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Ultrasonic Study of Molecular Interaction for 1-Propanol, 1-Butanol, 1-Pentanol with Toluene in a Non-Polar Solvent

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Ultrasonic velocity (U), density (ρ) and viscosity (η) measurement have been carried out in 3 ternary liquid mixtures of 1-propanol, 1-butanol and 1-pentanol with toluene in a non-polar of ethyl benzene at 303K. The experimental data have been used to calculate adiabatic compressibility, free length, free volume, internal pressure, available volume and their excess values. An attempt has been made to evaluate the experimental ultrasonic velocity with those predicted on the basis of free length theory (FLT) and collision factor theory (CFT) with a view to compare the merits of the above 3 ternary liquid mixtures. The modulus of percentage deviation in ultrasonic velocity is applied to check the validity of the theories. The relative interaction (RI) is also calculated and found to be useful in understanding molecular interaction in ternary liquid mixtures.

Key Words: Ultrasonic study, Molecular interaction, Toluene, Alkanol.

INTRODUCTION

Ultrasonic investigation finds extensive application in characterizing aspects of the physico-chemical behaviour of liquid mixtures, such as molecular interactions in pure liquids[1] binary and ternary mixtures[2,3]. The large numbers of studies have been made on the molecular interaction in liquid systems by various physical method like infrared[4,5], Raman effect[6], nuclear magnetic resonance[7], dielectric constant[8], ultraviolet[9] and ultrasonic methods[10,11]. The intermolecular forces that are responsible for the interaction can be classified as long range forces and short range forces. The long range forces are the electrostatic induction and dispersion forces which mainly depend on the intermolecular separation. The short range forces, which arise when molecules come close enough together for their electron clouds to overlap are often highly directional. The molecular interaction between the components in the mixture forms the basic for the deviation from the ideality Hildebrand[12] carried out extensive work on the role of intermolecular forces in the mixtures of liquids.

To study the property of liquid mixtures, viscosity is also other important parameters. The non-linearity in the variation of viscosity is explained in terms of hydrogen bond formation between components of mixtures.

In order to get more information on the nature and strength of molecular interaction, it is obvious to calculate the other related acoustical parameters, such as adiabatic compressibility, free length, free volume, internal pressure, available volume and their excess values. The non-linear variation of those derived acoustical parameters with concentration of the solute may be explained on the basis structural changes occurring in liquid mixtures.

In the present study, an attempt has been made to investigate the molecular interaction in typical ternary liquid systems of 1-propanol, 1 butanol and 1-pentanol with toluene in ethyl benzene with special references to the behaviour of adiabatic compressibility, free length, free volume, internal pressure and available volume and their excess properties.

In the recent years, there have been two main attempts have been made for calculating the sound velocity in ternary mixtures of organic liquids at different concentration on the basis of molecular models. The first one is based on Jacobson[13] concept of intermolecular free length in liquids known as free length theory (FLT). The second one is based upon the collision factor theory (CFT) of Schaaff's[14]. The temperature and pressure dependence of the above two theories have been undertaken by a few workers[15,16]. The ultrasonic velocities of the above 3 ternary liquid systems have been evaluated theoretically with a view to compare the two theories. Another aim of this work is to study the relative interactions in terms of R_{exp}/R_{id} in the mixtures in which interactions are known to exist.

EXPERIMENTAL

All the chemicals used in this present research works are AR and SR grades of minimum assay of 99.9 % obtained from E. Merck and Sd fines. Fresh conductivity water is used throughout the investigation. The purities of the above chemicals were checked by density determination at 303K ± 0.01K which almost agreed with in the accuracy of $\pm \times 10^{-4}$ gcm^{-3} with the available literature values.

