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ResearchArticle

EXPERIMENTAL STUDY OF THE THERMO PHYSICAL AND ACOUSTICAL PROPERTIES OF BINARY MIXTURES

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ARTICLE INFO	ABSTRACT
Article History: Received 13 th May, 2018 Received in revised form 11 th June, 2018 Accepted 8 th July, 2018 Published online 28 th August, 2018	Ultrasonic velocity (U), Density (ρ) and Viscosity (η) have been measured in binary liquid mixtures of Propylene glycol mono propyl ether (PGPE) + Aniline (ANI) and PGPE + 1-Amino-2-propanol (MIPA) at 303.15 K and 313.15 K. From these values, acoustical parameters such as Adiabatic compressibility (β), Free length (Lf), Acoustical impedance (Z) and Molar volume (V) were calculated using standard relations. Excess values such as ZE, LfE, η E, UE and VE were also determined. The results are interpreted in terms of molecular interaction such as dipole - dipole interaction or hydrogen bonding between the components of mixture.

Key Words:

Molecular interaction, Density, Viscosity, Adiabatic Compressibility, Free length, Acoustical Impedance, Molar Volume, Excess Value.

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INTRODUCTION

Ultrasonic investigation of liquid mixtures containing polar solvent and weakly interacting components are of considerable interest to researchers in order to understand the intermolecular interaction and binary forces prevailing between the atoms or molecules (1-3). Knowledge about the intermolecular interaction between the components is very much important in several industrial and technological processes (4–7). Ultrasonic velocity and its related parameterslike Adiabatic compressibility (β), Free length (L_f), Acoustical impedance (Z) and Molar volume (V) helps in characterizing thermodynamic and physico - chemical aspects of binary liquid mixtures such as molecular association and dissociation (8-9). Hence, the intermolecular interaction existing between the components of these binaries has to be revealed in order to understand any physico- chemical process in these mixtures.

The present work deals with the study of ultrasonic velocity and its related parameter like Adiabatic compressibility(β), Free length (L_f), Acoustical impedance (Z), Molar volume (V) and their excess values like Z^E, L_f^E, η^{E} , U^E and V^E in the binary system of propylene glycol mono propyl ether (PGPE) + aniline (ANI) and propylene glycol mono propyl ether (PGPE) + 1-Amino-2-propanol (MIPA) at two different temperatures viz., 303.15K and 313.15K.ANI is a popular industrial organic solvent and hence its interaction with neighbouring components in a mixture has to be exposed to know the significance of its compositional behaviour (10). PGPE is a polar solvent and the hydroxyl group of PGPE forms hydrogen bonding with the hydrogen atom of weak base components such as amino group of 1-Amino-2-propanol and aniline. The – OH group of PGPE forms hydrogen bonding with the presence of electronegative force between the components.

Experimental details

PGPE, ANI and MIPA were obtained from Sigma Aldrich Chemicals Ltd. Various concentration of the binary mixtures were prepared in terms of mole fractions. The ultrasonic velocities in liquid mixtures were measured using an Mittal type ultrasonic interferometer model M-84 at 2 MHz frequency with an accuracy of ± 0.1 ms⁻¹. Densities and Viscosities were determined using specific gravity bottle and Ostwald's

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viscometer with an accuracy of $\pm 0.1 \text{Kgm}^{-3} \& \pm 0.001 \text{NSm}^{-2}$ respectively. All the measurements were made at two different temperatures (303.15K and 313.15K) with the help of an electronically operated digital constant temperature bath (Guna make model) having the operating temperature range from 1°C to100°C with an accuracy of $\pm 0.1 \text{K}$.Each observation is repeated for five times and the average is taken as final value. Using the measured data, the acoustical parameter such as Acoustical impedance (Z), Adiabatic compressibility (β), Molar volume (V) and Free length (L_f) were calculated from the following standard thermophysical expressions

 $Z = U.\rho 1$

Where U = Ultrasonic velocity ρ = Density of liquid $\beta = 1/U^2 \rho$

$$V = M_{eff} / \rho_{mix}$$
 3

Where M_{eff}is effective molecular weight given as

$$M_{eff} = \sum M_i x$$

In the above expression, M_i & x_i are the molecularweight and mole fraction of individual constituents respectively.

2

$$L_f = K_T \sqrt{\beta}$$
 4

Where K_T is a Jacobson's constant.

