Research Article

Thermoacoustic, Volumetric, and Viscometric Investigations in Binary Liquid System of Cyclohexanone with Benzyl Benzoate at T = 308.15, 313.15, and 318.15 K

Sk. Md Nayeem,[1] M. Kondaiah,[2] K. Sreekanth,[3] and D. Krishna Rao[4]

[1]Department of Physics, KRK Govt. Degree College, Addanki 523 201, India
[2]Department of Physics, NM Govt. Degree College, Jogipet, Medak District, Telangana 502270, India
[3]Department of Physics, PBN College, Guntur District, Nidubrolu 522124, India
[4]Department of Physics, Acharya Nagarjuna University, Nagarjuna Nagar 522 510, India

Correspondence should be addressed to D. Krishna Rao; krdhanekula@yahoo.co.in

Ultrasonic velocities (u), densities (ρ), and viscosities (η) of binary liquid mixtures of cyclohexanone with benzyl benzoate, including pure liquids, over the entire composition range have been measured at 308.15 K, 313.15 K, and 318.15 K. Using the experimental results, parameters such as molar volume (V_m), isentropic compressibility (k_s), intermolecular free length (L_f), acoustic impedance (Z), internal pressure (π_i), enthalpy (H), Gibbs free energy of activation of viscous flow (G^{*E}), and excess/deviation properties of these including partial molar volumes ($\overline{V}_{m,1}$ and $\overline{V}_{m,2}$), excess partial molar volumes ($\overline{V}_{m,1}^E$ and $\overline{V}_{m,2}^E$), partial molar volume of the components at infinite dilution ($\overline{V}_{m,1}^\infty, \overline{V}_{m,2}^\infty$), and excess partial molar volume at infinite dilution ($\overline{V}_{m,1}^{E,\infty}$ and $\overline{V}_{m,2}^{E,\infty}$) have been computed. The observed negative values of V_m^E, Δk_s, L_f^E, and π_i^E and positive values of z^E, H^E, ΔG^{*E}, $\Delta \eta$, and Δu for all the liquid mixtures studied clearly indicate the presence of strong dipole-dipole-type interactions, fitting of smaller molecules into bigger molecules. Further theoretical values of sound velocity and viscosity in the mixtures have been evaluated using various theories and have been compared with experimental values to verify the applicability of such theories to the systems studied.

1. Introduction

Volumetric, viscometric, and ultrasonic investigations of liquid mixtures are of considerable importance in understanding the intermolecular interactions occurring among component molecules and they find application in several industrial and technological processes [1, 2]. The work on medicinally used chemical compounds requires the attention of the society in all aspects including ultrasonic behaviour. Benzyl benzoate is a carboxylate ester which is used in oily injections and as an insect repellent and as acaricide and pediculicide in veterinary hospitals. It is an effective and inexpensive topical treatment for human scabies. It is a polar (μ = 2.06 D) molecule ($C^+=O^-$) with the structure shown in Figure 12. Behaviour of benzyl benzoate in many liquids such as aliphatic alkanes, aromatic alkanes, aliphatic alcohols, substituted benzenes, acetates, ketones, and DMSO (super solvent) has been thoroughly studied ultrasonically [3–6].

Ketone is an organic compound that contains a carbonyl group and two aliphatic or aromatic substituents containing the chemical formula $RCOR^1$. Here, R and R^1 may be same or different incorporated into a ring (alkyl, aryl, and heterocyclic radicals). Cyclohexanone is a ketone liquid. The chemical reactivity of the carbonyl group ($C^+=O^-$) plays important role in chemical reactions and is influenced considerably by steric effects. The greater electronegativity of O^- and high dipole moment make ketones polar (μ = 3.25 D). The resonance structure shown in Figure 12 illustrates this polarity. Thus a study on thermophysical properties data of binary liquid mixtures has attracted considerable interest in the literature

[7–10]. Cyclohexanone and its derivatives are used for the synthesis of pharmaceuticals, dyes, herbicides, pesticides, plasticizers, and rubber chemicals.

The liquids under investigation have been chosen on the basis of their medicinal and industrial applications. These applications have greatly stirred the need for extensive information on the thermodynamic, acoustic, and transport properties of these solvents and their mixtures [11–13]. Ultrasonic and density data for binary mixtures of benzyl benzoate with Isomers of butanol have been studied previously in our laboratory [14]. The literature survey reveals that Madhuri et al. [15] have reported ultrasonic, volumetric, and viscometric studies of benzyl benzoate with acetonitrile and benzonitrile. Recently, Sri et al. [16] reported ultrasonic velocity and density in binary liquids of certain aldehydes and esters. In the present investigation, a detailed study of the binary mixture of benzyl benzoate (BB) with cyclohexanone (CH) at three temperatures 308.15 K, 313.15 K, and 318.15 K is aimed. From the experimentally measured data of ultrasonic velocity (u), densities (ρ), and viscosities (η), thermodynamic and other related parameters like molar volume, isentropic compressibility, acoustic impedance, free length, relaxation time, internal pressure, enthalpy, and Gibbs free energy of activation of viscous flow are computed and in terms of some of these excess/deviation parameters, the nature of molecular interactions is predicted in the binary mixtures. An evaluation of velocities and viscosities using three different empirical theories is also attempted at all the three temperatures.

2. Experimental Section

Benzyl benzoate (BB) and cyclohexanone (CH) used in the present study were the AR grade products from LOBA Chemicals, India, and were purified by standard methods described in the literature [17–19]. The mass fraction purity of liquids obtained is >0.995. Before use, the chemicals were stored over 0.4 nm molecular sieves approximately for 72 h to remove water content and degassed and latter kept in air tight bottles. The mass measurements are performed with a METTLER TOLEDO (Switzerland make) AB135-S/FACT digital balance with an accuracy of ±0.01 mg. The uncertainty in the mole fraction is 10^{-4}. The ultrasonic velocity of pure liquids and their binary mixtures has been measured by using a multifrequency ultrasonic interferometer (M-82 model) supplied by Mittal Enterprises, New Delhi, at a fixed frequency of 2 MHz with an accuracy of ±0.2%. The temperature of liquid sample in the interferometer cell is maintained constant by circulating water pumped from constant temperature water bath. In the present study, the constant temperature water bath (digital electronic) supplied by Concord Instruments Co., Ltd., Chennai (RAAGA type), has been used. The instrument can maintain temperature to ±0.01 K as per its specifications.

Densities and viscosities of pure liquids and liquid solutions are determined using 5 cm^3 two stem double walled Parker & Parker-type pycnometer [20] and Ostwald viscometer which is standardized as described by Naidu and Prasad

[21] using triply distilled water, respectively. The estimated accuracy in measuring the density is 3 in 10^5 parts. The uncertainty in the viscosity measurement is ±0.2%. The detailed description of measurement of density and viscosity is discussed in our previous papers [22–25]. The densities, velocities, and viscosities of pure liquids in this investigation at temperature of 308.15 K, 313.15 K, and 318.15 K are compiled in Table 1 together with the literature data [15, 26] available. These results are found to be in good agreement with reported data.

