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Chemistry

SPEED OF SOUND AND ISENTROPIC COMPRESSIBILITY OF BINARY MIXTURES AT 298.15 K

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ABSTRACT Speed of sound and isentropic compressibility of six polar-nonpolar cyclic liquid binary mixtures have been computed over the whole composition range at 298.15 K with the help of Prigogine-Flory-Patterson theory. Experimental surface tension and experimental density data were utilized in the prediction of sound velocity with the use of Auerbach relation. A comparison has then been carried out as regards the merit and demerits of the employed relations. An attempt has also been made to study the nature and magnitude of molecular interactions involved in the liquid mixture.

KEYWORDS : Ultrasonic Velocity, Isentropic Compressibility, Molecular Interactions, Prigogine-Flory-Patterson (PFP) model

INTRODUCTION:

Physicochemical behavior and molecular interactions occurring in a variety of liquid mixtures and solutions can be studied with the help of ultrasonic velocity. There has been an increasing interest in the study of molecular interactions and a number of experimental techniques have been used to investigate the same in binary liquid mixtures. Since data on sound velocity offers a convenient method for determining certain thermodynamical properties of liquids and liquid mixtures, which are not obtained by other methods. Ultrasonic investigations are important in elucidating internal structure of fluid mixtures involving heat transfer, mass transfer etc. which are applicable in many industrial applications

Extensive work has been carried out by Pereira et al. [1], Dzida and Ernest [2], Al-Kandary et al. [3], Pandey et al. [4], and Pan et al. [5], to investigate liquid state through analysis of ultrasonic propagation parameters and to correlate ultrasonic velocity with other physical and thermodynamic parameters. Substantial amount of work has also been done (Shukla et al. [6-9], successfully for the theoretical evaluation of ultrasonic velocity in binary and multicomponent non polar liquid mixtures by various empirical, semi-empirical and statistical mechanical concepts. Shukla et al. [10-11] have further applied these concepts to polar and ionic liquids. As a part of research concerning the thermochemical studies on new working fluid pair, we present here some useful data on speed of sound and isentropic compressibility for the mixture of trimethylbenzene with tetrahydrofuran, tetra chloromethane and dimethyl sulfoxide. These properties have been determined over the whole composition range. The major objective of the present work is to find out the applicability of Prigogine- Flory-Patterson (PFP) theory in polar-nonpolar cyclic liquid binary mixtures. The necessary parameters for computation have been taken from the work of Pan et al. [12], and CRC hand book of chemistry & physics [13]. To the best of our knowledge, this is the first report in which PFP model has been applied in polar-nonpolar cyclic liquid mixtures.

Theoretical:

In dealing with liquid state, Flory et al. [14-16], and Flory et al., defined an element (or segment) as an arbitrary chosen portion of the molecules. Considering such segments in a molecule and following Prigogine's treatment of (-mer) chain molecules and representing the number of external degree of freedom per segment by 3C, he was able to derive a partition function of the form

$$Z = Z_{comb}\left[\gamma(v^{1/3} - v^{*1/3})^3\right]^{rNC} \exp(-E_0/kT) \quad (1) \qquad (1)$$

Where k,N,T and E0 are respectively the Boltzmann constant, number of particles, absolute temeperature and excess intermonecular energy. Z_{comb} is combinatorial factor which takes into account the number of ways in which N elements intersperse among one another. Flory obtained the following experessions for the intermolecular free energy.

$$E_0 = \frac{Nrs\ \eta}{2v} \qquad (2)$$

Here is a constant characterising the energy of interaction for a pair of neighbouring sites, s is the number of intermolecular contact sites per segment and v is the volume per segment. The reduced partition function thus takes the form as;

$$z = z_{comb}(\gamma^*)^{rNC}(\tilde{v}^{1/3} - 1)^{3rNC}\exp(rNC/\tilde{v}\tilde{T}) \qquad (3)$$

the reduced equation of state obtained from the resulting partition function is given by

$$\frac{\tilde{P}\tilde{v}}{\tilde{T}} = \frac{\tilde{v}^{1/3}}{\tilde{v}^{1/3} - 1} - \frac{1}{\tilde{v}\tilde{T}} \qquad (4)$$

Changing from the molecular to molar units per segment for v, v* and , one gets,

$$\tilde{v} = v/v^* = V/V^* \qquad (5)$$

$$\tilde{T} = \frac{T}{T^*} = 2v^* ckT/S_\eta \quad \text{and (6)} \quad \tilde{P} = \frac{p}{p^*} = 2pv^*/S_\eta \quad (7)$$

