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Prediction Of Molecular Interactions In Binary System From 288.15 To 318.15 K By Ultrasonic Speed And Isentropic Compressibility.

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ABSTRACT

Ultrasonic speed for a mixture of dodecane and 1-butanol were calculated from (288.15 to 318.15K) over the whole composition range at atmospheric pressure, from the experimental work of J. Peleterio. Flory(non-associated), Ramaswamy and Glinski (associated) were used to predict the behaviour and molecular interactions of binary system. Deviation in ultrasonic speed (ΔU) was used in Redlich Kister polynomial to determine the numerical coefficients and standard deviation. Isentropic compressibility was also calculated over the entire composition range at various temperatures. Eyring's theory-based McAllister models were used to correlate the thermoacoustic properties. Calculation by these models were compared with the experimental values to test extent of the molecular interactions. Ramaswamy was found more consistent with experimental values in comparison to Flory.

Keywords: Ultrasonic speed, Isentropic compressibility, McAllister model, Redlich Kister

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INTRODUCTION

In recent years, ultrasonic studies provide a way to measure of various thermodynamic properties and to predict the molecular interaction of liquid mixtures. An exhaustive literature survey reveals that various experimental techniques have been used to investigate the interactions between the components of binary liquid mixtures [1-4] but sometimes theoretical prediction is required in the absence of experimental values. This paper is concerned with ultrasonic speed and isentropic compressibility of binary system which were calculated from the measured work of J. Peleteiro [5] by Ramaswamy and Anbananthan model [6] and Glinski model[7] which depend on associate of binary system along with adjusting parameter as an association constant and their calculation procedure was almost similar whereas Flory model[8-12] concern with non-associated process which explains the behaviour of non-ionic γ- merci spherical chain type molecules and deals with the additivity of liquids. In our previously published work [13] we have determined speed of sound and isentropic compressibility for weakly interacting liquids at different temperatures. Standard deviation and numerical coefficients were calculated by Redlich Kister equation [14] using deviation in ultrasonic speed (ΔU). McAllister model [15] obey the concepts of Eyring's theory was used to correlate the experimental results of ultrasonic speed and isentropic compressibility. The aim of this wok to understand the molecular interactions between the binary system and to estimate the various liquid state models.

Theoretical modelling

Flory model

Flory's statistical theory [8-12] was used to calculate the ultrasonic speed of binary system by well-known and well tested Auerbach relation which is expressed as

$$\mathrm{U} = \left(\frac{\sigma}{6.3 \times 10^{-4}\rho}\right)^{2/3} \qquad (1)$$

Where U is ultrasonic speed and σ is surface tension which can be calculated by the following equation

$$\sigma = \sigma^{*}\tilde{\sigma}(\mathrm{v}) \qquad (2)$$

where σ^* and $\tilde{\sigma}(v)$ are characteristic surface tension and reduced surface tension respectively.

$$\tilde{\sigma}(\mathrm{v}) = \mathrm{M}\tilde{\mathrm{v}}^{5/3} - \frac{\left(\tilde{\mathrm{v}}^{1/3} - 1\right)}{\left(\tilde{\mathrm{v}}^{2}\right)} \ln \frac{\left(\tilde{\mathrm{v}}^{1/3} - 0.5\right)}{\left(\tilde{\mathrm{v}}^{1/3} - 1\right)} \qquad (3)$$

Value of M lies in the range 0.25-0.29.

According to Patterson and Rastogi [12] there is a very close connection between corresponding state theory and Flory model. In order to determine characteristic surface tension (σ^*) Patterson and Rastogi extend the corresponding state theory by using the following equation

$$\sigma^{*} = \mathrm{k}^{1/3}\mathrm{p}^{*2/3}\mathrm{T}^{*1/3} \qquad (4)$$

Where k,P^* and $\mathrm{T}^*$are Boltzmann constant, Characteristic pressure and Characteristic temperature respectively

Ramaswamy model

Ramaswamy and Anbananthan [6] model depend on association process and deals with an assumption of linear relation between acoustic impedance and mole fractions of binary components. Above assumption when applied to ultrasonic speed in the same fashion we get the following equation

$$\mathrm{U}_{\mathrm{cal}} = \mathrm{X}_1\mathrm{U}_1 + \mathrm{X}_2\mathrm{U}_2 + \mathrm{X}_{12}\mathrm{U}_{12} \qquad (5)$$

X_1, X_2 are mole fractions of binary components, X_{12} is mole fraction of associate, U_1, U_2 are ultrasonic speeds of pure component 1&2 respectively. U_{12} is ultrasonic speed of associate and U_{cal} is calculated ultrasonic speed by Ramaswamy model.