3. Results and Discussion

The measured values of density, velocity, and viscosity and the calculated values of molar volume, isentropic compressibility, free length, acoustic impedance, internal pressure, relaxation time, enthalpy, and Gibbs free energy of activation of viscous flow are calculated using standard relations and are presented in Table 2 with mole fraction of cyclohexanone in benzyl benzoate. In the present investigation, the values of u and z increase and those of k_s and L_f decrease as the mole fraction increases. The variation of ultrasonic speed in a solution depends upon the increase or decrease of L_f after mixing the components. This is due to the fact that ultrasonic speed increases if the intermolecular free length decreases and vice versa. The decrease in the values of k_s and L_f with mole fraction in the present study indicates significant interactions between CH and BB molecules.

Study of deviation/excess properties plays vital role in the study of molecular interactions. The nonlinear variation of excess/deviation properties with mole fraction of BB is responsible for nonideality in the systems of binary liquid mixture [15]. The factors responsible for such departure from ideality may either be due to the presence of intermolecular forces between the constituents in the mixture or due to compound formation between solute and solvent, or as a result of association of either to form complex molecules [27]. These excess parameters throw light upon the strength of interaction and their variation with mole fraction finds application in typifying the physicochemical behaviour of liquid mixtures [28].

The deviation/excess properties have been calculated using the relation

$$Y^E = Y_{\text{real}} - Y_{\text{ideal}}, \tag{1}$$

where Y^E represent deviation/excess value of the parameter, Y_{real} is the experimental value of the parameter of liquid mixture, and Y_{ideal} is the ideal value of the parameter computed theoretically. The $Y^E = V_m^E$ is excess molar volume, Z^E is excess specific acoustic impedance, Δk_s is deviation in isentropic compressibility, L_f^E is excess intermolecular free length, π_i^E is excess internal pressure, H^E is excess enthalpy, ΔG^{*E} is excess Gibbs free energy of activation of viscous flow, $\Delta \eta$ is deviation in viscosity, and Δu is deviation in velocity.

The deviation in isentropic compressibility (Δk_s) has been calculated from

$$\Delta k_s = k_s - (\Phi_1 k_{s1} + \Phi_2 k_{s2}), \tag{2}$$

TABLE 1: Comparison of experimental values of ultrasonic velocity (u), density (ρ), and viscosity, (η) of pure liquids with the literature values concerned at 308.15, 313.15, and 318.15 K.

Liquid	Temp/K	$\rho/(kg \cdot m^{-3})$		$\eta/(10^{-3} \, N \cdot s \cdot m^{-2})$		$u/(m \cdot s^{-1})$	
		Exptl.	Lit.	Exptl.	Lit.	Exptl.	Lit.
Cyclohexanone	308.15	939.60	939.6 (26)	1.594	1.6013 (26)	1362.00	1362.0 (26)
	313.15	934.70	934.7 (26)	1.472	1.4561 (26)	1348.20	1348.2 (26)
	318.15	922.30	922.5 (26)	1.370	1.3101 (26)	1275.00	1272.8 (26)
Benzyl benzoate	308.15	1114.54	1114.5 (15)	7.118	—	1493.30	1493.3 (15)
	313.15	1109.70	1109.7 (15)	6.261	—	1471.40	1471.4 (15)
	318.15	1104.80	—	5.550	—	1440.00	—

where Φ_1, Φ_2 and k_{s1}, k_{s2} are the volume fractions and isentropic compressibilities of components 1 and 2, respectively. Since k_s is not additive on mole fraction but is additive on volume fraction, hence such values are calculated using volume fraction Φ_i:

$$\Phi_i = \frac{x_i V_i}{\sum x_i V_i}. \tag{3}$$

The excess/deviation properties have been fitted to a Redlich-Kister-type polynomial equation [29]:

$$Y^E = x(1-x) \sum_{i=0}^{j} A_i (1-2x)^i, \tag{4}$$

where x is the mole fraction of CH and for evaluating Δk_s, x is replaced by volume fraction (Φ_i) and A_i are the adjustable parameters of the function and are determined using the least square method. In the present investigation "i" values taken are from 0 to 4. The corresponding standard deviations $\sigma(Y^E)$ were calculated using the expression

$$\sigma\left(Y^E\right) = \left[\frac{\sum \left(Y_{exp}^E - Y_{cal}^E\right)^2}{(m-n)} \right]^{1/2}, \tag{5}$$

where "m" is the total number of experimental points and "n" is the number of coefficients in (4). The calculated values of the coefficients A_i along with the standard deviations (σ) are given in Table 3.

Figure 1 represents the variation of excess molar volume (V_m^E) with mole fraction of CH. The sign of V_m^E depends upon the contraction and expansion of volume of the liquids due to mixing. The excess molar volume is the resultant contribution from several opposing effects, namely, chemical, physical, and structural [30]. The chemical or specific interactions result in volume contractions, leading to negative excess molar volume, and these include charge-transfer complexes, dipole-dipole, dipole-induced dipole interactions and formation of H-bonding between component molecules. The physical interactions or nonspecific interactions are weak and these include breaking of the structure of one or both of the components in a solution, that is, the loss of dipolar association between the molecules (dispersion forces), steric hindrance of the molecules and H-bond rupture, and stretching of self-associated molecules (like alcohols). The structural contributions are mostly negative and arise from several effects such
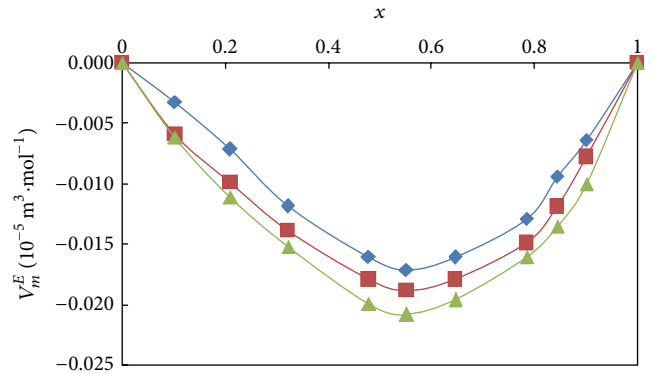

FIGURE 1: Variation of excess molar volume, V_m^E, with mole fraction, x, of CH in BB at (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.

as interstitial accommodation and geometrical fitting of one component into another due to the differences in the molar volume and free volume between components. In the present investigation, the sign of V_m^E is found to be negative over the entire composition range.

The nature of existing interaction in a binary liquid can also be analysed by knowing their individual chemical and physical properties. It is well known that BB and CH are polar and their dipole moment values follow: $\mu_{CH} = 3.25 \, D > \mu_{BB} = 2.06 \, D$. In the present study, possibility of Keesom dipole-dipole van der Waals forces which arise due to the dipole moment of the components gives strong interactions leading to negative values of V_m^E [31]. Moreover, the liquids BB and CH lack hydroxyl groups; hence the possibility for formation of intermolecular hydrogen bonds is trifling. Further, the molar volumes of CH and BB are 10.4459 and 19.0428 ($\times 10^{-5} \, m^3 \cdot mol^{-1}$), respectively, at 308.15 K. This implies that geometrical fitting of smaller molecules into the voids created by the bigger molecules is most favorable. The V_m^E values in binary mixture are thereby decreased with increasing temperature. Negative V_m^E values indicate strong interactions following the order 318.15 K > 313.15 K > 308.15 K. According to Rastogi et al. [32], such behaviour may arise due to the fact that, as temperature increases, thermal energy activates the molecule; this would increase the rate of association of unlike molecules. Similar type of trend was observed by Baragi et al. [33] in methyl cyclohexane with alkanes.

