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Study of ultrasonic and volumetric properties on non-aqueous-non electrolyte liquid mixtures

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Abstract
The ultrasonic velocities and densities for (MA or EA or BA or EHA + DMF) binary mixtures are measured and the values of Δu, V_m^E, Δk_s, L_f^E and Z^E are calculated. V_m^E, Δk_s and L_f^E values are positive where as Δu and Z^E values are negative in all the binary mixtures, indicating dispersion forces between the acrylate and DMF molecules and it follows the order MA+DMF>EA+DMF>BA+DMF>EHA>DM.

Keywords: Ultrasonic velocities, densities, acrylate and DMF molecules

Introduction
Direct uses in the pharmaceutical and biochemical industries are found in the study of the properties of liquid mixtures and solutions. Because of its ability to classify the physiochemical activity of the medium, ultrasonic technique is a valuable instrument in obtaining knowledge on the molecular behaviour of liquids. Acoustic and thermodynamic properties are used to explain various forms of association, molecular packing, and common types and strengths of intermolecular interactions [1]. Measuring the speed of sound in liquids allows some useful acoustic and thermodynamic parameters to be calculated, which are shown to be very sensitive to molecular interactions. These calculations are also helpful for studying the frequency of molecular interactions in liquid mixtures. Lagemann first studied the ultrasonic velocity method for the qualitative measurement of molecular interactions in solutions [2]. In order to better explain the nature and scale of the molecular aggregation patterns that occur in liquid mixtures arising from intermolecular interactions, ultrasonic experiments are of considerable significance. The high speed accuracy of sound measurements allows several other accurate parameters to be measured that provide details about device deviations from ideality. Qualitative knowledge about the type and strength of interactions in liquid mixtures is given by the parameters obtained from ultrasonic velocity calculation and the related excess functions. The acoustics the properties of binary liquid mixtures provide the requisite knowledge to evaluate current solution hypotheses [3, 4].
It provides an ultrasonic and volumetric analysis of molecular interactions in non-aqueous, non-electrolytic liquid binary/ternary (equimolar) mixtures. In the current investigation, the ultrasonic velocity and density are measured at a temperature of 308.15 K. Binary systems of N,N-dimethyl formamide liquid mixtures of methyl acrylate/ethyl acrylate/butyl acrylate/2-ethyl hexyl acrylate are being examined. A literature survey reveals that Kannappan *et al* [5] recorded measurements of velocity, density and viscosity at three temperatures of 303.15, 308.15 and 313.15 K. Density and viscosity data for binary mixtures of alkanol esters were stated by Gonozalez and Ortega [6], Francesconi and Comelli [7] and Liau *et al* [8]. Measured densities of ethyl acrylate, butyl acrylate, methyl methacrylate and styrene with toluene/cyclohexane/benzene/1, 4-dioxane at temperature 298.15 K. Wisnaik *et al* [9] and Peralta *et al* [10, 11]. Thirumaran and Sudha [12] have performed studies in recent years on the ternary mixture of benzene, chlorobenzene, nitrobenzene and N,N-dimethylformamide in cyclohexane at 303.15 K. Ultrasonic speed, density and viscosity values of the binary mixtures of ethanol + butyl acrylate and ethanol + butyl methacrylate were calculated by Arivazhagan *et al*[13] at 303 K. A.K. Nain [14] measured the densities of binary mixtures of butyl acrylate with 1-butanol, 2-butanol, 2-methyl-1-propanol, and 2-methyl-2-propanol at temperatures of (288.15, 293.15, 298.15, 303.15, 308.15, 313.15, and 318.15) K.

The densities and viscosities for binary mixtures of 2-butanol + iso-butanol, 2-butanol + tert-butanol, and iso-butanol + tert-butanol were recently presented by Micael G. Bravo-Sánchez *et al* [15] at temperatures between (308.15 and 343.15) K over the entire composition spectrum at atmospheric pressure.

