Studies of molecular interactions in binary mixtures using a temperature dependent relation of adiabatic compressibility

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Abstract

Making use of the definition of adiabatic compressibility, a temperature dependent relation has been deduced to estimate the adiabatic compressibility of binary mixtures at different temperatures from the knowledge of temperature coefficient of density and sound velocity of pure components. The validity of the new relation is examined by comparing the estimated values with those of experimental values for interacting as well as non-interacting binary mixtures at different temperatures. The results obtained were then explained on the basis of molecular interactions present in them.

Keywords: adiabatic compressibility, binary mixtures, molecular interactions, temperature.

Introduction

Liquid state is considered to be the most complicated among the three states of matter. Ultrasonic technique, due to its simplicity and accuracy, is widely used to study the liquid state, especially the molecular interactions in liquid mixtures [1-5]. A systematic study of liquid state using a few opto-acoustic parameters has been recently carried out in the laboratory [6-10]. As a continuation of the work, in the present paper, the deduction of a temperature dependent relation of adiabatic compressibility β_s has been reported from the knowledge of sound velocity and density of pure components of binary mixtures. This relation is then used to calculate β_s of six binary mixtures-interacting as well as non-interacting systems at different temperatures. The binary mixtures chosen for the present study are benzene + toluene, kerosene + diesel and kerosene + petrol, all falls under non-interacting systems and cyclohexane + benzene, ethyl acetate + cyclohexane and

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ethyl acetate + benzene, all come under interacting systems. The calculated values (estimated values) are then compared with the experimental values for these systems. .

Materials and Methods

Densities and sound velocities were determined experimentally at different temperatures and concentrations for the interacting as well as non-interacting systems under study. Chemicals of AR/BDH grade and distilled water were used for the experiment. The purity of all the liquids were tested (except petrol, diesel and kerosene, as there are no reliable standard values available for comparison) by comparing their densities with those in literature and found to be in good agreement [11,12]. The densities of these binary mixtures were measured using a 12 cm^3 double-stem pyknometer and the masses were recorded on an electronic balance having an accuracy of $\pm$ 0.1 mg. The ultrasonic velocities were determined using a single crystal ultrasonic interferometer supplied by Mittal Enterprises having a frequency of 2 MHz. The experiments were carried out at different temperatures using a thermostatically controlled water circulating arrangement having an accuracy of $\pm$ 0.1 K.

Theory

The adiabatic compressibility of a liquid is defined as

$$\beta_s = \frac{1}{U^2 \rho} \qquad (1)$$

where U is the sound velocity and ρ is the density of the liquid.

Both U and ρ are temperature sensitive parameters. Hence as temperature of the liquid changes, adiabatic compressibility also changes. So differentiating Eqn (1) with respect to temperature at constant pressure and dividing throughout by β_s, we get

$$\frac{1}{\beta_s} \left(\frac{\partial \beta_s}{\partial T}\right)_P = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P - \frac{2}{U}\left(\frac{\partial U}{\partial T}\right)_P$$

$$= \alpha + 2\beta \qquad (2)$$

where $\qquad \alpha = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P \quad$ and

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$$\beta = -\frac{1}{U}\left(\frac{\partial U}{\partial T}\right)_P$$

are the temperature coefficients of density and sound velocity of pure components respectively at the lowest temperature and at a particular concentration.

Re-arranging Eqn (2) and integrating we get,

$$\ln \beta_s = (\alpha + 2\beta)\,T + C \qquad (3)$$

where C is a constant of integration.

If β_s' is the adiabatic compressibility at a lower temperature T ′, then from eqn (3)

$$\ln \beta_s' = (\alpha + 2\beta)\,T' + C \qquad (4)$$

From Eqns (3) and (4), we get

$$\beta_s = \beta_s'\exp(\alpha + 2\beta)\,\Delta T \qquad (5)$$

where $\Delta T = T - T'$ is the difference in temperature.

This is the temperature dependent relation of adiabatic compressibility of a liquid.

Thus if the adiabatic compressibility β_s' of a liquid mixture at a temperature T ′ is known, the adiabatic compressibility at a higher temperature T can be estimated using Eq (5), knowing the values of α and β. In the case of a binary mixture, α and β were taken as the average of the component values $\alpha = (\alpha_1 + \alpha_2)/2$ and $\beta = (\beta_1 + \beta_2)/2$ where α_i and β_i are the temperature coefficients of density and sound velocity of the ith component respectively.

