

Research Article

Thermodynamic and Acoustic Study on Molecular Interactions in Certain Binary Liquid Systems Involving Ethyl Benzoate

B. Nagarjun,¹ A. V. Sarma,² G. V. Rama Rao,³ and C. Rambabu⁴

¹ Department of Physics, G.V.P. College of Engineering (A), Visakhapatnam 530048, Andhra Pradesh, India

² Department of Physics, Andhra University, Visakhapatnam 530003, Andhra Pradesh, India

³ Department of Physics, DAR College, Nuzvid 521201, Andhra Pradesh, India

⁴ Department of Chemistry, Acharya Nagarjuna University, Guntur 522510, Andhra Pradesh, India

Correspondence should be addressed to B. Nagarjun; au.nagarjun@gmail.com

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Speeds of sound and density for binary mixtures of ethyl benzoate (EB) with N,N-dimethylformamide (NNDMF), N,N-dimethylacetamide (NNDMAc), and N,N-dimethylaniline (NNDMA) were measured as a function of mole fraction at temperatures 303.15, 308.15 K, 313.15 K, and 318.15 K and atmospheric pressure. From the experimental data, adiabatic compressibility (β_{ad}), intermolecular free length (L_f), and molar volume (V) have been computed. The excess values of the above parameters were also evaluated and discussed in light of molecular interactions. Deviation in adiabatic compressibilities and excess intermolecular free length (L_f^E) are found to be negative over the molefraction of ethyl benzoate indicating the presence of strong interactions between the molecules. The negative excess molar volume V^E values are attributed to strong dipole-dipole interactions between unlike molecules in the mixtures. The binary data of $\Delta\beta_{ad}$, V^E , and L_f^E were correlated as a function of molefraction by using the Redlich-Kister equation.

1. Introduction

The thermophysical study of esters is of increasing interest due to their wide range in flavouring, perfumery, artificial essences, and cosmetics. Esters are also important solvents in pharmaceutical, paint, and plastic industries [1]. Recently, interest in the study of liquid mixtures containing esters as one of the components [2–5] has increased. These studies are of great significance because one can get information regarding structural changes that occur in pure ester because of mixing.

Among the different types of esters, ethyl benzoate (aromatic ester) is a polar solvent ($\mu = 1.8$ D) and is not a strongly associated liquid. On the other hand, NNDMF, to some extent, is associated by means of dipole-dipole interactions. Significant structural effects are absent due to the lack of hydrogen bonds. Therefore, it acts as an aprotic protophilic solvent [6]. NNDMAc is a dipolar aprotic solvent and is moderately structured [7]. The N,N-dialkylamides have no

significant intermolecular hydrogen bonding capability but are highly polar with high percentage of ionic character, making oxygen of C=O group strongly negative [8]. NNDMA is a unassociated liquid [9]. The interactions of ethyl benzoate with N,N-dialkylamides and N,N-dimethylaniline are important to enable analysis of their dissimilar geometric structures on mixture properties.

Rao [10] calculated the excess isentropic compressibilities for the binary mixtures of N,N-dimethylformamide and N,N-dimethylacetamide and various normal and branched esters. The negative excess values are attributed to the presence of n-n interactions between unlike molecules. Aminabhavi et al. [11] studied the physicochemical behavior of binary mixtures of ethyl benzoate with diethylene glycol and dimethyl ether. Rathnam et al. [12] reported the densities, viscosities, and refractive indices of ethyl benzoate with o-xylene, m-xylene, p-xylene, and ethyl benzene. Lien et al. [13] have measured the excess molar enthalpies of the binary mixture of ethyl benzoate with 1-Octanol at 298.15 K. Joshi et al. [14] measured

TABLE 1: Comparison of experimental velocities and densities of pure liquids with the literature values at 303.15 K.

Component	U (m/s)		$\rho \times 10^3$ (kg·m ⁻³)	
	Expt	Lit	Expt	Lit
Ethyl benzoate	1347.00	1346.50 ^a	1.0456	1.0455 ^a
N,N dimethyl formamide	1438.53	1438.32 ^b	0.9392	0.9393 ^b
N,N dimethyl acetamide	1438.00	1435.55 ^b	0.9319	0.9317 ^b
N,N dimethyl aniline	1470.50	1470.00 ^c	0.9482	0.94815 ^d

^aData taken from [18]. ^bData taken from [19]. ^cData taken from [20]. ^dData taken from [21].

the densities and viscosities of the binary mixture of bromoform with ethyl benzoate.

We report herein the experimental values of densities and ultrasonic velocities of mixtures of ethyl benzoate with NNDMF, NNDMAc, and NNDMA at temperatures 303.15, 308.15, 313.15, and 318.15 K covering the whole miscibility range to enable analysis of the effect of their dissimilar geometric structures on the mixture properties. The non-rectilinear behavior of ultrasonic velocity, compressibility, and other thermodynamic parameters of liquid mixtures reveal the strength of interactions. To get additional information about the nature and strength of molecular interactions, other parameters such as free length, adiabatic compressibility, and their excess parameters have been calculated in the liquid mixtures. These results were correlated with the Redlich-Kister polynomial equation [15] to derive the coefficients and standard deviation.

2. Materials and Experimental Details

Ethyl benzoate (SD Fine Chemicals, purity > 99%) was used without any further treatment. NNDMF, NNDMAc, and NNDMA were purchased from SD Fine or Merck and were purified by the recommended methods [16, 17]. Further, the purities were ascertained from their ultrasonic speed and density values at 303.15 K which agreed with the literature values (Table 1).

