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Ultrasonic Study of Dipole-Dipole Interactions in Ternary Liquid Mixtures at Different Temperatures

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The ultrasonic velocity, density and viscosity have been measured for the mixtures of primary alkanols, namely, 1-propanol, 1-butanol and 1-pentanol with aniline in toluene at 303, 308 and 313 K. The experimental data have been used to calculate the acoustical parameters namely adiabatic compressibility (β), free length (L_f), free volume (V_f), internal pressure (π_i), acoustic impedance (Z) and molar volume (V_m). The excess values of some of the above parameters are also evaluated and discussed in the light of molecular interaction present in the mixtures. It is observed that the alkanols form strong dipole-dipole interactions with aniline. However, no complex formation or charge-transfer reactions were observed in the present study. The elevation of temperature in the present study, which may probably, would be caused from the fact that thermal energy facilitates an increase in the molecular separation in the liquid mixtures.

Key Words: Dipole-dipole interaction, Free length, Internal pressure, Adiabatic compressibility.

INTRODUCTION

In recent years, the measurement of ultrasonic velocity has been adequately employed in understanding the nature of molecular interactions in pure liquids and liquid mixtures. The ultrasonic velocity measurements are highly sensitive to molecular interactions and can be used to provide qualitative information about the physical nature and strength of molecular interaction in the liquid mixtures[1-3]. Ultrasonic velocity of a liquid is fundamentally related to the binding forces between the atoms or the molecules and has been adequately employed in understanding the nature of molecular interaction in pure liquids[4], binary and ternary mixture[5-8]. The variation of ultrasonic velocity and related parameters throw much light upon the structural changes associated with the liquid mixtures having weakly interacting components[9] as well as strongly interacting components[10].

The study of molecular association in organic ternary mixtures having alcohol as one of the components is of particular interest, since alcohols are strongly self-associated liquid having a three dimensional network of hydrogen bond[11] and can be associated with any other group having some degree of polar attractions[12]. Accurate knowledge of thermodynamic mixing properties such as adiabatic compressibility, intermolecular free length, free volume, internal pressure and molar volume and their excess values for mixtures of protic, non-protic and associated liquids has a great importance in theoretical and applied areas of research. The deviations from ideality and specific or non-specific interactions have been revealed. Alcohols[1-7] and aniline[8-11] exist as associated structures in liquid state. Thus, upon mixing aniline with alcohols of varying chain lengths and provide interesting properties due to specific interactions arising from charge-transfer, dipole-dipole, donor-acceptor and hydrogen bond formation forces may be observed. Thermodynamic properties of aniline and alcohol mixtures are of particular interest because alcohols provide proton and aniline accepts it, forming donor-acceptor complexes. Such an interesting behaviour of these components which motivated the authors to carry out the present study.

The present ternary liquid systems taken up for present study are

System-I	1-propanol + toluene + aniline
System-II	1-butanol + toluene + aniline
System-III	1-pentanol + toluene + aniline

at three temperatures 303, 308 and 313 K.

EXPERIMENTAL

The liquid mixtures of various concentrations in mole fraction were prepared by taking AR grade chemicals, which were purified by standard methods[13]. In all systems, the mole fraction of the second component, toluene ($X_2 = 0.4$) was kept fixed while the mole fractions of the remaining two were varied from 0.0 to 0.6 so as to have the mixtures of different compositions. There is nothing significant on fixing the second component at the $X_2 = 0.4$. The ultrasonic velocity in liquid mixtures have been measured using an ultrasonic interferometer (Mittal type: Model: F81) working at frequency 3 MHz with an overall accuracy of ± 2 ms^{-1}. The density and viscosity are measured using a Pyckno meter and an Ostwald's Viscometer with an accuracy of $\pm$ 0.1 mg and $\pm$ 0.001 Nsm^{-2}, respectively. All the precautions were taken to minimise the possible experimental errors. The values obtained are compared with literature and found that they makes very well with each other. The chemicals used in the present work were analytical reagent (AR) and spectroscopic reagent (SR) grade with minimum assay of 99.9 % were obtained from SD Fine Chemicals, India and E-Merck, Germany.