Results and Discussion

The experimental and calculated values of β_s for non-interacting as well as interacting binary mixtures were calculated using Eqs. (1) and (5) and are tabulated in Tables 1 to 4.

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Table 1. Variation of U, ρ, β_s^{expt} and β_s^{cal} with mole fraction (x_1) for the non-interacting binary mixture benzene (x_1) + toluene ($1-x_1$) at different temperatures

x_1	U (ms⁻¹)			ρ (kgm⁻³)			$\beta_s^{expt} \times 10^{10}$ (m² N⁻¹)			$\beta_s^{cal} \times 10^{10}$ (m²N⁻¹)	
	303K	313K	323K	303K	313K	323K	303K	313K	323K	313K	323K
0.0	1289	1248	1207	857.8	848.6	840.0	7.016	7.566	8.172	7.561	8.149
0.2	1285	1245	1205	858.8	850.0	840.4	7.051	7.590	8.195	7.599	8.190
0.4	1282	1243	1202	859.5	850.4	841.6	7.079	7.611	8.224	7.629	8.222
0.6	1280	1240	1199	860.6	852.0	842.9	7.092	7.633	8.253	7.643	8.237
0.8	1278	1237	1195	861.9	853.0	845.0	7.104	7.661	8.287	7.656	8.251
1.0	1275	1234	1193	865.1	855.6	846.2	7.111	7.675	8.303	7.664	8.259

Table 2. Variation of U, ρ, β_s^{expt} and β_s^{cal} with volume fraction (Φ_1) for the non-interacting binary mixture kerosene (Φ_1) + diesel ($1-\Phi_1$) at different temperatures

Φ_1	U (ms⁻¹)					ρ (kgm⁻³)				
	298K	303K	308K	313K	318K	298K	303K	308K	313K	318K
0.0	1348.7	1331.4	1315.4	1299.2	1281.8	829.4	825.5	822.8	818.4	815.4
0.2	1339.6	1324	1305.1	1288.8	1272.1	823.5	820.0	816.1	812.6	809.4
0.4	1330.8	1313.1	1297.3	1280.7	1264.2	817.5	813.4	809.9	806.2	803.1
0.6	1320.8	1303.5	1286.9	1270.6	1254.7	810.9	807.7	803.1	799.0	796.0
0.8	1311.5	1295.2	1276.8	1260.1	1241.7	803.7	800.1	796.9	793.0	789.4
1.0	1298.3	1280.8	1264.1	1245.9	1227.0	796.0	792.6	789.2	784.7	781.8

Φ_1	$\beta_s^{expt} \times 10^{10}$ (m² N⁻¹)					$\beta_s^{cal} \times 10^{10}$ (m² N⁻¹)			
	298K	303K	308K	313K	318K	303K	308K	313K	318K
0.0	6.628	6.834	7.024	7.239	7.465	6.836	7.050	7.271	7.498
0.2	6.767	6.957	7.194	7.409	7.635	6.979	7.198	7.423	7.656
0.4	6.907	7.130	7.337	7.563	7.791	7.123	7.347	7.577	7.814
0.6	7.069	7.287	7.519	7.752	7.981	7.290	7.519	7.754	7.997
0.8	7.233	7.450	7.697	7.942	8.216	7.460	7.693	7.934	8.183
1.0	7.453	7.691	7.929	8.210	8.496	7.687	7.927	8.176	8.432

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Table 3: Variation of U, ρ, β_s^{expt} and β_s^{cal} with volume fraction (Φ_1) for the non-interacting binary mixture petrol (Φ_1) + kerosene (1-Φ_1) at different temperatures

Φ_1	U (ms^{-1})					ρ (kgm^{-3})				
	298K	303K	308K	313K	318K	298K	303K	308K	313K	318K
0.0	1298.3	1280.8	1264.1	1245.9	1227.0	796.02	792.60	789.23	784.72	781.84
0.2	1272.7	1254.4	1236.9	1218.0	1199.8	789.36	785.49	780.37	775.95	772.80
0.4	1246.7	1228.4	1210.9	1192.0	1173.8	768.27	764.31	760.63	756.16	753.39
0.6	1207.8	1189.7	1170.6	1149.8	1131.2	755.21	751.64	747.70	743.25	739.46
0.8	1176.3	1157.5	1137.7	1119.2	1099.7	741.22	736.97	733.32	727.92	724.63
1.0	1144.6	1127.2	1107.6	1085.0	1066.9	723.17	719.87	714.54	710.31	706.30

