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Ultrasonic Studies on Binary Liquid Mixtures of Triethylamine with Carbitols at 308.15 K

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ABSTRACT

Densities and ultrasonic velocities of binary liquid mixtures of triethylamine with carbitols, methyl carbitol, ethyl carbitol and butyl carbitol, have been measured at 308.15 K. The observed data have been utilized to calculate various acoustical parameters such as isentropic compressibility (K_S), intermolecular free length (L_f), and acoustic impedance (Z). The various excess properties such as excess ultrasonic velocity (u^E), excess acoustic impedance (Z^E), excess isentropic compressibility (K_S^E), and excess intermolecular free length (L_f^E) were calculated. The results were discussed in terms of the existence of intermolecular interactions between the components in the liquid mixtures under the study.

Key words: *Ultrasonic velocity, Triethylamine, Cellosolves, Carbitols, Excess isentropic compressibility, Excess intermolecular free length, Excess acoustic impedance.*

1. INTRODUCTION

Ultrasonic wave propagation affects the physical properties of the medium, and hence, can furnish information about molecular interactions of the liquid and liquid mixtures. The sign and magnitude of the non-linear deviations from ideal values of velocities and adiabatic compressibilities of liquid mixtures with compositions are attributed to the difference in molecular size and strength of interaction between unlike molecules [1-3]. Ultrasonic velocities have been adequately employed in understanding the nature of molecular interaction in pure liquids [4] and binary and ternary mixtures [5-7]. The method of studying the molecular interaction from the knowledge of variation of thermodynamic parameters and their excess values with composition gives an insight into the molecular process [8-16].

Triethylamine (TEA) is a weakly polar [17] liquid with a low dielectric constant (ε=2.42 at 298.15 K). Whereas, cellosolves and carbitols are polar and associative in nature. TEA shows more molecular interactions with other molecules due to the presence of a lone pair of electrons on N-atom. From the theoretical point of view, cellosolves and carbitols are drawn a special interest in the recent years [18-21]. The study of molecular interactions of cellosolves and carbitols is of interest because of investigating the effect of simultaneous presence of etheric and hydroxyl oxygen atoms in the same molecule. At the same time, it is also important to note that the presence of etheric oxygen enhances the ability of the –OH group to form hydrogen bonds with other organic solvents [22-25]. The binary liquid mixtures of TEA with organic liquids possessing hydroxyl group may be useful to study the molecular interactions. These interactions also form a basis to study the properties of these binary solvent systems, systematically. Thus, the different types of interactions between TEAs with cellosolves and carbitols are discussed from the present viscosity studies.

In view of the above, the present research aims to measure densities and ultrasonic velocity of binary mixtures of TEA with carbitols (methyl carbitol [MC], ethyl carbitol [EC], and butyl carbitol [BC]) at 308.15 K, and using this data, excess ultrasonic velocity (u^E), excess acoustic impedance (Z^E), excess isentropic

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compressibility (K_s^E), and excess intermolecular free length (L_f^E) were calculated. The results were discussed in terms of intermolecular interactions between unlike molecules.

2. MATERIALS AND METHODS

2.1. Materials

TEA, MC, EC, and BC were purchased from E-Merck, Mumbai, and used as recieved.

2.2. Methods

Mixtures were prepared by mixing weighed amounts of the pure liquids adopting the method of a closed system by using an electonic balance with the precision of ±0.01 mg. Mixtures were allowed to stand for sometimes before every measurement so as to avoid air bubbles. The purities of the liquids were checked by comparing the values of densities and ultrasonic velocities with literature data (Table 1) [17,26]. The measurements were made with proper care in an AC room to avoid evaporation loss.

The densities (ρ) of liquids and their mixtures were measured using bicapillary pycnometer having a capillary diameter of 0.85 mm, which was calibrated using double-distilled water. The necessary buoyancy corrections were applied. The density values were reproducible within ±0.2 kg m^{-3}. The ultrasonic velocity (u) measurements were made by a single frequency (2 MHz) variable path interferometer with an accuracy of ±0.03%. A thermostatically controlled, well-stirred constant temperature water bath, whose temperature was controlled to ±0.05 K, was used for all the measurements.