TABLE 2: Experimental values of densities (ρ), velocities (u), and viscosities (η) with calculated properties of molar volume, (V_m), acoustic impedance, (Z), isentropic compressibility, (k_s), free length, (L_f), internal pressure, (π_i), enthalpy, (H), and Gibbs free energy of activation of viscous flow, (G^{*E}), with mole fraction, x, of cyclohexanone (x) in benzyl benzoate at T = 308.15 K, 313.15 K, and 328.15 K.

x	$\rho/(\text{kg}\cdot\text{m}^{-3})$	$u/(\text{m}\cdot\text{s}^{-1})$	$\eta/(10^{-3}\ \text{N}\cdot\text{s}\cdot\text{m}^{-2})$	$V_m/(10^{-5}\ \text{m}^3\cdot\text{mol}^{-1})$	$Z^E/(10^6\ \text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1})$	$k_s/(10^{-10}\ \text{Pa}^{-1})$	$L_f/(10^{-10}\ \text{m})$	$\pi_i^E/(10^6\ \text{Pa})$	$H/(\text{J}\cdot\text{mol}^{-1})$	$G^{*E}/(\text{kJ}\cdot\text{mol}^{-1})$
308.15 K										
1.0000	939.60	1362.00	1.594	10.4459	1.2797	5.7372	0.5016	521.97	54525.20	−39.9867
0.9021	969.35	1385.94	1.878	11.2775	1.3435	5.3706	0.4853	505.62	57021.57	−39.3711
0.8443	984.74	1397.80	2.071	11.7709	1.3765	5.1973	0.4775	499.07	58745.42	−39.0100
0.7852	999.11	1409.21	2.289	12.2765	1.4080	5.0400	0.4702	493.94	60639.08	−38.6459
0.6470	1028.33	1433.09	2.879	13.4610	1.4737	4.7349	0.4557	486.22	65450.91	−37.8231
0.5522	1045.47	1447.20	3.347	14.2748	1.5130	4.5670	0.4476	483.19	68975.81	−37.2863
0.4789	1057.32	1456.83	3.746	14.9058	1.5403	4.4563	0.4421	481.64	71793.50	−36.8876
0.3232	1079.23	1473.61	4.696	16.2491	1.5904	4.2669	0.4326	479.88	77976.77	−36.0876
0.2112	1092.84	1482.60	5.466	17.2160	1.6203	4.1629	0.4273	479.51	82553.29	−35.5502
0.1035	1104.50	1488.79	6.276	18.1468	1.6444	4.0847	0.4233	479.63	87038.88	−35.0614
0.0000	1114.54	1493.30	7.118	19.0428	1.6643	4.0236	0.4201	479.96	91398.24	−34.6154
313.15 K										
1.0000	934.70	1348.89	1.472	10.5007	1.2608	5.8800	0.5078	502.22	52736.76	−40.1779
0.9021	964.24	1377.00	1.707	11.3373	1.3278	5.4694	0.4898	481.96	54641.93	−39.6016
0.8443	979.69	1390.87	1.870	11.8317	1.3626	5.2763	0.4811	473.72	56050.30	−39.2590
0.7852	994.09	1401.82	2.054	12.3385	1.3935	5.1190	0.4738	467.57	57692.00	−38.9103
0.6470	1023.27	1422.33	2.558	13.5275	1.4554	4.8306	0.4603	458.59	62036.14	−38.1127
0.5522	1040.41	1434.12	2.962	14.3443	1.4921	4.6733	0.4527	455.16	65289.63	−37.5869
0.4789	1052.26	1442.36	3.307	14.9774	1.5178	4.5680	0.4476	453.41	67909.90	−37.1940
0.3232	1074.26	1457.01	4.135	16.3243	1.5652	4.3849	0.4386	451.48	73702.04	−36.4016
0.2112	1087.89	1464.42	4.809	17.2944	1.5931	4.2863	0.4336	451.19	78030.81	−35.8666
0.1035	1099.62	1468.85	5.520	18.2273	1.6152	4.2150	0.4300	451.53	82303.37	−35.3788
0.0000	1109.70	1471.40	6.261	19.1259	1.6328	4.1623	0.4273	452.17	86483.25	−34.9328
318.15 K										
1.0000	933.20	1275.00	1.370	10.5176	1.1898	6.5918	0.5377	497.80	52356.76	−40.3579
0.9021	962.12	1309.97	1.567	11.3623	1.2604	6.0568	0.5154	472.78	53719.01	−39.8149
0.8443	977.21	1326.76	1.704	11.8617	1.2965	5.8133	0.5050	462.25	54831.30	−39.4904
0.7852	991.40	1342.23	1.860	12.3720	1.3307	5.5988	0.4956	453.82	56147.48	−39.1584
0.6470	1020.06	1372.34	2.290	13.5702	1.3999	5.2053	0.4778	440.75	59810.82	−38.3889
0.5522	1036.87	1388.73	2.639	14.3933	1.4399	5.0008	0.4683	435.57	62693.06	−37.8743
0.4789	1048.47	1399.38	2.940	15.0316	1.4672	4.8705	0.4622	432.95	65079.84	−37.4865
0.3232	1070.00	1417.30	3.668	16.3892	1.5165	4.6525	0.4517	430.03	70478.74	−36.6981
0.2112	1083.31	1427.23	4.265	17.3675	1.5461	4.5317	0.4458	429.22	74544.92	−36.1631
0.1035	1094.79	1435.30	4.895	18.3077	1.5714	4.4338	0.4410	428.90	78523.17	−35.6752
0.0000	1104.80	1440.00	5.550	19.2107	1.5909	4.3651	0.4376	429.05	82424.53	−35.2306

TABLE 3: Coefficients A_i of Redlich-Kister-type polynomial equation and the corresponding standard deviations, σ, of the system CH + BB at $T = 308.15$ K, 313.15 K, and 318.15 K.