Glinski model

Glinski [7] proposed an empirical equation between ultrasonic speed and volume fraction, based on an assumption of additivity.

$$U_{cal} = \frac{U_1U_2U_{12}}{U_1U_{12}\phi_2 + U_2U_{12}\phi_1 + U_1U_2\phi_{12}} \qquad (6)$$

Where ϕ_1 and ϕ_2 are volume fraction of pure components.

Isentropic compressibility (β_S) depend on ultrasonic speed (U) and density (ρ) of liquid mixture was calculated by the following equation

$$\beta_S = U^{-2}\rho^{-1} \qquad (7)$$

McAllister model

McAllister model [15] correlates the physicochemical properties with experimental results based on Eyring's absolute theory

McAllister-3- body

$$\ln U = x_1^3 \ln U_1 + 3x_1^2x_2 \ln U_{12} + 3x_1x_2 \ln U_{21} + x_2^3 \ln U_2 - \ln[x_1 + x_2M_2/M_1] + 3x_1^2x_2 \ln[(2 + M_2/M_1)/3] + 3x_1x_2^2 \ln[(1 + 2\,M_2/M_1)/3] + x_2^3 \ln[M_2/M_1] \qquad (8)$$

McAllister-4-body

If constituent molecules differ in size in large extent, then the four body McAllister model follow the same numerical procedure as three body McAllister model

$$\ln U = x_1^4 \ln U_1 + 4x_1^3x_2 \ln U_{1112} + 6x_1^2x_2^2 \ln U_{1122} + 4x_1x_2^2 \ln U_{222} + x_2^4 \ln U_2 - \ln[(x_1 + x_2M_2/M_1)] + 4x_1^3x_2 \ln[(3 + M_2/M_1)/4] + 6x_1^2x_2^2 \ln[(1 + M_2/M_1)/2] + 4x_1x_2^3 \ln[(1 + 3M_2/M_1)/4] + x_2^4 \ln(M_2/M_1) \qquad (9)$$

Where X_1, X_2 are mole fractions of binary components, U_1, U_2 are ultrasonic speed of individual components and M_1, M_2 are molecular weight of pure components.

RESULT AND DISCUSSION

Table 1 represents the coefficient derived from Redlich Kister equation and standard deviation. Coefficients calculated for ultrasonic speed and isentropic compressibility from McAllister 3 and 4 body model and their respective standard deviations at different temperatures are presented in table 2 and 3 respectively. Table4 represents the experimental density and percentage deviation for ultrasonic speed calculated for various models from (288.15-318.15K). Measured and calculated isentropic compressibility along with their percentage deviation are presented in table5. Deviation in ultrasonic speed (ΔU) calculated mathematically and applied to Redlich- Kister polynomials [14] using nonlinear least square method to determine the coefficients by the following equation

$$Y = X_1X_2 \sum_{p=0}^{n} A_p(2X_1 - 1)^p \qquad (10)$$

Where A_p represents the coefficient of Redlich Kister equation, X_1 and X_2 are mole fraction of respective liquids in binary system. Y represents deviation in ultrasonic speed. Coefficients and Standard deviation(δ) calculated from Redlich- Kister equation decreases with increase in temperature. Value of standard deviation lies in the range $0.23 < \delta < 0.53$.the highest value of standard deviation was found to be at 288.15K whereas the lowest value of standard deviation was found to be at 318.15K.