Mixtures were prepared by mixing appropriate volumes of liquids in airtight bottles and weighed in a single-pan Mettler balance to an accuracy of ± 0.001 mg. Preferential evaporation losses of solvents from mixtures were kept to a minimum as evidenced by repeated measurement of the physical properties over an interval of 2-3 days, during which no changes in physical properties were observed. The possible error in mole fractions is estimated to be around ± 0.0001 .

The densities of liquids and their mixtures were measured by a 25 mL specific gravity bottle, calibrated with redistilled water. The average uncertainty in measurement in the measured density is ± 0.05 kgm⁻³. With the fluctuation of ± 0.05 K, temperature was controlled by a water thermostat. Ultrasonic velocity was measured using the ultrasonic interferometer (Model M-83) provided by Mittal Enterprises, New Delhi. The values agree closely with the values given in the literature.

The experimental values of ultrasonic speeds (u), densities (ρ), adiabatic compressibilities (β_{ad}), molar volumes (V), and free length (L_f) of pure ethyl benzoate, NNDMF,

NNDMAc, NNDMA, and those of their binary mixtures over the entire composition range and at 303.15 K, expressed by mole fraction x_1 of ethyl benzoate, are listed in Table 2.

3. Results and Discussion

The experimental density (ρ) values of binary mixtures were used to calculate the excess molar volumes as

$$V^E = x_1 M_1 \left(\frac{1}{\rho} - \frac{1}{\rho_1} \right) + x_2 M_2 \left(\frac{1}{\rho} - \frac{1}{\rho_2} \right), \quad (1)$$

where M is the molar mass; subscripts 1 and 2 stand for pure components, ethyl benzoate and NNDMF, NNDMAc, and NNDMA, respectively. The uncertainty in V^E is estimated to be within $\pm 2 \times 10^{-6}$ m³ · mol⁻¹.

Assuming that ultrasonic absorption is negligible, adiabatic compressibility (β_{ad}) can be obtained from the density and velocity of ultrasonic sound (U) using the relation

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

Deviation in adiabatic compressibility was calculated using the relation

$$\Delta\beta = \beta_{ad} - (\beta_1 x_1 + \beta_2 x_2), \quad (3)$$

where β_{ad} is the compressibility of the mixture.

Intermolecular free length (L_f) has been calculated using the relation

$$L_f = K(\beta_{ad})^{1/2}, \quad (4)$$

where K is Jacobson's temperature-dependant constant and is equal to $(93.875 + 0.375T) \cdot 10^{-8}$.

The excess intermolecular free length (L_f^E) at a given mole fraction is the difference between mean free length and the sum of the fractional contributions of the two liquids given by

$$L_f^E = L_f - (L_{f1} x_1 + L_{f2} x_2), \quad (5)$$

where L_{f1} and L_{f2} are the individual intermolecular free length values of pure liquids in the binary mixtures.

The experimental values of ultrasonic velocity (U) and values of density (ρ), at the four temperatures, namely, $T = 303.15$ K, 308.15 K, 313.15 K, and 318.15 K, along with the derived values of adiabatic compressibility (β_{ad}), intermolecular free length (L_f), and molar volume (V) and their excess parameters are given in Tables 2, 3, and 4.

The excess properties were fitted to a Redlich, Kister-type [14] polynomial equation

$$Y^E = x_1 x_2 \sum_{i=0}^j A_i (1 - 2x_1)^i. \quad (6)$$

The optimum number of coefficients, j , was ascertained from an examination of the variation of the standard deviation σ . The values of coefficients, A_i , were evaluated by using the

TABLE 2: Experimental ultrasonic velocities, u , densities, ρ , and related thermodynamic parameters for ethyl benzoate + NNDMF system.