The ternary liquid mixtures of different known compositions were prepared in stoppered measuring flasks. The density, viscosity and velocity were measured as a function of composition of the ternary liquid mixture at 303K. The density was determined using a specific gravity bottle by relative measurement method. The weight of the sample was measured using electronic digital balance with an accuracy of ± 0.1 mg (Model: Shimadzu AX-200). An Ostwald's Viscometer (10 mL capacity) is used for the viscosity measurement. Efflux time was determined using a digital chronometer to within ± 0.01 s. An ultrasonic interferometer having the frequency 2 MHz with an overall accuracy of ± 0.02 ms^{-1} has been used for velocity measuremnt. An electronically digital operated constant temperature bath (RAAGA Industries) has been used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature. The accuracy in the temperature measurement is ± 0.01 K.

Theory and calculations

Various elastic and acoustical parameters were calculated from the measured data such as

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

Intermolecular free length $$L_f = K\sqrt{\beta} \tag{2}$$

where K is a temperature dependent constant. Its values are 631 × 10^{-6}, at 303K.

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

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.

Internal pressure $$\pi_i = bRT\left(\frac{K\eta}{U}\right)^{½}\left(\frac{\rho^{2/3}}{M_{eff}{}^{7/6}}\right) \tag{4}$$

where b is the cubic packing which is assumed to be 2 for all liquids and solution. R is a gas constant and T is absolute temperature.

Available volume can be calculated using the relation.

$$V_a = V_T\left(1 - \frac{U}{U_\infty}\right) \tag{5}$$

where V_T is the molar volume at T_K and U_∞ is the limiting ultrasonic velocity and is taken as 1600 m s^{-1}.

Excess values of the above parameters can be determined using the following relation as

$$A^E = A_{exp} - A_{id} \tag{6}$$

where $A_{id} = \Sigma A_i X_i$, A_i is any acoustical parameters and X_i the mole fraction of the liquid component.

Sound velocity is calculated using Jacobson's formula,

$$U_{mix} L_{mix} \rho_{mix}^{1/2} = K \tag{7}$$

where K is the temperature dependent constant U_{mix}, ρ_{mix} and L_{mix} are the velocity, density and free length in mixtures, respectively.

According to Schaaffs the ultrasonic velocity in ternary liquid mixtures as

$$U_{mix} = U_{\infty}\left(\frac{S_{mix} B_{mix}}{V_{mix}}\right) \tag{8}$$

where S is collision factor and B is the actual volume of molecule/mole. B can be computed from the knowledge of Vander Wall's Constant 'b' as it is related as follows

$$b = 4B \tag{9}$$

VanderWall's constant 'b' can be expressed as

$$b = \frac{M}{\rho}\left[1 - \frac{RT}{MU^2}\sqrt{1 + \frac{MU^2}{3RT}} - 1\right] \tag{10}$$

The modulus of percentage deviation in ultrasonic velocity between experimental and computed values are calculated as

$$\left|\frac{\Delta U}{U}\right| \% = \left(\frac{U_{exp} - U_{theo}}{U_{exp}}\right) \times 100 \tag{11}$$

Relative interaction for a liquid mixture is given by[17]

$$RI = \frac{R_{exp}}{R_{ideal}} \tag{12}$$

where R_{exp} evaluated using ultrasonic velocity U and effective molecular weight M were $R_{ideal} = R_1X_1 + R_2X_2 + R_3X_3$.

RESULTS AND DISCUSSION

The values of density, viscosity, ultrasonic velocity, adiabatic compressibility, free length, free volume, internal pressure and available volume of all the pure liquids and for the 3 ternary mixtures and summarized in Tables 1 and 2. The respective excess values of these parameters have been evaluated and presented in Table-3. The theoretical values of sound velocity in ternary liquid mixtures calculated on the basis of free length theory (FLT) and collision factor theory (CFT) along with the experimental values, the modulus of percentage deviations in velocity and the relative interaction parameters have been presented in Table-4.

