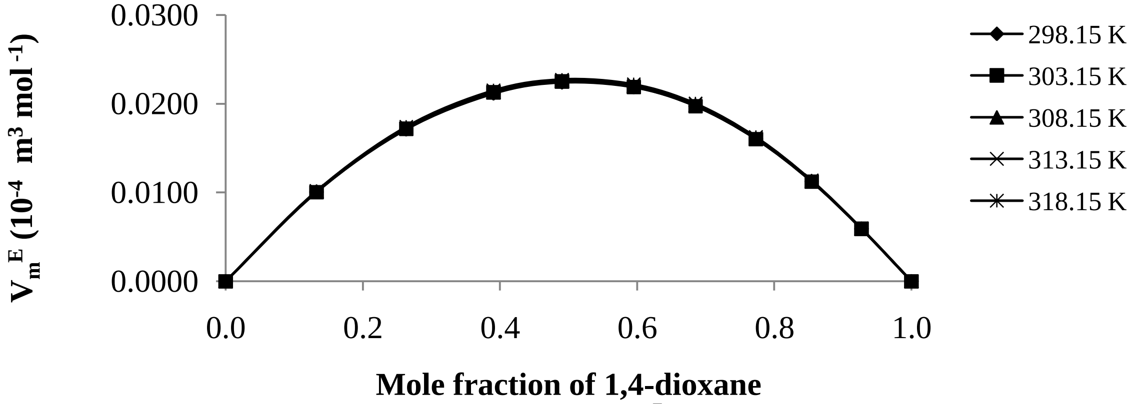

Fig. 2 - Variation of excess molar volumes V_m^E with mole fraction of 1,4-dioxane + pentanol system at different temperatures

Excess free length (L_f^E)

The values of 'L_f^E' for 1,4-dioxane +1-hexanol binary mixture studied at five different temperatures are presented in tables 2(a), 2(b) respectively, and there corresponding plots against mole fraction of 1,4-dioxane are shown in Fig. 3 shows more positive values at x_1=0.5over the entire range of composition of 1,4-dioxane at all five temperatures studied. According to Wankhede [26] the positive values of L_f^E is an indication of dispersive forces are operating in the liquid mixtures and it is showing the presence of weak interactions in the mixture .This also reveals the presence of week interactions in these mixtures.

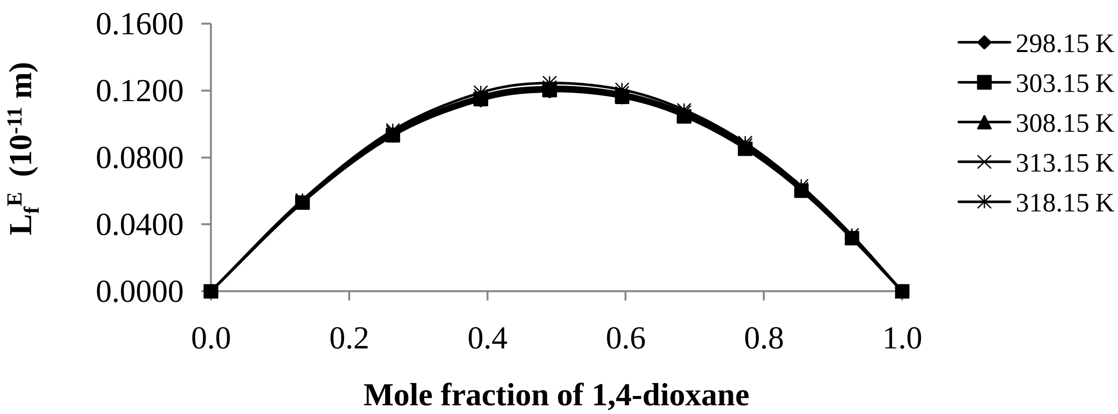

Fig. 3 - Variation of excess free length L_f^E with mole fraction of 1,4-dioxane + pentanol system at different temperatures

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Excess Acoustic Impedance (Z^E)

Further, the calculated values of excess acoustic impedance (Z^E) of binary mixture at all temperatures studied are negative. The corresponding plots are shown in fig- 4 shows; the excess acoustic impedance becomes more negative at 0.5947 mole fraction of the binary mixture. The negative values Z^E [6,27] suggests that breaking of hydrogen bond in 1-hexanol up to 0.5947 and after that it decreases and hence it leads to close packing of the structure. This shows weak molecular interactions between the components of the mixture exists.