x_1	$U \text{ ms}^{-1}$	$\rho \times 10^3 \text{ kgm}^{-3}$	$\beta_{\text{ad}} \times 10^{-12} \text{ m}^2 \text{N}^{-1}$	$V \times 10^{-6} \text{ m}^3 \text{mol}^{-1}$	$L_f \text{ \AA}$	$\Delta\beta_{\text{ad}} \times 10^{-12} \text{ m}^2 \text{N}^{-1}$	$V^E \times 10^{-6} \text{ m}^3 \text{mol}^{-1}$	$L_f^E \text{ \AA}$
$T = 303.15 \text{ K}$								
0.0000	1438.53	0.9392	51.4522	77.8216	0.4497	0.0000	0.0000	0.0000
0.0568	1433.50	0.9529	51.0688	81.2961	0.4481	-0.4549	-0.2620	-0.0019
0.1193	1427.94	0.9663	50.7512	85.1516	0.4467	-0.8511	-0.5200	-0.0037
0.1885	1421.80	0.9793	50.5133	89.4680	0.4456	-1.1761	-0.7540	-0.0051
0.2654	1414.94	0.9922	50.3414	94.2800	0.4449	-1.4448	-1.0030	-0.0063
0.3514	1407.25	1.0045	50.2715	99.7325	0.4446	-1.6230	-1.2130	-0.0071
0.4484	1398.50	1.0158	50.3334	105.9725	0.4448	-1.6831	-1.3510	-0.0073
0.5584	1388.25	1.0249	50.6279	113.3098	0.4461	-1.5271	-1.2520	-0.0066
0.6843	1376.50	1.0327	51.1051	121.8471	0.4482	-1.2084	-1.0000	-0.0052
0.8298	1362.89	1.0395	51.7198	131.8472	0.4512	-0.7048	-0.5770	-0.0030
1.0000	1347.00	1.0456	52.7108	143.6209	0.4552	0.0000	0.0000	0.0000
$T = 308.15 \text{ K}$								
0.0000	1420.69	0.9345	53.0178	78.2129	0.4598	0.0000	0.0000	0.0000
0.0568	1416.24	0.9498	52.4916	81.5612	0.4575	-0.5600	-0.3950	-0.0024
0.1193	1411.27	0.9639	52.0884	85.3661	0.4558	-1.0003	-0.7110	-0.0043
0.1885	1405.75	0.9773	51.7820	89.6557	0.4544	-1.3478	-0.9800	-0.0059
0.2654	1399.58	0.9900	51.5655	94.4878	0.4535	-1.6100	-1.2180	-0.0070
0.3514	1392.69	1.0023	51.4383	99.9475	0.4529	-1.7883	-1.4310	-0.0078
0.4484	1384.81	1.0134	51.4578	106.2300	0.4530	-1.8265	-1.5380	-0.0080
0.5584	1375.55	1.0226	51.6827	113.5634	0.4540	-1.6669	-1.4560	-0.0073
0.6843	1364.87	1.0304	52.0990	122.1266	0.4558	-1.3254	-1.1930	-0.0058
0.8298	1352.50	1.0368	52.7261	132.1880	0.4585	-0.7848	-0.7260	-0.0034
1.0000	1338.00	1.0419	53.6120	144.1309	0.4624	0.0000	0.0000	0.0000
$T = 313.15 \text{ K}$								
0.0000	1398.00	0.9294	55.0533	78.6421	0.4719	0.0000	0.0000	0.0000
0.0568	1394.18	0.9463	54.3682	81.8657	0.4690	-0.6765	-0.5300	-0.0029
0.1193	1389.85	0.9609	53.8719	85.6300	0.4668	-1.1633	-0.8980	-0.0050
0.1885	1385.04	0.9742	53.5109	89.9392	0.4652	-1.5137	-1.1600	-0.0065
0.2654	1379.64	0.9870	53.2289	94.7763	0.4640	-1.7840	-1.4070	-0.0077
0.3514	1373.59	0.9990	53.0536	100.2767	0.4632	-1.9462	-1.5950	-0.0084
0.4484	1366.70	1.0101	52.9994	106.5688	0.4630	-1.9856	-1.7100	-0.0086
0.5584	1358.48	1.0191	53.1703	113.9502	0.4638	-1.7980	-1.6000	-0.0078
0.6843	1349.00	1.0267	53.5206	122.5584	0.4653	-1.4285	-1.3150	-0.0061
0.8298	1337.99	1.0331	54.0707	132.6663	0.4677	-0.8562	-0.8280	-0.0037
1.0000	1325.00	1.0375	54.9010	144.7422	0.4712	0.0000	0.0000	0.0000
$T = 318.15 \text{ K}$								
0.0000	1380.74	0.9299	56.4079	78.5998	0.4810	0.0000	0.0000	0.0000
0.0568	1377.12	0.9487	55.5820	81.6576	0.4775	-0.8276	-0.7380	-0.0035
0.1193	1373.00	0.9638	55.0381	85.3744	0.4752	-1.3733	-1.2000	-0.0059
0.1885	1368.39	0.9770	54.6608	89.6770	0.4735	-1.7527	-1.5200	-0.0075
0.2654	1363.20	0.9888	54.4219	94.6046	0.4725	-1.9938	-1.7337	-0.0085
0.3514	1357.36	0.9998	54.2883	100.2006	0.4719	-2.1300	-1.8900	-0.0091
0.4484	1350.64	1.0098	54.2828	106.5998	0.4719	-2.1383	-1.9700	-0.0092
0.5584	1342.70	1.0184	54.4667	114.0329	0.4727	-1.9577	-1.8900	-0.0084
0.6843	1333.49	1.0256	54.8316	122.6897	0.4743	-1.5965	-1.6500	-0.0069
0.8298	1322.72	1.0310	55.4405	132.9388	0.4769	-0.9919	-1.1300	-0.0043
1.0000	1310.00	1.0325	56.4374	145.4431	0.4812	0.0000	0.0000	0.0000

TABLE 3: Experimental ultrasonic velocities, u , densities, ρ , and related thermodynamic parameters for ethyl benzoate + NNDMAc.