From the experimental results of ultrasonic velocity (u) and density (ρ) the following properties, deviation in ultrasonic velocity (Δu), deviation in isentropic compressibility (Δks), excess molar volume (E Vm), excess free length ( ) and excess acoustic impedance (ZE) are calculated. There are also measured percentage deviations of potential velocities from experimental values. In the binary systems studied, experimental ultrasonic velocity data were used to analyse molecular interactions. In addition, the experimental data are fitted into two special types of polynomial equations [16] which characterise the ultrasonic velocity data both quantitatively and qualitatively, including in the prevalent region of unique interaction in which the system's non-ideal behaviour is noticed.

Methodology

The derivatives of LOBA Chemicals, methyl acrylate (MA) (mass fraction purity 0.99), ethyl acrylate (EA) (mass fraction purity 0.99) and 2-ethyl hexyl acrylate (EHA) (mass fraction purity 0.99), obtained from KEMPHASOL Company, Bombay, are N,N-dimethyl formamide (DMF) (mass fraction purity 0.99) and butyl acrylate (BA) (mass fraction purity 0.99). Both are grades of analytical reagent (AR grade) used in the present analysis and are further purified by normal methods [17]. Solutions for binary mixtures of DMF of MA, EA, BA and EHA are prepared in specially made air-tight stopper glass bottles and sufficient steps are taken to reduce the loss of evaporation. METTLER TOLEDO (Switzerland make) ABB5-S/FACT digital balance with an accuracy of ±0.01 mg is used to measure the solutions. Using a multi-frequency ultrasonic interferometer (M-82 Model) at a fixed frequency of 2MHz, the ultrasonic velocity of pure liquids and their binary mixtures are measured. The density measurements are carried out using a two stem pyknometer of Parker & Parker type [18] of capacity of bulb volume of 5 cm^3.

Results and Discussion

Table 1 and 2 shows the experimental values of u and χ of pure liquids at 308.15 K, along with the values. It is found from the table and figure 1 that the ultrasonic velocity increased non-linearly and density first decreased to 0.3 mole fraction of DMF and then increased to 0.3 mole fraction of DMF in all binary mixtures except in the binary system of EHA+DMF.

Table 1: Comparison of experimental values of ultrasonic velocity (u) and density (ρ) of pure liquids with the corresponding literature values at 308.15 K

Liquid	*u* /m.s^{-1}		p/kg.m^{-3}	
	Present work	**Literature**	Present work	Literature
***N,N-d* imethyl**				
lb rmam i d e	1433.68	1434.40[1381]	935.88	935.717[13]'n
Methyl acrylate	1141.78	1140.00[1401]	937.13	937.70[1171]
Ethyl acrylate	1119.12	1118.70"	903.78	904.90[1171]
Butyl acrylate	1158.33	1159.30[1171]	885.40	884.60[1411]
2-Ethyl hexyl acrylate	1229.40		875.00	

Table 2: Experimental values of ultrasonic velocity (u) and density (ρ) for all the systems at 308.15 K with mole fraction (x) or volume fraction (Φ) of N,N-dimethyl formamide

	X1.1+ DM:				**E**	**IMF**			**13A+ Mir**				**EllA4DMI;**		
		***a*/as^4**	***p'* kgat^4**			***a*/**	***p*/ kg.s.^{4}**	**x**	**sa..0**	**kpo**	***p*/**	***x***	***m***	**a/as''**	***p*^{1} kg.a^4**
0.0000	0.0000	1141.78	937.13	0.0000	00000	1119.12	903.78	0.0030	00000	115833	885.40	0.0000	0.0000	1229.40	875.00
0.1175	0.1190	117328	924.97	0.1363	0.1378	115325	89704	0.1699	0.1713	119109	81293	02242	02251	124720	877.06
0.2284	02316	120126	915.96	0.2539	02581	1181.96	89506	03078	03117	1219.77	1113.92	03808	03829	1264.78	88130
0.3386	03473	122923	91330	03805	03879	121451	896.08	0.4456	0.4504	1247.60	891.03	03370	05393	12103.87	889.40
0.4401	0.4507	1254.45	914.67	0.4859	0,4944	1242.04	900.90	03488	05539	1270.63	89706	0.6344	0.6365	1306.10	895.86
0.5416	03534	1281.18	916.46	03914	0.6002	1272.94	90650	0.6528	0.6574	1297.96	904,42	0.7340	0.7356	132620	903.77
0.6367	0.6490	130819	918.70	0.6793	0.6873	130121	912.27	0.7328	0.7367	1323.00	910.66	0.7993	0.8006	1345.77	909.72
0.7322	0.7433	133824	92232	0.7675	0.7744	133256	917.97	01132	0.8163	135138	91738	08645	08652	1368.61	91700
0.8258	08347	1369.79	926.11	0.8.475	08518	1362.70	924.68	08876	01891	1380.00	924.71	01980	0.8986	1378.98	924.93
0.9195	0.9238	1403.15	931.65	0.9276	0.9299	1397.18	93043	0.9428	0.9435	140332	93000	0.9609	0.9611	1408.96	929.69
1.0000	1.0000	14331.8	935.88	1.0000	1.0000	1433.68	93588	1.0000	10000	1433.68	93588	1.0000	1.0000	1433.68	93588