Theory: Using the measured data, the following acoustical parameters have been calculated:

$$\text{Adiabatic compressibility } \beta = \frac{1}{U^2\rho} \tag{1}$$

Intermolecular free length (L_f) has been calculated from relation,

$$L_f = K_T\sqrt{\beta} \tag{2}$$

where K_T is a temperature dependent constant.

Free volume (V_f) has been calculated from relation,

$$V_f = \left(\frac{M_{eff}U}{K\eta}\right)^{3/2} \tag{3}$$

where M_{eff} is the effective molecular weight ($M_{eff} = \Sigma m_i x_i$, in which m_i and x_i are the molecular weight and the mole fraction of the individual constituents respectively). K is a temperature independent constant which is equal to 4.28×10^9 for all liquids.

The internal pressure (π_i) can be found out as,

$$\pi_i = bRT\left(\frac{K\eta}{U}\right)^{1/2}\left(\frac{\rho^{2/3}}{M_{eff}^{7/6}}\right) \tag{4}$$

K is a constant, T the absolute temperature, η the viscosity in Nsm^{-2}, U the ultrasonic velocity in ms^{-1}, ρ the density in Kgm^{-3}, M_{eff} the effective molecular weight.

The acoustic impedance can be related as

$$Z = U\rho \tag{5}$$

where U is a velocity of ultrasound in medium and ρ is density.

Excess parameter (A^E) has been calculated by using the relation

$$A^E = A_{exp} - A_{id} \quad (6)$$

where $A_{id} = \sum_{i=1}^{n} A_i X_i$, A_i is any acoustical parameters and X_i the mole fraction of the liquid component.

RESULTS AND DISCUSSION

The experimentally determined values of the density (ρ), viscosity (η) and ultrasonic velocity (U) for the three liquid systems at 303, 308 and 313 K are reported in Table-1. The values of adiabatic compressibility (β), intermolecular free length (L_f), free volume (V_f) and internal pressure (π_i), for the three liquid systems at the above said temperatures are presented in Table-2. Similarly, the values of molar volume (V_m) and acoustic impedance (Z) for the above three liquid systems are furnished in Table-3. The excess values of the some of the above parameters have been evaluated and are presented in the Tables 4 and 5.

In all the mixtures, the ultrasonic velocity (U) decreases with increase in mole fraction of alcohols. It is observed that as the number of hydrocarbon group or chain-length of alcohol increases, a gradual decrease in sound velocity is noticed. This behaviour at such concentrations is different from the ideal mixtures behaviour can be attributed to intermolecular interactions in the systems studied[14,15]. As toluene is weakly polar molecule, interactions are possible between 1-alkanols and aniline, whereas the interactions of toluene with any one of the other two is not so favoured. This is further supported by the decreasing velocity trend with increasing mole fraction of 1-alkanols.