x_1	$U \text{ ms}^{-1}$	$\rho \times 10^3 \text{ kgm}^{-3}$	$\beta_{\text{ad}} \times 10^{-12} \text{ m}^2 \text{N}^{-1}$	$V \times 10^{-6} \text{ m}^3 \text{mol}^{-1}$	$L_f \text{ \AA}$	$\Delta\beta_{\text{ad}} \times 10^{-12} \text{ m}^2 \text{N}^{-1}$	$V^E \times 10^{-6} \text{ m}^3 \text{mol}^{-1}$	$L_f^E \text{ \AA}$
$T = 303.15 \text{ K}$								
0.0000	1438.00	0.9319	51.8935	93.4864	0.4517	0.0000	0.0000	0.0000
0.0674	1432.95	0.9480	51.3740	96.3878	0.4494	-0.5747	-0.4800	-0.0025
0.1400	1427.32	0.9635	50.9473	99.5830	0.4475	-1.0606	-0.9200	-0.0047
0.2181	1421.01	0.9782	50.6271	103.1217	0.4461	-1.4447	-1.3000	-0.0063
0.3026	1414.06	0.9925	50.3887	107.0031	0.4451	-1.7521	-1.6553	-0.0077
0.3943	1406.27	1.0058	50.2747	111.3334	0.4446	-1.9410	-1.9200	-0.0085
0.4940	1397.57	1.0175	50.3173	116.2342	0.4448	-1.9800	-2.0200	-0.0087
0.6030	1387.19	1.0274	50.5796	121.7970	0.4459	-1.8068	-1.9200	-0.0079
0.7225	1375.40	1.0358	51.0348	128.0889	0.4479	-1.4492	-1.6200	-0.0063
0.8542	1362.17	1.0419	51.7277	135.3109	0.4510	-0.8639	-1.0000	-0.0037
1.0000	1347.00	1.0456	52.7108	143.6209	0.4552	0.0000	0.0000	0.0000
$T = 308.15 \text{ K}$								
0.0000	1425.00	0.9282	53.0553	93.8591	0.4600	0.0000	0.0000	0.0000
0.0674	1414.43	0.9533	52.4328	96.6298	0.4573	-0.6600	-0.6200	-0.0029
0.1400	1407.10	0.9757	51.7632	99.7749	0.4543	-1.3700	-1.1200	-0.0060
0.2181	1400.06	0.9955	51.2467	103.3044	0.4521	-1.9300	-1.5200	-0.0085
0.3026	1392.49	1.0127	50.9238	107.2326	0.4506	-2.3000	-1.8400	-0.0101
0.39 43	1385.16	1.0275	50.7248	111.5802	0.4498	-2.5500	-2.1000	-0.0112
0.4940	1378.23	1.0390	50.6703	116.4747	0.4495	-2.6600	-2.2200	-0.0117
0.6030	1370.45	1.0463	50.8910	122.0725	0.4505	-2.5000	-2.1000	-0.0109
0.7225	1362.13	1.0499	51.3375	128.3908	0.4525	-2.1200	-1.7900	-0.0093
0.8542	1351.69	1.0489	52.1808	135.6309	0.4562	-1.3500	-1.1700	-0.0059
1.0000	1338.00	1.0419	53.6120	144.1309	0.4624	0.0000	0.0000	0.0000
$T = 313.15 \text{ K}$								
0.0000	1405.50	0.9238	54.7974	94.3061	0.4708	0.0000	0.0000	0.0000
0.0674	1403.10	0.9419	53.9280	97.0079	0.4671	-0.8764	-0.7000	-0.0038
0.1400	1400.00	0.9589	53.2064	100.0549	0.4639	-1.6055	-1.3100	-0.0069
0.2181	1396.00	0.9743	52.6688	103.5372	0.4616	-2.1512	-1.7700	-0.0093
0.3026	1390.60	0.9883	52.3255	107.4594	0.4601	-2.5033	-2.1100	-0.0109
0.3943	1384.40	1.0015	52.0988	111.8120	0.4591	-2.7395	-2.3800	-0.0119
0.4940	1377.50	1.0138	51.9853	116.6629	0.4586	-2.8632	-2.5600	-0.0124
0.6030	1368.80	1.0237	52.1359	122.2385	0.4592	-2.7239	-2.4800	-0.0118
0.7225	1358.10	1.0314	52.5672	128.6366	0.4611	-2.3051	-2.1100	-0.0100
0.8542	1345.00	1.0365	53.3302	136.0082	0.4645	-1.5557	-1.3800	-0.0067
1.0000	1325.00	1.0375	54.9010	144.7422	0.4712	0.0000	0.0000	0.0000
$T = 318.15 \text{ K}$								
0.0000	1385.00	0.9194	56.7017	94.7575	0.4823	0.0000	0.0000	0.0000
0.0674	1384.98	0.9396	55.4839	97.2461	0.4771	-1.2000	-0.9300	-0.0051
0.1400	1381.08	0.9573	54.7647	100.2213	0.4740	-1.9000	-1.6300	-0.0082
0.2181	1376.75	0.9726	54.2441	103.7130	0.4717	-2.4000	-2.1000	-0.0103
0.3026	1371.70	0.9871	53.8417	107.5884	0.4700	-2.7800	-2.5078	-0.0120
0.3943	1366.33	1.0002	53.5575	111.9618	0.4687	-3.0400	-2.7800	-0.0131
0.4940	1359.66	1.0121	53.4475	116.8575	0.4683	-3.1236	-2.9400	-0.0135
0.6030	1352.00	1.0220	53.5323	122.4504	0.4686	-3.0100	-2.8700	-0.0130
0.7225	1342.03	1.0295	53.9307	128.8682	0.4704	-2.5800	-2.5100	-0.0111
0.8542	1330.16	1.0320	54.7659	136.6042	0.4740	-1.7100	-1.4485	-0.0074
1.0000	1310.00	1.0325	56.4374	145.4431	0.4812	0.0000	0.0000	0.0000

TABLE 4: Experimental ultrasonic velocities, u , densities, ρ , and related thermodynamic parameters for ethyl benzoate + NNDMA system.

