Asian Journal of Chemistry Vol. 21, No. 8 (2009), 6359-6368

Thermo Acoustical Studies of Some Aliphatic Alcohols in Toluene with Aniline at 303 K

S. THIRUMARAN* and K. RAMYA
Department of Physics (D.D.E.), Annamalai University, Annamalainagar-608 002, India
E-mail: thiru_phymaran@yahoo.co.in

The ultrasonic velocity, density and viscosity have been measured for the ternary mixtures of aliphatic alcohols such as 1-propanol, 2-propanol, 1-butanol and 2-butanol with aniline in toluene at 303 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) and acoustic impedance (Z). The excess values of some the above parameters are also evaluated and discussed in the light of molecular interaction in the mixtures. It is observed that the alkanols form strong dipole-dipole interactions with aniline. It is also interesting to note that the specific interaction between them affects, on going to higher alcohol due to less proton-donating tendency of aniline. The donor-acceptor complexation reactions were also observed in the present study.

Key Words: Adiabatic compressibility, Free length, Free volume, Acoustic impedance.

INTRODUCTION

Velocity of sound waves in a medium is fundamentally relates to the binding forces between the atoms or the molecules. The variation of ultrasonic velocity and related parameters throw much light upon the structural changes associated with the liquid mixtures having weakly interacting components[1-3] as well as strongly interacting components[4-6]. Though, a number of investigations are carried out in liquid mixtures having alcohol as one of the components, ternary systems with iso-alcohols as one of the components has been scarcely reported. Further, ternary mixtures with secondary alcohols as one of the component are indispensable for the industrial rectification column to avoid the formation of azeotropes.

Further, accurate knowledge of thermodynamic mixing properties such as adiabatic compressibility, intermolecular free length, free volume, molar volume, acoustic impedance, *etc.* and their excess values for mixtures of protic, aprotic and associated liquids has a great importance in theoretical and applied areas of research. The deviation from ideality and specific or non-specific interactions have been revealed. Alcohols[7-13] and aniline[14-17] exist as associated structures in liquid state. Thus upon mixing aniline with alcohols of varying chain lengths and branching, interesting properties due to specific interactions arising from charge-transfer, dipole-dipole interactions, donor-acceptor and hydrogen bonding formation forces may be observed.

Thermodynamic properties of aliphatic alcohols + toluene + aniline mixtures are of particular interest, because alcohols provide protons and aniline accepts it forming donor-acceptor type complexes. To the best of knowledge of the authors, there are no reports of acoustic behaviour of aliphatic alcohols with a base such as aniline. Hence, in the present study, the authors had performed a thorough study on the interaction of aliphatic alcohols with weakly polar component toluene and with strongly polar aniline, using ultrasonic velocity data at 303 K.

The main purpose of this study is to characterize the molecular interactions in these systems by the ultrasonic velocity values of these mixtures.

The present work deals with the measurement of ultrasonic velocity and evaluation of the related parameters in the following ternary liquid systems at 303 K.

System-I	1-propanol + toluene + aniline
System-II	2-propanol + toluene + aniline
System-III	1-butanol + toluene + aniline
System-IV	2-butanol + toluene + aniline

EXPERIMENTAL

The liquid mixtures of various concentrations in mole fraction were prepared by taking AR grade chemicals, which were purified by standard methods[18]. 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 ± 0.2 ms^{-1}. The density and viscosity are measured using a Pycknometer and an Ostwald's viscometer of an accuracy of ± 0.1 kg m^{-3} and ± 0.001 Nsm^{-2}, respectively. All the precautions were taken to minimize the possible experimental error. The set-up is checked for known liquids. 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

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

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

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

where K_T is a temperature dependent constant.

Free volume (V_f) has been calculated from relation:

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

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:

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

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:

$$Z = U\rho \quad (5)$$

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

Excess value (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 δi.

RESULTS AND DISCUSSION

The experimentally determined values of the density (ρ), viscosity (η) and ultrasonic velocity (U) of four liquid systems at 303 K are tabulated in Table-1.