TABLE-1
VALUES OF DENSITY (ρ) VISCOSITY (η) AND ULTRASONIC VELOCITY (U) FOR

m/(mol kg^{-1})		ρ (kg/m^3)			η ($\times 10^{-3}$ Nsm^{-2})			U (m/s)		
X_1	X_3	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
System I: 1-propanol + toluene + aniline										
0.0000	0.5945	935.74	924.57	912.08	1.2674	1.1217	0.9584	1465.9	1462.3	1446.6
0.1000	0.5000	922.68	916.35	910.69	1.1356	1.0157	0.9209	1426.8	1408.3	1387.7
0.1999	0.4000	915.22	910.99	904.42	1.0695	0.9687	0.8669	1399.2	1395.7	1386.0
0.3000	0.2999	890.82	885.74	882.45	0.9776	0.9250	0.8150	1350.5	1347.5	1342.8
0.3676	0.2646	874.77	871.45	864.21	0.8995	0.8214	0.7417	1344.2	1335.3	1312.9
0.4998	0.0999	850.56	842.89	832.28	0.8439	0.7660	0.6988	1294.5	1282.9	1250.8
0.5999	0.0000	827.01	813.98	807.16	0.8371	0.7476	0.6792	1233.9	1226.1	1215.9
System II: 1-butanol + toluene + aniline										
0.0000	0.5999	935.74	924.57	912.08	1.2674	1.1217	0.9584	1465.9	1462.3	1446.6
0.0999	0.4999	934.06	920.94	910.64	1.1327	1.0428	0.9329	1417.0	1407.0	1397.7
0.1999	0.3999	913.82	908.13	905.14	1.0579	0.9763	0.8866	1403.7	1399.6	1394.5
0.2999	0.3000	891.33	886.84	872.13	1.0202	0.9345	0.8411	1358.0	1340.0	1327.3
0.4000	0.1999	863.45	853.62	846.34	0.9822	0.8846	0.8006	1314.0	1310.7	1308.3
0.4999	0.1000	859.80	838.40	832.08	0.9565	0.8551	0.7802	1269.1	1264.9	1248.4
0.5988	0.0000	798.25	775.38	766.24	0.9441	0.8471	0.7684	1246.3	1234.5	1221.0
System III: 1-pentanol + toluene + aniline										
0.0000	0.5999	935.74	924.57	912.08	1.2675	1.1217	0.9584	1465.90	1462.3	1446.6
0.0999	0.4999	934.87	911.11	902.61	1.2027	1.0658	0.9363	1437.80	1426.1	1408.1
0.2000	0.4000	932.98	890.81	888.09	1.1483	1.0119	0.9296	1403.40	1391.0	1384.5
0.3000	0.3000	895.78	887.26	880.79	1.1043	1.0078	0.9113	1357.80	1349.8	1333.7
0.4000	0.2000	875.40	868.66	850.99	1.0835	0.9926	0.8828	1315.90	1298.6	1288.4
0.4999	0.1000	847.89	843.64	835.81	1.0792	0.9856	0.8684	1285.01	1280.4	1276.1
0.5999	0.0000	827.76	813.47	804.78	1.0200	0.9763	0.8461	1268.60	1250.7	1242.5

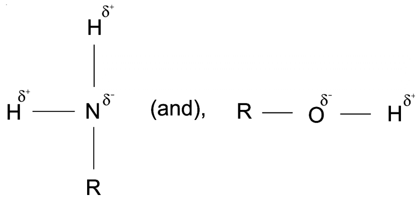
The primary alkanols are having characteristic carbo cations. The stability of a charged system is increased by the dispersal of the charge. Hence, any factor that tends to spread out the positive charge of the electron deficient carbon and distribute it over the rest of the ion must stabilize the carbo cation. Further, these carbon cations will be stabilized by electron donating substituents and will be rendered less stable by electron withdrawing substituents. The alkyl group, attached to the carbon cation bearing positive charge, exerts an electron releasing inductive effect and thus reduces the positive charge of the carbon atom to which it is attached and hence, the alkyl group itself becomes positive. Therefore, dipole-dipole interactions or H-bonding is expected between the molecules. Further the increase in chain length involves more number of alkyl groups, these interactions are expected to be more pronounced in higher numbers.

However, the value of β shows an inverse behaviour as compared to the ultrasonic velocity (U). The adiabatic compressibility (β) increases with increase of concentration of alcohols as shown in Table-2. It is primarily the compressibility that increases due to structural changes of molecules in the mixture leading to a decrease in ultrasonic velocity[16-18]. Such a continuous increase in adiabatic compressibility with respect to the increase of molar concentration of alcohols has been qualitatively ascribed to the effect of hydrogen bonding or dipole-dipole interactions[19]. The increase in adiabatic compressibility (β) with increasing concentration of alcohols indicate significant interaction between aniline and alcohols forming dipole-dipole interactions.