TABLE-1
VALUES OF DENSITY (ρ), VISCOSITY (η), ULTRASONIC VELOCITY (U) ADIABATIC COMPRESSIBILITY (β), FREE LENGTH (L_f), FREE VOLUME (V_f), INTERNAL PRESSURE (π_i) AND AVAILABLE VOLUME (V_a) OF PURE LIQUIDS AT 303K

Liquids	Density ρ/kg m^{-3}	Viscosity $\eta/(\times 10^{-3} Nsm^{-2})$	Ultrasonic velocity $U/(ms^{-1})$	Adiabatic compressibility $\beta/(\times 10^{-10} m^2 N^{-1})$	Free length $L_f/(\times 10^{-10} m)$	Free volume $V_f/(\times 10^{-8} m^3 Mol^{-1})$	Internal pressure $\pi_i^{/}(\times 10^6 Nm^{-2})$	Available volume $V_a/(\times 10^{-6} m^3 mol^{-1})$
Ethyl benzene	859.7	0.5952	1304.0	0.6840	0.5231	0.4006	2721.17	22.85
Toluene	856.5	0.5256	1297.0	0.6940	0.5269	0.3872	3016.20	20.37
1-Propanol	803.4	1.6372	1199.0	0.8658	0.5884	0.0330	8734.06	18.75
1-Butanol	804.6	2.0670	1233.2	0.8173	0.5714	0.0332	7584.18	21.18
1-Pentanol	808.2	2.9580	1268.0	0.7695	0.5548	0.0262	7326.57	22.63

The measurement of viscosity in ternary mixture provides some reliable information in the study of molecular interaction. Table-2 shows that the viscosity increases with increase in concentration of the third component (1-propanol, 1 butanol and 1-pentanol).

According to Fort[18], the excess viscosity gives the strength of the molecular interaction between the interacting molecules. For systems were dispersion, induction and dipolar forces are operating the values of excess viscosity are found to be negative, whereas the existence of specific interactions leading to the formation of complexes in liquid mixtures tend to make excess viscosity positive. The excess viscosity is negative through the whole range of concentration in all systems are studied and decreases with increase in concentration of the third component. The large negative values of excess viscosity for all systems can be attributed to the presence of dispersion, induction and dipolar forces between the components.

It is observed from the Table-2, that the value of adiabatic compressibility and free length increases as the concentration of third component increases. Further, the Table-3 shows that the values of excess adiabatic compressibilities are positive (except system-I at 0.4 and 0.5 mol fraction) for all the systems. The value of excess adiabatic compressibility for the first 2 systems decreases with the increase in concentration, whereas in 3rd system the same increases with the increase in concentration. Fort[18] found that the increasing negative value of excess compressibility indicates greater interaction between the components of the mixtures. A positive value in excess properties corresponds mainly to the existence of dispersive forces. Dispersive forces, which are greatly present in all the types of system, would make positive contribution.

TABLE-2
VALUES OF DENSITY (ρ), VISCOSITY (η) AND ULTRASONIC VELOCITY (U), ADIABATIC COMPRESSIBILITY (β), FREE LENGTH (L_f), FREE VOLUME (V_f), INTERNAL PRESSURE (π_i) AND AVAILABLE VOLUME (V_a) AT 303K