3. RESULTS AND DISCUSSION

From the measured densities (ρ) and ultrasonic velocities (u), the various acoustical parameters such as K_S, Z, L_f, and Y^E were calculated using the following equations 1, 2, 3, and 4, respectively, and are incorporated in Table 2 for the binary systems under the study.

$$K_S=1/u^2\rho \quad (1)$$

$$Z=\rho u \quad (2)$$

$$L_f=K(K_s)^{1/2} \quad (3)$$

Where K is Jacobson's constant [27].

The excess functions Y^E have been calculated using the relation:

$$Y^E=Y_{mix}-(X_1Y_1+X_2Y_2) \quad (4)$$

Where Y denotes u, Z, K_S, and L_f, respectively, X is the mole fraction of TEA, and suffixes 1 and 2 denote the components 1 and 2 in binary mixtures and the values are given in Table 2. The dependence of u^E, Z^E, K_S^E, and L_f^E on the mole fraction of TEA (X_{TEA}) for all the six systems were calculated and given in Table 2, where Y^E is u^E, Z^E, K_S^E, and L_f^E parameters.

The variations of u, Z, K_s, L_f, and R_A with a mole fraction of TEA for all the mixtures are shown in Table 2. From the Table 2, it is clear that K_s and L_f increase with an increase in X_{TEA} for all the mixtures studied, while u, R_A, and Z exhibit reverse trends. The decrease in u and the corresponding increase in the K_s and L_f with X_{TEA} observed in the present systems are in accordance with the view proposed by Erying and Kincaid [28].

The increase in K_S and L_f with X_{TEA} in the present investigation indicates the presence of specific

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

Liquid	Density ρ × 10^{-3} (kg m^{-3})		Ultrasonic velocity (u) (m s^{-1})	
	Experimental	Literature	Experimental	Literature
TEA	0.7144	0.7144 [26]	1079.9	1070.0 [26]
2-(2-methoxyethoxy) ethanol (MC)	1.0073	1.0065 [17]	1414.5	1414.5 [17]
2-(2-ethoxyethoxy) ethanol (EC)	0.9838	0.9839 [17]	1385.8	1385.2 [17]
2-(2-butoxyethoxy) ethanol (BC)	0.9480	0.9479 [17]	1357.0	1356.4 [17]

MC=Methyl carbitol, EC=Ethyl carbitol, BC=Butyl carbitol

Table 2: Values of density (ρ), ultrasonic velocity (u), excess ultrasonic velocity(u^E), acoustic impedance (Z), excess acoustic impedance (Z^E), isentropic compressibility (K_S), excess isentropic compressibility (K_S^E), intermolecular free-length (L_f), ecess intermolecular free-length (L_f^E) and relative association (R_A) for the binary liquid mixtures of triethyamine (TEA) with carbitols at 308.15 K.