The excess values of the parameter are computed from the following expressions

$$\eta^{\rm E} = \eta_{\rm mix} - (x_1\eta_1 + x_2\eta_2) \qquad 5$$
$$Z^{\rm E} = (\rho_{\rm mix}U_{\rm mix}) - (X_1\rho_1U_1 + X_2\rho_2U_2) \qquad 6$$

$$V^{E} = (X_{1}M_{1} + X_{2}M_{2}/\rho_{mix}) - (X_{1}M_{1}/\rho_{1} + X_{2}M_{2}/\rho_{2}) 7$$

Where

 ρ_{mix} = Density mixtures

 ρ_1 = Density of first components

 ρ_2 = Density of second components

 M_1 = Molecular weight of first components

 M_2 = Molecular weight of second components

$$L_{\rm f}^{\rm E} = K/(U_{\rm mix}^2 \rho_{\rm mix})^{1/2} - (X_1 K/(U_1^2 \rho_1)^{1/2} + (X_2 K/(U_2^2 \rho_2)^{1/2}) 8$$

Where

Z K = Jacobson's constant = 200×10^{-8} at 303.15K = 203×10^{-8} at 313.15K

$$U^{E} = U - (X_{1}U_{1} + X_{2}U_{2})$$
9

From the review of literature, the values for the Density and Speed of MIPA, ANI and PGPE are given in table 1 (11).

Table 1 Density and Speed of MIPA, ANI and PGPE

T (K)	P (Kgm ⁻³)	U (Ms ⁻¹)
	MIPA	
303.15	951	1526
505.15	952.39	
313.15	944	1493
515.15	944.30	
	ANILINE	
303.15	1011	1612
303.15	1010.9	1614
313.15	1006	1581
515.15		
	PGPE	
303.15	879	1215
303.15		
313.15	872	1186
313.15		

RESULTS AND DISCUSSION

The experimental density (ρ), viscosity (η), and Ultrasonic velocity (U) values and computed parameters β ,Z, V & L_fof PGPE in ANI and PGPE in MIPA at 303.15K & 313.15K are given in Table 2 and 3 respectively. All the excess values of the parameter V^E, L_f^E, Z^E, η^{E} and U^E at 303.15K & 313.15K are given in Table 4 and 5.

Table 2Experimental Density(ρ), Viscosity(η) and Ultrasonicvelocity(U) Data and Computed Parameters (β , Z, V and L_f)values for the PGPE + ANI binary mixture

Mole fractionX ₁	ρ Kgm ⁻⁽	3 η×10 ⁻³ Nsm ⁻¹	Ums ⁻¹	β× 10 ⁻¹⁰	Z×10 ³	V×10 ⁶ M ³ mol ⁻¹	$L_f \times 10^{-11}$
	1	2	3	4	5	6	7
			30)3.15K			
1.0000	879	1.5587	1215	7.7065	1067.9	134.448	5.5521
0.9196	893	1.5835	1254	7.1212	1119.8	130.0335	5.3371
0.8371	906	1.6468	1293	6.6019	1171.4	126.0816	5.1388
0.7470	918	1.7093	1335	6.1121	1225.5	121.5359	4.9445
0.6549	933	1.7786	1374	5.6773	1281.9	117.3311	4.7654
0.5592	946	1.8453	1416	5.2720	1339.5	106.8816	4.5921
0.4580	958	1.9536	1455	4.9306	1393.5	109.1022	4.4409
0.3522	972	2.0253	1494	4.6092	1452.1	104.9588	4.2938
0.2402	987	2.0565	1537	4.2887	1517.0	100.4660	4.1418
0.1235	1000	2.1280	1577	4.0210	1577.0	96.1771	4.0104
0.0000	1011	2.1984	1612	3.8064	1629.7	92.1167	3.9019
			313.15H	Κ			
1.0000	872	1.3917	1186	8.1529	1034.1	135.5275	5.7960
0.9196	885	1.5301	1223	7.5544	1082.3	131.2090	5.5795
0.8371	898	1.5924	1261	7.0031	1132.3	127.2048	5.3720
0.7470	913	1.6595	1300	6.4810	1186.9	122.2015	5.1679
0.6549	926	1.6831	1338	6.0322	1238.9	118.2181	4.9857
0.5592	948	1.7651	1379	5.5470	1307.2	106.6561	4.7810
0.4580	953	1.8167	1418	5.2186	1351.3	109.6747	4.6373
0.3522	967	1.9291	1458	4.8647	1409.8	105.5015	4.4773
0.2402	982	2.0026	1501	4.5198	1473.9	100.9775	4.3157
0.1235	995	2.0732	1542	4.2267	1534.2	96.6532	4.1734
0.0000	1006	2.0961	1581	3.9768	1590.4	92.5754	4.0482