TABLE-2
VALUES OF ADIABATIC COMPRESSIBILITY (β),
FREE LENGTH (L_f) AND FREE VOLUME (V_f) FOR

m (mol kg^{-1})		β (×10^{-10} m^2 N^{-1})			L_f (×10^{-10} m)			V_f (×10^{-7} m^3 mol^{-1})		
X_1	X_3	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
System I: 1-propanol + toluene + aniline										
0.0000	0.5945	4.9732	5.0581	5.2392	0.4450	0.4523	0.4647	1.5283	1.7167	1.9248
0.1000	0.5000	5.3236	5.5044	5.7025	0.4604	0.4719	0.4848	1.4911	1.6940	1.8545
0.1999	0.4000	5.5812	5.6357	5.7559	0.4714	0.4775	0.4871	1.4231	1.6287	1.8247
0.3000	0.2999	6.1550	6.2181	6.2851	0.4950	0.5015	0.5089	1.3820	1.5905	1.8000
0.3676	0.2646	6.3265	6.4354	6.7131	0.5019	0.5102	0.5260	1.3620	1.5610	1.7995
0.4998	0.0999	7.0157	7.2095	7.6797	0.5285	0.5400	0.5626	1.3507	1.5294	1.7668
0.5999	0.0000	7.9421	8.1721	8.3800	0.5623	0.5749	0.5877	1.3452	1.4885	1.6441
System II: 1-butanol + toluene + aniline										
0.0000	0.5999	4.9371	5.0581	5.2392	0.4450	0.4523	0.4647	1.1526	1.2496	1.5365
0.0999	0.4999	5.3320	5.4851	5.6211	0.4607	0.4687	0.4785	1.1207	1.2376	1.4328
0.1999	0.3999	5.5538	5.6214	5.6813	0.4702	0.4768	0.4839	1.1061	1.2271	1.4267
0.2999	0.3000	6.0836	6.2798	6.5085	0.4921	0.5040	0.5179	1.0842	1.2169	1.4107
0.4000	0.1999	6.7076	6.8191	6.9348	0.5168	0.5252	0.5346	1.0473	1.1854	1.3805
0.4999	0.1000	7.2212	7.4548	7.7113	0.5362	0.5491	0.5638	1.0248	1.1765	1.3644
0.5988	0.0000	8.0652	8.4625	8.7540	0.5667	0.5851	0.6007	1.0066	1.1329	1.2964
System III: 1-pentanol + toluene + aniline										
0.0000	0.5999	4.9731	5.0581	5.2392	0.4450	0.4523	0.4647	1.0242	1.2271	1.5366
0.0999	0.4999	5.1740	5.3966	5.5875	0.4539	0.4672	0.4799	1.0232	1.2161	1.4623
0.2000	0.4000	5.4420	5.8014	5.8568	0.4655	0.4844	0.4913	1.0124	1.2159	1.3762
0.3000	0.3000	6.0550	6.1859	6.3829	0.4910	0.5002	0.5129	0.9912	1.1331	1.3090
0.4000	0.2000	6.5968	6.8271	7.0786	0.5125	0.5255	0.5401	0.9510	1.0791	1.2828
0.4999	0.1000	7.1424	7.2302	7.3478	0.5332	0.5408	0.5503	0.9376	1.0619	1.1662
0.5999	0.0000	7.5072	7.8587	8.0495	0.5467	0.5638	0.5760	0.9018	1.0096	1.1479

Since, alcohols are liquids which are associated through hydrogen bonding and in the pure state, they exhibit an equilibrium between the monomer and polymer species. Futher, when aniline is mixed with alcohols, the mechanism of reactions can be explained as the dipoles in aniline and alcohols arise due to difference in electronegatives of nitrogen, oxygen and hydrogen. They are in the order of nitrogen > oxygen > hydrogen. Hence dipolar aniline and alcohols are viewed as[20,21].

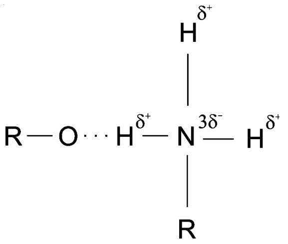


There are three possibilities of dipole-dipole interactions, which are as follows: (a) linkage between $N^{\delta-}$ aniline and $H^{\delta+}$ alcohols, hence alcohols being proton donor. (b) linkage between $O^{\delta-}$ of alcohol with $H^{\delta+}$ of amine, hence amine acts as proton donor. (c) linkage between $O^{\delta-}$ of alcohol with $N^{\delta+}$ of amine, alcohol being proton donor.

However, the third possibility is less likely, since electronegative of nitrogen (3.5) and oxygen (3) are very close together. Therefore, aniline-alkanol complexation may arise due to first two possibilities.

The second possibility is remote because of steric hindrance of hydrogen groups from aniline to oxygen to alcohol.

However, the first possibility of dipole-dipole interaction is more likely which can be represented as:



From Table-2, it is noticed that the free length (L_f) increases in all the three liquid systems with the increasing concentration of alkanols. The additions of interacting molecules break up the molecular clustering of the other, releasing several dipoles for interaction. In view of greater force of interaction between the alkanols and toluene, there will be an increase in cohesive energy and hence an increase in free length (L_f) is observed in all systems. Similar observation was noticed by earlier workers[22].

