Computational and experimental study of molecular interaction in a ternary liquid mixture of cyclohexylamine+ethanol+benzene

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In the present investigation, we undertake the study of molecular interaction in a ternary mixture of cyclohexylamine (CHA), ethanol (Eth) and benzene (B) theoretically as well as experimentally. We study the interaction between CHA dimer and complexes (Eth-CHA, B-CHA, CHA-Eth-B) theoretically using DFT (Density functional theory) in gas phase. It includes the quantum chemical calculations of interaction energy, bond length, and identification of the intermolecular interaction from structural parameter analysis and molecular orbital analysis. Further, we experimentally determine the excess thermo-acoustic parameters of the mixture. For this, we have measured the ultrasonic velocity (u) and density (ρ) of the ternary mixture of CHA+Eth+B and of pure components at 303 K, 308 K and 313 K temperature. From these experimental values of u and ρ, excess thermo-acoustic parameters have been estimated using standard relations. The variations in excess acoustic parameters with the concentration of CHA (x_1) are discussed in terms of the interactions and molecular geometry of the liquid mixture. At lower concentration, weak interactions dominate while on increasing x_1 strong forces between molecules take place. The strong H-bonding interaction between CHA-ethanol molecules and molar volume differences of pure components leads to interstitial accommodation of molecules. The experimentally determined excess properties behaviour is supported by the theoretical calculation.

The study of structural and excess thermodynamic properties of mixed solvents is an interesting area of chemistry of liquids. Mixing of liquids introduces some extra degree of freedom and results into new phenomena that are not present in pure liquids, such as complex formation, association, dissociation etc[1]. Douheret *et al*.[2] pointed out the importance of examination of consequences of the mixing process. The process of mixing is considerably affected by various molecular interactions[3,4]. Also, the nature of the intermolecular interaction presented in the system determines the thermodynamic properties, the kinetics and mechanics of chemical activities[5].

Rawat *et al*.[6] explained that the important light aromatics like benzene, toluene, xylene can be obtained from liquid-liquid extraction of petroleum-based aromatics. According to Deal *et al*.[7] solvents with high concentration of non-hydrogen bonded polar groups, such as the oxygen in ketones and sulphones; nitrogen in nitriles and alkylated or cyclic amides or amines, possess high selectivity as a solvent for the extraction process. This motivated us to study the behaviour of the mixture of cyclic amine i.e., cyclohexylamine (CHA) mixture with benzene (B) and ethanol (Eth). Various researchers[8-15] have studied the binary mixture theoretically as well as experimentally but the work on the ternary mixture is still fragmentary.

Coupled-cluster single-, double-, and perturbative triple-excitations [CCSD(T)] method and Moller-Plesset second-order perturbation (MP2) method has been widely used in literature to estimate the XH-π (X=O, N, C) interaction due to their accuracy[16-19]. The decomposition of the total energy of XH-π interaction into various components i.e., electrostatic energy, dispersive energy, exchange repulsion energy and induction energy studies show that the dispersion is essential to the binding energy of XH-π interaction[18]. So, the popular density functional theory (DFT) methods such as B3LYP fail to predict the correct binding energy for XH-π interaction. Although, methods that include dispersion such as wB97X-D, B3LYP-D, BLYP-D, M05-2X, M06-2X, etc. give promising results for non-covalent XH-π interaction[16]. We optimized the geometry by DFT/B3LYP method and calculated the single-point energy by DFT/ B3LYP and DFT/ wB97X-D method.

Thus, we study the liquid mixture theoretically by quantum chemical calculation using DFT[20] and experimentally by ultrasonic method[21]. We choose DFT for the study of intermolecular interaction as it is computationally simple and phenomenally successful in describing the structural and electronic properties of molecular systems[13]. The standard basis sets 6-311++G(d,p) provide sufficiently accurate description of the alcohol/π-complexes[9] and CHA/alcohol complexes[10,11].

The ultrasonic method is directly related to large numbers of thermo-dynamical properties and excess thermodynamic properties. Prigogine *et al.*[22] have shown that excess parameters give relative strength of the interaction A-A, B-B and A-B in the mixture of A and B liquids. The thermodynamic parameters are necessary for the development of thermodynamical models which are used to optimize the process of chemical, pharmaceutical, and other industries. Accurate knowledge of various thermophysical properties is essential for the right design of several types of relevant industrial equipment. Also, various liquid state theories are based upon thermodynamic consideration.