Φ_1	$\beta_s^{expt} \times 10^{10}$ (m^2 N^{-1})					$\beta_s^{cal} \times 10^{10}$ (m^2 N^{-1})			
	298K	303K	308K	313K	318K	303K	308K	313K	318K
0.0	7.453	7.691	7.929	8.210	8.496	7.704	7.963	8.231	8.508
0.2	7.821	8.091	8.376	8.687	8.989	8.084	8.356	8.638	8.929
0.4	8.375	8.671	8.966	9.308	9.634	8.657	8.948	9.250	9.561
0.6	9.077	9.400	9.760	10.177	10.568	9.383	9.698	10.025	10.362
0.8	9.750	10.128	10.535	10.967	11.411	10.078	10.418	10.768	11.131
1.0	10.555	10.933	11.408	11.959	12.438	10.910	11.278	11.657	12.050

Table 4. Variation of U, ρ, β_s^{expt} and β_s^{cal} with mole fraction (x_1) for the interacting binary mixtures at different temperatures

x_1	U (ms^{-1})			ρ (kgm^{-3})			$\beta_s^{expt} \times 10^{10}$ (m^2 N^{-1})			$\beta_s^{cal} \times 10^{10}$ (m^2 N^{-1})	
	298K	303K	323K	298K	303K	323K	298K	303K	323K	303K	323K
cyclohexane (x_1) + benzene (1- x_1)											
0.0	1300	1276	1180	874.0	868.5	846.7	6.770	7.072	8.482	7.096	8.567
0.2	1278	1253	1155	847.2	842.0	821.4	7.227	7.565	9.126	7.575	9.145
0.4	1262	1238	1142	823.7	818.6	798.2	7.623	7.971	9.606	7.990	9.646
0.6	1252	1228	1133	804.4	799.3	779.2	7.931	8.296	9.997	8.313	10.036

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	303K	313K	323K	303K	313K	323K	303K	313K	323K	313K	323K
0.8	1251	1225	1127	788.0	783.1	763.5	8.109	8.510	10.312	8.500	10.261
1.0	1256	1228	1130	774.0	769.1	749.6	8.190	8.622	10.448	8.585	10.364
	303K	**313K**	**323K**	**303K**	**313K**	**323K**	**303K**	**313K**	**323K**	**313K**	**323K**
ethyl acetate (x_1) + cyclohexane (1- x_1)										
0.0	1228	1177	1130	769.1	759.4	749.6	8.622	9.506	10.448	9.465	10.391
0.2	1184	1139	1094	786.5	775.1	763.6	9.070	9.945	10.942	9.957	10.930
0.4	1155	1105	1056	805.4	795.1	784.4	9.307	10.300	11.432	10.217	11.216
0.6	1134	1086	1038	828.2	819.7	807.7	9.389	10.344	11.491	10.307	11.315
0.8	1123	1078	1032	856.3	844.9	832.9	9.260	10.185	11.273	10.165	11.159
1.0	1118	1075	1025	888.5	876.1	863.7	9.004	9.877	11.020	9.884	10.851
	303K	**313K**	**323K**	**303K**	**313K**	**323K**	**303K**	**313K**	**323K**	**313K**	**323K**
ethyl acetate (x_1) + benzene (1- x_1)										
0.0	1276	1228	1180	868.5	857.6	846.7	7.072	7.732	8.482	7.733	8.456
0.2	1242	1194	1147	872.1	861.2	850.0	7.434	8.145	8.942	8.129	8.888
0.4	1210	1162	1117	876.0	864.9	853.4	7.797	8.563	9.392	8.526	9.322
0.6	1180	1132	1086	880.0	868.8	857.1	8.161	8.982	9.893	8.924	9.758
0.8	1150	1104	1056	884.1	873.3	861.1	8.553	9.395	10.414	9.352	10.226
1.0	1118	1075	1025	888.5	876.1	863.7	9.005	9.877	11.020	9.847	10.767

The variations of β_s with concentration for the non-interacting systems at different temperatures were plotted in Fig. 1 (a to c).