From Table-2, it is understood that in all the three liquid systems, as the concentration of alkanols increases, the free volume (V_f) decreases, whereas the internal pressure (π_i) increases. This suggests that the close packing of molecules inside the shield, which may brought about by the increasing magnitude of interactions[23,24].

Further, in all three liquid systems, the values of acoustic impedance (Z) is found to be decreased, which are listed in Table-3. When an acoustic wave travels in a medium, there is a variation of pressure from particle to particle. The ratio of the instantaneous pressure excess at any particle of the medium to the instantaneous velocity of that particle is known as 'specific acoustic impedance' of the medium.

This factor is governed by the inertial and elastic properties of the medium. It is important to examine specific acoustic impedance in relation to concentration and temperature. When a plane ultrasonic wave is set up in a liquid, the pressure and hence density and refractive index show specific variations with distance from the source along the direction of propagation. In the present investigation, it is observed that these acoustic impedance (Z) values decrease with increasing concentration of alcohols. Such a decreasing values of acoustic impedance (Z) further supports the possibility of strong molecular interactions due to dipole-dipole interaction between aniline-alkanols.

TABLE-3
VALUES OF ACOUSTIC IMPEDENANCE (Z) AND MOLAR VOLUME (V_m) FOR

m (mol kg^{-1})		Z (×10^6)			V_m (×10^{-5} m^3 mol^{-1})		
X_1	X_3	303 K	308 K	313 K	303 K	308 K	313 K
System I: 1-propanol + toluene + aniline							
0.0000	0.5945	1.3717	1.3519	1.3194	3.9431	3.9916	4.0452
0.1000	0.5000	1.3164	1.2914	1.2637	4.6503	4.6824	4.7123
0.1999	0.4000	1.2805	1.2715	1.2529	5.3432	5.3682	5.4126
0.3000	0.2999	1.2030	1.1935	1.1849	6.1643	6.1972	6.2227
0.3676	0.2646	1.1785	1.1636	1.1346	6.4008	6.4248	6.4784
0.4998	0.0999	1.1010	1.0813	1.0410	7.8671	7.9394	8.0417
0.5999	0.0000	1.0205	0.9980	0.9814	8.8164	8.9573	9.0336
System II: 1-butanol + toluene + aniline							
0.0000	0.5999	1.3717	1.3519	1.3377	3.9446	3.9923	4.0469
0.0999	0.4999	1.3236	1.2958	1.2728	4.7441	4.8116	4.8655
0.1999	0.3999	1.2827	1.2712	1.2622	5.6592	5.6943	5.7135
0.2999	0.3000	1.2104	1.1884	1.1576	6.6312	6.6643	6.7770
0.4000	0.1999	1.1345	1.1184	1.1047	7.7043	7.7936	7.8605
0.4999	0.1000	1.0912	1.0605	1.0388	8.5974	8.8175	8.8832
0.5988	0.0000	0.9948	0.9572	0.9356	10.1933	10.4939	10.6191
System III: 1-pentanol + toluene + aniline							
0.0000	0.5999	1.3717	1.3520	1.3194	3.9441	3.9924	4.0463
0.0999	0.4999	1.3442	1.2994	1.2709	4.8901	5.0183	5.0835
0.2000	0.4000	1.3093	1.2391	1.2296	5.8432	6.1204	6.1384
0.3000	0.3000	1.2163	1.1976	1.1746	7.0695	7.1374	7.1893
0.4000	0.2000	1.1519	1.1280	1.0964	8.2391	8.3036	8.4575
0.4999	0.1000	1.0865	1.0793	1.0665	9.5451	9.5938	9.6837
0.5999	0.0000	1.0501	1.0174	0.9999	10.8413	11.0315	11.1516

It is seen from the Table-3, the values of molar volume (V_m) increases with increasing concentration of alkanols in all the three liquid systems. Moreover, molar volume (V_m) is also increases with rise in temperature in the present study, which may probably would be caused from the fact that thermal energy facilitates an increase in the molecular seperation[25] in the liquid mixtures which leads to an increase in molar volume (V_m) with elevation of temperature.