Table 3Experimental Density(ρ), Viscosity(η) and Ultrasonicvelocity(U) Data and Computed Parameters (β , Z, V and L_f)values for the PGPE + MIPA binary mixture

Mole fraction X ₁	ρKgm ⁻³	η×10 ⁻³ Nsm ⁻¹	U ms ⁻¹	β× 10 ⁻¹⁰	Z×10 ³	V×10 ⁶ M ³ mol ⁻¹	$L_f \times^{10-11}$
	1	2	3	4	5	6	7
			303	3.15K			
1.0000 0.9338	879	1.5587	1215	7.7065	1067.9	134.4482	5.5521
0.8626	885	1.8048	1232	7.4444	1090.3	130.2259	5.4568
0.7853	892	2.1354	1250	7.1748	1115.0	125.8632	5.3571
0.7018	899	2.6703	1270	6.8965	1141.7	121.0789	5.2522
0.6114	905	2.7683	1308	6.4585	1183.7	116.2872	5.0827
0.5171	912	3.3558	1343	6.0792	1224.8	111.2280	4.9312
0.4043	919	4.5223	1380	5.7138	1268.2	106.5832	4.7807
0.3161	927	5.5480	1418	5.3649	1314.4	99.8489	4.6324
0.1485	934	7.0806	1455	5.0573	1358.9	94.9464	4.4976
0.0000	942	9.0206	1490	4.7816	1403.5	86.4755	4.3733
0.0000	951	11.7207	1526	4.5155	1451.2	78.9800	4.2499
			315	5.15K			
1.0000	872	1.3917	1186	8.1529	1034.1	135.5275	5.7963
0.9338	879	1.4808	1213	7.7319	1066.2	131.1149	5.6446
0.8626	885	1.9617	1242	7.3250	1099.1	126.8587	5.4941
0.7853	892	2.5704	1271	6.9397	1133.7	122.0291	5.3476
0.7018	898	2.6275	1302	6.5690	1169.1	117.1937	5.2029
0.6114	905	3.0893	1331	6.2372	1204.5	112.0883	5.0698
0.5171	912	3.8814	1361	5.9195	1241.2	107.4013	4.9389
0.4043	920	4.8944	1394	5.5935	1282.4	100.6086	4.8010
0.3161	928	6.2946	1425	5.3066	1322.4	95.5603	4.6763
0.1485	935	7.8758	1457	5.0381	1362.2	87.1229	4.5564
0.0000	944	10.6300	1493	4.7523	1409.3	79.5656	4.4253

 $\begin{array}{l} \textbf{Table 4} \ Excess \ acoustic \ impedance \ (Z^E), excess \ viscosity(\eta^E), \\ excess \ Molar \ volume(V^E), \ excess \ free \ lenth(L_f^E) \ and \ excess \\ Ultrasonic \ velocity(U^E) \ values \ of \ the \ PGPE \ + \ ANI \ binary \\ mixture \end{array}$