Benzene, toluene, petrol, diesel and kerosene are all hydrocarbons. It can be seen that a change in temperature of hydrocarbons may change its physical properties like density, sound velocity, refractive index etc.

Fig. 1(a) shows the variation of β_s with mole fraction for the binary mixture benzene + toluene at temperatures 303, 313 and 323K respectively.

The lowest temperature is taken as the standard and the values of β_s for higher temperatures have been estimated. The present system has been chosen for the study because it has been accepted as an ideal mixture [13].
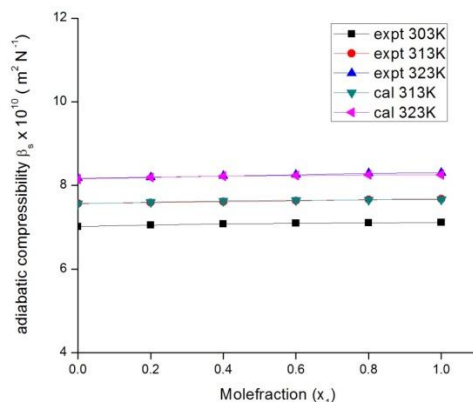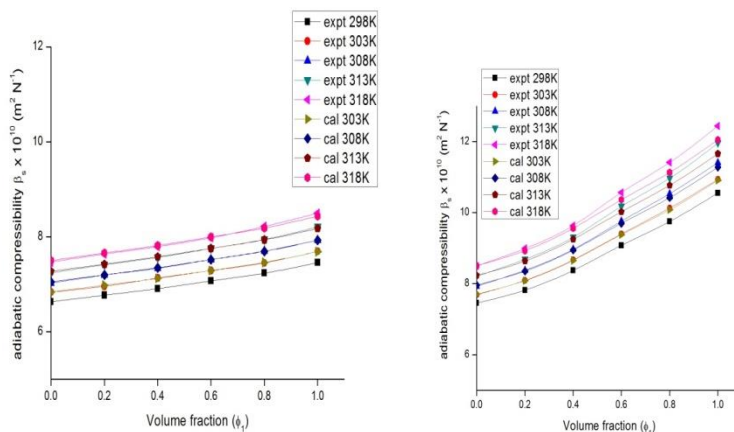
Fig 1(a) benzene + toluene

Fig.1 (b) kerosene + diesel

Fig.1 (c) kerosene + petrol

Fig.1 Variation of β_s with concentration for the non-interacting systems at different temperatures

From the figure it can be seen that the estimated values (calculated values) agree well with the experimental ones within experimental error for the higher temperatures 313K and 323K, over the whole composition range. The nature of variation of both β_s^{expt} and β_s^{cal} is nearly a straight line, almost parallel to the X-axis for both the temperatures. Further, with change in composition, there is no appreciable change in the estimated values of β_s from the experimental values. ie; both experimental and calculated values remain a constant for the whole composition range

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Also, in benzene + toluene system, there is no appreciable shift in the values with rise in temperature or increase in concentration. This clearly indicates that the system acts as an ideal mixture having no molecular interaction, which is in agreement with the accepted fact.

Fig 1(b) and 1(c) shows the variation of β_s^{expt} and β_s^{cal} with volume fraction for the binary mixtures of kerosene + diesel and kerosene + petrol at 298K, 303K, 308K, 313K and 323K respectively. Here also we can see that there is agreement between the experimental and calculated values of β_s within experimental error throughout the concentration range. This is clear from Tables 2 and 3. For both the binary systems, at low temperatures, there is no appreciable deviation between β_s^{expt} and β_s^{cal} values with increase in concentration. But for kerosene + diesel mixture, at temperature 318K, a slight deviation occurs at higher mole fraction range. This may be due to the volatile nature of the components of the mixture. Both kerosene and diesel evaporate quickly as temperature increases. For kerosene + petrol mixture, though there is agreement between experimental and calculated values of β_s at lower temperatures, slight deviation occurs at higher temperatures. This is because petrol being more volatile evaporates quickly than kerosene and diesel at high temperatures in spite of the precautions taken to prevent evaporation during experiment. The absence of deviation between experimental and calculated values of β_s at lower temperatures for kerosene + diesel and kerosene + petrol mixtures shows that these mixtures act as ideal mixtures having no molecular interaction. Thus it can be seen that for non-interacting systems, the calculated values of β_s using the temperature dependent relation agree well with the experimental values.