Coefficients and standard deviation calculated from McAllister 3 and 4 body model for ultrasonic speed are presented in table2 shows that coefficients and standard deviation decreases with increase in temperature. Standard deviation for ultrasonic speed lies in range $0.54 < \delta < 0.74$ and $0.24 < \delta < 0.31$ respectively. McAllister 4 body model show more accurate result than McAllister 3 body model at higher temperature whereas for isentropic compressibility, standard deviation calculated for McAllister 3 and 4 body model (table3) increases with increase in temperature which is shows inverse relation of ultrasonic speed and isentropic compressibility. for McAllister 3 body model standard deviation lies in the range $1.89 < \delta < 2.50$ whereas standard deviation calculated for McAllister 4 body lies in the range $0.45 < \delta < 0.48$.which is more consistent with the observed value. A close examination of table 4 reveals that ultrasonic speed decreases with increase in the concentration of dodecane except few points for all the models at different temperatures as shown in figure 1. Ultrasonic speed decreases with increase in the isentropic compressibility which indicate a weak interaction between the liquid components. Extent of liquid-liquid interactions can be determined with the help of density. Density of binary system increases with increase in the concentration of dodecane. which indicate the association between the binary components but with increase in temperature density decreases which confirm the weak liquid-liquid interactions at higher temperatures. Lesser percentage deviation was observed in Ramaswamy and Glinski model in comparison to Flory model. Decreasing order of percentage deviation was Ramasawamy < Glinski < Flory. Isentropic compressibility was used to predict the intermolecular interactions in term of association or repulsion. calculated results of isentropic compressibility presented in table 5 reveals that isentropic compressibility increases with increase in the concentration of dodecane. the theoretical values calculated from Ramaswamy and Glinski model are very close to experimental values than values calculated from Flory model. Which confirm that Ramaswamy (associated) model gave an excellent result than non-associated (Flory) model. The percentage deviation obtained from Ramaswamy and Glinski model are positive over the entire concentration range for all the temperature except Flory model as shown in figure2.Since isentropic compressibility increases with increase in temperature while ultrasonic speed decreases which clearly indicates that molecular association is stronger at lower temperatures.

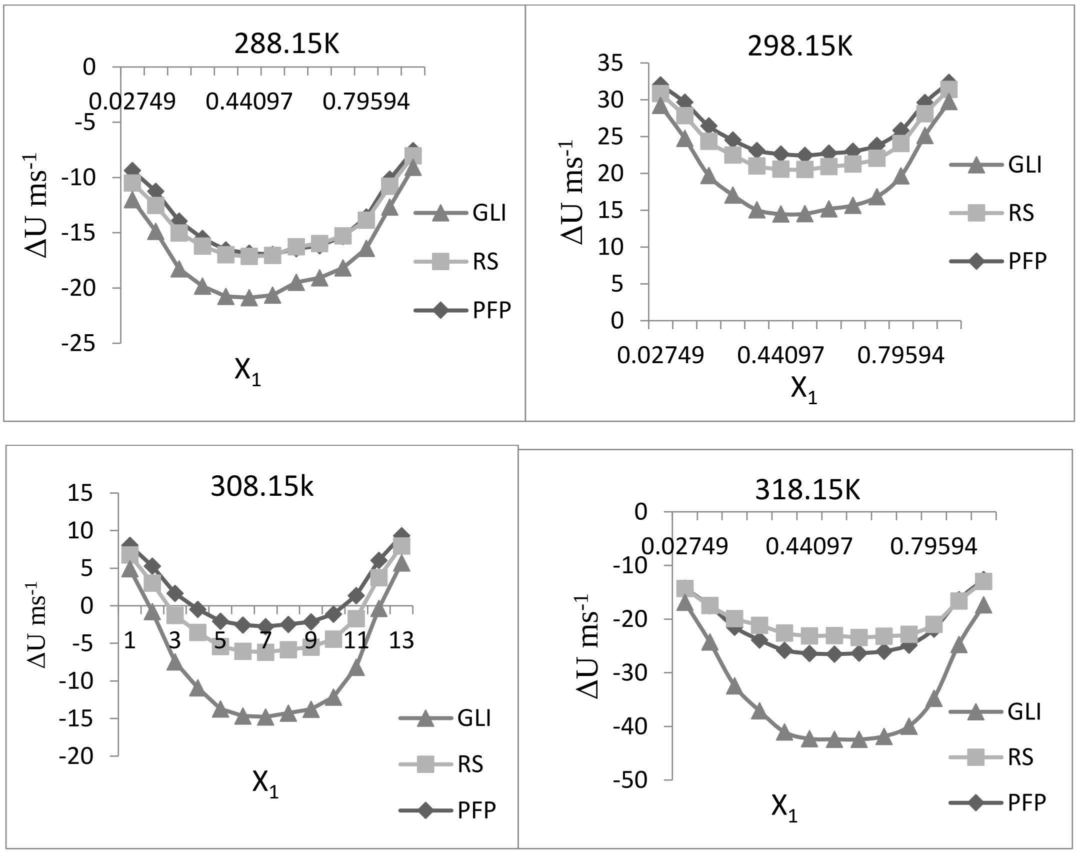

Figure 1: Deviation in ultrasonic speed(ΔU)