		303.15 K			
Mole fraction X ₁	Z ^E X10 ³	η ^E X10 ⁻³	V ^E X10 ⁶ m ³ mol ⁻¹	L _f ^E X10 ⁻ ⁹ m	U ^E ms ⁻¹
1.0000	0	0	0	0	0
0.9196	6.510	-0.0267	-1.0200	-0.0261	6.920
0.8371	9.521	-0.0193	-1.6088	-0.0475	10.912
0.7470	20.151	-0.0048	-1.9351	-0.0565	24.234
0.6549	21.240	0.0008	-2.4438	-0.0678	23.125
0.5592	24.423	0.0054	-2.5471	-0.0732	26.486
0.4580	22.743	0.0501	-2.3285	-0.0676	26.118
0.3522	18.980	0.0506	-2.1405	-0.0608	20.534
0.2402	22.557	0.0122	-1.8419	-0.0498	20.598
0.1235	17.459	0.0099	-1.1213	-0.0295	14.836
0.0000	0	0	0	0	0
		313.15K			
1.0000	0	0	0	0	0
0.9196	3.279	0.0816	-0.8117	-0.0241	5.085
0.8371	5.181	0.0836	-1.4543	-0.0459	8.283
0.7470	16.579	0.0958	-2.1878	-0.0551	18.649
0.6549	13.933	0.0499	-2.4214	-0.0646	16.792
0.5592	28.363	0.0636	-3.3524	-0.0769	19.358
0.4580	16.924	0.0450	-2.4893	-0.0659	19.174
0.3522	14.055	0.0795	-2.2676	-0.0600	14.854
0.2402	17.456	0.0762	-1.9378	-0.0485	15.116
0.1235	13.305	0.0653	-1.1724	-0.0280	10.573
0.0000	0	0	0	0	0

 $\begin{array}{l} \textbf{Table 5} Excess \ acoustic \ impedance \ (Z^E), excess \ viscosity(\eta^E), \\ excess \ Molar \ volume(V^E), \ excess \ free \ lenth(L_f^E) \ and \ excess \\ Ultrasonic \ velocity(U^E) \ values \ of \ the \ PGPE + MIPA \ binary \\ mixture \end{array}$

X10 ³ η ^E X1 0 0 .726 -0.416 .931 -0.821 .084 -1.058 0.70 -1.807 .759 -2.152 .994 -2.032 .334 -2.071	m mol 0 0 50 -0.4665 18 -0.9714 84 -1.3726 77 -1.5186 29 -1.6615 38 -1.6768		ms ⁻¹ 0 -2.214 -8.036 -10.245 1.938
.726 -0.416 .931 -0.821 .084 -1.058 .070 -1.807 .759 -2.152 .994 -2.033	60 -0.4665 18 -0.9714 84 -1.3726 77 -1.5186 29 -1.6615 38 -1.6768	0 -0.0016 -0.0053 -0.0050 -0.0240	-2.214 -8.036 -10.245 1.938
.931 -0.821 .084 -1.058 .070 -1.807 .759 -2.152 .994 -2.033	18 -0.9714 84 -1.3726 77 -1.5186 29 -1.6615 38 -1.6768	-0.0053 -0.0050 -0.0240	-8.036 -10.245 1.938
.084 -1.058 .070 -1.807 .759 -2.152 .994 -2.033	84 -1.3726 77 -1.5186 29 -1.6615 38 -1.6768	-0.0050 -0.0240	-10.245 1.938
.070 -1.807 .759 -2.152 .994 -2.033	77 -1.5186 29 -1.6615 38 -1.6768	-0.0240	1.938
.759 -2.152 .994 -2.033	29-1.661538-1.6768		
.994 -2.033	-1.6768	-0.0364	
			6.992
334 -2.07		-0.0555	3.067
	-1.5952	-0.0463	16.821
.468 -1.423	-1.5200	-0.0512	27.917
.847 -1.186	63 -0.6927	-0.0216	10.793
0 0	0	0	0
313.15	δK		
.421 -1.893 .005 -2.053	13 -0.9873 41 -1.5011 72 -1.5428 33 -1.6887 32 -1.7040 68 -1.6063 09 -1.6475	$\begin{array}{c} 0\\ -0.0179\\ -0.0362\\ -0.1641\\ -0.0567\\ -0.0613\\ -0.0725\\ -0.0573\\ -0.0573\\ -0.0223\\ 0\end{array}$	$\begin{array}{c} 0\\ 8.020\\ 13.519\\ 20.580\\ 26.094\\ 25.550\\ 16.253\\ 24.224\\ 29.639\\ 10.186\\ 0\end{array}$
	4.671 -1.50' 4.421 -1.89' 5.005 -2.05' 9.937 -2.000' 2.173 -1.410'	.671 -1.5072 -1.5428 .421 -1.8933 -1.6887 .005 -2.0532 -1.7040 .937 -2.0068 -1.6063 .173 -1.4109 -1.6475 .185 -1.3779 -0.7043	$\begin{array}{cccccccccccccccccccccccccccccccccccc$

From the tables 2 and 3, it is found that the ultrasonic velocity (U), density (ρ) and viscosity (η) are increasing with increasing in mole fraction of ANI and MIPA. The increase in velocity perhaps is due to structural changes occurring in the mixture, resulting in weakening of intermolecular forces (12). However, in both the binary systems, ultrasonic velocity (U) decreases with increase of temperature at all noted concentration. This is because of the increase in spacing between molecules and increase in enthalpy of its structural arrangement. Other

parameters such as density (ρ) and viscosity (η) decreases with increase of temperature. This indicates that the intermolecular forces decreases due to increases in thermal vibrations of the system, which also causes an increase in volume expansion and hence increase in free path length.

