



## Evaluation of Ultrasonic Velocity Theories on Binary Liquid Mixtures of Propiophenone with Isomeric Xylenes at Temperatures ( $T = 303.15$ to $318.15$ K)

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Densities ( $\rho$ ), ultrasonic speeds of sound ( $u$ ) of binary mixtures containing propiophenone with *o*-xylene, *m*-xylene and *p*-xylene were measured over the entire composition range at temperatures from 303.15-318.15 K and at atmospheric pressure 0.1 MPa. Experimental data of ultrasonic velocity was used to compute the theoretical velocities by using the various theories like Nomoto's relation ( $U_{\text{NOM}}$ ), impedance relation ( $U_{\text{IMP}}$ ), Van Dael and Vangeel's relation ( $U_{\text{VDV}}$ ), Rao's specific velocity relation ( $U_{\text{RAO}}$ ), Jouyban-Acree's ( $U_{\text{JOE}}$ ) and Junjie's theory ( $U_{\text{JUN}}$ ). The results are in good agreement with the experimental data. The relative percentage error, chi square test for goodness of fit and the molecular interaction parameter ( $\alpha$ ) values for non-ideality in the binary mixtures were computed and analyzed in terms of intermolecular interactions between the molecules of the binary mixtures.

**Keywords:** Ultrasonic speed of sound, Molecular interaction parameter, Velocity theories, Chi square test, Relative percentage error.

### INTRODUCTION

Ultrasonic velocity of the binary liquid mixtures is the vital tool to measure the mechanical stability of liquids and to understand the molecular interactions, physico-chemical behaviour in pure, binary and higher order multi component liquid mixtures [1]. The ultrasonic velocity ( $u$ , m/s) in a liquid or a liquid mixture is mainly determined by its intermolecular properties [2]. It is clear that the measurement of speed of sound can supplement and replace measurements on other physical properties in studies of molecular structure [3]. Theoretical evaluation of ultrasonic velocity values of binary mixtures with the experimental data indicates the behaviour of molecular interactions and it is used to develop theoretical models for liquids. Literature survey reveals that some studies were reported by other researchers on the measurement of ultrasonic speeds, densities, viscosities, relative permittivity, etc., of binary mixtures containing xylenes with other components at different temperatures [4-11]. No study has been reported on the theoretical evaluation of ultrasonic velocity by using velocity theories such as Nomoto's relation, Van Dael and Vangeel's relation,

Junjie's relation, Impedance dependence relation, Jouyban-Acree relation and Rao's specific velocity relation for the propiophenone with isomeric xylene (*ortho*, *meta* and *para*) binary mixtures at temperatures from 303.15 to 318.15 K. The results were discussed further in terms of molecular interaction parameter ( $\alpha$ ) values for the non-ideality in the liquid mixture, relative deviation percentage error with respect to experimental data and chi square ( $\chi^2$ ) test for the goodness of fit. Propiophenone is widely used component in perfumes industries [11]. Xylenes with other components used as chemical intermediates. Xylenes are also used as solvent in leather, rubber, dyes, pesticides, printing industries, etc. [12]. The current study compared the experimental values of ultrasonic velocities with the calculated values from the different velocity theories and studied further on the deviations in molecular interaction parameter ( $\alpha$ ) values, relative percentage error and chi square test values for the goodness of fit were analyzed.

### EXPERIMENTAL

Propiophenone is taken as a primary component while *o*-xylene, *m*-xylene and *p*-xylene are taken as secondary compo-

nents. These chemicals purchased from S.D. Fine chemicals Ltd, India. Purification of the solvents was done by using the standard methods [13,14] and their purities were as follows, propiophenone 98%, *o*-xylene 99.7%, *m*-xylene 99.5% and *p*-xylene 99.5%. Mettler Toledo (ME204) balance with the precision of  $\pm 0.1$  mg was used to prepare the mixtures by mixing weighed amounts of the pure liquids adopting the method of closed system. Anton Paar (DSA 5000 M) oscillating u-tube densimeter having automatically thermostatic within  $\pm 0.01$  K was used to calculate the densities and speed of sounds of the pure components and the binary mixtures over the whole composition range at ( $T = 303.15$  to  $318.15$  K). The standard uncertainties associated with the measurement of density and speeds of sound are  $\pm 0.001$  kg m<sup>-3</sup>,  $\pm 0.01$  m s<sup>-1</sup>, respectively.