In the present paper, we have studied the molecular interaction in a ternary liquid mixture of CHA, B and Eth by the theoretical method and by analysing the trend of change of excess thermo-acoustic parameters with concentration. We have optimized the structure of monomer, dimer and complexes; and calculated the single point energy on these fully optimized geometries using DFT. Then the intermolecular interaction energy was calculated using 'Supermolecular method'[23]. These interaction energies and geometries are helpful to investigate the interaction in the mixture. We have reported the ultrasonic velocity (u) and density (ρ); and some excess thermo-acoustic parameters like excess adiabatic compressibility (β^E or K_s^E), excess intermolecular free length (L_f^E), excess molar volume (V_m^E) and excess acoustic impedance (Z^E) of ternary mixtures of CHA+Eth+B at (303, 308, 313) K temperature. The variations in these excess acoustic parameters were interpreted in terms of the interaction and structural geometry of the liquid mixtures. It is observed that at lower concentration of CHA (x_1) weak interaction dominates while at high concentration the forces between molecules become strong. The individual molar volume of each pure component is quite different. Our results show that

when the ternary mixture is formed then the interstitial accommodation of molecules of components into voids of each other take place; also, the strong H-bonding interaction between CHA-Eth molecules is there which gives a compact structure to the system.

Materials and Methods

Experimental

The chemicals of AR (analytic reagent) grade with minimum assay of 99.9% were used without further purification. The mixtures of various concentrations in mole fraction were prepared by mass using a digital balance (manufactured by Aczet) with a precision of ± 1 mg. In all systems, mole fraction of second component, Eth (x_2 =0.4) was kept fixed while mole fractions of other two (x_1 of CHA and x_3 of B) were varied from 0.0 to 0.6; to have the mixture of different compositions. The ultrasonic velocities in liquid mixtures have been measured using a Mittal type (M-81D) ultrasonic interferometer working at 4 MHz frequency with an accuracy of ± 0.1 m s^{-1} with a provision for temperature constancy. Density of solution is measured with relative density (RD) bottle of 10 mL by weighing the firstly empty RD bottle first and then bottle with solution using digital electronic balance to an accuracy of ± 1 mg. The complete method was described elsewhere[24].

Computational

All quantum chemical calculations including geometry optimization and single point energy calculation were done in GAMESS program[25,26] using a web-based interface facility www.chemcompute.org[27,28]. The initial molecule(s) was built in Avogadro[29] program and then fully optimized using GAMESS calculation. The common practice of running a high-level single point energy calculation on the geometry computed by the use of a cheaper method is as effective as performing all calculations at a higher level of theory[18]. The geometry of monomer and dimer were optimized at B3LYP level theory (Cheng *et al.*[18] have shown that DFT/B3LYP theory optimized the geometry with good accuracy). The single point energy calculation and geometry optimization of B, Eth and CHA; CHA dimer and their complex (Eth-CHA, B-CHA, CHA-Eth-B) were done using DFT with B3LYP/6-311++G(d, p) and wB97X-D/6-311++G(d, p) level. The wB97X-D functional includes the dispersive interaction.

Results and Discussion

Theoretical study: Interaction energy and structural changes on dimerization

Molecular geometry has a decisive effect on interaction energy. The structures of monomer and dimer were optimized with DFT with B3LYP level of theory with 6-311G++(d, p) basis. The optimized structure of monomer(s), dimer and complexes are shown in Figs. 1-3. On these optimized structures, the single point energy was calculated with the B3LYP and wB97X-D level of theory. The interaction energy of the benzene-ammonia complex calculated by the wB97X-D/6-311++G(d,p) level of theory is -2.94 kcal/mol (Supplementary Data) which is higher than the reported value of -2.40 kcal/mol with MP2 level of theory[18,30]. There is a variation of 22.5% which shows that the wB97X-D theory is suitable to study the NH/π interaction. Interaction energies were determined by using the "Supermolecular" method. Single point energy, interaction energy and dipole moment are summarized in Table 1.

The dimers are a simple prototype to study the interaction between the molecules in the ternary liquid mixture. The most stable structure depends on the equilibrium forces between the molecules. As a result of dimerization, there is interaction between structural subunits, which modifies the bond length, bond angle or leads to some preferred geometry. So, these structural changes and interaction energies give most fundamental information to understand the intermolecular interaction. The most significant structural changes on dimerization are as follows.