In PGPE + ANI system, when mole fraction of aniline is increased, an increasing trend is observed in density, viscosity and velocity. It reveals that the addition of aniline increases the effective molecular space. This can be attributed to both relatively higher dielectric constant of aniline (13,14) and its electron donor nature. When ANI is mixed with PGPE, ANI exhibits significant interactions such as dipole – dipole interaction and hydrogen bonding in the liquid mixture.

However, Adiabatic compressibility (β) and Free length (L_f) of PGPE + MIPA decreases with increase in concentration of MIPA. This shows significantinteraction between solute and solvent molecules and is found also in other binary system. Acoustic impedance (Z) is the ratio of the effective sound pressure at a point to the effective particular velocity at that point. In both the binary systems, on increasing the mole fraction of ANI and MIPA, the acoustic impedance (Z) also increases. This shows the weakening of intermolecular forces (15).

The decrease in molar volume (V), adiabatic compressibility (β) and free length (L_f) and increase in acoustic impedance (Z) with the mole concentration of both binary systems reveals the presence of specific interactions between the components in these mixtures.

An examination of data in tables 4 & 5 suggests that the excess acoustic impedance (Z^{E}) of PGPE + MIPA was initially negative and increases with increase in the mole fraction of MIPA and finally becomes positive. When temperature is increased, the entire range of excess acoustic impedance (Z^{E}) becomes positive. Meanwhile the excess acoustic impedance (Z^{E}) of PGPE + ANI is positive in the entire range of composition. Positive value of Z^E in mole concentration of both binary systems reveals the possibility of the presence of strong attractive force between the components. The n^E value of PGPE + ANI was initially negative and on increasing the mole fraction of ANI, the value of η^E becomes positive. This reveals the presence of both strong attraction forces between the molecules and specific interaction such as formation of hydrogen bonding between OH and N. The negative value of η^E of PGPE + MIPA suggest the mutual loss of specific interactions between unlike molecules (16,17). Negative deviation in viscosity may also occur where dispersion forces are dominant, particularly for the system having different molecular sizes (17,18). It reveals the presence of weak interaction between the molecules.

It is noticed from Table 4 and 5 that the behaviour of excess acoustic impedance (Z^E) is similar to excess ultrasonic velocity U^E . The positive value of U^E also indicates strong attraction among the components of a mixture (22).

The behaviour of excess values of Z^E , L_f^E , V^E , η^E and U^E with mole fraction of binary system at 303.15K and 313.15K were shown in figure 1to10.