The values of adiabatic compressibility (β), intermolecular free length (L_f), free volume (V_f), internal pressure (π_i) and acoustic impedance (Z) of the four systems are presented in Table-2. The excess values of the some of the above parameters for the four liquid systems are given in the Table-3.

In all the mixtures, the ultrasonic velocity 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[19,20]. It is well known that alcohols are highly associative in nature. The C-O bond is weaker in the case of secondary alcohols due to the +I (electron repelling) effect of the alkyl groups, while the O-H bond is weaker in primary alcohols, as the electron density increases between the O-H bond and the hydrogen, which tends to separate as a

TABLE-1

VALUES OF DENSITY (ρ), VISCOSITY (η) AND ULTRASONIC VELOCITY (U) FOR

X_1	X_3	ρ (kg/m^3)	η ($\times 10^{-3}$ NSm^{-2})	U (m/s)
System I: 1-propanol + toluene + aniline				
0.0000	0.6001	951.382	1.3661	1457.4
0.0998	0.5001	948.400	1.2811	1452.4
0.1999	0.4000	941.700	1.1211	1444.0
0.3000	0.2999	937.800	0.9899	1439.7
0.4000	0.1999	929.400	0.8911	1431.4
0.4999	0.1000	918.400	0.8722	1429.4
0.5999	0.0000	904.700	0.7911	1422.9
System II: 2-propanol + toluene + aniline				
0.0000	0.6000	949.200	1.2495	1461.60
0.1000	0.5000	941.310	1.1495	1459.70
0.2000	0.4000	936.220	0.9248	1449.20
0.3000	0.3000	931.895	0.8905	1432.80
0.4000	0.1999	924.200	0.8245	1422.40
0.5000	0.0999	918.201	0.7491	1415.40
0.5999	0.0000	910.186	0.7166	1409.70
System III: 1-butanol + toluene + aniline				
0.0000	0.6000	952.047	1.3495	1462.65
0.0999	0.5001	938.400	1.2195	1444.70
0.2000	0.3999	906.100	1.1020	1420.90
0.3001	0.2999	881.400	1.0053	1387.65
0.4000	0.1999	868.400	0.9879	1374.70
0.5000	0.1001	842.400	0.9632	1364.80
0.6000	0.0000	820.021	0.9321	1352.40
System IV: 2-butanol + toluene + aniline				
0.0000	0.6001	950.867	1.3742	1458.26
0.1000	0.5001	932.821	1.1849	1439.40
0.2000	0.4000	902.870	1.1219	1395.60
0.3000	0.3000	886.912	1.0286	1352.04
0.4001	0.1999	868.228	0.8951	1328.40
0.5000	0.1000	842.684	0.8145	1308.40
0.6000	0.0000	822.916	0.7811	1279.95

proton. Thus primary alcohols are most reactive, when there is cleavage of the O-H bond, while secondary alcohols are most reactive, when there is cleavage of C-O bond. During O-H cleavage, a proton is given out, showing that alcohols are acidic in nature. The acidic nature increases from tertiary to secondary and from secondary to primary as the +1 effect decreases. When primary and secondary alcohol molecules are mixed with aniline molecule, they exhibit complex behaviour, which lead to decrease of ultrasonic velocity[21].

TABLE-2
VALUES OF ADIABATIC COMPRESSIBILITY (β), FREE LENGTH (L_f) FREE VOLUME (V_f) AND INTERNAL PRESSURE (π_i) AND ACOUSTIC IMPEDANCE (Z) FOR