CHA dimer: CHA molecule (Fig. 1a) is a non-planar molecule due to strained C-ring and capable of formation of H-bonding due to -NH$_2$ group. The optimized structure of CHA dimer (Fig. 2a) shows that two CHA molecules interact via intermolecular H-bonding between N and H of length 2.348 Å. In the acceptor molecule, the bond length
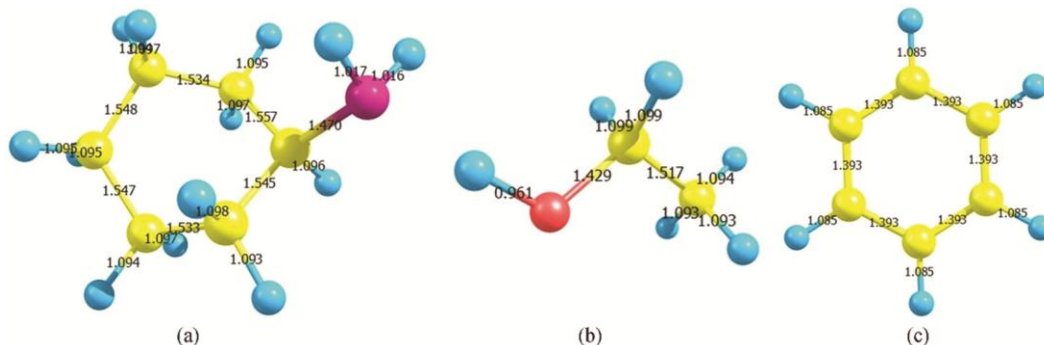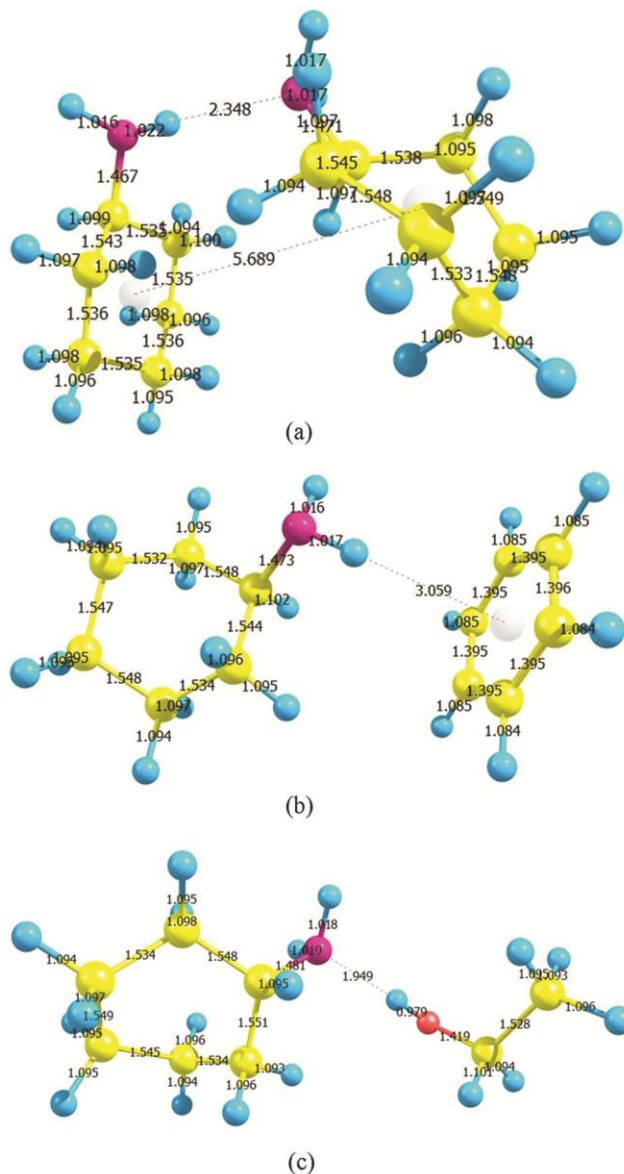

Fig. 2 — Optimized geometry of (a) cyclohexylamine dimer, (b) cyclohexylamine+benzene complex and (c) cyclohexylamine+ ethanol complex obtained by DFT/B3LYP/6-311++G(d, p)

Fig. 1 — Optimized geometry of (a) cycloexylamine, (b) ethanol and (c) benzene monomer obtained by DFT/B3LYP/6-311++G(d, p)

r(N-H) of CHA gets increased by 0.005 Å and r(C-N) gets decreased by 0.003 Å.