x_1	$U \text{ ms}^{-1}$	$\rho \times 10^3 \text{ kgm}^{-3}$	$\beta_{\text{ad}} \times 10^{-12} \text{ m}^2 \text{ N}^{-1}$	$V \times 10^{-6} \text{ m}^3 \text{ mol}^{-1}$	$L_f \text{ \AA}$	$\Delta\beta_{\text{ad}} \times 10^{-12} \text{ m}^2 \text{ N}^{-1}$	$V^E \times 10^{-6} \text{ m}^3 \text{ mol}^{-1}$	$L_f^E \text{ \AA}$
$T = 303.15 \text{ K}$								
0.0000	1470.50	0.9482	48.7719	127.8106	0.4379	0.0000	0.0000	0.0000
0.0900	1463.99	0.9605	48.5763	128.8888	0.4370	-0.5500	-0.3445	-0.0025
0.1820	1455.87	0.9724	48.5187	130.0539	0.4367	-0.9700	-0.6340	-0.0043
0.2761	1446.91	0.9841	48.5394	131.2837	0.4368	-1.3200	-0.8920	-0.0058
0.3724	1436.85	0.9957	48.6486	132.5568	0.4373	-1.5900	-1.1410	-0.0070
0.4709	1425.42	1.0070	48.8766	133.9043	0.4383	-1.7500	-1.3510	-0.0077
0.5717	1413.22	1.0176	49.2038	135.3745	0.4398	-1.8200	-1.4750	-0.0080
0.6750	1398.88	1.0268	49.7705	137.0818	0.4423	-1.6600	-1.4000	-0.0072
0.7807	1383.14	1.0347	50.5169	138.9865	0.4456	-1.3300	-1.1670	-0.0058
0.8890	1366.09	1.0412	51.4636	141.1360	0.4498	-0.8100	-0.7300	-0.0035
1.0000	1347.00	1.0456	52.7108	143.6209	0.4552	0.0000	0.0000	0.0000
$T = 308.15 \text{ K}$								
0.0000	1453.40	0.9439	50.1538	128.3928	0.4472	0.0000	0.0000	0.0000
0.0900	1448.57	0.9571	49.7950	129.3529	0.4456	-0.6700	-0.4560	-0.0029
0.1820	1441.85	0.9691	49.6332	130.4900	0.4449	-1.1500	-0.7670	-0.0051
0.2761	1433.31	0.9810	49.6186	131.6920	0.4448	-1.4900	-1.0460	-0.0066
0.3724	1423.92	0.9925	49.6915	132.9731	0.4452	-1.7500	-1.2800	-0.0077
0.4709	1413.61	1.0038	49.8522	134.3235	0.4459	-1.9300	-1.4800	-0.0085
0.5717	1402.59	1.0144	50.1109	135.8035	0.4470	-2.0200	-1.5870	-0.0089
0.6750	1389.28	1.0236	50.6179	137.5103	0.4493	-1.8700	-1.5050	-0.0082
0.7807	1374.46	1.0316	51.3136	139.4123	0.4524	-1.5400	-1.2670	-0.0067
0.8890	1358.23	1.0383	52.2081	141.5350	0.4563	-1.0200	-0.8490	-0.0044
1.0000	1338.00	1.0419	53.6120	144.1309	0.4624	0.0000	0.0000	0.0000
$T = 313.15 \text{ K}$								
0.0000	1435.20	0.9398	51.6583	128.9530	0.4571	0.0000	0.0000	0.0000
0.0900	1431.88	0.9534	51.1601	129.8537	0.4549	-0.7900	-0.5200	-0.0035
0.1820	1425.32	0.9658	50.9684	130.9465	0.4541	-1.2800	-0.8800	-0.0056
0.2761	1417.25	0.9777	50.9236	132.1433	0.4539	-1.6300	-1.1690	-0.0071
0.3724	1408.35	0.9892	50.9658	133.4173	0.4540	-1.9000	-1.4150	-0.0083
0.4709	1398.80	1.0004	51.0852	134.7768	0.4546	-2.1000	-1.6110	-0.0092
0.5717	1388.45	1.0109	51.3122	136.2689	0.4556	-2.2000	-1.7110	-0.0096
0.6750	1375.74	1.0199	51.8070	138.0100	0.4578	-2.0400	-1.6000	-0.0088
0.7807	1361.81	1.0279	52.4598	139.9134	0.4606	-1.7300	-1.3660	-0.0075
0.8890	1345.54	1.0345	53.3911	142.0506	0.4647	-1.1500	-0.9390	-0.0049
1.0000	1325.00	1.0375	54.9010	144.7422	0.4712	0.0000	0.0000	0.0000
$T = 318.15 \text{ K}$								
0.0000	1412.00	0.9356	53.6093	129.5319	0.4690	0.0000	0.0000	0.0000
0.0900	1409.96	0.9497	52.9638	130.3486	0.4661	-0.9000	-0.6150	-0.0040
0.1820	1404.50	0.9624	52.6740	131.4026	0.4649	-1.4500	-1.0250	-0.0064
0.2761	1397.05	0.9743	52.5901	132.6049	0.4645	-1.8000	-1.3200	-0.0079
0.3724	1389.02	0.9857	52.5824	133.8966	0.4645	-2.0800	-1.5600	-0.0091
0.4709	1380.54	0.9967	52.6410	135.2781	0.4647	-2.3000	-1.7460	-0.0100
0.5717	1371.38	1.0071	52.7962	136.7845	0.4654	-2.4300	-1.8440	-0.0106
0.6750	1359.31	1.0160	53.2681	138.5332	0.4675	-2.2500	-1.7380	-0.0098
0.7807	1345.99	1.0239	53.9072	140.4535	0.4703	-1.9100	-1.5000	-0.0083
0.8890	1330.79	1.0303	54.8035	142.6280	0.4742	-1.3200	-1.0490	-0.0057
1.0000	1310.00	1.0325	56.4374	145.4431	0.4812	0.0000	0.0000	0.0000

TABLE 5: Coefficients, A_i , of (6) and standard deviations, σ , for binary systems at different temperatures.