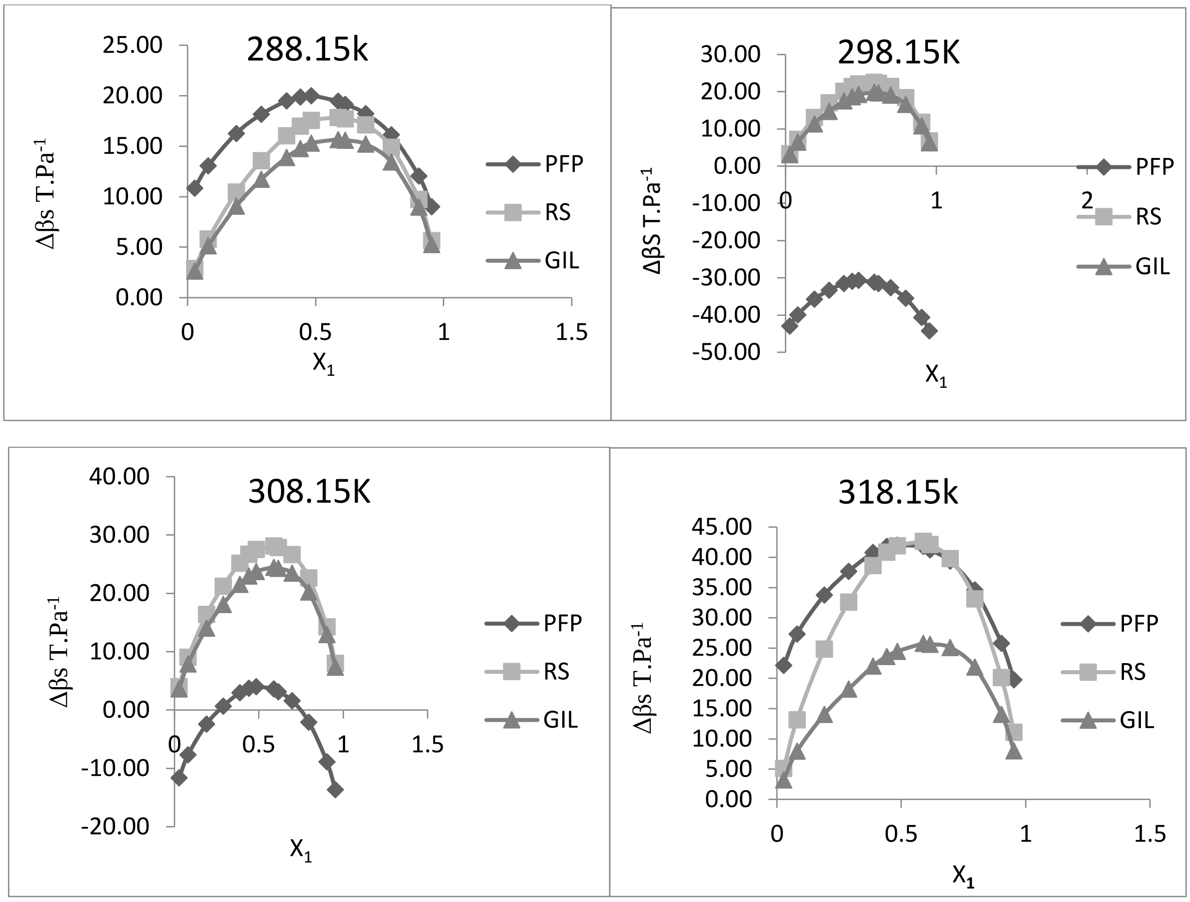

Figure 2: Deviation in isentropic compressibility ($\Delta\beta_S$)

Table 1: Redlich Kester's coefficients and standard deviation(δ) for ultrasonic speed

T/K	A_0	A_1	A_2	A_3	(δ)
288.15	-5.9515	-2.2594	-26.0363	18.0136	0.53
298.15	-14.4089	-3.1439	-26.2173	14.5411	0.45
308.15	-22.8886	-5.2692	-26.9655	13.7852	0.40
318.15	-31.3878	-5.2018	-22.7796	2.6484	0.23