CHA+B (Fig. 2b): On clustering of CHA molecule with B molecule, the bond length of B r(C-C) gets increased by 0.002 to 0.003 Å while the bond length r(N-C) of CHA molecule is increased by 0.003 Å. In CHA-B cluster, N-H bond of CHA takes a geometry perpendicular to π- ring of B.

CHA+Eth (Fig. 2c): On clustering, the bond length r(N-C) and r(N-H) of CHA get increased by 0.011 Å and 0.002 Å, respectively, while the bond length in ethanol, r(O-H) is increased by 0.010 Å, r(C-O) decreased by 0.010 Å and r(C-C) increased by 0.011 Å. A similar increment in bond length r(O-H) of alcohol on clustering with CHA was reported by Ranjbar *et al.*[10]. They reported that in CHA-iso-butanol cluster, the bond length r(O-H) of iso-butanol get increased from 0.956 Å to 0.982 Å. The CHA and Eth molecule interact via H-bonding with interaction length of 1.949 Å.

CHA+Eth+B (Fig. 3): In CHA+Eth+B complex, CHA molecule interacts with Eth via H-bonding with interaction distance 1.926 Å. while N-H bond of CHA interact with π-ring of B with interaction distance 3.053 Å. The complex is equilibrated via H-bonding and NH/π bonding.

The single point energy, dipole moment and interaction energy from both B3LYP and wB97X-D method are presented in Table 1. It is seen that the wB97X-D functional gives larger interaction energies compared to B3LYP functional. A rather higher interaction energies for wB97X-D functional than B3LYP functional indicated that wB97X-D functional include dispersive interactions and improves interaction energy results qualitatively, such as for CHA-B complex and CHA-Eth-B complex.

Eth is a polar molecule (Table 1) and has the tendency of self-association through H-bonding. It also has a strong H-bond formation tendency with polar molecules. Due to being polar, it can also interact with non-polar molecules via dipole-induced dipole interaction[31]. On the other hand, benzene is a non-polar molecule (μ=0, Table 1) but has a π-electron density and can interact with a molecule that has the capability to accept this. In B dimer[19,32], dispersive forces cause benzene dimer to take face to face π-stacking structure while weak CH/π interaction causes edge-to-face T-shaped structure. The distance between the centres of two CHA molecules is 5.689 Å. The interaction energy of CHA dimer is -12.448 kcal/mol calculated by wB97X-D level theory which is in the order of H-bonding interaction energies. Despite having large interaction energy, the distance is found to be large which is because of the repulsion of two non-polar C-rings of CHA molecules.
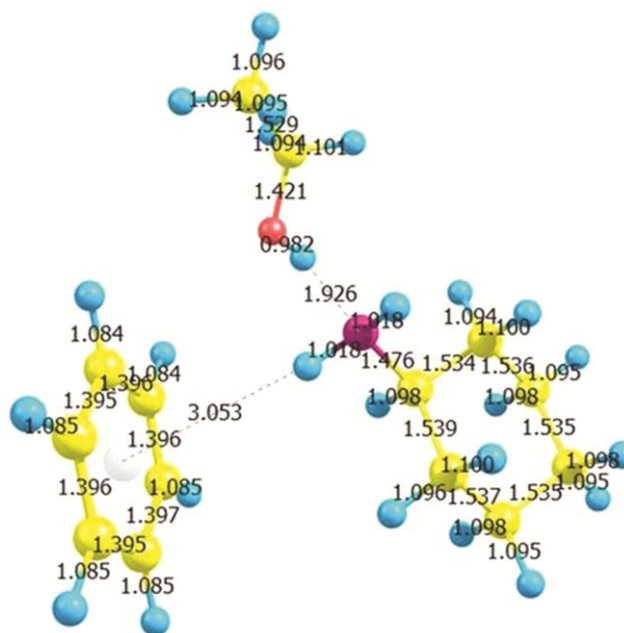

Fig. 3 — Optimized geometry of cyclohexylamine+ethanol+benzene complex obtained by DFT/B3LYP/6-311++G(d, p)

Table 1 — The values of single point energy and dipole moment of monomer and dimer obtained by DFT with B3LPY/6-311++G(d, p) and wB97X-D/6-311++G(d,p) level calculation and the interaction energies (ΔE) calculated by Supermolecular method