In order to understand the nature of molecular interactions between the components of the liquid mixtures, it is of interest to discuss the same in term of excess parameter rather than actual values. Non-ideal liquid mixtures show considerable deviation from linearity in their physical behaviour with respect to concentration and these have been interpreted as arising from the presence of strong or weak interactions. The extent of deviation depends upon the nature of the constituents and composition of the mixtures.

The excess values of adiabatic compressibility (β^E), free length (L_f^E) and internal pressure (π_i^E) for all the three liquid systems are furnished in Table-4. It is learnt that the dispersion forces are responsible for possessing positive excess values, while dipole-dipole, dipole-induced dipole, charge transfer interaction and hydrogen bonding between unlike molecules are responsible for possessing negative excess values. In the present investigation, the excess adiabatic compressibility (β^E) and the excess free length (L_f^E) exhibit negative values over the entire range composition in all the three liquid systems clearly indicating the presence of strong interactions

TABLE-4
EXCESS VALUES OF ADIABATIC COMPRESSIBILITY (β^E), FREE LENGTH (L_f^E) AND INTERNAL PRESSURE (π_i^E) FOR

m (mol kg^{-1})		β^E ($\times 10^{-10}$ m^2 N^{-1})			L_f^E ($\times 10^{-10}$ m)			π_i^E ($\times 10^6$ Nm^{-2})		
X_1	X_3	303 K	308 K	313 K	303 K	308 K	313 K	303 K	308 K	313 K
System I: 1-propanol + toluene + aniline										
0.0000	0.5945	-0.3449	-0.7371	-1.6032	-0.0074	-0.0252	-0.0367	-90	-84	-72
0.1000	0.5000	-0.5093	-0.7776	-1.0733	-0.0142	-0.0262	-0.0372	-106	-99	-81
0.1999	0.4000	-0.7434	-1.1073	-1.4838	-0.0230	-0.0388	-0.0529	-121	-104	-91
0.3000	0.2999	-0.6627	-0.9202	-1.4199	-0.0194	-0.0331	-0.0492	-133	-108	-99
0.3676	0.2646	-0.7041	-0.9663	-1.1746	-0.0209	-0.0319	-0.0391	-150	-126	-118
0.4998	0.0999	-0.7870	-0.9192	-0.9549	-0.0258	-0.0310	-0.0316	-152	-127	-119
0.5999	0.0000	-0.3529	-0.4182	-0.7191	-0.0119	-0.0143	-0.0246	-159	-134	-120
System II: 1-butanol + toluene + aniline										
0.0000	0.5999	-0.4028	-0.7617	-1.0713	-0.0096	-0.0276	-0.0392	-106	-95	-83
0.0999	0.4999	-0.4438	-0.7442	-1.0912	-0.0118	-0.0277	-0.0413	-108	-97	-86
0.1999	0.3999	-0.6587	-1.0183	-1.4338	-0.0204	-0.0360	-0.0519	-114	-99	-87
0.2999	0.3000	-0.5653	-0.7700	-1.0089	-0.0165	-0.0252	-0.0338	-119	-103	-89
0.4000	0.1999	-0.3792	-0.6423	-0.9866	-0.0099	-0.0205	-0.0332	-125	-105	-90
0.4999	0.1000	-0.3018	-0.4166	-0.6124	-0.0085	-0.0131	-0.0199	-130	-107	-91
0.5988	0.0000	-0.1039	-0.1789	-0.0249	-0.0038	-0.0063	-0.0008	-131	-109	-92
System III: 1-pentanol + toluene + aniline										
0.0000	0.5999	-0.3668	-0.7617	-1.0713	-0.0096	-0.0276	-0.0392	-111	-98	-83
0.0999	0.4999	-0.5684	-0.7910	-1.0971	-0.0175	-0.0277	-0.0389	-109	-93	-80
0.2000	0.4000	-0.7025	-0.7538	-1.2014	-0.0227	-0.0254	-0.0425	-108	-89	-75
0.3000	0.3000	-0.4920	-0.7372	-1.0494	-0.0141	-0.0246	-0.0359	-106	-84	-71
0.4000	0.2000	-0.3527	-0.4639	-0.7277	-0.0094	-0.0143	-0.0236	-101	-80	-67
0.4999	0.1000	-0.2096	-0.4286	-0.8326	-0.0055	-0.0139	-0.0284	-100	-73	-67
0.5999	0.0000	-0.2473	-0.1680	-0.5050	-0.0089	-0.0059	-0.0177	-98	-68	-58