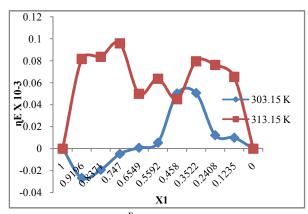


Fig 1 Excess viscosity η^{E} - mole fraction of PGPE + Aniline

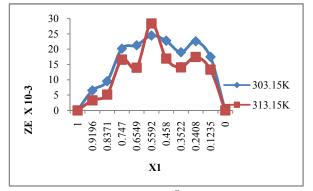


Fig 2 Excess Acoustic Impedance Z^E - mole fraction of PGPE + Aniline

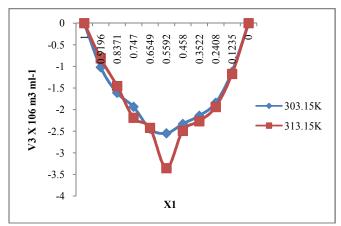


Fig 3 molar volume V^E - mole fraction of PGPE + Aniline

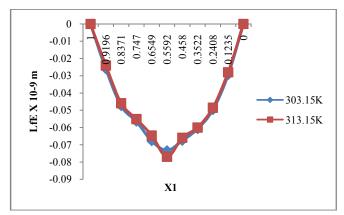


Fig 4 Free Length L_{f}^{E} - mole fraction of PGPE + Aniline

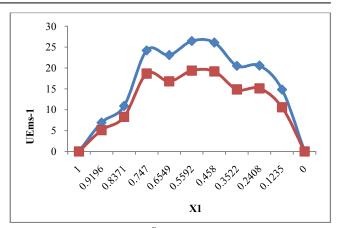


Fig 5 ultrasonic velocity U^{E} - mole fraction of PGPE + Aniline

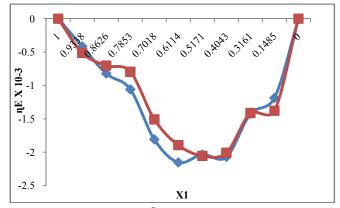


Fig 7 Excess viscosity η^{E} - mole fraction of PGPE + MIPA

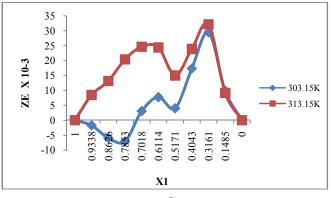


Fig 6 Excess Acoustic Impedance \mathbf{Z}^{E} - mole fraction of PGPE + MIPA

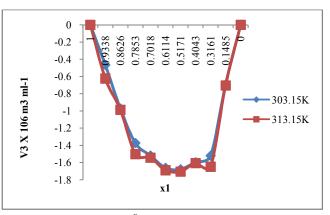


Fig 8 molar volume $V^{E}\,$ - mole fraction of PGPE + MIPA

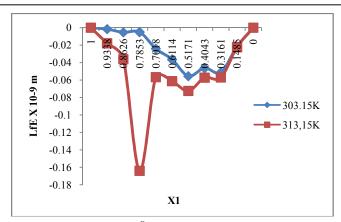


Fig 9 Free Length L_{f}^{E} - mole fraction of PGPE + MIPA e

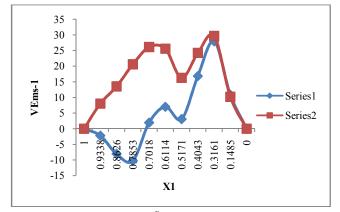


Fig10 ultrasonic velocity U^{E} - mole fraction of PGPE + MIPA

The value of V^E is the resultant contribution from several opposing effects namely chemical, physical and structural. The chemical or specific interaction result in volume concentration, leading to negative excess volume and these interactions include charge transfer complexes, dipole – dipole and dipole – induced dipole interaction, proton acceptor interaction and H – bonding between unlike molecules.

The more negative V^E value of both systems viz., PGPE + Aniline and PGPE + MIPA at 303.15K and 313.15K reveals the presence of two CH₃ groups increases the electron density at the oxygen atom of hydroxyl group. Hence it may be concluded that the interaction between PGPE with ANI and MIPA under study increases when hydroxyl group is attached to a carbon atom with greater number of CH₃ groups.

The numerical values of V^E become more negative as the temperature is increased and they vary in the following order PGPE + Aniline < PGPE + MIPA. The excess intermolecular free length L_f^E are negative in both binary systems, at over the entire range of compositions. The negative excess values are due to the associated molecules, which accounts for the existence of strong molecular interaction (19), whereas positive excess values causes weak interaction between unlike molecules. The sign of excess intermolecular free length L_f^E play a vital role in assessing the interaction between the components due to hydrogen bonding, charger transfer, dipole dipole interaction and dipole-induced dipole interaction, intertial accommodation and orientationalordering (20). Therefore negative value of excess intermolecular free length L_{f}^{E} in present work suggest that dipole – dipole interaction are prevailing through H – bonding in the liquid mixtures (21).

CONCLUSION

The densities, viscosities, speed of sound, acoustic impedance, molar volume, intermolecular free length, and adiabatic compressibility at 303.15 K & 313.15K have been measured for two binary mixtures. From these measured data excess acoustic impedance, excess molar volume, deviation in viscosities, excess ultrasonic velocity and excess intermolecular free length have been calculated. These results have been used to discuss the nature of interaction between unlike molecules in terms of hydrogen bonding, dipole – dipole interaction and proton acceptor interaction of molecules.

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