System	B3LYP				wB97X-D			
	Energy (au)	ΔE (au)	ΔE (kcal/ mol)	Dipole moment (D)	Energy (au)	ΔE (au)	ΔE (kcal/mol)	Dipole moment (D)
CHA	-291.1013024884			1.326	-291.2168832777			1.357
Eth	-155.0014209619			1.750	-155.0421486608			1.774
B	-232.1598925415			0.000	-232.2233040759			0.000
CHA dimer	-582.2174433827	-0.014838	-9.311	2.357	-582.4536044602	-0.019838	-12.448	2.375
CHA - B	-523.2627560132	-0.001561	-0.980	1.492	-523.4462704057	-0.006083	-3.817	1.516
CHA - Eth	-446.1129117058	-0.010188	-6.393	3.465	-446.2729661767	-0.013934	-8.744	3.474
CHA – Eth - B	-678.2870328705	-0.024417	-15.322	3.017	-678.5160252641	-0.033689	-21.140	3.027

The interaction energy between CHA-B molecules is -3.817 kcal/mol. The optimized geometry of CHA+B (Fig. 2b) depicts that NH_2 group of CHA takes a preferred geometry, N-H bond takes position perpendicular to π-plane of benzene similar to the OH/π interaction. The geometry suggests the H atom of NH_2 group of CHA act as π-electron density acceptor. The distance of polar hydrogen of CHA and benzene ring is 3.059 Å, slightly higher than the OH/π interaction distance 2.635 Å (in Supplementary Data, Fig. S1). This indicates that the interaction between CHA and B is weaker than OH/π interaction. Tsuzuki et al.[39] called it NH/π interaction and explained that long-ranged electrostatic interaction plays an important role in determining the magnitude and the directionality of this interaction. They have reported that OH/π interaction is stronger than NH/π interaction. The CHA-Eth-B geometry is equilibrated by the combined effect of NH/π and H-bonding interaction and the total interaction energy is -21.140 kcal/mol by wB97X-D level theory. In CHA-

CHA and CHA+Eth dimer there is H-bonding with different interaction strengths. The interaction in CHA+B cluster is NH/π interaction. The order of strength of the interaction of different complexes is: CHA dimer>CHA+Eth>CHA+B (Table 1).

Molecular orbital analysis

To study the orbital interaction contribution in NH/π and H-bonding and to better understand their nature, orbital analysis of CHA-B, CHA-Eth complexes and CHA-CHA dimer is done at wB97X-D level theory. Molecular orbitals are formed by the overlap of atomic orbitals. Such orbital interactions not only exist between atoms but also within special organic molecules. Orbital interaction occurs between close orbitals in which the distance between orbital centres is usually less than 3.0Å [Ref. 40,41]. Schematic diagrams of orbital interaction are presented in Figs. 4-6. HOMO (highest occupied molecular orbital) and LUMO (lowest unoccupied molecular orbital) levels of each complex represent that these are made of by the mixing of molecular orbital of
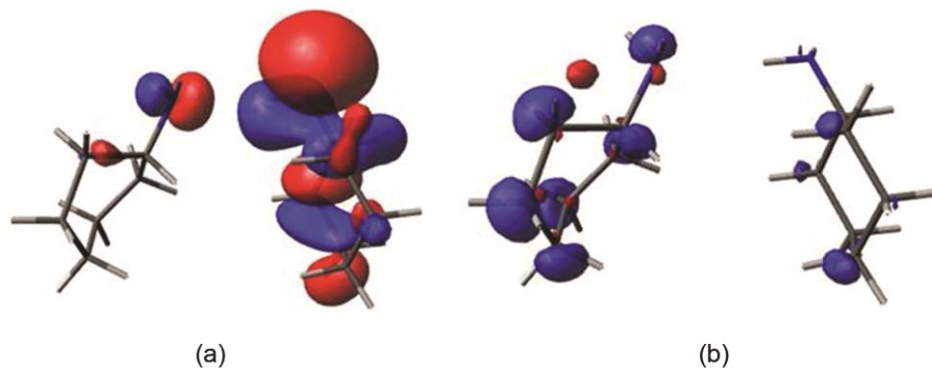

(a)   (b)