between unlike molecules. The strength of the interaction between the component molecules increase, when excess values tend to become increasingly negative. Further, the excess internal pressure (π_i^E) which is usually described in terms of molecular interaction, whose negative excess values for all the liquid systems clearly suggest that strong molecular association between the unlike molecules. Followed by, the excess molar volume (V_m^E) values which are furnished in Table-5 whose negative deviation clearly supporting the existence of specific interactions. It is again well confirmed by the excess values of acoustic impedance (Z^E) which are furnished in Table-5, are all positive in all the liquid systems over the entire range of composition. The positive excess values of[27] Z^E clearly suggest that there is a strong molecular interaction existing between the aniline-alknaols. Similar observations were observed by earlier workers[22,26] supports the present investigation.

TABLE-5
EXCESS VALUES OF ACOUSTIC IMPEDANCE (Z^E) AND MOLAR VOLUME (V_m^E) FOR

m (mol kg^{-1})		Z^E (×10^6)			V^E (×10^{-5} m^3 mol^{-1})		
X_1	X_3	303 K	308 K	313 K	303 K	308 K	313 K
		System I: 1-propanol + toluene + aniline					
0.0000	0.5945	0.0005	0.0630	0.0915	-6.1381	-6.3399	-6.5635
0.1000	0.5000	0.0012	0.0507	0.0803	-5.3084	-5.5229	-5.7600
0.1999	0.4000	0.0314	0.0876	0.1221	-4.4402	-4.6576	-4.8691
0.3000	0.2999	0.0199	0.0663	0.1067	-3.4444	-3.6497	-3.8689
0.3676	0.2646	0.0186	0.0579	0.0751	-3.0180	-3.2161	-3.3903
0.4998	0.0999	0.0499	0.0672	0.0678	-1.3932	-1.5509	-1.6711
0.5999	0.0000	0.0354	0.0407	0.0608	-0.2680	-0.3528	-0.4878
		System II: 1-butanol + toluene + aniline					
0.0000	0.5999	0.0093	0.0547	0.1019	-6.1877	-6.3908	-6.6143
0.0999	0.4999	0.0052	0.0522	0.0856	-5.3830	-5.5603	-5.7724
0.1999	0.3999	0.0268	0.0810	0.1241	-4.4634	-4.6673	-4.9021
0.2999	0.3000	0.0174	0.0516	0.0602	-3.4868	-3.6867	-3.8159
0.4000	0.1999	0.0036	0.0349	0.0641	-2.4105	-2.5483	-2.7112
0.4999	0.1000	0.0228	0.0304	0.0459	-1.5130	-1.5141	-1.6662
0.5988	0.0000	0.0115	0.0197	0.0077	-0.0814	-0.1661	-0.0849
		System III: 1-pentanol + toluene + aniline					
0.0000	0.5999	0.0093	0.0548	0.0836	-6.1882	-6.3907	-6.6149
0.0999	0.4999	0.0234	0.0532	0.0823	-5.4088	-5.5297	-5.7318
0.2000	0.4000	0.0487	0.0438	0.0882	-4.6219	-4.5921	-4.8304
0.3000	0.3000	0.0159	0.0532	0.0804	-3.5621	-3.7399	-3.9334
0.4000	0.2000	0.0118	0.0346	0.0494	-2.5590	-2.7385	-2.8192
0.4999	0.1000	0.0096	0.0368	0.0667	-1.4196	-1.6132	-1.7470
0.5999	0.0000	0.0305	0.0259	0.0473	-0.2899	-0.3403	-0.4331

Conclusion

By exhaustively analyzing all the related parameters and some of their excess values, it is obvious that there exist a strong molecular interaction existing between the unlike molecules. The 1-alkanols form strong dipole-dipole interactions with aniline. However, no complex formation or charge-transfer reactions were observed in the present study. The elevation of temperature in the present study, which may probably, would be caused from the fact that thermal energy facilitates an increase in the molecular separation in the liquid mixtures.

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