**Theory:** The theoretical values of ultrasonic velocities of the binary mixtures Propiophenone with xylenes at  $T = 303.15$  to  $318.15$  K were calculated by using the velocity theories like Nomoto's relation, Vandael and Vangeel's relation, Impedance relation, Junjie's relation, Jouyban-Acree's relation and Rao's specific velocity relations. The relations are as shown below.

**Nomoto's relation [15]:** Nomoto's empirical formula for the sound velocity  $U$  is given by

$$U_{\text{Nomoto}} = \left( \frac{x_1 R_1 + x_2 R_2}{x_1 V_1 + x_2 V_2} \right)^3 \quad (1)$$

where  $R_1 = \frac{(M_1 u_1^{1/3})}{\rho_1}$ ;  $R_2 = \frac{(M_2 u_2^{1/3})}{\rho_2}$ ;  $V_1 = \frac{M_1}{\rho_1}$ ;  $V_2 = \frac{M_2}{\rho_2}$ .

In above equations  $x_1$  and  $x_2$  are the mole fraction of component liquids.  $M_1, M_2, u_1, u_2, V_1$  and  $V_2$  are the molecular weights, ultrasonic velocity and molar volumes of components of binary liquids mixtures.

**Van Dael and Vangeel's relation (ideal mixing relation) [16,17]:**

$$U_{\text{VDV}} = \left[ \left( \frac{x_1}{M_1 U_1^2} + \frac{x_2}{M_2 U_2^2} \right) (x_1 M_1 + x_2 M_2) \right]^{-1/2} \quad (2)$$

where  $M_1, M_2$  and  $U_1, U_2$  are molecular weights and ultrasonic velocities of the compounds **1** and **2**, respectively.

**Impedance relation [18]:**

$$U_{\text{IMP}} = \left( \frac{\sum x_i Z_i}{\sum x_i \rho_i} \right) \quad (3)$$

where  $x_i$  is the mole fraction,  $\rho_i$  the density of the mixture and  $Z_i$  is the acoustic impedance.

**Rao's specific velocity relation [19]:** Using the ratio of the temperature coefficient of velocity and expansion coefficient, Rao derived a formula for ultrasonic velocity ( $U$ ):

$$U_{\text{RAO}} = \left( \sum (x_i r_i \rho_i) \right)^3 \quad (4)$$

where 'r' is Rao's constant or molar sound velocity, which is constant for a liquid at a temperature.

**Junjie's relation [20]:** This relation derived by Junjie's for the ultrasonic velocity of the mixture in terms of the mole fraction, molecular weight and density of the mixture.

$$U_{\text{JUN}} = \frac{\sum_{i=1}^n x_i V_i}{\left( \sum_{i=1}^n x_i M_i \right)^{1/2} \times \left( \sum_{i=1}^n \frac{x_i V_i}{\rho_i U_i^2} \right)^{1/2}} \quad (5)$$

where the symbols have their usual meaning.

**Jouyban-Acree equation for velocity [21]:**

$$\ln u = x_1 \ln u_1 + x_2 \ln u_2 + A_0 \left( \frac{x_1 x_2}{T} \right) + A_1 \left( \frac{x_1 x_2 (x_1 - x_2)}{T} \right) + A_2 \left( \frac{x_1 x_2 (x_1 - x_2)^2}{T} \right) \quad (6)$$

where  $u$  is the ultrasonic velocity of the binary liquid mixture and  $x_1, u_1, x_2, u_2$  are the mole fractions and velocities of components **1** and **2**, respectively.  $A_0, A_1$  and  $A_2$  are the adjustable interaction parameters.

**Chi square test for goodness of fit:** According to Karl Pearson, chi square value is evaluated for the binary liquid mixtures under study using the formula

$$\chi^2 = \sum_{i=1}^n \left( \frac{U_{\text{obs}} - U_{\text{cal}}}{U_{\text{cal}}} \right)^2 \quad (7)$$

where 'n' is the number of data used,  $U_{\text{obs}}$  = experimental values of ultrasonic velocities,  $U_{\text{cal}}$  = computed values of ultrasonic velocities,  $\chi^2$  = chi square.