Fig. 4 — (a) HOMO and (b) LUMO of CHA dimer
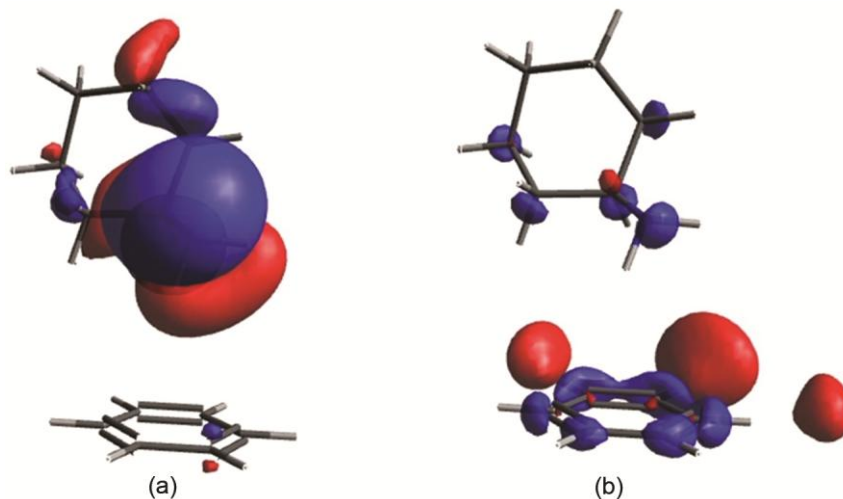

(a)   (b)

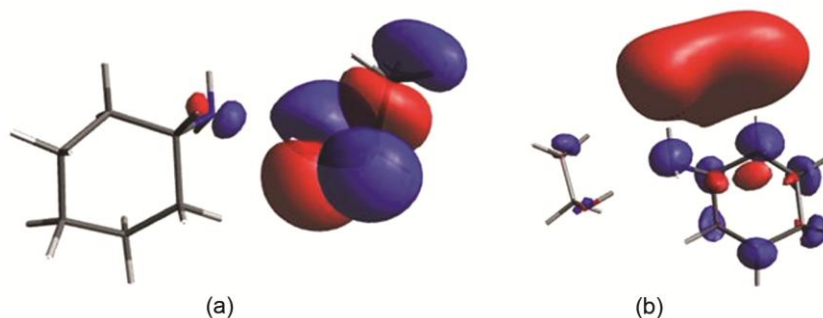Fig. 5 — (a) HOMO and (b) LUMO of CHA-B complex

Fig. 6 — (a) HOMO and (b) LUMO of CHA-Eth complex

Table 2 — Experimental speed of sounds and densities and calculated molar volume of pure liquids at (303, 308 and 313) K temperature

Liquid	Temperature (K)	Velocity (m s^{-1})	Density (kg m^{-3})	Molar volume ($V_m \times 10^{-5}$ m^3 mol^{-1})
Cyclohexylamine	303	1371.57	857.60	11.56361
	308	1351.25	852.60	11.63149
	313	1332.68	848.82	11.68333
Ethanol	303	1124.64	782.25	5.88930
	308	1106.47	778.52	5.91749
	313	1088.00	774.77	5.94619
Benzene	303	1262.90	868.06	8.99867
	308	1235.02	859.76	9.08554
	313	1212.95	855.18	9.13427

component molecules. This indicates that the orbital interaction is involved in all such interaction. The amount of such orbital mixing is different for different complexes. The similar orbital interaction is also reported in literature for conventional H-bonded complexes and NH/π bonded complexes[18-42].

Bulk behaviour study: Excess thermoacoustic properties

The experimental values of ultrasonic velocity (u) and density (ρ) of pure components at temperature (303, 308, 313) K were summarised in Table 2. From the experimental values of u and ρ the excess parameters were calculated by using the standard formule[24]. In Table 3, the experimental values of ultrasonic velocity, density; and calculated excess thermoacoustic parameters of the ternary mixture of CHA+Eth+B at temperature (303, 308, 313) K were summarised. The variation of excess velocity (u^E), excess density (ρ^E), excess adiabatic compressibility (β^E or K_s^E), excess intermolecular free length (L_f^E), excess molar volume (V_m^E) and excess acoustic impedance (Z^E) with mole fraction of CHA (x_1) are also shown in Supplementary Data, Figs. S2-S7, respectively. The variations in the sign and values of excess acoustic parameters help in determining the type and strength of the interaction between the unlike molecules of the mixture.