Mole fraction		Density ρ/(Kg m^{-3})	Viscosity η/(× 10^{-3} Ns m^{-2})	Ultrasonic velocity U/(m s^{-1})	Adiabatic compressibility β/(× 10^{-10} m^2 N^{-1})	Free length L_f/(× 10^{-10} m)	Free volume V_f/(× 10^{-8} m^3 mol^{-1})	Internal pressure π_i/(× 10^6 N m^{-2})	Available volume V_a/ (× 10^{-6} m^3 mol^{-1})
X_2	X_3								
System-I [Ethyl benzene (X_1) + toluene (X_2) + 1 - propanol (X_3)]									
0.60	0.00	857.6	0.8350	1289.9	0.7008	0.5294	0.2096	3560.56	22.09
0.50	0.10	852.6	0.5936	1273.4	0.7233	0.5379	0.3262	3129.70	22.64
0.40	0.20	849.0	0.6272	1269.7	0.7306	0.5406	0.2843	3341.62	22.22
0.30	0.30	845.3	0.6581	1258.6	0.7467	0.5465	0.2472	3576.16	22.25
0.20	0.40	840.9	0.7119	1255.7	0.7542	0.5492	0.2073	3871.96	21.75
0.10	0.50	836.5	0.7879	1243.4	0.7732	0.5561	0.1655	4269.11	21.78
System-II [Ethyl benzene (X_1) + toluene (X_2) + 1 - butanol (X_3)]									
0.60	0.00	857.6	0.8350	1289.9	0.7008	0.5294	0.2096	3560.56	22.09
0.50	0.10	853.5	0.5975	1276.9	0.7185	0.5361	0.3316	3084.11	22.70
0.40	0.20	849.0	0.6794	1273.2	0.7266	0.5391	0.2646	3355.38	22.66
0.30	0.30	843.8	0.7154	1266.5	0.7388	0.5436	0.2364	3512.17	22.85
0.20	0.40	839.1	0.7851	1261.0	0.7494	0.5475	0.1994	3755.12	22.93
0.10	0.50	836.5	0.8906	1258.1	0.7557	0.5498	0.1585	4098.60	22.69
System-III [Ethyl benzene (X_1) + toluene (X_2) + 1 - pentanol (X_3)]									
0.60	0.00	857.6	0.8350	1289.9	0.7008	0.5294	0.2096	3560.56	22.09
0.50	0.10	852.8	0.6113	1285.8	0.7094	0.5327	0.3309	3054.77	22.42
0.40	0.20	848.0	0.6648	1278.9	0.7211	0.5371	0.2879	3196.74	22.95
0.30	0.30	841.6	0.7696	1274.9	0.7311	0.5407	0.2284	3445.09	23.31
0.20	0.40	837.6	0.8661	1266.4	0.7445	0.5457	0.1882	3673.00	23.94
0.10	0.50	832.9	0.9897	1262.8	0.7529	0.5488	0.1525	3936.36	24.23

Negative value of β^E is associated[19] with a structure forming tendency while a positive value is taken to indicate a structure breaking tendency due to hetero-molecular interaction between the component molecules of the mixtures. The positive excess adiabatic compressibility which indicates loosely packed molecules in the mixture due to the shape and size. In the present investigation the positive deviation in β^E in ternary systems have been attributed to dispersive forces that show weak interaction between the unlike molecules. Table-3 shows that the values of excess free length are positive in all 3 systems. The value of excess intermolecular free length L_f^E for the system **I** and **II** decreases with increase in concentration

TABLE-3

VALUES OF EXCESS VISCOSITY (η^E), ADIABATIC COMPRESSIBILITY (β^E), FREE LENGTH (L_f^E), FREE VOLUME (V_f^E), INTERNAL PRESSURE (π_i^E) AND AVAILABLE VOLUME (V_a^E) AT 303K