Property	A_0	A_1	A_2	A_3	A_4	σ
At 308.15 K						
$V_m^E/(10^{-5}\,\mathrm{m^3 \cdot mol^{-1}})$	−0.1852	−1.2716	−0.6378	5.1475	1.5786	0.0442
$Z^E/(10^6\,\mathrm{kg \cdot m^{-2} \cdot s^{-1}})$	0.2425	−0.0386	0.0089	0.0046	0.1483	0.0001
$\Delta k_s/(10^{-10}\,\mathrm{Pa^{-1}})$	−1.5704	0.4932	−0.1605	0.0213	−0.1559	0.0003
$L_f^E/(10^{-10}\,\mathrm{m})$	−0.0727	0.0009	0.1215	0.2317	−0.3916	0.0060
$\pi_i^E/(10^6\,\mathrm{Pa})$	−81.96	72.84	222.75	−575.16	−400.46	6.1774
$H^E/(\mathrm{J \cdot mol^{-1}})$	−7945.70	3556.51	−1661.40	519.50	−462.21	0.4439
$\Delta G^{*E}/(\mathrm{kJ \cdot mol^{-1}})$	1.2005	0.0541	−0.1314	0.0720	−0.0263	0.0001
$\Delta \eta/(\mathrm{mN \cdot s \cdot m^{-2}})$	−2.912	−0.001	0.001	0.003	−0.002	0.0002
$\Delta u/(\mathrm{m \cdot s^{-1}})$	106.13	−6.91	0.50	3.72	11.64	0.03
At 313.15 K						
$V_m^E/(10^{-5}\,\mathrm{m^3 mol^{-1}})$	−0.0473	−0.0514	−0.1583	0.4146	0.1953	0.0046
$Z^E/(10^6\,\mathrm{kg \cdot m^{-2} \cdot s^{-1}})$	0.255	−0.0511	0.0606	−0.0826	−0.0177	0.0002
$\Delta k_s/(10^{-10}\,\mathrm{Pa^{-1}})$	−1.6905	0.6082	−0.5890	0.6564	0.0583	0.0012
$L_f^E/(10^{-10}\,\mathrm{m})$	−0.0738	0.0240	−0.0262	0.0258	0.0073	0.0001
$\pi_i^E/(10^6\,\mathrm{Pa})$	−93.42	55.97	−36.68	24.88	−2.77	0.0259
$H^E/(\mathrm{J \cdot mol^{-1}})$	−9845.27	4398.17	−3040.88	2073.36	435.13	3.1835
$\Delta G^{*E}/(\mathrm{kJ \cdot mol^{-1}})$	0.9980	0.1547	−0.1695	0.0906	−0.0263	0.0001
$\Delta \eta/(\mathrm{mN \cdot s \cdot m^{-2}})$	−2.644	−0.001[	0.001	0.001	−0.001	0.0003
$\Delta u/(\mathrm{m \cdot s^{-1}})$	119.47	−21.79	58.41	−74.26	−24.87	0.15
At 318.15 K						
$V_m^E/(10^{-5}\,\mathrm{m^3 mol^{-1}})$	−0.0656	0.0232	0.0195	0.0245	0.0006	0.0002
$Z^E/(10^6\,\mathrm{kg \cdot m^{-2} \cdot s^{-1}})$	0.2772	−0.0793	−0.0024	0.0463	0.0444	0.0003
$\Delta k_s/(10^{-10}\,\mathrm{Pa^{-1}})$	−2.2868	1.0124	−0.3349	0.1037	−0.3589	0.0006
$L_f^E/(10^{-10}\,\mathrm{m})$	−0.0952	0.0378	−0.0091	−0.0011	−0.0163	0.0001
$\pi_i^E/(10^6\,\mathrm{Pa})$	−119.32	72.47	−29.02	9.38	−10.37	0.0120
$H^E/(\mathrm{J \cdot mol^{-1}})$	−10268.50	8865.00	−13730.10	−17705.21	15648.81	2.5820
$\Delta G^{*E}/(\mathrm{kJ \cdot mol^{-1}})$	0.7885	0.2677	−0.1434	0.0399	0.0029	0.0001
$\Delta \eta/(\mathrm{mN \cdot s \cdot m^{-2}})$	−2.436	0.007	0.068	−0.001	0.003	0.0005
$\Delta u/(\mathrm{m \cdot s^{-1}})$	156.09	−49.22	2.17	4.21	37.00	0.06

The strong molecular interactions in the presented binary system are well reflected in the properties of partial molar volumes. The partial molar volumes $\overline{V}_{m,1}$ of component 1 (CH) and $\overline{V}_{m,2}$ of component 2 (BB) in the mixtures have been calculated by using the following equations:

$$\overline{V}_{m,1} = V_m^E + V_1^* + x_2 \left(\frac{\partial V_m^E}{\partial x} \right)_{T,P},$$

$$\overline{V}_{m,2} = V_m^E + V_2^* + x_1 \left(\frac{\partial V_m^E}{\partial x} \right)_{T,P}, \tag{6}$$

where V_1^* and V_2^* are the molar volumes of the pure components of CH and BB, respectively. By differentiating (4) and then rearranging one can get the following equations for $\overline{V}_{m,1}$ and $\overline{V}_{m,2}$.

The derivates in the above equations are obtained by differentiating Redlich-Kister equation (8) which leads to the following equations for $\overline{V}_{m,1}$ and $\overline{V}_{m,2}$:

$$\overline{V}_{m,1} = V_1^* + x_2^2 \sum_{i=0}^{j} A_i (x_2 - x_1)^i - 2x_1 x_2^2 \sum_{i=1}^{j} A_i (x_2 - x_1)^{i-1}, \tag{7}$$

$$\overline{V}_{m,2} = V_2^* + x_1^2 \sum_{i=0}^{j} A_i (x_2 - x_1)^i - 2x_2 x_1^2 \sum_{i=1}^{j} A_i (x_2 - x_1)^{i-1}. \tag{8}$$

The excess partial molar volumes are then given by

$$\overline{V}_{m,1}^E = \overline{V}_{m,1} - V_1^*,$$

$$\overline{V}_{m,2}^E = \overline{V}_{m,2} - V_2^*. \tag{9}$$

The values of $\overline{V}_{m,1}$ and $\overline{V}_{m,2}$ are tabulated in Table 4. This table reproduces the values of $\overline{V}_{m,1}$ and $\overline{V}_{m,2}$ for both the

components in the mixtures are smaller than their individual molar volumes in the pure state; this indicates that shrinkage of volume takes place on mixing CH with BB. Figures 2 and 3 represent the disparity of excess partial molar volumes of $\overline{V}^E_{m,1}$ and $\overline{V}^E_{m,2}$, respectively, in the binary mixture. Examination of these figures not only reveals the existence of strong forces between the unlike molecules but also supports the deductions drawn from excess molar volume.

The partial molar volumes and excess partial molar volumes of the components at infinite dilution, $\overline{V}^{\infty}_{m,1}$, $\overline{V}^{\infty}_{m,2}$, $\overline{V}^{E,\infty}_{m,1}$, and $\overline{V}^{E,\infty}_{m,2}$, respectively, were obtained by putting $x = 0$ in (7) and $x = 1$ in (8):

$$\overline{V}^{E,\infty}_{m,1} = A_0 + A_1 + A_2 + A_3 + \cdots = \overline{V}^{\infty}_{m,1} - V^*_1,$$
$$\overline{V}^{E,\infty}_{m,2} = A_0 - A_1 + A_2 - A_3 + \cdots = \overline{V}^{\infty}_{m,2} - V^*_2. \quad (10)$$

The pertinent values of $\overline{V}^{\infty}_{m,1}$, $\overline{V}^{\infty}_{m,2}$, $\overline{V}^{E,\infty}_{m,1}$, and $\overline{V}^{E,\infty}_{m,2}$ are reported in Table 5. This table shows that these values are negative, from which we conclude that strong interactions exist among the unlike molecules of the mixtures, which support the trends of V^E_m values observed in these mixtures.