**Relative percentage error:**

$$\sigma = \left( \frac{1}{n} \right) \times \sum \left( \frac{U_{\text{obs}} - U_{\text{cal}}}{U_{\text{obs}}} \right) \times 100\% \quad (8)$$

where 'n' is the number of data used,  $U_{\text{obs}}$  = experimental values of ultrasonic velocities,  $U_{\text{cal}}$  = computed values of ultrasonic velocities.

**Molecular interaction parameter:**

$$\infty = \left[ \left( \frac{U_{\text{obs}}}{U_{\text{cal}}} \right)^2 - 1 \right] \quad (9)$$

$U_{\text{obs}}$  = experimental values of ultrasonic velocities,  $U_{\text{cal}}$  = computed values of ultrasonic velocities.

## RESULTS AND DISCUSSION

Evaluation of ultrasonic studies for the binary mixtures of propiophenone with xylenes by using velocity theories are very useful to understand the intermolecular interactions, which takes place in the binary mixtures.

The experimental 'U' values and the calculated ultrasonic velocity values by using the various velocity theories for the studied three binary mixtures over the entire mole fraction range at temperatures ( $T = 303.15$  to  $318.15$  K) are presented in Tables 1-3. The relative percentage errors for all the mathematical velocity models, chi square values and average interaction parameter values of the studied binary mixtures at temperatures from 303.15 to 318.15 K are tabulated in Table-4.

Figs. 1-3 shows the relative percentage deviations in the ultrasonic velocities with respect to experimental data are positive in  $U_{\text{NOM}}$ ,  $U_{\text{VDV}}$ ,  $U_{\text{JUN}}$ ,  $U_{\text{JOU}}$  and  $U_{\text{RAO}}$  and negative in  $U_{\text{IMP}}$



TABLE-3  
EXPERIMENTAL AND THEORETICAL ULTRASONIC VELOCITY VALUES OF PPH + *p*-XYLENE  
BINARY MIXTURE OVER THE ENTIRE MOLE FRACTION RANGE AT 303.15-318.15 K