X_1	X_3	β ($\times 10^{-10}$ $m^2 N^{-1}$)	L_f ($\times 10^{-10}$ m)	V_f ($\times 10^{-7}$ $m^3 mol^{-1}$)	π_i ($\times 10^6$ Nm^{-2})	Z ($\times 10^6$ Kg $m^{-2} s^{-1}$)
System I: 1-propanol + toluene + aniline						
0.0000	0.6001	4.949	0.4439	1.1113	511.37	1.3865
0.0998	0.5001	4.999	0.4461	1.1229	508.00	1.3775
0.1999	0.4000	5.093	0.4503	1.3195	506.38	1.3598
0.3000	0.2999	5.145	0.4526	1.4485	501.38	1.3500
0.4000	0.1999	5.251	0.4572	1.5303	498.97	1.3303
0.4999	0.1000	5.329	0.4606	1.6013	489.31	1.3128
0.5999	0.0000	5.459	0.4662	1.6959	476.12	1.2873
System II: 2-propanol + toluene + aniline						
0.0000	0.6000	4.9316	0.4431	1.2758	482.91	1.3874
0.1000	0.5000	4.9859	0.4455	1.3666	480.82	1.3740
0.2000	0.4000	5.0859	0.4499	1.7226	478.33	1.3567
0.3000	0.3000	5.2270	0.4562	1.7371	474.04	1.3352
0.4000	0.1999	5.3479	0.4614	1.8144	467.41	1.3146
0.5000	0.0999	5.4363	0.4652	1.9515	458.18	1.2996
0.5999	0.0000	5.5285	0.4692	1.9993	449.31	1.2831
System III: 1-butanol + toluene + aniline						
0.0000	0.6000	4.9097	0.4421	1.1378	502.68	1.3925
0.0999	0.5001	5.1057	0.4509	1.2606	487.81	1.3557
0.2000	0.3999	5.4663	0.4665	1.3313	468.18	1.2875
0.3001	0.2999	5.8921	0.4843	1.3922	465.55	1.2369
0.4000	0.1999	6.0255	0.4898	1.4559	460.86	1.2073
0.5000	0.1001	6.3729	0.5037	1.4964	459.68	1.1497
0.6000	0.0000	6.5872	0.5121	1.5275	458.15	1.1225
System IV: 2-butanol + toluene + aniline						
0.0000	0.6001	4.9455	0.4437	1.1026	507.49	1.3866
0.1000	0.5001	5.1742	0.4539	1.3090	479.78	1.3427
0.2000	0.4000	5.6866	0.4758	1.3139	475.51	1.2601
0.3000	0.3000	6.1679	0.4955	1.3816	468.77	1.1991
0.4001	0.1999	6.5269	0.5098	1.6037	446.23	1.1534
0.5000	0.1000	6.9319	0.5253	1.7156	431.67	1.1020
0.6000	0.0000	7.4175	0.5434	1.7472	429.15	1.0533

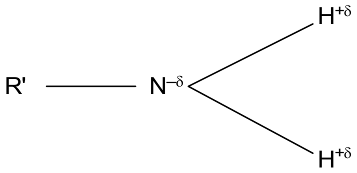
It is clear from the Table-2 that the value of β (adiabatic compressibility) shows an inverse behaviour as compared to the ultrasonic velocity (U). The adiabatic compressibility (β) increases with increase of concentration. It is primarily the compressibility that increases due to structural changes of molecules in the mixture leading to a decrease in ultrasonic velocity[22-24]. Such a continuous increase in adiabatic compressibility with respect to the solute concentration has been qualitatively ascribed to the effect

TABLE-3
EXCESS VALUES OF ADIABATIC COMPRESSIBILITY (β^E), FREE LENGTH (L_f^E), INTERNAL PRESSURE (π_i^E) AND ACOUSTIC IMPEDANCE (Z^E) FOR