Mole fraction of THF (X_{THF})	$\rho\times 10^{-3}$ (kg m^{-3})	u (m s^{-1})	u^E (m s^{-1})	$Z\times10^{-4}$ (kg m^{-2} s^{-1})	$Z^E\times10^{-4}$ (kg m^{-2} s^{-1})	$K_S\times10^{11}$ (m^2 N^{-1})	$K_S^E\times10^{11}$ (m^2 N^{-1})	$L_f\times10^{12}$ (m)	L_f^E x 10^{12} (m)	Relative association (R_A)
TEA+MC										
0.0000	1.0073	1382.7	0.0000	1.3928	0.0000	51.93	0.0000	4.7724	0.0000	1.0000
0.1006	0.9792	1355.0	3.6671	1.3268	−0.2827	55.62	−3.3587	4.9393	−0.8903	0.9787
0.1997	0.9504	1327.3	6.8665	1.2615	−0.5968	59.72	−6.2062	5.1182	−1.6227	0.9565
0.3001	0.9210	1297.3	8.1712	1.1948	−0.9595	64.51	−8.4574	5.3195	−2.1645	0.9340
0.4003	0.8915	1265.7	7.8135	1.1284	−1.3137	70.02	−9.9801	5.5418	−2.4912	0.9115
0.5002	0.8627	1233.3	6.5624	1.0640	−1.4829	76.21	−10.7969	5.7815	−2.6357	0.8897
0.5998	0.8340	1201.3	5.6176	1.0019	−1.4389	83.09	−10.9037	6.0368	−2.6172	0.8677
0.7004	0.8047	1168.6	4.2847	0.9404	−1.2750	91.00	−10.0469	6.3177	−2.3680	0.8449
0.8001	0.7760	1136.0	2.7712	0.8815	−0.9001	99.86	−8.1797	6.6181	−1.9009	0.8225
0.9003	0.7462	1103.5	1.5135	0.8234	−0.4205	110.05	−5.0119	6.9477	−1.1542	0.7986
1.0000	0.7144	1070.9	0.0000	0.7651	0.0000	122.06	0.0000	7.3168	0.0000	0.7723
TEA+EC										
0.0000	0.9748	1345.3	0.0000	1.3114	0.0000	56.68	0.0000	4.9861	0.0000	1.0000
0.1003	0.9545	1322.7	4.9223	1.2625	0.5917	59.88	−3.3565	5.1250	−0.9493	0.9847
0.1999	0.9314	1299.9	9.4526	1.2107	0.8543	63.54	−6.2109	5.2791	−1.7290	0.9665
0.3001	0.9071	1276.8	13.8474	1.1582	1.0746	67.62	−8.6772	5.4462	−2.3941	0.9469
0.4002	0.8822	1252.7	17.2149	1.1051	1.2382	72.23	−10.6114	5.6287	−2.9014	0.9268
0.5001	0.8573	1226.7	18.6274	1.0516	1.3480	77.52	−11.8600	5.8309	−3.2079	0.9069
0.6002	0.8313	1198.3	17.6949	0.9961	1.2666	83.77	−12.1454	6.0617	−3.2327	0.8863
0.7001	0.8047	1168.4	15.2074	0.9402	1.1311	91.03	−11.4210	6.3188	−2.9907	0.8652
0.8009	0.7769	1137.3	11.7670	0.8836	0.9740	99.51	−9.5263	6.6067	−2.4609	0.8429
0.9008	0.7475	1105.2	7.0795	0.8261	0.6888	109.52	−6.0479	6.9310	−1.5462	0.8188
1.0000	0.7144	1070.9	0.0000	0.7651	0.0000	122.06	0.0000	7.3168	0.0000	0.7908
TEA+BC										
0.0000	0.9397	1325.3	0.0000	1.2454	0.0000	60.59	0.0000	5.1550	0.0000	1.0000
0.1005	0.9262	1314.7	14.9672	1.2177	2.0564	62.47	−4.2993	5.2343	−1.3796	0.9883
0.2009	0.9095	1304.8	30.6090	1.1867	3.7830	64.58	−8.3548	5.3223	−2.6708	0.9729

(*Contd...*)

Table 2: Continued...

Mole fraction of THF (X_{THF})	$\rho\times 10^{-3}$ (kg m^{-3})	u (m s^{-1})	u^E (m s^{-1})	$Z\times10^{-4}$ (kg m^{-2} s^{-1})	$Z^E\times10^{-4}$ (kg m^{-2} s^{-1})	$K_S\times10^{11}$ (m^2 N^{-1})	$K_S^E\times10^{11}$ (m^2 N^{-1})	$L_f\times10^{12}$ (m)	L_f^E x 10^{12} (m)	Relative association (R_A)
0.3004	0.8917	1288.7	39.8218	1.1491	4.8042	67.53	−11.5257	5.4423	−3.6217	0.9578
0.3998	0.8725	1265.3	41.7091	1.1040	5.0627	71.59	−13.5735	5.6036	−4.1574	0.9429
0.5002	0.8521	1238.8	40.7509	1.0556	5.0460	76.47	−14.8614	5.7916	−4.4481	0.9274
0.6004	0.8299	1210.6	38.0418	1.0047	4.7685	82.22	−15.2743	6.0052	−4.4776	0.9102
0.6999	0.8057	1181.3	34.0546	0.9518	4.2574	88.94	−14.6677	6.2459	−4.2217	0.8909
0.7993	0.7796	1149.3	27.5022	0.8960	3.4843	97.11	−12.6489	6.5264	−3.5657	0.8700
0.9005	0.7501	1113.9	17.6872	0.8355	2.2692	107.45	−8.4947	6.8649	−2.3679	0.8458
1.0000	0.7144	1070.9	0.0000	0.7651	0.0000	122.06	0.0000	7.3168	0.0000	0.8162