The reduced equation of state at zero pressure becomes,

$$\tilde{T} = \frac{\tilde{v}^{1/3} - 1}{\tilde{v}^{4/3}} \qquad (8$$

$$\left(\frac{\alpha T}{3+3\alpha T}+1\right)^3 \qquad (9)$$

Where α is the coefficient of thermal expansion at P=0. Thus the reduced volume $\tilde{v}$ and reduced temperature $\tilde{T}$ can be obtained from the experimental values of α. The values of $\tilde{v}$, $\tilde{T}$, v* and T* can be computed using eqs (5) & (6). From the reduced equation of state, it follows that;

$$P^* = \frac{\alpha}{\beta_T} T\tilde{v}^{-2} = \gamma T\tilde{v}^{-2} \qquad (10)$$

where $_P$=(P/T)$_V$ is the thermal pressure coefficient at P=0. Characteristic pressure P* is evaluated from this equation for binary liquid mixtures with the comonent subscript 1 and 2, Flory obtained the following expressions for binary liquid mixtures as

$$\tilde{T} = \frac{T}{T^*} = \left[\frac{\Psi_1 P_1^* T_1 + \Psi_2 P_2^* T_2}{\Psi_1 P_1^* + \Psi_2 P_2^*}\right]\left[1 - \frac{\Psi_1 \theta P_2 X_{12}}{\Psi_1 P_1^* + \Psi_2 P_2^*}\right] \qquad (11)$$

and

$$P^* = \Psi_1 P_1^* + \Psi_2 P_2^* - (\Psi_1 \theta_2 X_{12}) = \frac{\tilde{T}\tilde{v}^{-4/3}}{(\tilde{v}^{-1/3} - 1)}\left[\frac{\Psi_1 P_1^*}{T_1^*} + \frac{\Psi_2 P_2^*}{T_2^*}\right] \qquad (12$$

where Ψ_1 & Ψ_2 are the segment fractions and θ_1 and θ_2 are the site fractions given by

$$\theta_2 = \frac{x_2 v_2 \tilde{v}_2^{-1}}{(x_1 v_1 \tilde{v}_1^{-1} + x_2 v_2 \tilde{v}_2^{-1})} = \left[\frac{s_2 \Psi_2}{s_1 \Psi_1 + s_2 \Psi_2}\right] \qquad (13)$$

where x_1 and x_2 are the mole fractions, X_{12} is the interaction parameter

and v_1 and v_2 are the molar volumes of the components 1 and 2 respectively. The ration $(s_1/s_2) = (v^*_1/ v^*_2)^{-1/3} = (r_1/ r_2)^{-1/3}$ for spherical molecules. The interaction parameter (X_{12}) can be expressed as

$$X_{12} = P_1^* \left[1 - \left(\frac{P_2^*}{P_1^*}\right)^{1/2} \left(\frac{v_2^*}{v_1^*}\right)^{1/6}\right]^2 \quad (14)$$

X_{12} is the energy parameter and it is measured of the difference of interaction energy between the unlike pairs and the mean of the like pairs.

The surface tension of binary liquid mixture in terms of Flory's statistical theory can be described by the expression

$$\sigma = \sigma^* \tilde{\sigma}(\tilde{v}) \quad (15)$$

Where σ*, σ̃, & ῡ are the characteristic and reduced parameters involved in the eqn.

Patterson and Rastogi [17] in their extension of the corresponding state theory dealt with the surface tension by using as a reduction parameter

$$\sigma^* = k^{1/3} P^{*2/3} T^{*1/3} \quad (16)$$

called the characteristic surface tension of the liquid. Here *k* is Boltzmann constant and P* and T* are the characteristic pressure and temperature respectively. Starting from the work of Prigogine and Saraga [18] and Prigogine and Bellmas [19] , they derived a reduced surface equation for vander Waals liquids

$$\tilde{\sigma}(\tilde{v}) = \left[M \tilde{v}^{-5/3} - \left(\frac{\tilde{v}^{-1/3} - 1.0}{\tilde{v}^2}\right) \ln\left(\frac{\tilde{v}^{-1/3} - 0.5}{\tilde{v}^{-1/3} - 1.0}\right)\right] \quad (17)$$

Where M is the fraction of the nearest neighbour that a molecule loses on moving from the bulk of the liquid to the surface. Its most suitable value ranges from 0.25 to 0.29

Although Flory's statistical theory is not directly related with ultrasonic velocity but its evaluation needed the use of well-known and well-tested Auerbach relation which can be successfully used for the evaluation of ultrasonic velocity in binary liquid mixtures represented as

$$u = \left(\frac{\sigma}{0.3 \times 10^{-3} \rho}\right)^{3/2} \quad (18)$$

where and are the surface tension and density of the liquid mixture respectively.