Figures 4 and 5 represent the deviation in isentropic compressibility (Δk_s) and excess intermolecular free length (L^E_f), respectively. These values are found to be negative for the system under investigation at all temperatures. The nature of Δk_s, L^E_f plays vivacious role in assessing the compactness due to molecular rearrangement. The extent of molecular interactions in liquid mixtures may be due to charge transfer, dipole-induced-dipole, dipole-dipole interactions, and interstitial accommodation, leading to more compact structure making Δk_s and L^E_f negative. The values of the excess functions Δk, L^E_f depend upon several physical and/or chemical contributions [34–37]. The physical contribution consists of dispersion forces or weak dipole-dipole interaction that leads to positive values of Δk_s and L^E_f. Another factor, which involves a physical contribution, is the geometrical effect allowing the fitting of molecules of two different sizes into each other's structure resulting in negative Δk_s, L^E_f values. The strength of the interactions increases with temperatures as observed from more negative values of the excess parameters. They are negative throughout and become more negative at all concentrations as the temperature is increased. In heteromolecular interaction between the component molecules of the mixtures, Fort and Moore [38] found that the negative value of excess compressibility indicates greater interaction between the components of the mixtures due to the formation of hydrogen bond. Thus the negative Δk_s values for binary mixtures indicate strong interactions between BB and CH.

According to Singh et al. [39], negative values of excess intermolecular free length L^E_f indicate that sound waves cover longer distances due to decrease in intermolecular free length ascribing the dominant nature of hydrogen bond interaction between unlike molecules. Fort and Moore indicated that the positive values of excess free length should be attributed to the dispersive forces, and negative excess values should be due
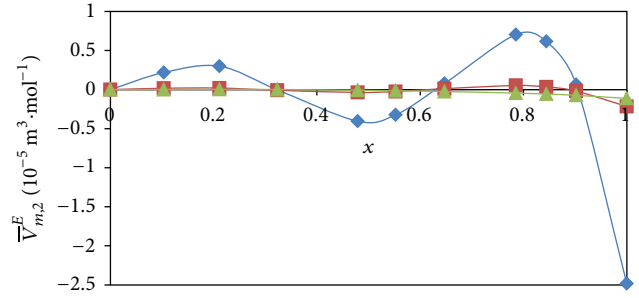

FIGURE 2: Variation of excess partial molar volume, $\overline{V}^E_{m,2}$, with mole fraction, x, of CH in BB (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.
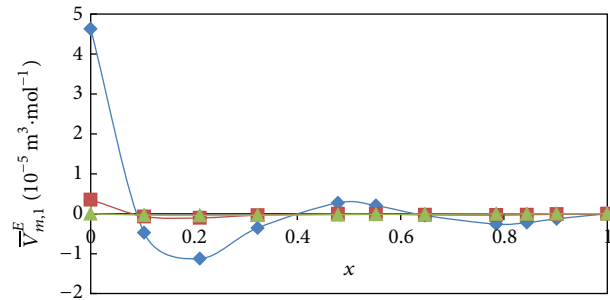

FIGURE 3: Variation of excess partial molar volume, $\overline{V}^E_{m,1}$, with mole fraction, x, of CH in BB (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.
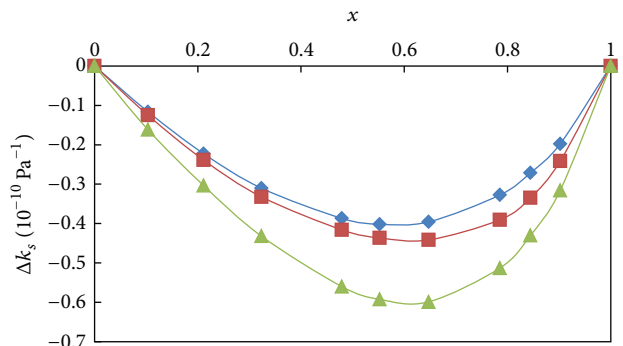

FIGURE 4: Variation of deviation in isentropic compressibility, Δk_s, with mole fraction, x, of CH in BB (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.
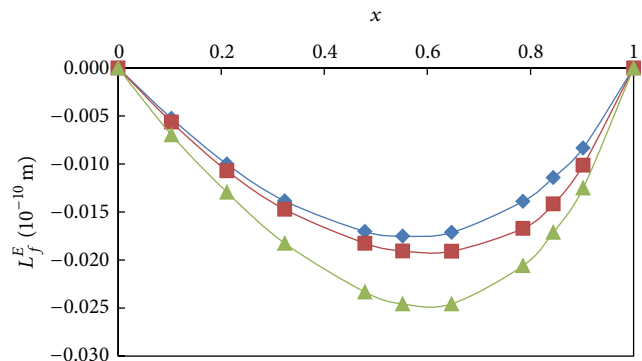

FIGURE 5: Variation of excess free length, L^E_f, with mole fraction, x, of CH in BB (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.

TABLE 4: Mole fraction (x) and values of partial molar volume $\overline{V}_{m,2}/(10^{-5}\ m^3 \cdot mol^{-1})$, $\overline{V}_{m,1}/(10^{-5}\ m^3 \cdot mol^{-1})$ of the binary system at 308.15, 313.15, and 318.15 K.

x	$T = 308.15$ K		$T = 313.15$ K		$T = 318.15$ K	
	$\overline{V}_{m,2}$	$\overline{V}_{m,1}$	$\overline{V}_{m,2}$	$\overline{V}_{m,1}$	$\overline{V}_{m,2}$	$\overline{V}_{m,1}$
1.0000	16.5603	10.4459	18.9107	10.5007	19.0979	10.5176
0.9021	19.1068	10.3190	19.1066	10.4880	19.1381	10.5161
0.8443	19.6593	10.2239	19.1608	10.4759	19.1540	10.5144
0.7852	19.7480	10.1852	19.1786	10.4667	19.1663	10.5123
0.6470	19.1208	10.4029	19.1386	10.4730	19.1864	10.5058
0.5522	18.7192	10.6493	19.1017	10.4931	19.1966	10.4996
0.4789	18.6383	10.7183	19.0898	10.5035	19.2030	10.4935
0.3232	19.0412	10.0898	19.1175	10.4635	19.2123	10.4792
0.2112	19.3430	9.3218	19.1459	10.3961	19.2141	10.4735
0.1035	19.2619	9.9694	19.1433	10.4335	19.2124	10.4826
0.0000	19.0428	15.0774	19.1259	10.8536	19.2107	10.5198

TABLE 5: Values of partial molar volume of the components at infinite dilution ($\overline{V}_{m,1}^{\infty}$, $\overline{V}_{m,2}^{\infty}$) and excess partial molar volume at infinite dilution ($\overline{V}_{m,1}^{E,\infty}$, $\overline{V}_{m,2}^{E,\infty}$) for the system CH + BB at 308.15, 313.15, and 318.15 K.