THF=Tetrahydrofuran, TEA=Triethylamine, MC=Methyl carbitol, EC=Ethyl carbitol, BC=Butyl carbitol

interactions between TEA and carbitols molecules. A similar observation was made by Pal and Das [24] from their ultrasonic velocity studies for the binary liquid mixtures. The added TEA to cellosolves tends to cause breaking of associates in the cellosolve and carbitol molecules which leads to an increase in K_s and L_f. In the present investigation, the relative association (R_A) is found to decrease with X_{TEA} for all the systems studied. This is due to the dissociation of cellosolve-cellosolve associates. The decrease in the values of Z for the mixtures under the study is in agreement as per the requirement given by the equation $Z=\rho u$ [11,17].

To have a better understanding of intermolecular interactions between unlike molecules, excess functions are considered to be more sensitive than those discussed above. With this view, the variation of the excess functions u^E, Z^E, K_s^E, and L_f^E with X_{TEA} have been shown in Figures 1-4, respectively. In general, the negative K_s^E and L_f^E values are shown by the systems involving hydrogen bonding where one of the components is associated in pure state [17]. The negative K_s^E and L_f^E values (Figures 3 and 4) may be regarded as evidence for the formation of hydrogen bonds between the lone-pair electrons of nitrogen atom of TEA and the hydrogen atom of the hydroxyl group of cellosolves [17].

From the Figures 1 and 2, it is seen that for all the mixtures studied K_s^E and L_f^E are negative over the whole range of composition and show a minima at ≈ 0.6 X_{TEA}. The negative values of K_s^E and L_f^E fall in the following order.

TEA + BC > TEA + EC > TEA + MC

The above sequence indicates that the decrease in compressibility increases with the chain length of the alkoxy group of carbitols. Similar observation was made by Pal *et al*. [17,29]. The result further suggests A-B type of forming (Scheme 1).

Figures 3 and 4 show the variation of u^E and Z^E, respectively. u^E values are positive over the whole range of composition for all the systems. The positive deviations observed in u^E are due to the close packing of molecules present in the mixture and may further be explained due to strong interactions, formed as a result of hydrogen bonding between unlike molecules. Whereas, in the case of Z^E, the experimental results are positive for the systems TEA + EC and TEA + BC but negative for the system TEA + MC. Negative values of Z^E for the system TEA + MC indicate less interactions between the component molecules when compared to the other systems. u^E and Z^E values support our earlier view that the interactions increase with an increase in chain length of the alkoxy group of carbitols. The

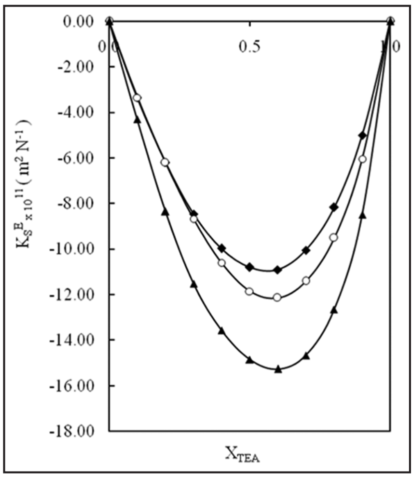


Figure 1: Plots of excess isentropic compressibility (K_S^E) versus mole fraction of triethylamine (TEA) (XTEA) at 308.15 K for the binary mixtures of TEA with methyl carbitol (−♦−), ethyl carbitol (−○−), and butyl carbitol (−▲−).