Isentropic compressibility is related to speed of sound and density by the relation

$$\beta_s = u^{-2} \rho^{-1} \quad (19)$$

RESULT AND DISCUSSION:

Experimental surface tension and experimental density data have been utilized to evaluate experimental sound velocity with the help of well-known and well tested relation of Auerbach for six cyclic binary liquid mixture over the whole composition range at 298.15K. Parameters of pure components are listed in table 1 along with the literature values. Values of density (), molar volume (V_m), theoretical and experimental speed of sound (u_{cal} & u_{exp}), percent deviation in speed of sound (% u) and isentropic compressibility ($_s$) for six binary mixtures; tetrahydrofuran (THF)+ 1,2,4-trimethyl benzene (TMB), tetrahydrofuran (THF)+ 1,2,3-trimethyl benzene (TMB), tetrachloromethane (TCM)+ 1,2,4- trimethyl benzene (TMB), terachloromethane (TCM)+1,3,5-trimethylbenzene, dimethyl sulfoxide (DMSO)+ 1,2,4- trimethyl benzene + dimethyl sulfoxide (DMSO)+ 1,3,5- trimethyl benzene at 298.5 K are presented in Tables 2-7. All the necessary data for computation were taken from the work of Pan et al., 2004. A close observation of all the tables reveal that experimental and calculated values of speed of sound are very close to each other indicating the success of PFP model in TMB with THF, TCM and DMSO liquid mixtures.

Table 1: Parameters of pure components at 298.15 K

Liquids	ρ[a](exp)	ρ[a](lit)	10^4α/K-1	β_T/TPa-1	V_m/cm^{3}mol-1	u /ms-1(exp)	u /ms-1(cal)	u /ms-1(lit)
1,2,4-trimethylbenzene	0.87164	087174	11.168	81.445	137.893	1415.7	1412.3	1421b
1,3,5-trimethylbenzene	0.86103	0.86109	11.320	85.961	139.592	1389.3	1390.8	1397c
Tetrahydrofuran	0.88206	0.88197	11.464	90.440	81.752	1291.2	1283.6	1278d
Tetrachloromethane	1.58380	1.58429	11.504	91.704	97.121	921.6	916.6	926e
Dimethylsulfoxide	1.09554	1.095560	9.8922	50.134	71.316	1480.9	1498.4	1487d

a- ref 5, b- ref 20, c- ref21, d- ref 22, e- ref7

Table 2: Values of Mole fraction (X1) Density (ρ), Molar volume (Vm), Theoretical and Experimental Speed of sound (u-cal,u-exp) , % Δu and Isentropic Compressibility of Tetrahydrofuran and 1,2,4-trimethyl benzene

x1	ρm /g.cm-3	Vm/ c.c.mol-1	u(cal)/ ms-1	u(exp)/ ms-1	(% Δu)	βs / Mpa-1
0.0988	0.8727	132.3	1404.5	1410.1	0.390	0.5809
0.2018	0.8737	126.6	1396.5	1405.3	0.632	0.5869
0.3003	0.8748	121.0	1388.8	1400.4	0.828	0.5927
0.3995	0.8758	115.5	1381.1	1394.4	0.958	0.5986
0.5002	0.8769	109.8	1373.3	1386.8	0.974	0.6047
0.6005	0.8779	104.2	1365.7	1377.9	0.885	0.6107
0.6986	0.8789	98.67	1358.5	1367.8	0.677	0.6165
0.7996	0.8800	93.00	1351.4	1355.9	0.336	0.6223
0.9006	0.8810	87.33	1344.6	1343.7	-0.067	0.6278

Table 3: Values of Mole fraction (X_1) Density (ρ), Molar volume (V_m), Theoretical and Experimental Speed of sound (u- cal,u-exp) , % Δu and Isentropic Compressibility of Tetrahydrofuran and 1,3,5-trimethyl benzene

x1	ρm /g.cm-3	Vm/ c.c.mol-1	u(cal)/ ms-1	u(exp)/ ms-1	(% Δu)	βs / Mpa-1
0.0481	0.8620	136.8	1388.0	1381.3	-0.484	0.60217
0.2021	0.8653	127.9	1379.0	1372.6	-0.473	0.60769
0.3002	0.8673	122.2	1373.4	1365.4	-0.587	0.61121
0.4000	0.8694	116.5	1367.8	1358.9	-0.653	0.61475
0.5004	0.8716	110.6	1362.3	1352.8	-0.702	0.61826
0.6010	0.8737	104.8	1356.9	1347.6	-0.687	0.62167
0.7003	0.8758	99.09	1351.8	1342.9	-0.665	0.62488
0.7989	0.8778	93.38	1347.0	1338.8	-0.615	0.62784
0.9000	0.8800	87.54	1342.5	1335.6	-0.515	0.63054