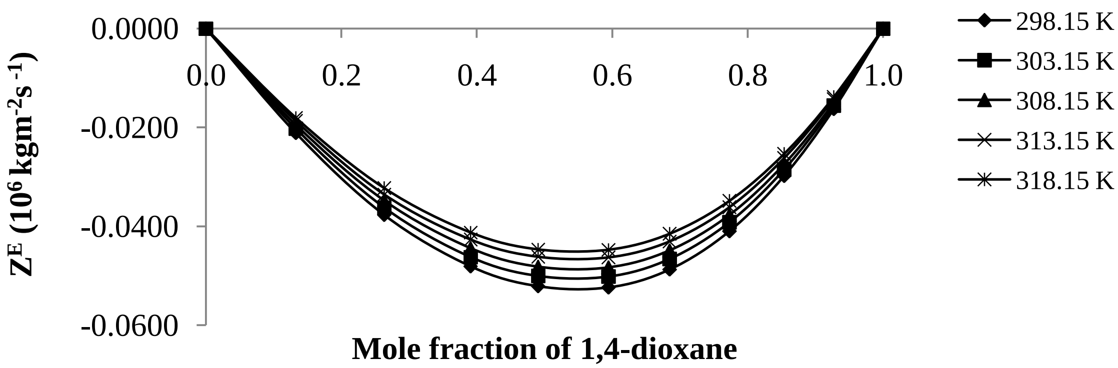

Fig. 4 - Variation of excess acoustic impedance Z^E with mole fraction of 1,4-dioxane + pentanol system at different temperatures

Excess Ultrasonic Velocity (u^E)

Figs 4,5 show that behaviors of Z^E and u^E support each other both exhibit negative deviations over the entire composition range of 1,4-dioxane in the mixture at all five temperatures. The negative deviations suggested dispersion forces are operative in the system. Similar observations are reported by A Ali et al [8] on the binary mixtures 2,2,4-trimethyl pentane with n-hexane and cyclohexane.

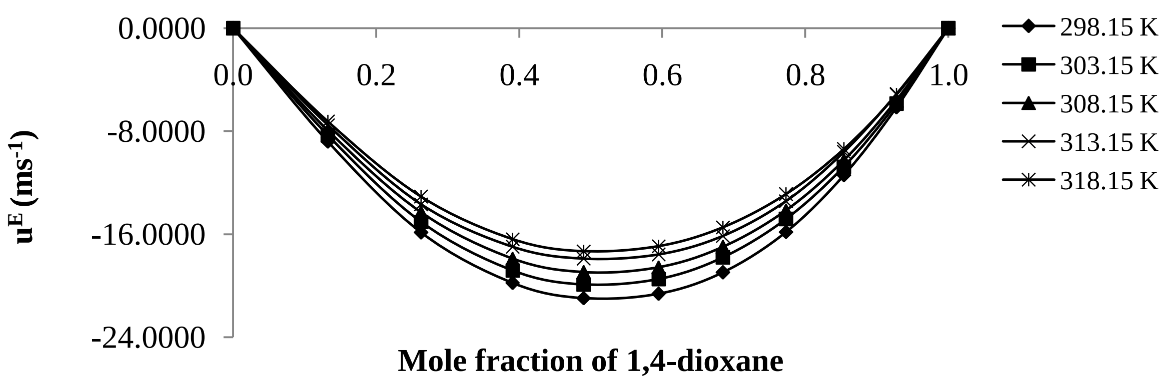

Fig. 5 - Variation of excess ultrasonic velocity with mole fraction of 1,4-dioxane + pentanol system at different temperatures

On close observation of Table 2(a), 2(b) the values of ΔK_s and V_m^E are positive at all temperatures studied. An increase in ΔK_s denotes highly compressible, weakening of inter-molecular interactions i.e., the hetro molecular (1,4-dioxane + 1-

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hexanol) not only disturb the homo molecular (1,4-dioxane molecules), (1-hexanol molecules) interactions in components liquids but also causes a re-arrangement in the geometry of the clusters of molecules in such a way volume of the mixture i.e., excess molar volume would be positive [28]. The sign and magnitude of ΔK_s and V_m^E play a vital role in assessing the molecular interactions in the liquid mixtures. In general negative values of ΔK_s and V_m^E indicates strong interaction in the mixture which include charge-transfer, diploe-dipole, dipole-induced dipole interactions and interstitial accommodation of the smaller molecules into the spaces created by bigger molecules, while positive signs of these parameters are indicative of weakening of interactions between the component molecules [29].

Also, the values of ΔK_s & L_f^E (Fig 1,3) are found to be positive over the entire range of mole fraction of 1,4-dioxane in the mixture at all studied temperatures. The sign of excess properties plays a vital role in assessing the compactness or extent of molecular interactions. The various types of interactions that are operating between the molecules are dispersion forces, which should make a positive contribution to excess values and charge transfer, H-bonding, dipole-dipole interaction and dipole-induced dipole interactions expected to make positive contributions. In the present mixture as ΔK_s & L_f^E are positive suggesting dispersive forces are present in the mixture [26].

V. CONCLUSION

Sound velocity and density for binary mixture consist of 1,4-dioxane with 1-hexanol system are measured at T=(298.15 to 318.15) K using Anton-Paar. The calculated excess parameters are discussed and concluded the presence of weak dispersion forces in the mixture.

VI. ACKNOWLEDGEMENT

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