The variations of β_s with concentration for the interacting systems under study at different temperatures were plotted in Fig. 2 (a to c).
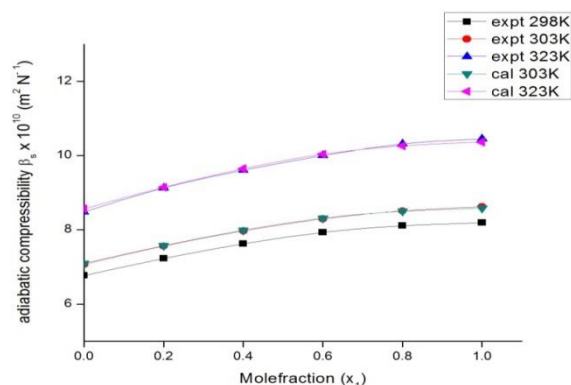
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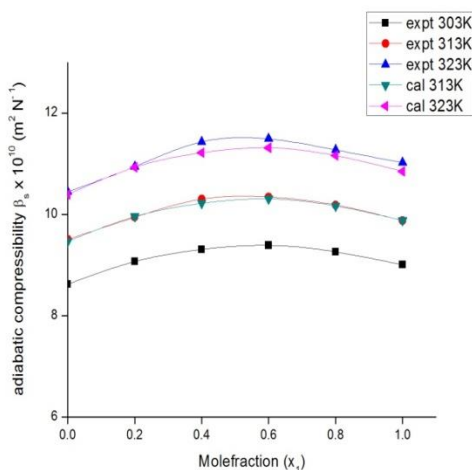
Fig 2(a) cyclohexane + benzene

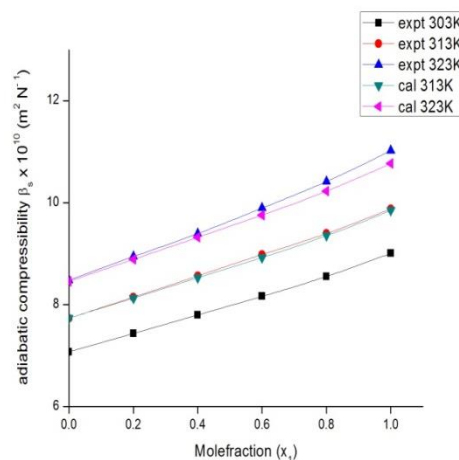**Fig.2 (b) ethyl acetate + cyclohexane**

Fig.2 (c) ethyl acetate + benzene

Fig.2 Variation of β_s with concentration for the interacting systems at different temperatures

Fig. 2(a) shows the variation of β_s^{expt} and β_s^{cal} with mole fraction for the cyclohexane + benzene mixture. From the figure it is quite clear that at lower temperature 303K, the experimental and calculated values of β_s agree well upto $x_1 = 0.9$ mole fraction and thereafter shows very slight deviation. But at higher temperature 323K, the deviation starts from lower mole fraction itself. Oswal and Palsanawala [14] who studied this system using ideal mixing relation found that the excess adiabatic compressibility (β_s^E) which is the difference between the experimental value and ideal value shows positive deviation. They explained this positive deviation as the result of disruption of locally ordered state of benzene by cyclohexane. Now, on

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analyzing this system based on the temperature dependent relation, it has been found that β_s^E which is the difference between the experimental and calculated values is negative upto $x_1 = 0.7$ mole fraction at 323K and positive beyond this concentration. Similar to experimental curve, the calculated curve of β_s also bends downwards. It can be seen that the slope of the curve decreases with increase in concentration.

It has been reported by several workers [15,16] that the positive values of β_s^E indicate weak interaction between unlike molecules whereas the negative values indicate strong interaction between them. The change of sign of β_s^E from negative to positive with increase in concentration of a component of a binary mixture can be taken as an indication of weak interaction in the mixture which is attributed to the dispersion forces existing between the components of the mixture. Thus for cyclohexane + benzene binary mixture, the downward bending of the curve, decrease in slope and the change of sign of β_s^E with increase in concentration can be attributed due to the presence of weak interaction in the system. The reason is that as the concentration of cyclohexane increases, the dispersion forces become dominant which are responsible for the disruption of the locally ordered state of liquid benzene by cyclohexane.