	T/K	A_0	A_1	A_2	σ
Ethyl benzoate + NNDMF					
$\Delta\beta_{ad} \times 10^{-12}/m^2N^{-1}$	303.15	-6.4467	2.2499	-0.0638	0.0171
	308.15	-6.8775	2.8689	-1.3139	0.0391
	313.15	-7.2181	3.6083	-2.8140	0.0767
	318.15	-7.6186	4.2860	-5.1293	0.1145
$V^E \times 10^{-6}/m^3mol^{-1}$	303.15	-5.1947	0.8178	1.3098	0.0259
	308.15	-5.8404	1.3461	-0.4321	0.0456
	313.15	-6.1729	2.0602	-2.4103	0.0899
$L_f^E/\text{\AA}$	318.15	-7.0606	-2.4146	-5.8222	0.1234
	303.15	-0.0285	0.0096	0.0020	0.0001
	308.15	-0.0301	0.0123	-0.0047	0.0002
	313.15	-0.0312	0.0154	-0.0118	0.0003
	318.15	-0.0328	0.0177	-0.0215	0.0005
	Ethyl benzoate + NNDMAc				
$\Delta\beta_{ad} \times 10^{-12}/m^2N^{-1}$	303.15	-7.8289	1.3767	-0.1647	0.0155
	308.15	-10.7235	-0.2065	0.0599	0.0681
	313.15	-11.2398	0.4350	-3.1080	0.0260
	318.15	-11.9293	2.3285	-6.8757	0.1496
$V^E \times 10^{-6}/m^3mol^{-1}$	303.15	-8.0967	-0.1873	0.3985	0.0196
	308.15	-8.7131	0.0699	-1.4562	0.0315
	313.15	-9.9358	-0.2638	-1.9087	0.0390
$L_f^E/\text{\AA}$	318.15	-11.1717	1.5314	-3.0602	0.1497
	303.15	-0.0342	0.0064	0.0000	0.0001
	308.15	-0.0471	-0.0007	0.0004	0.0003
	313.15	-0.0488	0.0022	-0.0131	0.0001
	318.15	-0.0516	0.0093	-0.0287	0.0007
	Ethyl benzoate + NNDMA				
$\Delta\beta_{ad} \times 10^{-12}/m^2N^{-1}$	303.15	-7.0776	-0.9608	-0.6333	0.0256
	308.15	-7.8226	-1.4455	-2.2954	0.0322
	313.15	-8.4985	-1.4000	-3.4138	0.0474
	318.15	-9.3139	-1.6851	-4.5462	0.0618
$V^E \times 10^{-6}/m^3mol^{-1}$	303.15	-5.5381	-2.0172	-0.4794	0.0310
	308.15	-6.0491	-1.9707	-1.6874	0.0364
	313.15	-6.5792	-2.0734	-2.1865	0.0427
$L_f^E/\text{\AA}$	318.15	-7.1185	-2.0844	-3.1230	0.0440
	303.15	-0.0311	-0.0032	-0.0031	0.0001
	308.15	-0.0344	-0.0061	-0.0089	0.0001
	313.15	-0.0372	-0.0049	-0.0143	0.0003
	318.15	-0.0404	-0.0064	-0.0203	0.0003

method of least squares, with all points weighted equally. The coefficients A_0 , A_1 , and A_2 along with standard deviations, σ of fit for all the mixtures are listed in Table 5.

The $\Delta\beta_{ad}$, V^E , and L_f^E values plotted against mole fraction of ethyl benzoate (x_1) are shown in Figures 2, 3, and 4.

In this investigation, the values of ultrasonic velocity decrease with increase in the concentration of ethyl benzoate and decrease with increase in temperature at any particular

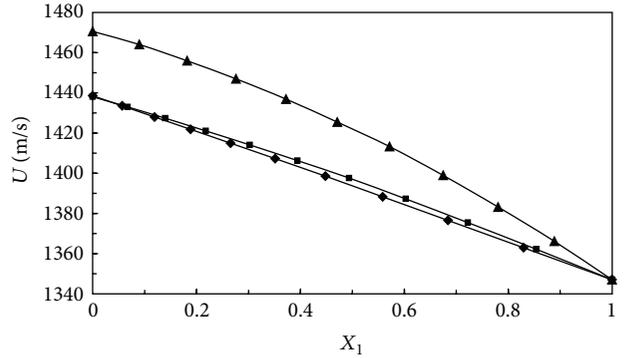


FIGURE 1: Ultrasonic velocities (U) plotted against the mole fractions of ethyl benzoate (X) in binary mixtures of ethyl benzoate + NNDMF (\blacklozenge), ethyl benzoate + NNDMAc (\blacksquare), and ethyl benzoate + NNDMA (\blacktriangle).

concentration for all the three systems (Figure 1). The ultrasonic velocity values decrease with increase of temperature due to the breaking of hetero- and homomolecular clusters at high temperatures [22]. Lagemann and Duban [23] were the first to point out the ultrasonic velocity approach for qualitative estimation of the interaction in liquids.