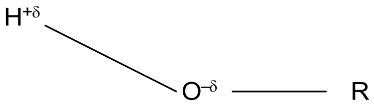
X_1	X_3	β^E ($\times 10^{-10}$ $m^2 N^{-1}$)	L_f^E ($\times 10^{-10}$ m)	π_i^E ($\times 10^6$ Nm^{-2})	Z^E ($\times 10^6$ kg $m^{-2} s^{-1}$)
System I: 1-propanol + toluene + aniline					
0.0000	0.6001	-0.0619	-0.0020	-49.24	0.0462
0.0998	0.5001	-0.5376	-0.0168	-73.81	0.0156
0.1999	0.4000	-0.9719	-0.0339	-96.83	0.0685
0.3000	0.2999	-1.4481	-0.0529	-123.24	0.1293
0.4000	0.1999	-1.8686	-0.0696	-147.02	0.2332
0.4999	0.1000	-2.3178	-0.0873	-178.05	0.2783
0.5999	0.0000	-2.7151	-0.1029	-212.62	0.4148
System II: 2-propanol + toluene + aniline					
0.0000	0.6000	-0.0990	-0.0018	-77.63	-0.0451
0.1000	0.5000	-0.8967	-0.0222	-96.95	0.0202
0.2000	0.4000	-1.6522	-0.0438	-116.66	0.0815
0.3000	0.3000	-1.8057	-0.0634	-138.18	0.1387
0.4000	0.1999	-2.3766	-0.1488	-161.98	0.1969
0.5000	0.0999	-2.9403	-0.1791	-194.43	0.2606
0.5999	0.0000	-3.5259	-0.1927	-214.51	0.3227
System III: 1-butanol + toluene + aniline					
0.0000	0.6000	-0.1009	-0.0002	-57.86	-0.0115
0.0999	0.5001	-0.3481	-0.0093	-80.72	0.0099
0.2000	0.3999	-0.4312	-0.0121	-108.27	0.0375
0.3001	0.2999	-0.4498	-0.0126	-118.98	0.0449
0.4000	0.1999	-0.7593	-0.0255	-131.57	0.0500
0.5000	0.1001	-0.8563	-0.0301	-140.90	0.0645
0.6000	0.0000	-1.0851	-0.0400	-150.34	0.0838
System IV: 2-butanol + toluene + aniline					
0.0000	0.6001	-0.1066	-0.0019	-53.13	-0.0142
0.1000	0.5001	-0.5191	-0.0065	-94.83	0.0015
0.2000	0.4000	-0.8233	-0.0082	-113.02	0.0375
0.3000	0.3000	-1.2864	-0.0123	-133.77	0.0480
0.4001	0.1999	-1.3190	-0.0164	-170.31	0.0590
0.5000	0.1000	-1.3629	-0.0195	-198.86	0.0654
0.6000	0.0000	-1.6410	-0.0201	-215.38	0.0868

of hydrogen bonding or dipole-dipole interactions[25]. Such an increase in adiabatic compressibility (β) with increasing concentration of alcohols indicate significant interaction between aniline and alcohols forming dipole-dipole interactions. The other parameter intermolecular free length (L_f) show reverse trend as adiabatic compressibility, which is also found to be increased in all the four liquid systems with increase in molar concentration of alcohols. This is attributed to the addition of interacting molecules break up the molecular clustering of the other, releasing several dipoles for interaction, especially in the system-II, between 2-propanol and toluene, there will be an increase of cohesive energy and hence the free length (L_f) increases in the mixture after mixing. On the basis of sound propagation in liquid[26], the increase in free length after mixing results in decrease in ultrasonic velocity. The intermolecular free length is found to be predominant factor on determining the nature of sound velocity variation in liquid mixtures[27]. 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. The mechanism of reaction between aniline and alkanols can be explained as follows:

The dipoles in aniline and alcohol arise due to difference in electro negativities of nitrogen, oxygen and hydrogen. They are in the order nitrogen > oxygen > hydrogen. Hence, dipolar molecules are pictured as:

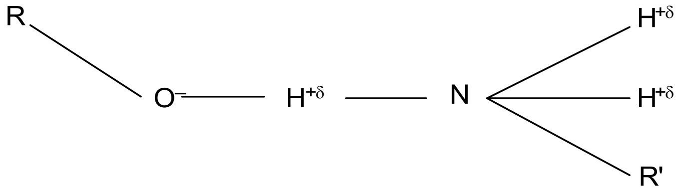


and



There are three possibilities of dipole-dipole interactions: (1). Linkage between $N^{-\delta}$ of amine with $H^{+\delta}$ alcohol, alcohol being a proton donor, (2) Linkage between $O^{-\delta}$ of alcohol with $H^{+\delta}$ of amine, here amine acts as a proton donor, (3) Linkage between $O^{-\delta}$ of alcohol with $N^{-\delta}$ of amine, alcohol being proton donor.