Table 2: Standard deviation and coefficient of McAllister model for ultrasonic speed (U)

	McAllister 3 body(U/ms$^{-1)}$					McAllister 4 body(U/ms$^{-1)}$		
T/K	a	b	(δ)		a	b	c	(δ)
288.15	709.10	969.36	0.74		1334.02	1322.44	1315.44	0.31
298.15	687.34	939.90	0.68		1294.01	1282.65	1275.35	0.30
308.15	665.82	910.90	0.60		1254.29	1243.67	1235.75	0.30
318.15	644.36	882.82	0.54		1215.21	1204.58	1197.64	0.24

Table 3: Standard deviation and coefficient of McAllister model for isentropic compressibility(β_S)

	McAllister 3 body(β_S/TPa^{-1})					McAllister 4 body(β_S/TPa^{-1})		
T/K	a	b	(δ)		a	b	c	(δ)
288.15	435.75	582.16	1.89		823.60	786.15	793.93	0.45
298.15	468.35	625.51	2.08		883.85	844.08	853.08	0.48
308.15	504.10	672.95	2.31		949.99	907.18	917.92	0.47
318.15	543.89	724.17	2.50		1022.72	977.11	987.70	0.45

Table 4: Percentage deviation (%Δ) of ultrasonic speed calculated for different models

X_1	ρ^{mix}	U^{Exp}	U^{Flory}	U^{RS}	U^{GLI}	% ΔF^{lory}	% Δ^{RS}	% Δ^{GLI}
				T=288.15 K				
0.02749	0.75275	1314.74	1324.12	1315.86	1316.24	-0.71	-0.09	-0.11
0.08048	0.75382	1311.88	1323.13	1313.17	1314.22	-0.86	-0.10	-0.18
0.19017	0.7565	1306.67	1320.62	1307.74	1309.92	-1.07	-0.08	-0.25
0.28812	0.75947	1302.34	1317.85	1303.06	1305.94	-1.19	-0.05	-0.28
0.38654	0.76298	1298.07	1314.64	1298.50	1301.83	-1.28	-0.03	-0.29
0.44097	0.76523	1295.76	1312.62	1296.04	1299.50	-1.30	-0.02	-0.29
0.4853	0.76714	1293.98	1310.96	1294.08	1297.57	-1.31	-0.01	-0.28
0.58849	0.77265	1289.78	1306.21	1289.62	1293.00	-1.27	0.01	-0.25
0.61515	0.77427	1288.69	1304.86	1288.50	1291.80	-1.25	0.01	-0.24
0.69605	0.77963	1285.21	1300.56	1285.15	1288.09	-1.19	0.00	-0.22
0.79594	0.78776	1280.87	1294.43	1281.17	1283.42	-1.06	-0.02	-0.20
0.90295	0.79912	1276.41	1286.55	1277.07	1278.28	-0.79	-0.05	-0.15
0.95321	0.8057	1274.76	1282.34	1275.21	1275.82	-0.59	-0.04	-0.08
				T=298.15 K				
0.02749	0.74549	1275.72	1243.70	1276.90	1277.34	2.51	-0.09	-0.13
0.08048	0.74651	1272.6	1242.96	1274.43	1275.65	2.33	-0.14	-0.24
0.19017	0.74912	1267.38	1240.95	1269.47	1272.01	2.09	-0.16	-0.37
0.28812	0.75204	1263.17	1238.65	1265.22	1268.59	1.94	-0.16	-0.43
0.38654	0.75551	1259.03	1235.94	1261.11	1264.99	1.83	-0.17	-0.47
0.44097	0.75773	1256.85	1234.22	1258.91	1262.94	1.80	-0.16	-0.48
0.4853	0.75962	1255.23	1232.81	1257.16	1261.24	1.79	-0.15	-0.48
0.58849	0.76509	1251.4	1228.68	1253.21	1257.15	1.82	-0.14	-0.46
0.61515	0.76672	1250.47	1227.48	1252.22	1256.07	1.84	-0.14	-0.45
0.69605	0.77204	1247.52	1223.72	1249.29	1252.72	1.91	-0.14	-0.42
0.79594	0.7802	1244.06	1218.25	1245.83	1248.45	2.08	-0.14	-0.35
0.90295	0.79157	1240.79	1211.19	1242.31	1243.72	2.39	-0.12	-0.24
0.95321	0.79815	1239.77	1207.41	1240.73	1241.44	2.61	-0.08	-0.13
				T=308.15K				
0.02749	0.73818	1237.32	1229.27	1238.58	1239.15	0.65	-0.10	-0.15
0.08048	0.73913	1234	1228.72	1236.25	1237.81	0.43	-0.18	-0.31
0.19017	0.74167	1228.68	1227.03	1231.60	1234.84	0.13	-0.24	-0.50
0.28812	0.74451	1224.6	1225.05	1227.68	1231.99	-0.04	-0.25	-0.60
0.38654	0.74793	1220.59	1222.63	1223.95	1228.92	-0.17	-0.28	-0.68
0.44097	0.75012	1218.52	1221.09	1221.98	1227.13	-0.21	-0.28	-0.71
0.4853	0.752	1217.02	1219.80	1220.42	1225.64	-0.23	-0.28	-0.71