Fig. 2(b) shows the variation of β_s with mole fraction for the binary mixture ethyl acetate + cyclohexane. For ethyl acetate + cyclohexane mixture, the β_s value increases with increase in concentration upto a particular mole fraction and thereafter decreases. The maximum value of β_s is at around the mole fraction say $x_1 = 0.5$ for all the temperatures. This is true for experimental as well as calculated values of β_s. Further, the β_s values increase with increase in temperature. The deviation between experimental and calculated values of β_s is greater at higher temperature than at lower temperature.

According to Oswal and Palsanawala [14], the β_s^E value of ethyl acetate + cyclohexane mixture is positive and its value is 2.5 times greater than that of cyclohexane + benzene mixture. They explained this large positive value of β_s^E as the result of disruption of ethyl acetate molecules by cyclohexane which is stronger than the disruption of cyclohexane in benzene. Also the β_s^E values are positive throughout the concentration range. Moreover, similar to cyclohexane

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+ benzene mixture, the curve for ethyl acetate + cyclohexane also bends downwards. Here, the slope of the curve decreases with increase in concentration, reaches zero at about 0.5 mole fraction and then increases with increase in concentration. Though the slope is positive at lower mole fraction range, it is negative at higher mole fraction range. Thus, the positive excess values of β_s, downward bending of the curve and change in the slope indicate that weak interactions due to dispersion forces are prominent in ethyl acetate + cyclohexane mixture. At higher temperature 323K, since the magnitude of β_s^E in ethyl acetate + cyclohexane mixture is large when compared to cyclohexane + benzene, it can be seen that the disruption of local dipolar order in ethyl acetate molecules by cyclohexane is large when compared to the disruption of order in benzene. This fact is evident if we compare the slopes of the curves for the above mixtures. Thus the strength of weak interaction is greater in ethyl acetate + cyclohexane system than in cyclohexane + benzene system.

The variation of β_s with mole fraction for the binary mixture ethyl acetate + benzene is depicted in Fig. 2(c). At 313K, the deviation between experimental and calculated values of β_s is small whereas it is high at higher temperature 323K. Also the deviation increases with increase in mole fraction at higher temperature. A perusal of the figure shows that the curve of β_s for ethyl acetate + benzene bends upwards with increase in concentration of ethyl acetate. Further the slope of the curve increases with increase in concentration. It is interesting to note that for cyclohexane + benzene and ethyl acetate + cyclohexane mixtures, appreciable deviation starts from only a particular mole fraction at higher temperatures. But for ethyl acetate + benzene mixture, the deviation starts from lower mole fraction itself at higher temperatures. Thus these analysis points to the fact that specific interaction may exists in ethyl acetate + benzene mixture. Compared to cyclohexane + benzene and ethyl acetate + cyclohexane mixtures, the opposite nature in shape and slope of the curve in ethyl acetate + benzene mixture reveals that the destruction of the local dipolar order in the pure ethyl acetate molecules has been counter-balanced by the specific interaction between ethyl acetate and benzene molecules.

Thus from these analysis, it can be concluded that structure breaking effect is prominent in cyclohexane + benzene and ethyl acetate + cyclohexane mixtures whereas specific interaction

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exists in ethyl acetate + benzene mixture. Hence our findings are in agreement with that of Oswal and Palsanawala [14].

Conclusion

An estimation of adiabatic compressibility β_s of any binary liquid mixture at moderately higher temperatures can be done using the derived temperature dependent relation, if its value at a lower temperature and temperature coefficients of sound velocity and density of pure components are known. The results show that the estimated values agree well with the experimental ones, if there is no molecular interaction, as in the case of non-interacting systems such as benzene + toluene, kerosene + diesel etc mixtures and deviation between experimental and calculated values occurs if there is molecular interaction between the components of the mixture. Further, this deviation in a binary mixture can be used to interpret the molecular interaction in the system. From the interpretation, it has been found that structure breaking effect is evident in cyclohexane + benzene and ethyl acetate + cyclohexane mixtures whereas specific interaction exists in ethyl acetate + benzene mixture.

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