The intermolecular free length is the distance between the surfaces of the neighboring molecules. The variation of ultrasonic velocity in a solution depends upon the increase or decrease of intermolecular free length after mixing the components. The interdependence of intermolecular free length and ultrasonic velocity was evolved from a model for sound propagation proposed by Kincaid and Eyring [24]. The ultrasonic velocity should decrease if the intermolecular free length increases as a result of mixing of components. This fact is observed in the present investigation for ethyl benzoate + NNDMF, ethyl benzoate + NNDMAc, and ethyl benzoate + NNDMA systems. Figure 2 represents the variation of excess intermolecular free length with mole fraction of ethyl benzoate. The excess intermolecular free length values are negative, and the curves appear to reach negative peak value at about 0.45 mole fraction of ethyl benzoate. According to Ramamurthy and Sastry [25], the negative L_f^E values indicate that sound wave has to travel a longer distance. This may be attributed to the dominant nature of interactions between unlike molecules.

The compressibility behavior of solutes, which is the second derivative of the Gibbs energy, is a very sensitive indicator of molecular interactions and can provide useful information about this phenomenon [26–28]. The structural change of molecules takes place due to the existence of electrostatic field between interacting molecules. The change in adiabatic compressibility value in liquids and liquid mixtures may be ascribed to the strength of intermolecular attraction. The effect of depolymerization increases the compressibility of the system [29]. Electrostatic attraction and association decrease the compressibility. The compressibility of the mixtures is the result of these two effects depending upon the predominance [30]. According to Jacobson [31, 32],

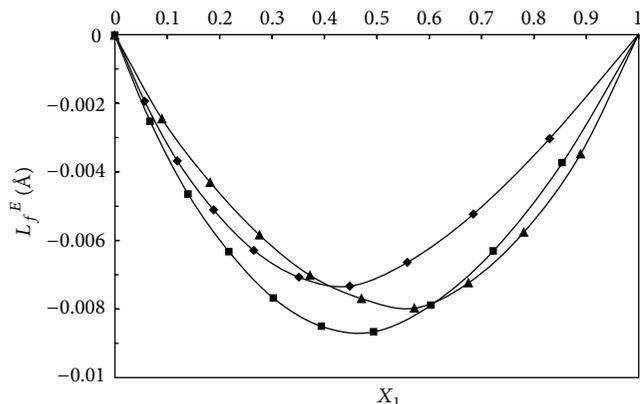


FIGURE 2: Excess intermolecular free lengths (L_f^E) plotted against the mole fractions of ethyl benzoate (X) in binary mixtures of ethyl benzoate + NNDMF (◆), ethyl benzoate + NNDMAc (■), and ethyl benzoate + NNDMA (▲).

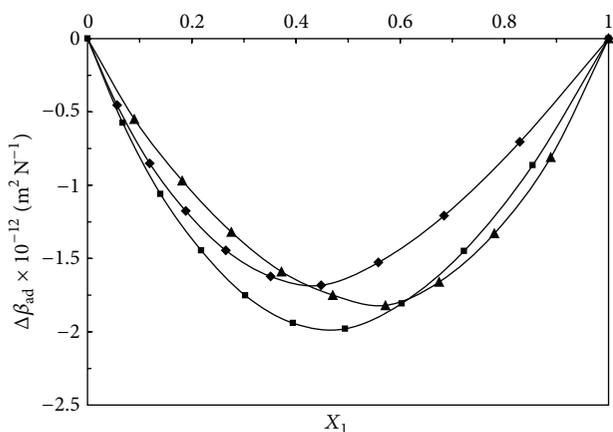


FIGURE 3: Deviation in adiabatic compressibilities ($\Delta\beta_{ad}$) plotted against the mole fractions of ethyl benzoate (X) in binary mixtures of ethyl benzoate + NNDMF (◆), ethyl benzoate + NNDMAc (■), and ethyl benzoate + NNDMA (▲).

the adiabatic compressibility can be studied more through intermolecular free length.

Figure 3 represents the deviation in adiabatic compressibility with the mole fraction of ethyl benzoate. In the present investigation, deviation in adiabatic compressibilities is found to be negative over the mole fraction of ethyl benzoate indicating the presence of strong interactions between the molecules. The negative deviation in adiabatic compressibility reaches a peak at about 0.45 mole fraction of ethyl benzoate in all the three systems chosen.

The deviation in adiabatic compressibility can be explained by taking into consideration the following factors.

- Loss of dipolar association and difference in size and shape of component molecules which lead to decrease in velocity and increase in compressibility.
- Dipole-dipole interaction or hydrogen-bonded complex formation between unlike molecules which lead to increase in sound velocity and decrease of compressibility.