Mole fraction		Excess viscosity η^E/ (× 10^{-3} poise)	Excess adiabatic compressibility β^E/(× 10^{-10} $m^2 N^{-1}$)	Excess free length L_f^E/(× 10^{-10} m)	Excess free volume V_f^E/(× 10^{-8} $m^3 mol^{-1}$)	Excess internal pressure (atm.) π_i^E/(× 10^6 N m^{-2})	Excess available volume V_a^E/(× 10^{-6} $m^3 mol^{-1}$)
X_2	X_3						
System-I [Ethyl benzene (X_1) + toluene (X_2) + 1-propanol (X_3)]							
0.60	0.00	0.2816	0.0108	0.0040	-0.1834	662.37	0.7310
0.50	0.10	-0.0710	0.0161	0.0063	-0.0309	78.96	1.4384
0.40	0.20	-0.1486	0.0062	0.0029	-0.0373	-2509.86	1.1868
0.30	0.30	-0.2589	0.0052	0.0027	-0.0391	-1037.38	1.3722
0.20	0.40	-0.2862	-0.0045	-0.00078	-0.0436	-1313.37	1.0396
0.10	0.50	-0.3214	-0.0017	0.00003	-0.0499	-1488.00	1.2260
System-II [Ethyl benzene (X_1) + toluene (X_2) + 1-butanol (X_3)]							
0.60	0.00	0.2816	0.0108	0.0040	-0.1834	662.37	0.7310
0.50	0.10	-0.1100	0.0162	0.0063	-0.0256	-270.88	1.2597
0.40	0.20	-0.1823	0.0119	0.0048	-0.0572	-456.40	1.1344
0.30	0.30	-0.3005	0.0118	0.0047	-0.0499	-756.41	1.2431
0.20	0.40	-0.3849	0.0100	0.0042	-0.1277	-970.66	1.2458
0.10	0.50	-0.4345	0.0040	0.0020	-0.1332	-1084.07	0.9215
System-III [Ethyl benzene (X_1) + toluene (X_2) + 1-pentanol (X_3)]							
0.60	0.00	0.2816	0.0108	0.0040	-0.1834	662.37	0.7310
0.50	0.10	-0.1854	0.0118	0.0045	-0.0255	-274.45	0.8350
0.40	0.20	-0.3751	0.0160	0.0061	-0.0324	-563.52	1.1370
0.30	0.30	-0.5135	0.0184	0.0069	-0.0558	-746.21	1.2730
0.20	0.40	-0.6603	0.0243	0.0091	-0.0599	-949.34	1.6740
0.10	0.50	-0.7201	0.0251	0.0094	-0.0596	-1117.01	1.7370

of the third component. But in the case of system **III** the values of L_f^E increases with increase in concentration. The positive excess value clearly indicate that the existence of molecular interaction in the mixture. According to Ramamoorthy *et al.*[20] negative value of excess intermolecular free length L_f^E indicates that sound waves cover longer distances due to decrease in intermolecular free length describing the dominant nature of hydrogen bond interaction between unlike molecules. Fort *et al.*[18], indicated that the positive values of excess free length should be attributed to the dispersive forces and negative excess values should be due to charge transfer. In the present case the positive values of excess parameters show a weak interaction for all the three stystems. Spencer *et al.*[21] have also reported a similar observation on the basis of excess values of free length.

Table-3 gives a qualitative picture of the excess free volume for the ternary liquid mixtures. This indicates the extent of deviation from ideal with the molar composition of the mixtures. The excess values for all the three systems are found to be negative. In all the systems studied excess free volume is negative and gradually decreases with increases in concentration of third component. For some binary liquid mixtures, Fort *et al.*[18], noticed the negative excess free volume tends to decrease as the strength of the interaction between the unlike molecules increases, although they do not parallel with the excess compressibilities. However, in the present investigation, the reverse trend is observed in excess free volume *vs.* mole fraction which indicates that there is a decrease in the strength of interaction between components of these mixtures with the increase in the mole fraction of the third component.

In the study of liquid mixtures the variation of the internal pressure may give some information regarding the nature and strength of the forces existing between the molecules. The excess internal pressures are negative and non-linear in system **I** and the same decrease with the increase in concentration of third component in system **II** and **III**. The negative values of π_i^E indicate that only dispersion and dipolar forces are operating with complete absence of specific interaction.

Fort and Moore[18] observed that when interaction occurs between the molecules of the two species, the excess available volume becomes increasingly negative. This is due to closer approach of unlike molecules. The positive excess available volume obtained in the present study reflects the absence of stronger interaction. Dissociation also leads to positive contribution.

In the system containing ethyl benzene-toluene-1-propanol theoretical sound velocity obtained from CFT shows a deviation of 1.38 to 1.86 per cent where as those calculated using FLT deviate around 1.47 to 1.84 per cent. In the case of ethyl benzene-toluene-1-butanol, CFT shows a deviation of 0.78 to 1.29 per cent where a FLT shows a deviation of 1.41 to 1.59 per cent. For the system ethyl benzene-toluene-1-pentanol, the deviation in CFT is 0.86 to 1.78 per cent where as in FLT it is 0.97 to 2.02 per cent.