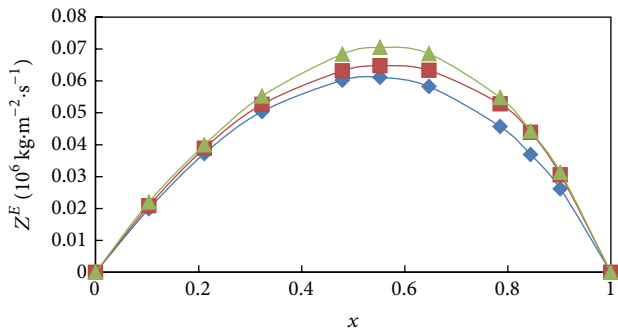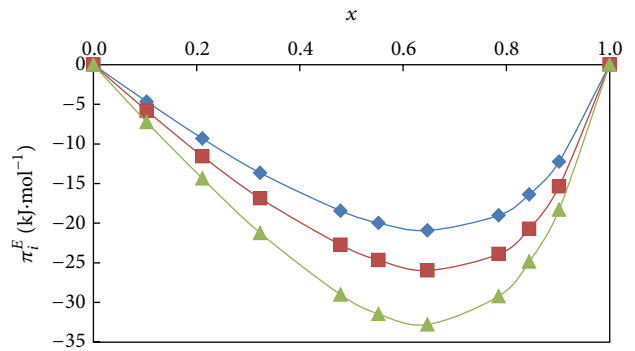| Temp. (K)	$\overline{V}_{m,1}^{E,\infty}$	$\overline{V}_{m,2}^{E,\infty}$	$\overline{V}_{m,1}^{\infty}$	$\overline{V}_{m,2}^{\infty}$
		$/(10^{-5}\ m^3 \cdot mol^{-1})$		
308.15	4.6315	−3.1203	15.0774	15.9225
313.15	0.3529	−0.3735	10.8536	18.7524
318.15	0.0022	−0.0933	10.5197	19.1173

FIGURE 7: Variation of excess internal pressure, π_i^E, with mole fraction, x, of CH in BB (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.
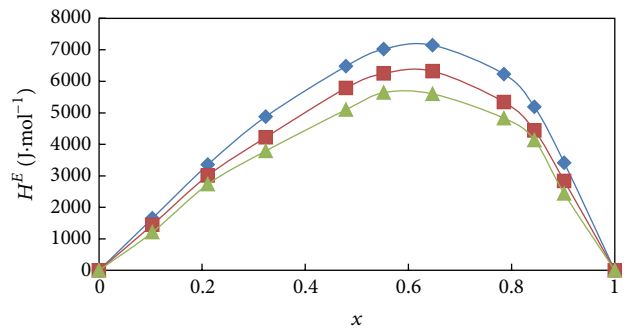

FIGURE 6: Variation of excess acoustic impedance, Z^E, with mole fraction, x, of CH in BB (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.

FIGURE 8: Variation of excess enthalpy, H^E, with mole fraction, x, of CH in BB (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.

to hydrogen bond formation or dipole-dipole interactions between solute and solvent. In the present study the negative contribution in all the systems prevails in the existence of greater interactions. Nain et al. [40] also concluded a similar observation. From Figure 6, the excess values of Z^E are found to be positive for the system at all temperatures under investigation. According to Kondaiah and Rao [41], the nature of the positive values of Z^E is attributed to specific interaction between the heteromolecules. This further supports our earlier finding.

The variations of excess internal pressure (π_i^E) and excess enthalpy (H^E) are presented in Figures 7 and 8 for the present

system at all the three temperatures, respectively. From these figures it is observed that π_i^E is negative and H^E is positive over the entire composition range of CH. In accordance with Kondaiah et al. [42], the negative values of π_i^E indicate the existence of strong specific interactions and according to Kumar and Rao [43], positive value of ΔH suggests strong bonding and exothermic reactions. Thus, it can be concluded that strong specific interactions are operative in the systems
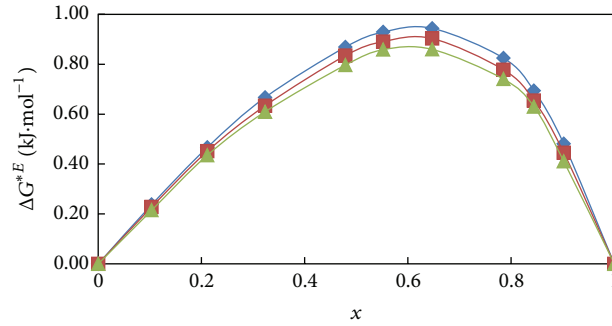
FIGURE 9: Variation of excess Gibbs free energy of activation of viscous flow, ΔG^{*E}, with mole fraction, x, of CH in BB (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.
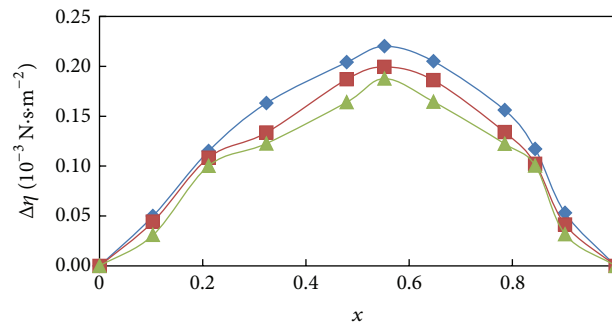

FIGURE 10: Variation of deviation in viscosity, $\Delta \eta$, with mole fraction, x, of CH in BB (◆) 308.15 K, (■) 313.15 K, and (▲) 318.15 K.

investigated. These conclusions support our earlier findings of strong interactions. The variation of excess Gibbs free energy of activation of viscous flow ΔG^{*E} with mole fraction of CH has been presented in Figure 9 at all temperatures. These values are positive over the entire range of composition of CH. According to Iloukhani et al. [44, 45], negative values of ΔG^{*E} indicate the presence of weak physical forces such as dispersive forces in the system. On the other hand positive values of it suggest strong specific interactions. This also supports the conclusions drawn from our earlier findings.

According to Fort and Moore [46], deviation in viscosity tends to become more positive as the strength of the interaction increases. The deviation in viscosity variation gives a qualitative estimation of the strength of the intermolecular interactions. The deviation in viscosities may be generally explained by considering the following factors. (i) The difference in size and shape of the component molecules and the loss of dipolar association in pure component may contribute to a decrease in viscosity and (ii) specific interactions between unlike components such as hydrogen bond formation or dipole-dipole interactions or charge-transfer complexes may cause increase in viscosity in mixtures compared to in pure components [47]. The former effect produces negative deviation in viscosity and latter effect produces positive deviation in viscosity. A perusal of Figure 10 shows that the values of $\Delta \eta$ are positive for the present system at three temperatures. The positive $\Delta \eta$ values decrease with increasing temperature. The positive $\Delta \eta$ values indicate specific interactions while the negative $\Delta \eta$ values indicate dispersion forces [48–50].

In the present investigation, the positive values of all binary systems may be attributed to the dipole-dipole interactions. The sign and magnitude of Δu play important roles in describing molecular rearrangements as a result of molecular interactions occurring among the component molecules in the mixtures. The excess ultrasonic speed with mole fraction of CH is shown in Figure 11. Here we observed that the Δu values are positive for all binary systems over the entire range of composition at all the studied temperatures. Positive deviations indicate the increasing strength of interaction between component molecules of binary liquid mixtures. According to Reddy et al. [51], strong interactions among the components of a mixture lead to the formation of molecular aggregates and more compact structures; then sound will travel faster through the mixture by means of longitudinal waves and hence the ultrasonic speed deviations with respect to a linear behaviour will be positive while if the structure-breaking factor in the mixture predominates resulting expansion, then the speed of sound through the mixture will be slower resulting in negative deviation in the speed of sound. According to Ali et al. [52], more positive values mean much more strong interactions between the molecules. Therefore, based on the above references for the present scenario, interactions are found to be in the order 318.15 K > 313.15 K > 308.15 K; this again emphasizes our view regarding the interaction between CH and BB.