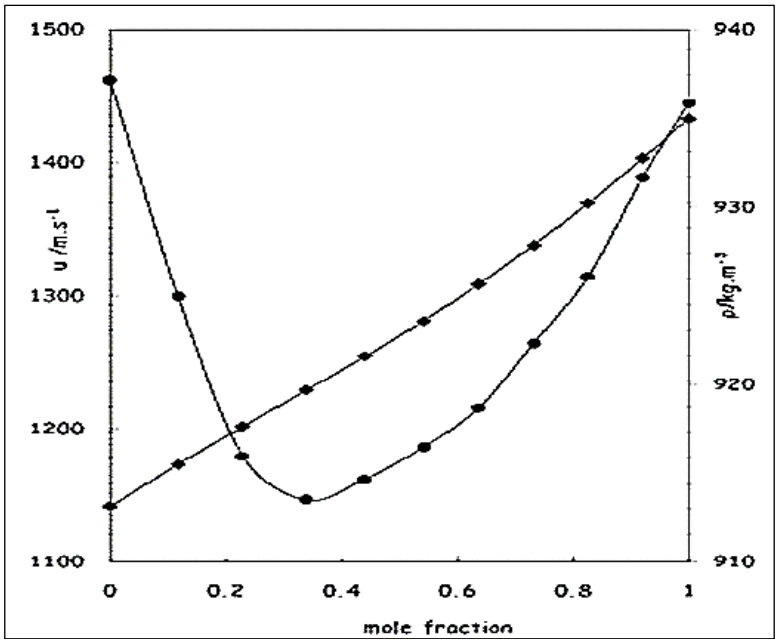

Fig 1: Variation of ultrasonic velocity

The deviation/excess properties, Δu, E Vm, Δks, E Lf and ZE are fitted to Redlich-Kister type polynomial equation 3.28. The co-efficients in the RedlichKister polynomial and the corresponding standard deviations calculated are compiled in Table 3

Table 3: Coefficients Ai of Redlich-Kister type polynomial equation and the corresponding Standard deviations of all the systems under investigation