Table 4: Values of Mole fraction(X_1) Density (ρ), Molar volume (V_m), Theoretical and Experimental Speed of sound (u- cal,u-exp) , % Δu, Isentrtopic Compressibility of Tetrachloromethane and 1,2,4-trimethyl benzene

x1	ρm /g.cm-3	Vm/ c.c.mol-1	u(cal)/ ms-1	u(exp)/ ms-1	(% Δu)	βs / Mpa-1
0.0496	0.9070	135.9	1371.8	1374.9	0.222	0.58587
0.0996	0.9426	133.8	1333.6	1337.3	0.277	0.59654
0.1986	1.0131	129.8	1264.4	1270.1	0.449	0.61742
0.3001	1.0854	125.7	1201.2	1208.3	0.588	0.63854
0.4006	1.1569	121.6	1145.1	1152.0	0.604	0.65921
0.5987	1.2980	113.5	1049.6	1060.5	1.029	0.69938
0.6986	1.3692	109.4	1007.6	1014.9	0.721	0.71937
0.7985	1.4403	105.3	969.2	973.0	0.394	0.73918
0.9011	1.5134	101.2	932.9	932.1	-0.088	0.75929

Table 5: Values of Mole fraction(X_1) Density (ρ) , Molar volume (V_m), Theoretical and Experimental Speed of sound (u- cal,u-exp) , % Δu, Isentropic Compressibility of Tetrachloromethane and 1,3,5-trimethyl benzene

x1	ρm /g.cm-3	Vm/ c.c.mol-1	u(cal)/ ms-1	u(exp)/ ms-1	(% Δu)	βs / Mpa-1
0.1009	0.9340	137.5	1311.8	1306.0	-0.445	0.62221
0.2014	1.0066	133.3	1242.8	1235.2	-0.615	0.64319
0.2994	1.0774	129.0	1183.2	1173.6	-0.818	0.66299
0.4007	1.1506	124.7	1128.2	1118.0	-0.912	0.68274
0.4998	1.2223	120.5	1080.1	1071.0	-0.847	0.70131
0.5995	1.2943	116.2	1036.5	1028.8	-0.744	0.71914
0.6988	1.3661	112.0	997.3	991.3	-0.613	0.73592
0.7970	1.4371	107.8	962.4	957.6	-0.494	0.75134
0.9012	1.5124	103.5	929.0	925.1	-0.418	0.76616

Table 6: Values of Mole fraction(X1), Density(ρ), Molar volume (Vm), Theoretical and Experimental Speed of sound (u- cal,u-exp) , % Δu, Isentropic Compressibility of Dimethylsulfoxide and 1,2,4-trimethyl benzene

x1	ρm /g.cm-3	Vm/ c.c.mol-1	u(cal)/ ms-1	u(exp)/ ms-1	(% Δu)	βs / Mpa-1
0.1007	0.8942	131.2	1259.5	1367.2	7.88	0.70503
0.2007	0.9166	124.5	1264.7	1354.0	6.59	0.68209
0.3007	0.9390	117.9	1270.4	1333.6	4.74	0.65984
0.3995	0.9611	111.3	1276.6	1321.9	3.42	0.63841
0.5011	0.9838	104.5	1283.7	1312.2	2.17	0.61685
0.6012	1.0062	97.9	1291.4	1289.1	-0.18	0.59588
0.6992	1.0282	91.3	1300.0	1271.0	-2.28	0.57552
0.7992	1.0506	84.7	1309.9	1272.4	-2.95	0.55474
0.9013	1.0734	77.9	1321.7	1407.5	6.10	0.53331

Table 7: Values of Mole fraction(X_1) Density (ρ) , Molar volume (V_m), Theoretical and Experimental Speed of sound (u- cal,u-exp) , % Δu, Isentropic Compressibility of Dimethylsulfoxide and 1,3,5-trimethyl benzene

x1	ρm /g.cm-3	Vm/ c.c.mol-1	u(cal)/ ms-1	u(exp)/ ms-1	(% Δu)	βs / Mpa-1
0.1023	0.8850	132.6	1399.7	1338.4	-4.58	0.57675
0.1987	0.9076	126.0	1408.4	1303.8	-8.02	0.55548
0.3027	0.9320	118.9	1418.1	1274.0	-11.31	0.53355
0.401	0.9551	112.2	1427.7	1275.5	-11.93	0.51371
0.4999	0.9783	105.5	1437.7	1285.3	-11.86	0.49452
0.6003	1.0018	98.6	1448.5	1314.8	-10.17	0.47575
0.6990	1.0250	91.9	1459.7	1350.6	-8.08	0.45791
0.8003	1.0487	85.0	1471.9	1404.3	-4.81	0.44016
0.8998	1.0720	78.2	1484.6	1466.3	-1.25	0.42321