In the present study, theoretical sound velocities have been evaluated by considering CH as one component and BB as the other component in the binary mixture at all
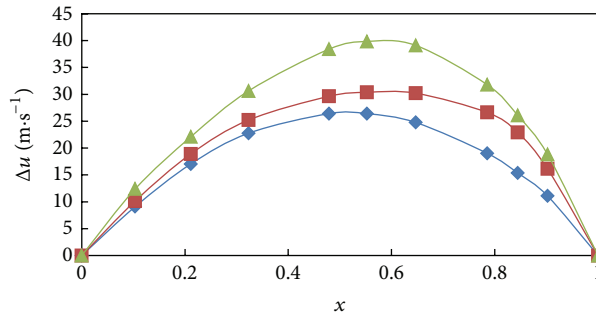
FIGURE 11: Variation of deviation in ultrasonic velocity, Δu, with mole fraction, x, of CH in BB ($\blacklozenge$) 308.15 K, ($\blacksquare$) 313.15 K, and ($\blacktriangle$) 318.15 K.

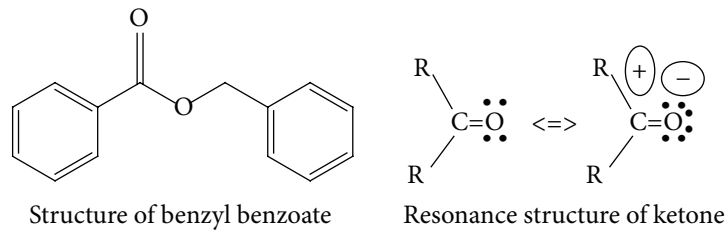Structure of benzyl benzoate   Resonance structure of ketone

FIGURE 12

the three temperatures. Such an evaluation of theoretical sound velocity is useful to verify the applicability of various assumes of the theories of liquid mixtures and to arrive at some useful implications regarding the (strength of) molecular interactions between component liquids in some cases. The theoretical values of ultrasonic velocity obtained using various theories along with the experimental velocity and percentage deviation are summarized in Table 6. Nomoto [53] established the following relation for sound velocity based on the supposition of the linearity of the molecular sound velocity and the additives of molar volume:

$$U_N = \left\{ \frac{\left(\sum x_i R_i \right)}{\left(\sum x_i V_i \right)} \right\}^3. \tag{11}$$

Van Dael [54] obtained the ideal mixture relation

$$\sum \left(\frac{x_i M_i}{u_i^2} \right) = \left\{ \frac{1}{\sum x_i M_i} \right\} \left\{ \frac{1}{U_V} \right\}^2. \tag{12}$$

The impedance dependence relation used by Baluja and Parsania [55] is given below:

$$U_{\text{imp}} = \frac{\sum x_i Z_i}{\sum x_i \rho_i}, \tag{13}$$

where x_i is mole fractions, M_i is molecular weight, R is the molar sound speed, and Z_i is acoustic impedance of CH and BB, respectively, at three temperatures.

Percentage deviation in ultrasonic speed is given by

$$\%\Delta u = 100 * \left[1 - \frac{U_{\text{cal}}}{u_{\text{expt}}} \right]. \tag{14}$$

On observing Table 6, among all the empirical theories, Nomoto's relation gives the best estimate of experimental values of sound velocity in the system followed by impedance's relation. In the present binary systems, the difference between experimental and theoretical velocities is greater where the mole fraction of CH varies in the region 0.4 to 0.7. Hence it can be qualitatively inferred that the strength of interaction in the binary mixtures is more in this range of composition of binary mixtures.

The dynamic viscosities of the liquid mixtures have been calculated using the several empirical relations.

Grunberg and Nissan [56] proposed the following equation for the measurement of viscosity of liquid mixtures:

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 G_{12}, \tag{15}$$

where G_{12} is an interaction parameter and it is the function of components 1, 2 and temperature.

Hind et al. [57] suggested an equation for the viscosity of binary liquid mixtures as

$$\eta = x_1^2 \eta_1 + x_2^2 \eta_2 + 2 x_1 x_2 H_{12}, \tag{16}$$

where H_{12} is an interaction parameter and is attributed to unlike pair interaction.

Katti and Chaudhri [58] proposed the following equation:

$$\ln (\eta V) = x_1 \ln (\eta_1 V_1) + x_2 \ln (\eta_2 V_2) + \frac{x_1 x_2 W_{\text{vis}}}{\text{RT}}, \tag{17}$$

where $(W_{\text{vis}}/\text{RT})$ is an interaction parameter.

The theoretical viscosity values using (15) to (17) along with the percentage error are compiled in Table 7. The evaluated values of parameters G_{12}, H_{12}, and W_{vis}/RT and standard deviations σ (10^{-3} kg^{-1}·s^{-1}) are presented in Table 8.

Table 6: Experimental and theoretical velocities from various empirical relations and percentage deviation in velocities with molefraction, x, of CH with BB at T = 308.15, 313.15, and 318.15 K.

x	u_{expt}	U_N	U_V	U_I	$\%U_N$	$\%U_V$	$\%U_I$
308.15 K							
1.0000	1362.00	1362.00	1362.00	1361.96	0.00	0.00	0.00
0.9021	1385.94	1383.13	1331.26	1376.93	−0.20	−3.95	−0.65
0.8443	1397.80	1394.27	1318.02	1385.52	−0.25	−5.71	−0.88
0.7852	1409.21	1404.79	1307.85	1394.13	−0.31	−7.19	−1.07
0.6470	1433.09	1426.46	1296.25	1413.55	−0.46	−9.55	−1.36
0.5522	1447.20	1439.35	1297.68	1426.33	−0.54	−10.33	−1.44
0.4789	1456.83	1448.39	1304.03	1435.94	−0.58	−10.49	−1.43
0.3232	1473.61	1465.38	1333.84	1455.57	−0.56	−9.49	−1.22
0.2112	1482.60	1476.03	1371.26	1469.08	−0.44	−7.51	−0.91
0.1035	1488.79	1485.25	1423.35	1481.61	−0.24	−4.40	−0.48
0.0000	1493.30	1493.30	1493.30	1493.26	0.00	0.00	0.00
313.15 K							
1.0000	1348.89	1348.89	1348.89	1348.88	0.00	0.00	0.00
0.9021	1377.00	1368.65	1318.18	1362.86	−0.61	−4.27	−1.03
0.8443	1390.87	1379.07	1304.90	1370.88	−0.85	−6.18	−1.44
0.7852	1401.82	1388.91	1294.64	1378.91	−0.92	−7.65	−1.63
0.6470	1422.33	1409.16	1282.67	1397.04	−0.93	−9.82	−1.78
0.5522	1434.12	1421.20	1283.67	1408.97	−0.90	−10.49	−1.75
0.4789	1442.36	1429.65	1289.58	1417.93	−0.88	−10.59	−1.69
0.3232	1457.01	1445.52	1318.07	1436.24	−0.79	−9.54	−1.43
0.2112	1464.42	1455.48	1354.08	1448.84	−0.61	−7.53	−1.06
0.1035	1468.85	1464.09	1404.28	1460.53	−0.32	−4.40	−0.57
0.0000	1471.40	1471.60	1471.61	1471.38	0.00	0.00	0.00
318.15 K							
1.0000	1275.00	1275.00	1275.00	1275.22	0.00	0.00	0.00
0.9021	1309.97	1301.37	1247.66	1293.99	−0.66	−4.76	−1.22
0.8443	1326.76	1315.31	1236.19	1304.75	−0.86	−6.83	−1.66
0.7852	1342.23	1328.49	1227.68	1315.54	−1.02	−8.53	−1.99
0.6470	1372.34	1355.68	1219.56	1339.89	−1.21	−11.13	−2.36
0.5522	1388.73	1371.88	1223.20	1355.93	−1.21	−11.92	−2.36
0.4789	1399.38	1383.27	1231.24	1367.99	−1.15	−12.02	−2.24
0.3232	1417.30	1404.69	1265.00	1392.63	−0.89	−10.75	−1.74
0.2112	1427.23	1418.15	1306.03	1409.60	−0.64	−8.49	−1.23
0.1035	1435.30	1429.81	1362.97	1425.35	−0.38	−5.04	−0.69
0.0000	1440.00	1440.00	1440.00	1439.98	0.00	0.00	0.00