The strong interaction between unlike particles enhances the wave propagation in the medium while weak interaction leads to slow propagation of wave[43]. At x_1=0, the negative value of u^E (Table 3, Fig. S2) indicates that the interaction between B and Eth is weak. On adding CHA into the B-Eth complex, the interaction energy gets increased (Table 1) and results into an increased value of u^E. Further increasing x_1, CHA-CHA and CHA-Eth type interaction dominate over B-B or B-Eth interaction. The positive and increasing value of u^E with increasing x_1 indicates that the strength of interaction increases. Thus, the interaction between CHA-CHA and CHA-Eth will be stronger than B-B and B-Eth interaction.

Excess acoustic impedance Z^E is the measure of the propagation of wave through the medium. Strong interaction between molecules causes the pressure vibration to propagate easily and makes Z^E positive and increasing whereas weak interaction leads to less propagation of wave and will decrease the value of Z^E. In CHA+Eth+B mixture, positive and increasing values of Z^E (Fig. S3) indicate that strong interaction is present in the mixture. The variation of u^E and Z^E with temperature reveals that the temperature increment strengthens the interaction at lower x_1 while decreases the interaction strength at higher x_1.

Table 3 — Experimentally determined velocity(u) and density (ρ); and calculated excess thermodynamic parameter: excess velocity (u^E), excess density (ρ^E), excess adiabatic compressibility (β^E or K_s^E), excess intermolecular free length (L_f^E), excess molar volume (V_m^E) and excess acoustic impedance (Z^E) of the ternary mixture of cyclohexylamine +ethanol +benzene at (303, 308 and 313) K temperature and at 4 MHz frequency

x_1	u (m s^{-1})	ρ (kg m^{-3})	u^E (m s^{-1})	ρ^E (kg m^{-3})	$K_s^E \times 10^{-10}$ (N^{-1} m^2)	$L_f^E \times 10^{-11}$ (m)	$V_m^E \times 10^{-6}$ (m^3 mol^{-1})	$Z^E \times 10^4$ (kg m^{-2} s^{-1})
				303 K				
0.6	1349.47	853.04	76.671	25.579	-1.325	-0.474	-1.584	9.350
0.5	1332.00	851.25	70.068	22.740	-1.243	-0.440	-1.300	8.420
0.4	1312.29	850.33	61.221	20.771	-1.138	-0.398	-1.115	7.421
0.3	1289.36	848.99	49.160	18.393	-0.984	-0.338	-0.894	6.099
0.2	1263.85	847.86	34.515	16.219	-0.788	-0.264	-0.700	4.591
0.1	1235.48	844.58	17.014	11.893	-0.517	-0.165	-0.310	2.580
0.0	1201.00	838.33	-6.597	4.593	-0.107	-0.018	0.339	-0.283
				308 K				
0.6	1328.94	847.69	75.605	24.720	-1.371	-0.485	-1.527	9.072
0.5	1310.46	845.23	68.748	21.548	-1.282	-0.449	-1.205	8.086
0.4	1290.51	843.95	60.418	19.553	-1.177	-0.407	-1.018	7.137
0.3	1268.15	842.67	49.682	17.557	-1.032	-0.352	-0.838	5.990
0.2	1242.11	841.14	35.266	15.306	-0.826	-0.275	-0.641	4.508
0.1	1212.41	838.38	17.183	11.828	-0.537	-0.170	-0.334	2.577
0.0	1178.18	832.24	-5.422	4.973	-0.116	-0.022	0.275	-0.113
				313 K				
0.6	1305.83	846.35	71.026	27.159	-1.412	-0.494	-1.808	8.930
0.5	1288.91	844.71	66.071	24.882	-1.347	-0.467	-1.573	8.225
0.4	1270.24	842.87	59.379	22.400	-1.251	-0.429	-1.325	7.353
0.3	1248.45	841.43	49.565	20.328	-1.111	-0.376	-1.129	6.277
0.2	1223.06	839.59	36.145	17.846	-0.905	-0.300	-0.901	4.853
0.1	1193.88	835.79	18.942	13.415	-0.605	-0.192	-0.495	2.890
0.0	1158.79	828.00	-4.175	4.985	-0.136	-0.028	0.270	-0.007

mole fraction x_1: cyclohexylamine, x_2: ethanol (0.4), x_3: benzene

The theoretical study shows that at higher concentration conventional H-bonding takes place while at lower concentration weak interaction such as non-conventional H-bonding and π-stacking between the molecules taken place. Although, H-bonding interaction is less affected by temperature variation as compared to Hp-π interaction (suggested by Du et al.[31]) but due to C-ring in CHA molecule there is a decrease in the strength of interaction with the temperature at higher concentration.