The percent deviations in speed of sound at 298.15 K from Figures 1&2 shows that they are positive for THF and TCM with 1, 2, 4 TMB and are negative for THF, TCM and DMSO with 1, 3, 5 TMB while DMSO with 1, 2, 4 TMB has both positive and negative values. The maximum positive value of percent deviation in speed of sound follow the order DMSO+ 1,2,4 TMB>TCM + 1,2,4 TMB>THF + 1,2,4 TMB and maximum negative values of percent deviation in speed of sound follow the order DMSO+ 1,3,5 TMB> TCM+ 1,3,5 TMB> THF + 1,3,5 TMB. In both the cases, the trend is similar except for the system DMSO+1, 2, 4 TMB where both positive and negative deviations are observed.

The possible reason may be that there are no strong intermolecular forces between DMSO and TMB because 1, 3, 5 TMB is a symmetrical nonpolar molecule. Conversely, stronger forces exist for the systems with 1, 2, 4 TMB, so the values are positive. Negative values of the systems with 1,3,5- TMB are possibly owing to the greater repulsion due to bulky methyl group as compared to 1,2,4-TMB.

These short range repulsive forces are responsible for less compact structure of 1,3,5-TMB.With the increase of concentration, density increases and molar volume decreases due to shrinkage in the volume of DMSO and TCM. Hence, an increase in the speed of sound may be correlated to the structure former of the 1, 3, 5-TMB. Positive deviations are due to dipolar-dipolar interactions because 1, 2, 4 TMB is a polar molecule.

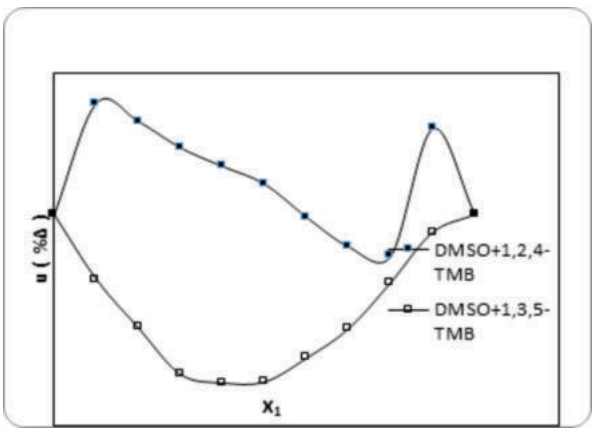

Fig. 1-variation of deviation in speed of sound (%Δu) against mole fraction(X_1) of binary liquid mixture at 298.15 K

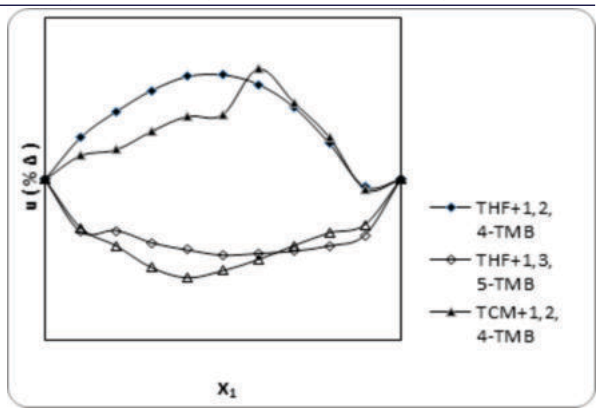

Fig. 2- variation of deviation in speed of sound (%Δ u) against mole fraction of binary liquid mixture at 298.15 K

The lack of smoothness in deviations is due to the interaction between the component molecules. Isentropic compressibility increases regularly with the increase of mole fraction. Increment becomes larger with 1, 2, 4 TMB. Strong discrepancy is observed in Table 6 where the value of isentropic compressibility decreases. Possibly, it is due to dipolar repulsion which lowers the energy hence regular trend is not observed, while density and speed of sound show regular behaviour.

CONCLUSION:

Thus it can be concluded from the preceding discussion that PFP model can be applied successfully to polar-nonpolar cyclic liquid binary mixtures.

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