The estimated values of σ are indicating that the viscosities are well correlated by all the three viscosity models. Prolongo et al. [59] reported positive values of interaction parameters corresponding to systems with negative excess molar volumes. This is consistent with our results. Among all of these models, the viscosity representations obtained from the Hind model are in best agreement with the experimental viscosity data.

4. Conclusions

(i) Ultrasonic velocities, densities, and viscosities for binary liquids of CH with BB are determined exper-imentally at T = 308.15 K, 313.15 K, and 318.15 K over the entire composition range.

(ii) The values of V_m^E, Δk_s, L_f^E, Z^E, π_i^E, H^E, ΔG^{*E}, and Δu are calculated from experimental results at all three temperatures. The excess/deviation properties are fitted to Redlich-Kister-type polynomial and corresponding standard deviations are evaluated. The observed negative values of V_m^E, Δk_s, L_f^E, and π_i^E and positive values of Z^E, H^E, ΔG^{*E}, $\Delta\eta$, and Δu for all the liquid mixtures studied clearly indicate dipole-dipole-type interactions and the geometrical fitting

TABLE 7: Experimental and theoretical values of viscosity from various empirical relations and percentage deviation in viscosity in CH + BB at T = 308.15, 313.15, and 318.15 K.

T (K)	Exptl. viscosity	Gruenberg	Hind	Katti	%G	%H	%K
	1.594	1.594	1.594	1.595	0.000	0.000	0.000
	1.878	1.891	1.878	1.891	0.698	0.005	0.692
	2.071	2.086	2.071	2.086	0.685	−0.014	0.676
	2.289	2.302	2.289	2.302	0.537	−0.009	0.524
	2.879	2.877	2.879	2.877	−0.052	0.007	−0.069
308.15 K	3.347	3.333	3.347	3.333	−0.445	−0.006	−0.448
	3.746	3.721	3.746	3.721	−0.649	0.003	−0.667
	4.696	4.658	4.695	4.658	−0.794	−0.002	−0.809
	5.460	5.430	5.466	5.431	−0.662	−0.009	−0.658
	6.276	6.253	6.276	6.253	−0.362	0.006	−0.366
	7.118	7.118	7.118	7.118	0.000	0.000	0.000
	1.472	1.472	1.472	1.472	0.000	0.000	0.000
	1.707	1.724	1.707	1.724	0.966	−0.023	0.937
	1.870	1.890	1.870	1.890	1.075	0.021	1.070
	2.054	2.073	2.055	2.073	0.886	0.005	0.876
	2.558	2.560	2.559	2.560	0.070	0.004	0.039
313.15 K	2.962	2.947	2.963	2.947	−0.513	0.007	−0.540
	3.307	3.279	3.308	3.278	−0.871	0.006	−0.907
	4.135	4.084	4.135	4.084	−1.214	0.010	−1.233
	4.809	4.757	4.809	4.756	−1.094	−0.006	−1.123
	5.520	5.484	5.520	5.484	−0.665	−0.005	−0.670
	6.261	6.261	6.261	6.262	0.000	0.000	0.000
	1.370	1.370	1.370	1.370	0.000	0.000	0.000
	1.567	1.585	1.566	1.586	1.122	−0.128	1.148
	1.704	1.726	1.703	1.728	1.344	−0.059	1.408
	1.860	1.883	1.860	1.884	1.237	0.000	1.290
	2.290	2.298	2.294	2.301	0.380	0.175	0.480
318.15 K	2.639	2.629	2.644	2.632	−0.345	0.189	−0.265
	2.940	2.914	2.945	2.917	−0.867	0.170	−0.782
	3.668	3.612	3.671	3.615	−1.548	0.055	−1.472
	4.265	4.202	4.265	4.205	−1.496	−0.023	−1.430
	4.895	4.848	4.893	4.850	−0.974	−0.061	−0.940
	5.550	5.550	5.550	5.550	0.000	0.000	0.000

TABLE 8: Various interaction parameters calculated from various empirical relations and the corresponding standard deviations ($\sigma/10^{-3}$ N·s·m^{-2}) of the present system CH + BB at T = 308.15, 313.15, and 318.15 K.

Temp. (K)	G_{12}	σ	H_{12}	σ	W_{vis}/RT	σ
308.15	0.272	0.020	0.003	0.003	0.004	0.020
313.15	0.186	0.028	0.003	0.003	0.003	0.028
318.15	0.103	0.032	0.002	0.030	0.003	0.031

of molecules leading to strong molecular interactions between CH and BB. These strong interactions increase with temperature.

(iii) The values of $\overline{V}_{m,1}$, $\overline{V}_{m,2}$ and $\overline{V}^E_{m,1}$, $\overline{V}^E_{m,2}$ are calculated from experimental data. The observed lower partial molar volumes in the liquid mixture when compared to the respective molar volumes of pure components indicate that strong interactions present in the system support the trends of V^E_m values very well.

(iv) The ultrasonic velocities/viscosities computed from three different velocity/viscosity theories were correlated with the experimentally measured ultrasonic velocities and their percentage deviations were evaluated. Among all the empirical theories Nomoto's velocity/Hind viscosity relation gives the best estimate of experimental values of sound velocity/viscosity in the system at all temperatures.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.
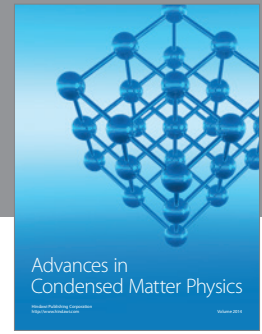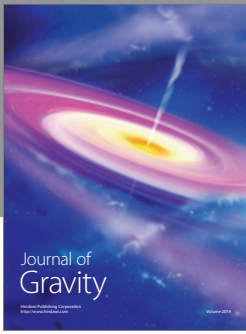
Acknowledgments

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