However, the third possibility is less likely, since electronegativities of N (3.5) and oxygen (3) are very close. Therefore, aniline-alcohol complexation may arise due to first two possibilities. The second possibility of linkage between $O^{-\delta}$ alcohol and $H^{+\delta}$ of aniline is remote, because of the satiric hindrance of hydrogen groups from aniline to oxygen in alcohol. However, the first possibility of dipole-dipole interaction between $N^{-\delta}$ group of aniline with $H^{+\delta}$ group of alcohol is most likely, which can be represented as follows:



The N-atom is *sp*^{3} hybridized and the shape of the amine is pyramidal[28], there is a lot of space on the outside of the apex of pyramid (N atom position) for the OH to penetrate and enter into complexation. It is further assumed that complex formation between aniline and alcohols is due to polarization effect and not due to charge transfer interactions[29].

The free volume (V_f) is also found to be increased with increasing concentration of alcohols. The hydrogen bonded association of alcohols breaks up gradually with the addition of toluene lead to expansion of volume[30]. Further, the decrease in internal pressure (π_i) in all the four liquid systems with increasing molar concentration clearly confirms this prediction.

It is observed that in all four liquid systems, the value of acoustic impedance (Z) is found to be decreased, which are furnished in Table-2. 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 molecular interactions between the unlike molecules.

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 four liquid systems are presented in Table-3.

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[31]. In the present study, the excess adiabatic compressibility (β^E) and excess free length (L_f^E), exhibit negative deviations in all the four liquid systems over the entire composition range. These are tabulated in Table-3. The strength of the interaction between the component molecules increase, when excess values tend to become increasingly negative. Or otherwise the increasing of negative values with increasing concentration of alcohols, thereby indicating strong molecular association between the aniline and alcohol molecules[32,33]. It may also be interpreted as due to dipole-induced dipole or dipole-dipole interactions between the unlike molecules. The maximum value of excess adiabatic compressibility (β^E) as well as excess free length (L_f^E) is observed in the case of system-II (2-propanol + toluene + aniline).

Further with higher alcohols, however β^E and L_f^E become less negative suggesting less of specific interaction of aniline with higher alcohols due to their less proton donating tendencies. It is to be noted here that β^E and L_f^E values of branched chain alcohols are smaller than those of their straight chain homologues probably due to more steric repulsion between -NH_2 group of aniline and alkyl chain of the alcohol. Such effects have also been observed earlier[34,45]. The perusal of Table-3 shows that the excess internal pressure (π_i^E) which is often described in terms of molecular interactions, whose negative values clearly supporting this.

Further, it is again well supported by the excess values of acoustic impedance (Z^E) which are furnished in Table-3 are all positive in all the four liquid systems over the entire range of composition. The almost positive excess values of Z^E clearly suggest that there is a strong molecular interaction[30] existing between the aniline-alkanols. Similar observations were observed by earlier workers[36,37] supports the present investigation.

Conclusion

By exhaustively analyzing all the related parameters and some of their excess values, it is obvious that there exist a strong molecular interaction between the unlike molecules. The alkanols form strong dipole-dipole interactions with aniline. However, on going to higher alcohols, the specific interaction between them decreases due to less proton donating tendency of aniline. Followed by, the donor-acceptor complex formation reactions were also observed in the present study.

ACKNOWLEDGEMENT

The authors are thankful to Dr. A.N. Kannappan, Professor & Head, Department of Physics, Annamalai University for encouragement.