Property	A.	AI A2		A.	Ai	*a*
MA+DM F						
fu/ms"	-70.65	51.80	33.77	-14.57	-26.92	0.1418
VI_I /10^4 m/.mor^i	7.7672	5.2878	6.1153	-23331	-8.3997	0.0273
mite Pa$^{\pi}$	3.1448	0.2320	0.2445	-0.7328	-3.5493	0.0181
L^E /10'^{11} m	0.2639	-0.0146	0.0059	-0.0229	-0.1360	0.0007
Z^E/10^s kg.M^2.1^1	-1.7172	-0.0071	-0.4304	0.1672	0.8269	0.0027
		EA+DMF				
	-121.15	64.34	11.04	16.11	-47.61	03252
V_m^E /10^4 m^i.mo1^{-1}	6.5798	53956	1.7741	-1.6620	-2.0397	0.0329
Ak_s/10'"	6.3556	-0.8276	-2.5701	-2.2872	2.1178	0.0133
L^E /10^{-II} m	0.3915	-0.0528	-0.0747	-0.0966	0.1129	0.0006
en 05 kg.m^4.s^{-I}	-2.1356	0.2600	0.0452	0.3133	-0.2168	0.0037
		BA+DMF				
Au/ms"	-147.83	101.88	38.83	-42.07	-128.35	0.3655
Vm /10' m^3.ma^l	4.5147	6.3454	4.9313	-4.2326	-8.0567	0.0327
WO*" Pa'	10.4690	-4.3212	-3.5343	3.2173	7.1040	0.0413
-Lf^E /10^{it} m	0.5491	-0.2209	-0.1283	0.1241	0.3374	0.0017
2^E/10^5 kg.m^4.s$^{'s}$	-2.3219	0.6886 -0.0646		0.0217	-0.4498	0.0056
		EHA+DMF				
Du/ms" -197.29		94.34	-68.43	52.13	-46.32	0.3252
3 -I Vm/10 M.mol	2.7985	4.3423	1.5024	-4.1783	-4.3331	0.0329
Mile Pa'	19.1319	-4.8672	7.8463	-5.4316	-7.1208	0.0133
L^E /10'^{11} m	0.8634	-0.2717	0.3433	-0.2288	-0.2156	0.0006
ZE/105 kg.m.2.s.1	-2.8381	1.0964	-1.1525	0.7624	0.6946	0.0037

A theoretical sound velocity assessment is useful to validate the applicability of different postulates of liquid mixture theories and in some situations, to arrive at some useful inferences about the (strength of) molecular interactions between component liquids. Theoretical sound velocities are measured in the present analysis by considering acrylate as one factor and DMF as the other ingredient in the binary mixture. The theoretical values of ultrasonic velocity obtained using various theories along with the experimental velocity are summarized in Tables 4-6

Table 4: Molar volumes (Vm) and partial molar volumes (Vm,1 andVm,2) of all the binary systems with mole fraction (x) of N,Ndimethyl formamide

X	MA+DMF				EA+DMF			BA+DMF					EHA+DMF		
	V. 17.1		V,,,a	x	V. V..1		VC.:	x	V.	i7,..1	V.2	x	V.	ir.0	*17.2*
	10'sm^3.mor'				10^{3}m^3.gnor'				10^{4}m^3.1nori				lems.mor'		
0.0000	9.1866	8.6546	9.1866	0.0000	11.0779	8.8156	11.0779	0.0000	14.4759	8.1610	14.4759	0.0000	21.0606	7.8240	21.0606
0.1175	9.1423	8.7863	9.1915	0.1363	10.7507	85295	11.1039	0.1699	13.4569	8.4150	14.4899	0.2242	18.1689	8.1040	21.0778
0.2284	9.075	8.3642	9.2818	0.2539	10.4195	8.1910	11.1859	0.3078	12.5828	7.9694	14.6290	0.3808	16.1065	7.8383	21.1902
03386	8.9427	8.0271	9.4125	03805	10.0258	7.9448	11.2978	0.4456	11.6304	7.7744	14.7394	03370	14.0063	7.7707	212407
0.4401	8.7871	7.8948	9.4939	0.4859	9.6559	7.8477	11.3702	05488	10.9189	7.7729	14.7386	0.6344	12.6966	7.7845	21.2202
05416	8.6261	7.8691	9.5169	03914	9.2820	7.8139	11.4082	0.6528	10.1969	7.7998	14.6982	0.7340	11.3603	7.7997	21.1879
0.6367	8.4706	7.8649	9.5235	0.6793	8.9628	7.8074	11.4189	0.7328	9.6430	7.8063	14.6844	0.7993	10.4885	7.8037	21.1748
0.7322	83028	7.8418	9.5757	0.7675	8.6477	7.8064	11.4214	0.8132	9.0895	7.8011	14.7031	0.8645	9.6140	7.8053	21.1670
0.8258	8.1376	7.8109	9.6856	0.8475	83510	7.8067	11.4200	0.8876	83746	7.7992	14.7114	0.8980	9.1685	7.8064	21.1592
0.9195	7.9585	7.8021	9.7321	0.9276	8.0668	7.8088	11.4018	0.9428	8.1992	7.8048	14.6445	0.9609	8.3301	7.8095	21.1130
1.0000	7.8108	7.8108	94394	1.0000	7.8108	7.8108	113360	1.0000	7.8108	7.8108	14.4036	1.0000	7.8108	7.8108	21.0410