The topological information can be accessed by the study of ρ^E, L_f^E, V_m^E and K_s^E. Molar volume in solution represents the aggregate of molecular size or the volume of a cluster of molecules which act as a single entity. Structure and molar volume of pure components highly affect the values and sign of V_m^E in the mixture as these factors are responsible for interstitial accommodation of molecules into each other[44]. For all temperatures, the molar volume of pure components follows the order CHA>B>Eth (Table 2). The sign and values of V_m^E depend upon the two main effects (i) positive contribution due to

disruption of intermolecular interaction on mixing and due to weak/ dispersive interaction between unlike molecules and (ii) negative contribution due to strong interaction between unlike molecules and due to interstitial accommodation of molecules of different sizes into each other's structure[8].

The positive and increasing values of ρ^E (Table 3 and Supplementary Data, Fig. S4) with increasing x_1 represent the strong interaction and well packing of molecules into voids of each-other. On moving from x_1=0.0 to 0.1, the heavier CHA molecules replace B molecules and the H-bonds between CHA and Eth cause an increase in the values of ρ^E. After x_1=0.1 to onward the excess density increases slowly and at higher concentrations the CHA-CHA interaction dominates. It is clear from geometry analysis that CHA-CHA interaction distance is large, and this makes the system compressible due to the non-polar ring of CHA molecules. But the presence of Eth (smaller size) and H-bonding of CHA-Eth and Eth-B molecules causes intermolecular fitting. These two-opposing features, H-bonding between molecules and

large space between CHA molecules in mixture cause a slow but steady increase in excess density.

L_f^E (Supplementary Data, Fig. S5) and K_s^E (Supplementary Data, Fig. S6) follow the same trends and become more negative with increase in x_1. The L_f^E, V_m^E and K_s^E values shows the well packing of molecules as x_1 increases. As discussed earlier, the H-bonding between CHA and Eth molecules leads to more negative values of L_f^E, V_m^E and K_s^E with increasing x_1. Simultaneously, the non-polar nature of C-ring of CHA molecules creates space between molecules due to repulsion and provides a suitable condition for the geometrical fitting of ethanol molecules. Similarly, molar volume difference (molecular size difference) between molecules makes the system more compact. Radovic *et al.*[18] and Ranjbar *et al.*[11] also reported the negative values of V_m^E in CHA+alcohol system due to strong H-bonding and interstitial accommodation. With increasing temperature, dispersive forces strengthen while strong forces get weak. Both these effects are found to be responsible for increase in the space between molecules with x_1 at the higher temperatures. At lower values of x_1 the change in L_f^E and K_s^E become less negative whereas at higher values of x_1 it becomes more negative.

Conclusions

In the present investigation, we study the molecular interactions in a ternary mixture of CHA, Eth and B with quantum chemistry method (theoretically) and with the ultrasonic method (experimentally). Quantum chemistry calculation reveals that in CHA-CHA dimer, interaction is the strong with interaction energy of -12.448 kcal/mol and has a lot of intermolecular spacing due to C-ring repulsion. The interaction between CHA-Eth molecules is the strong H-bonding with interaction energy -8.744 kcal/mol and between CHA-B is comparatively weak NH/π interaction with interaction energy -3.817 kcal/mol and it prefers orientation effect like OH/π interaction. The molecular orbital analysis shows that orbital interaction is also taking place in the complexes. We determine the ultrasonic velocity and density of CHA, Eth, B, and their ternary mixture. With the help of these parameters calculated the excess parameters viz. u^E, ρ^E, L_f^E, V_m^E, K_s^E and Z^E using standard relations. The values and sign of u^E and Z^E indicate that at lower values of x_1 weak interactions dominates while with increasing x_1 strong forces between molecules take place. The trends of ρ^E, L_f^E, V_m^E, and K_s^E indicates that strong

H-bonding interaction between CHA-Eth molecules and molar volume difference of pure components leads to interstitial accommodation of molecules into each other's structure. Temperature increase causes dispersive forces to increase in strength. Despite of the fact that H-bonding is less affected by temperature variation, H-bonding interaction between cyclohexylamine and ethanol molecules is more affected by temperature variation due to the dispersive forces of C-ring of cyclohexylamine.

Supplementary Data

Supplementary data associated with this article are available in the electronic form at http://nopr.niscair.res.in/jinfo/ijca/IJCA_60A(08)1072 -1080_SupplData.pdf.

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