REFERENCE

1. S.L. Oswal, P. Oswal and R.P. Phalak, *J. Sol. Chem.*, **27**, 507 (1998).
2. J. Rajasekar and P.R. Naidu, *J. Chem. Engg. Data*, **41**, 373 (1996).
3. G. Arul and L. Palaniappan, *J. Acoust. Soc. (India)*, **28**, 393 (2000).
4. M. Kalidoss and R. Srinivasamoorthy, *J. Pure Appl. Ultrason.*, **19**, 9 (1997).
5. J.D. Pandey and A.K. Shukla, *J. Pure Appl. Ultrason.*, **15**, 37 (1997).
6. P.S. Nikam, T.R. Mahala and M. Hassan, *J. Pure Appl. Phys.*, **37**, 92 (1999).
7. R. Battino, *Chem. Rev.*, **1,** 23 (1971).
8. J.S. Rowlison, Liquids & Liquids Mixtures, Butterworths, London, edn. 2, p. 159 (1969).
9. R. Mecke, *Faraday Soc.*, **9**, 161 (1950).
10. G.N. Swami, G. Dharmaraju and G.K. Raman, *Can. J. Chem.*, **58**, 229 (1980).
11. R.E. Verall, D.K. Jacobson, M.V. Hershberger and R.W. Humry, *J. Chem. Engg. Data*, **24**, 289 (1979).
12. S. Sarkar and R.N. Joarder, *J. Chem. Phys.*, **99**, 2032 (1993).
13. S. Sarkar and R.N. Joarder, *J. Chem. Phys.*, **100**, 5118 (1994).
14. C.N.R. Rao, B.D.N. Rao, P. Venkateswarlu and A.S.N. Murthy, *Can. J. Chem.*, **40**, 963 (1962).
15. J. Freeney and L.H. Sutaliffe, *J. Chem. Soc., Part-I*, 123 (1962).
16. R.K. Nigam, P.P. Singh and K.C. Singh, *Can. J. Chem.*, **57**, 2211 (1979).
17. H. Wolf and D. Mathias, *J. Phys. Chem.*, **77**, 2081 (1973).

18. J.A. Riddick and W.B. Bunker, Organic Solvents, Wiley New York (1986).
19. A.S. Rao, B.V. Naidu and K.J.C. Rao, *Acoust. Soc. (India)*, **28**, 303 (2000).
20. K. Tiwari, C. Patra and V. Chakravorthy, *Acoust. Lett.*, **19**, 53 (1995).
21. S. Singh, I. Vibhu, M. Gupta and J.P. Shukla, *Chin. J. Phys.*, **45**, 412 (2007).
22. P.S. Nikam, V.M. Kapade and M. Hassan, *J. Pure Appl. Phys.*, **38**, 170 (2000).
23. M. Rastogi, A. Awasthi, M. Guptha and J.P. Shukla, *Asian J. Phys.*, **7**, 739 (1998).
24. U. Sridevi, K. Samatha and A. Viswanadasarma, *J. Pure Appl. Ultrason.*, **26**, 1 (2004).
25. S. Hyder, A.K. Nain and A. Ali, *Indian J. Phys.*, **74B**, 63 (2000).
26. K. Tewari, C. Patra, C. Padhy and V. Chakaravorthy, *Phys. Chem. Liq.*, **32**, 149 (1996).
27. M. Kaulgud, *Z. Phys. Chem.*, **36**, 365 (1963).
28. P.S. Nikam, M. Hassan and R.B. Pathak, *J. Pure Appl. Ultrason.*, 18, 19(1996).
29. S. Tripathi, G.S. Roy and B.B. Swain, *J. Pure Appl. Phys.*, **31**, 828 (1993).
30. R. Mehra and M. Pancholi, *Indian J. Pure Appl. Phys.*, **45**, 580 (2007).
31. M.A. Palofax, *Indian J. Pure Appl. Phys.*, **31**, 90 (1993).
32. R.J. Fort and W.R. Moore, *Trans. Faraday Soc.*, **61**, 2102 (1993).
33. B.K. Rout and V. Chakravorthy, *Indian J. Chem.*, **33A**, 303 (1994).
34. J. Karunakaran, K.D. Reddy and M.V.P. Rao, *J. Chem. Engg. Data*, **27**, 348 (1982).
35. T.M. Aminabhavi, M. Arulaguppi and R.H. Balundgi, *J. Chem. Engg. Data*, **38**, 31 (1993).
36. G. Arul and L. Palaniappan, *J. Pure Appl. Phys.*, **39**, 561 (2001).
37. P.S. Nikam, M. Hassan and V.V. Patil, *Indian J. Pure Appl. Phys.*, **38**, 693 (2000).

(*Received*: 18 December 2008; *Accepted*: 28 May 2009) AJC-7619