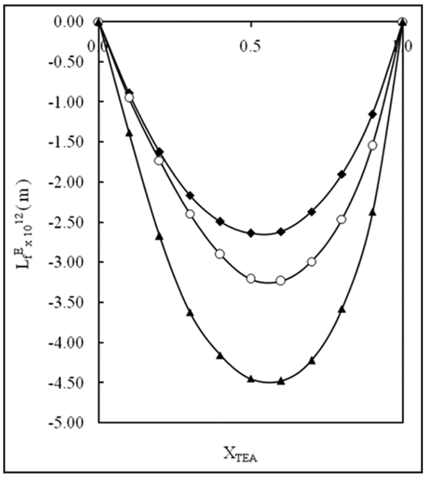
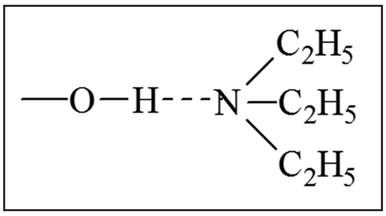


Figure 2: Plots of excess intermolecular free length (Lf^E) versus mole fraction of triethylamine (X_{TEA}) with methyl carbitol (−♦−), ethyl carbitol (−○−), and butyl carbitol (−▲−).



Scheme 1: Schematic representation of interaction between triethylamine and carbitols.

values of u^E and Z^E for the systems under study are in the following order:

TEA + BC > TEA + EC > TEA + MC

It has been suggested that the concentration at which the excess functions exhibiting maxima or minima

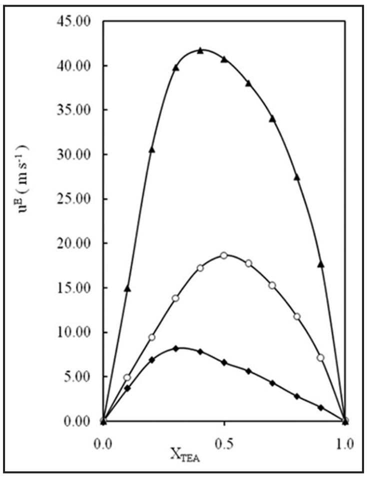


Figure 3: Plots of excess ultrasonic velocity (u^E) versus mole fraction of triethylamine (TEA) (X_{TEA}) at 308.15 K for the binary mixtures of TEA with methyl carbitol (−♦−), ethyl carbitol (−○−), and butyl carbitol (−▲−).

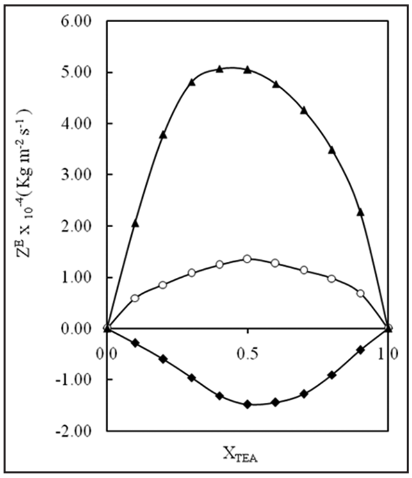


Figure 4: Plots of excess acoustic impedance (Z^E) versus mole fraction of triethylamine (TEA) (X_{TEA}) at 308.15 K binary mixtures of TEA with methyl carbitol (−♦−), ethyl carbitol (−○−), and butyl carbitol (−▲−).

indicates strong interactions between the component molecules [11,13], which in turn suggests the complex formation at this composition between the unlike molecules. The results further suggest A-B type of interactions forming as shown in Scheme 1. The above trend suggests an increase in hydrogen bonding between unlike molecules with an increase in chain length of alkoxy group of carbitols.

4. CONCLUSION

Ultrasonic method is a powerful probe for characterizing the physicochemical properties and existence of molecular interaction in the mixture. In addition, the density, ultrasonic velocity, and the derived excess acoustical parameters provide evidence of confirmation. It is concluded that the dependence of ultrasonic velocity on composition of the mixtures is satisfactorily explained. The trends in the variation of the parameters derived from ultrasonic velocity and the sign and extent of deviation of the excess function from rectilinear dependence on composition of these mixtures suggest the presence of molecular interaction between the components of binary mixtures. The interactions are primarily due to the electron donor-acceptor interactions existing between the components of the mixtures.

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