0.58849	0.7574	1213.58	1216.04	1216.97	1222.01	-0.20	-0.28	-0.69
0.61515	0.75903	1212.77	1214.91	1216.11	1221.04	-0.18	-0.28	-0.68
0.69605	0.76436	1210.28	1211.39	1213.61	1218.00	-0.09	-0.28	-0.64
0.79594	0.7725	1207.63	1206.25	1210.72	1214.08	0.11	-0.26	-0.53
0.90295	0.78388	1205.58	1199.55	1207.87	1209.68	0.50	-0.19	-0.34
0.95321	0.79048	1205.26	1195.93	1206.62	1207.53	0.77	-0.11	-0.19
				T=318.15K				
0.02749	0.7308	1199.74	1213.94	1199.84	1202.29	-1.18	-0.01	-0.21
0.08048	0.73165	1196.12	1213.61	1196.10	1202.89	-1.46	0.00	-0.57
0.19017	0.73412	1190.76	1212.25	1189.18	1203.28	-1.80	0.13	-1.05
0.28812	0.73686	1186.7	1210.60	1183.92	1202.68	-2.01	0.23	-1.35
0.38654	0.7402	1182.73	1208.52	1179.53	1201.17	-2.18	0.27	-1.56
0.44097	0.74236	1180.76	1207.15	1177.48	1199.96	-2.23	0.28	-1.63
0.4853	0.74423	1179.44	1205.99	1176.01	1198.76	-2.25	0.29	-1.64
0.58849	0.74954	1176.25	1202.61	1173.29	1195.28	-2.24	0.25	-1.62
0.61515	0.75118	1175.56	1201.54	1172.75	1194.23	-2.21	0.24	-1.59
0.69605	0.75646	1173.49	1198.32	1171.49	1190.64	-2.12	0.17	-1.46
0.79594	0.76459	1171.64	1193.51	1170.77	1185.42	-1.87	0.07	-1.18
0.90295	0.77598	1170.77	1187.16	1171.00	1178.87	-1.40	-0.02	-0.69
0.95321	0.7826	1171.1	1183.71	1171.47	1175.47	-1.08	-0.03	-0.37

Table 5: Percentage deviation (%Δ) of isentropic compressibility calculated for different models

X_1	βs^{Exp}	βF^{lory}	βs^{RS}	βs^{GLI}	% ΔF^{lory}	% Δ^{RS}	% Δ^{GLI}
			T=288.15K				
0.02749	768.55	757.69	765.65	765.89	1.41	0.38	0.35
0.08048	770.80	757.75	765.01	765.68	1.69	0.75	0.67
0.19017	774.21	757.94	763.76	765.15	2.10	1.35	1.17
0.28812	776.32	758.16	762.74	764.60	2.34	1.75	1.51
0.38654	777.84	758.36	761.81	763.95	2.50	2.06	1.79
0.44097	778.32	758.46	761.33	763.55	2.55	2.18	1.90
0.4835	778.52	758.51	760.95	763.21	2.57	2.26	1.97
0.58849	778.01	758.56	760.16	762.34	2.50	2.29	2.01
0.61515	777.70	758.54	759.97	762.10	2.46	2.28	2.01
0.69605	776.54	758.32	759.43	761.34	2.35	2.20	1.96
0.79594	773.74	757.61	758.86	760.31	2.08	1.92	1.74
0.90295	768.08	756.02	758.34	759.12	1.57	1.27	1.17
0.95321	763.78	754.78	758.13	758.53	1.18	0.74	0.69
			T=298.15K				
0.02749	824.23	867.21	820.87	821.16	-5.21	0.41	0.37
0.08048	827.14	867.06	819.89	820.71	-4.83	0.88	0.78
0.19017	831.06	866.84	817.96	819.68	-4.30	1.58	1.37
0.28812	833.37	866.69	816.36	818.65	-4.00	2.04	1.77
0.38654	835.00	866.50	814.86	817.50	-3.77	2.41	2.10
0.44097	835.45	866.36	814.07	816.82	-3.70	2.56	2.23
0.4835	835.52	866.23	813.46	816.24	-3.68	2.64	2.31
0.58849	834.63	865.78	812.12	814.81	-3.73	2.70	2.38
0.61515	834.10	865.64	811.79	814.42	-3.78	2.67	2.36