Table 5: Theoretical values of ultrasonic velocities for MA+DMF mixture as a function of mole fraction (x) of N,N-dimethyl formamide

	UN	Uv	**LI hop**	**U.Iffis**	**UJ**	**UR**
0.0000	1141.78	1141.78	1141.68	1141.78	114135	114138
0.1175	116947	116956	1175.96	1163.66	1173.31	112927
0.2284	1196.93	1197.07	120830	1186.27	1201.80	1125.85
0.3386	1225.66	1225.81	1240.49	1210.99	1229.25	1146.19
0.4401	1253.41	125355	1270.11	1235.99	1254.50	1178.19
0.5416	1282.52	1282.62	1299.75	1263.51	1281.12	1213.31
0.6367	1311.13	1311.19	1327.54	1291.97	1308.89	124927
03322	1341.22	134123	1355.45	1323.59	1338.26	1291.96
0.8258	1372.09	1372.08	1382.79	1358.02	1369.81	1335.98
0.9195	1404.53	140451	1410.19	1396.62	1403.16	1389.09
1.0000	1433.68	1433.68	143333	1433.68	1433.70	1433.68

Table 6: Theoretical values of ultrasonic velocities from eqn.s [(3.38)-(3.43)] for EA+DMF mixture as a function of mole fraction (x) of N,N-dimethyl formamide

x	**Us**	**Ur**	**Ulm,**	**U.**	**Uj**	**UR**
0.0000	1119.12	1119.12	1119.08	1119.12	1119.1	1119.12
0.1363	1148.30	115336	1163.26	1140.88	1153.32	1116.25
0.2539	1175.97	1184.69	1201.06	1162.54	1181.88	1127.96
03805	1208.74	1220.46	1241.40	1189.59	1214.42	1152.68
0.4859	1238.71	1251.99	1274.72	1215.81	1242.00	1189.23
0.5914	1271.50	1285 26	1307.80	124628	1273.05	1229.91
0.6793	1301.28	1314.45	1335.20	1275.76	1301.17	1269.31
0.7675	1333.64	1345.11	1362.50	1309.98	1332.63	1309.47
0.8475	136530	137429	1387.15	1346.17	1362.73	1353 39
0.9276	1399.98	1404.83	1411.68	1388.56	1397.22	139439
1.0000	1433.68	1433.68	1433.73	1433.68	1433.70	1433.68

Generally, Δks and values depend upon several contributions arising from physical, chemical and structural effects [19, 20]. These observation supports the prediction that fitting of molecules of one component into the voids in the structure of second component is less favourable if the difference in molar volumes of component liquids is less. Peralta *et al.* [19] reported similar studies on positive values

Conclusions

i) The ultrasonic velocities and densities for (MA or EA or BA or EHA + DMF) binary mixtures are measured and the values of Δu, E Vm, Δks, E Lf and ZE are calculated.

ii) The E Vm, Δks and E Lf values are positive where as Δu and ZE values are negative in all the binary mixtures, indicating dispersion forces between the acrylate and DMF molecules and it follows the order MA+DMF>EA+DMF>BA+DMF>EHA>DMF.

iii) The ultrasonic velocities calculated from various hypotheses of velocity are compared with the ultrasonic velocities determined experimentally. The Jacobson equation establishes a strong consensus between the theoretical and experimental values of ultrasonic velocity.

iv) In addition, the ultrasonic velocity is suited to two forms of polynomials that characterise the ultrasonic velocity data both quantitatively and qualitatively, including in the prevalent area of real interaction where non-ideal device activity is noticed.

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