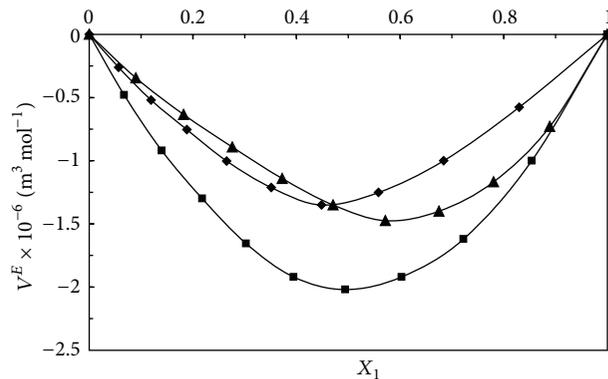


FIGURE 4: Excess molar volumes (V^E) plotted against the mole fractions of ethyl benzoate (X) in binary mixtures of ethyl benzoate + NNDMF (◆), ethyl benzoate + NNDMAc (■), and ethyl benzoate + NNDMA (▲).

The actual deviation depends on the resultant effect. The strength of the interaction between the components increases when excess values tend to become increasingly negative. This may be qualitatively interpreted in terms of closer approach of unlike molecules leading to reductions in compressibility and volume [33, 34]. This type of interactions for the binary mixtures has been already reported previously [35]. The deviations in adiabatic compressibilities are found to increase with increasing temperature which is in agreement with the previously reported results [36]. The $\Delta\beta_{ad}$ and L_f^E minima occur at the same concentrations further strengthen the occurrence of molecular associations [37].

The values of V^E calculated by using (1) are included in Tables 2–4.

The sign of V^E of a system depends upon the relative magnitude of expansion and contraction of the two liquids due to mixing [38].

The negative V^E arises due to dominance of the following factors.

- Chemical interaction between constituent molecules such as heteromolecular association through the formation of H-bond, often termed as strong specific interaction.
- Association through weaker physical forces such as dipolar force or any other forces of this kind.
- Accommodation of molecules of one component into the interstitial positions of the structural network of molecules of the other component.
- Geometry of the molecular structure that favors fitting of the component molecules with each other.

The V^E values are negative over the entire mole fraction range and at all temperatures investigated for all binary systems under study (Figure 4). The observed negative values of V^E for the three mixtures indicate the presence of specific interactions between ethyl benzoate and amide molecules. The negative V^E values are attributed to strong dipole-dipole interactions between unlike molecules in the mixtures. The

V^E values are more negative for ethyl benzoate + DMAc than those for other two systems.

The electron density at oxygen atom of the carbonyl atom of DMAc is greater than that of DMF due to the presence of methyl group at carbon atom of carbonyl group in DMF resulting in stronger interaction in the system [39, 40]. The V^E values of ethyl benzoate + DMA are in between these two. The strength of interactions between ethyl benzoate + DMA is stronger than ethyl benzoate + DMF due to the fact that negative charge on nitrogen in DMA is more than that of nitrogen in DMF due to conjugation with C=O group (this is also evident from the fact that the DMA is more basic than DMF).

Rao and Reddy [41] reported increased negative values of excess molar volumes with the increase in carbon chain length of the ester, in the binary mixtures of DMF, and aliphatic esters. The observed higher negative values of V^E ($-1.35 \text{ cm}^3 \text{ mol}^{-1}$) for aromatic ester with DMF in the present investigation, when compared to that of ethyl acetate and DMF ($V^E = -0.375$) at the same temperature (303.15 K), indicate much stronger interactions between the unlike molecules of components in this binary mixture due to the formation of not only dipole-dipole interactions but also of induced polar interaction like dipole-induced dipole and dipole interactions between aromatic ester and DMF. Similar interactions are reported in the mixtures of DMF and polycyclic aromatic hydrocarbons by Nikam and Kharat [42, 43] and by Ramadevi and Prabhakara Rao [44]. Also, formation of induced polar interactions like dipole-induced dipole interactions between polycyclic aromatic hydrocarbons and NMP (acyclic amide) is reported by Sugiura and Ogawa [45]. This further supports that amides react with aromatic compounds more strongly than the corresponding aliphatic counter parts.

The V^E values decrease (become more negative) with increase in temperature for all the 3 systems. This is attributed due to the dissociation of self-associated amide molecules resulting in the more favorable fitting of the smaller molecules into large voids of bigger ethyl benzoate molecules leading to contraction in volume and hence resulting in more negative V^E values with rise in temperature.

4. Conclusions

The adiabatic compressibility (β_{ad}) and intermolecular free length (L_f) both have an inverse relationship with ultrasonic velocity (U). Occurrence of U maxima, β_{ad} , and L_f minima at the same concentrations indicates the strong interaction through dipole-dipole interactions between the components. The negative V^E values are attributed to strong dipole-dipole interactions between unlike molecules in the mixtures.

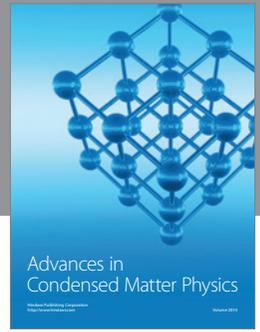
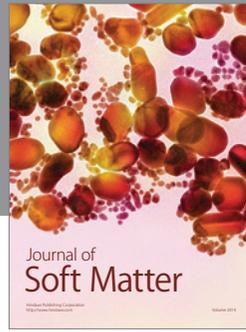
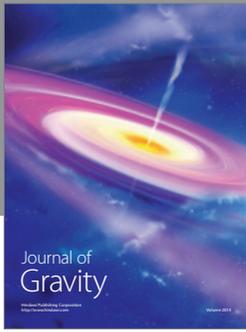
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