0.69605	832.27	864.96	810.85	813.19	-3.93	2.57	2.29
0.79594	828.15	863.62	809.79	811.58	-4.28	2.22	2.00
0.90295	820.57	861.17	808.77	809.74	-4.95	1.44	1.32
0.95321	815.14	859.43	808.34	808.84	-5.43	0.83	0.77
			T=308.15K				
0.02749	884.86	896.48	880.83	881.24	-1.31	0.46	0.41
0.08048	888.48	896.14	879.44	880.57	-0.86	1.02	0.89
0.19017	893.12	895.53	876.70	879.05	-0.27	1.84	1.58
0.28812	895.66	895.00	874.41	877.53	0.07	2.37	2.02
0.38654	897.43	894.43	872.26	875.86	0.33	2.80	2.40
0.44097	897.85	894.08	871.14	874.87	0.42	2.98	2.56
0.4835	897.81	893.77	870.25	874.04	0.45	3.07	2.65
0.58849	896.47	892.86	868.32	871.98	0.40	3.14	2.73
0.61515	895.74	892.60	867.85	871.43	0.35	3.11	2.71
0.69605	893.16	891.53	866.48	869.67	0.18	2.99	2.63
0.79594	887.63	889.67	864.94	867.38	-0.23	2.56	2.28
0.90295	877.72	886.58	863.46	864.77	-1.01	1.63	1.48
0.95321	870.86	884.50	862.82	863.49	-1.57	0.92	0.85
			T=318.15K				
0.02749	950.66	928.56	945.56	947.44	2.33	0.54	0.34
0.08048	955.32	927.98	942.15	947.35	2.86	1.38	0.83
0.19017	960.69	926.94	935.86	946.66	3.51	2.59	1.46
0.28812	963.68	926.00	931.11	945.47	3.91	3.38	1.89
0.38654	965.78	925.00	927.16	943.74	4.22	4.00	2.28
0.44097	966.19	924.41	925.34	942.55	4.32	4.23	2.45
0.4835	965.92	923.92	924.04	941.47	4.35	4.34	2.53
0.58849	964.29	922.48	921.67	938.52	4.34	4.42	2.67
0.61515	963.31	922.10	921.20	937.67	4.28	4.37	2.66
0.69605	959.96	920.60	920.16	934.85	4.10	4.15	2.62
0.79594	952.76	918.17	919.66	930.90	3.63	3.47	2.29
0.90295	940.17	914.38	920.06	926.11	2.74	2.14	1.50
0.95321	931.69	911.94	920.59	923.66	2.12	1.19	0.86

Standard deviation

Standard deviation (δ) of acoustical parameters are calculated by:

$$\delta = \sqrt{\sum_{i=1}^{k} \frac{(\Delta P)^2}{(EP - AP)}} \qquad (11)$$

EP and AP are no of observational point and adjustable coefficient respectively. ΔP represent difference between experimental and calculated acoustical parameters.

CONCLUSION

Ultrasonic speed and isentropic compressibility are two important parameters which provide a way to understand the molecular interactions and internal structure of associates. In above discussion it can be concluded that the calculated values of ultrasonic speed decreases with increase in temperatures while isentropic compressibility increases with increase in temperatures which indicate the distance between the surface of neighbouring molecules increases confirm the weak interaction between the binary components. Ramaswamy model(associated) gave excellent result than Glinski and Flory model (non- associated).

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