From the above, it is clear that the collision factor theory shows better agreement than the free length theory. Such a conclusion was also suggested by Kannappan *et al.*[22] for different ternary mixtures.

The relative interaction term (Table-4) for the systems of ethyl benzene-toluene -1-propanol and ethyl benzene-toluene-1-pentanol is around 0.9 and for the system ethyl benzene-toluene-1 butanol is unity except at 0.1 mol fraction of the third component. For the mixtures behaving

TABLE-4
VALUES OF ULTRASONIC VELOCITY OF EXPERIMENTAL AND CALCULATED ON THE BASIS OF FLT AND CFT, THE MODULUS OF PERCENTAGE DEVIATION OF VELOCITY AND RELATIVE INTERACTION (RI) AT 303K

Mole fraction		Ultrasonic velocity (ms^{-1})			$\left\|\frac{\Delta U}{U}\right\|$ %		Relative interaction
X_2	X_3	U_{exp}	CFT	FLT	CFT	FLT	RI
System-I [Ethyl benzene (X_1) + toluene (X_2) + 1-propanol (X_3)]							
0.50	0.10	1273.4	1294.2	1294.8	1.63	1.68	0.9947
0.40	0.20	1269.7	1287.9	1288.4	1.44	1.47	0.9956
0.30	0.30	1258.6	1281.1	1281.5	1.79	1.82	0.9947
0.20	0.40	1255.7	1273.0	1274.2	1.38	1.47	0.9959
0.10	0.50	1243.4	1265.6	1266.3	1.86	1.84	0.9950
System-II [Ethyl benzene (X_1) + toluene (X_2) + 1-butanol (X_3)]							
0.50	0.10	1276.9	1293.4	1296.1	1.29	1.51	0.9974
0.40	0.20	1273.2	1287.0	1291.3	1.00	1.42	1.0002
0.30	0.30	1266.5	1280.6	1286.3	1.12	1.56	1.0023
0.20	0.40	1261.0	1274.3	1281.1	1.05	1.59	1.0046
0.10	0.50	1258.1	1267.9	1275.8	0.78	1.41	1.0079
System-III [Ethyl benzene (X_1) + toluene (X_2) + 1-pentanol (X_3)]							
0.50	0.10	1285.8	1296.8	1298.3	0.86	0.97	0.9967
0.40	0.20	1278.9	1294.0	1295.8	1.18	1.32	0.9959
0.30	0.30	1274.9	1291.1	1293.3	1.27	1.44	0.9956
0.20	0.40	1266.4	1288.2	1290.8	1.72	1.93	0.9946
0.10	0.50	1262.8	1285.3	1288.4	1.78	2.02	0.9940

in an ideal manner, the relative interaction term is to be taken as unity[23]. Since this parameter takes a value less than unity in system **I** and **III** and equal to unity in system **II**, it may be confirmed that weak molecular interaction exists in all the cases. From magnitude of the interaction term, it is inferred that the strength of interaction in the system **I** and **III** is more than that in the system **II**. Nickam *et al.*[24] studied existence of intermolecular interaction based on the relative interaction.

Conclusion

The excess ultrasonic properties reveal that the presence of molecular interaction in all the systems were studied. The mixtures of ethyl benzene and toluene with different alkanols the strength of intermolecular interaction decreases with increase in size of alkanol-molecules. It also confirms the absence of the complex formation in mixtures. However, from the magnitude as well as the tendency in sign of the excess parameters, it is concluded that a weak molecular interaction exists in the mixtures which may be due to dispersion forces and dipolar interaction between the unlike molecules.

It is interesting to note that from the trend of excess parameters, dispersion forces are dominant in all the mixtures, which strongly confirms the existence of weak interaction among them.

The CFT is best suited than FLT for the evaluation of sound velocities in ternary systems. The relative interaction term is found to be a useful factor in the elucidation of molecular interaction.

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