$x_1$	$U_{EXP}$ ( $m s^{-1}$ )	$U_{NOM}$ ( $m s^{-1}$ )	$U_{UDV}$ ( $m s^{-1}$ )	$U_{IMP}$ ( $m s^{-1}$ )	$U_{JUN}$ ( $m s^{-1}$ )	$U_{RAO}$ ( $m s^{-1}$ )	$U_{JOU}$ ( $m s^{-1}$ )	$U_{EXP}$ ( $m s^{-1}$ )	$U_{NOM}$ ( $m s^{-1}$ )	$U_{UDV}$ ( $m s^{-1}$ )	$U_{IMP}$ ( $m s^{-1}$ )	$U_{JUN}$ ( $m s^{-1}$ )	$U_{RAO}$ ( $m s^{-1}$ )	$U_{JOU}$ ( $m s^{-1}$ )
303.15 K								308.15 K						
0.0000	1289.49	1289.49	1289.49	1289.49	1289.49	1289.49	1289.49	1269.05	1269.05	1269.05	1269.05	1269.05	1269.05	1269.05
0.0934	1305.05	1304.51	1296.56	1306.54	1299.25	1303.64	1305.04	1282.98	1281.99	1274.92	1283.73	1277.36	1281.26	1282.97
0.1882	1320.60	1319.68	1305.02	1323.27	1310.05	1318.11	1320.61	1296.59	1295.06	1282.05	1298.13	1286.61	1293.74	1296.59
0.2844	1336.20	1334.99	1315.01	1339.69	1321.98	1332.90	1336.19	1310.04	1308.25	1290.55	1312.25	1296.85	1306.49	1310.05
0.3820	1351.77	1350.45	1326.68	1355.78	1335.11	1348.02	1351.78	1323.44	1321.55	1300.55	1326.09	1308.15	1319.51	1323.43
0.4811	1367.41	1366.06	1340.23	1371.59	1349.56	1363.49	1367.41	1336.84	1334.98	1312.23	1339.68	1320.60	1332.81	1336.85
0.5817	1383.10	1381.82	1355.89	1387.10	1365.44	1379.31	1383.09	1350.33	1348.52	1325.76	1353.01	1334.29	1346.41	1350.33
0.6839	1398.84	1397.74	1373.96	1402.34	1382.89	1395.51	1398.83	1363.88	1362.20	1341.39	1366.10	1349.35	1360.32	1363.88
0.7876	1414.62	1413.80	1394.75	1417.29	1402.07	1412.07	1414.62	1377.43	1375.98	1359.38	1378.95	1365.86	1374.53	1377.43
0.8930	1430.47	1430.03	1418.73	1431.98	1423.17	1429.04	1430.48	1390.86	1389.91	1380.10	1391.56	1384.01	1389.08	1390.86
1.0000	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41	1446.41	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95	1403.95
313.15 K								318.15 K						
0.0000	1278.80	1278.80	1278.80	1278.80	1278.80	1278.80	1278.80	1259.22	1259.22	1259.22	1259.22	1259.22	1259.22	1259.22
0.0934	1293.77	1293.00	1285.41	1294.92	1287.99	1292.19	1293.74	1271.98	1270.84	1264.29	1272.37	1266.60	1270.19	1271.98
0.1882	1308.53	1307.34	1293.35	1310.74	1298.18	1305.88	1308.55	1284.33	1282.57	1270.54	1285.26	1274.85	1281.39	1284.33
0.2844	1323.29	1321.82	1302.76	1326.25	1309.44	1319.86	1323.29	1296.45	1294.39	1278.07	1297.91	1284.01	1292.82	1296.44
0.3820	1338.01	1336.42	1313.77	1341.46	1321.85	1334.15	1338.02	1308.44	1306.31	1286.99	1310.31	1294.13	1304.49	1308.44
0.4811	1352.78	1351.17	1326.59	1356.39	1335.51	1348.77	1352.77	1320.45	1318.33	1297.44	1322.47	1305.30	1316.41	1320.45
0.5817	1367.60	1366.06	1341.41	1371.04	1350.52	1363.71	1367.59	1332.52	1330.45	1309.60	1334.41	1317.59	1328.58	1332.52
0.6839	1382.46	1381.09	1358.51	1385.43	1367.03	1379.01	1382.46	1344.65	1342.69	1323.67	1346.13	1331.09	1341.02	1344.65
0.7876	1397.37	1396.26	1378.20	1399.55	1385.16	1394.64	1397.37	1356.73	1355.01	1339.88	1357.62	1345.90	1353.73	1356.74
0.8930	1412.26	1411.58	1400.88	1413.42	1405.10	1410.66	1412.26	1368.63	1367.46	1358.54	1368.92	1362.17	1366.72	1368.63
1.0000	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04	1427.04	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00	1380.00

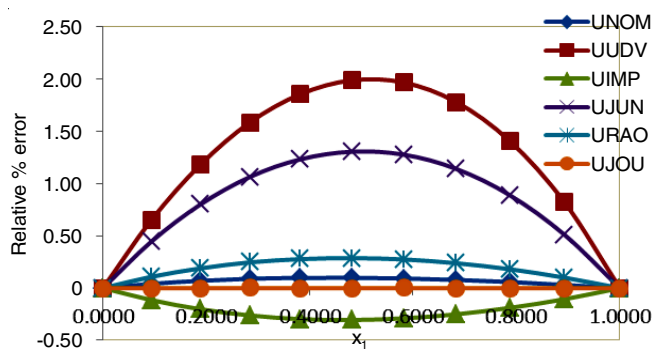


Fig. 3. Relative percentage deviations of  $U$  values at the whole mole fraction range for the binary mixture propiophenone + *p*-xylene at 303.15 K

relations at 303.15 K for the studied binary mixtures (similar trend is observed at all remaining temperatures). Percentage deviation values of  $U_{JOU}$  followed by  $U_{NOM}$ ,  $U_{IMP}$  and  $U_{RAO}$  values are in good agreement with the experimental values, whereas  $U_{VDV}$  and  $U_{JUN}$  values are deviating away from the experimental values. It indicates the non-ideal behaviour in the liquid mixtures and it is also supported by the Interaction parameter ( $\alpha$ ) whose values are having the parabolic trend which indicates the non-ideality. Positive values of average ' $\alpha$ ' are observed for all the theories except Impedance relation over the entire composition range at all four temperatures and they are decreasing with increase in temperature. Similarly, the ultrasonic velocity values are also decreasing at higher temperatures, hence, the rate of association of unlike molecules increases due to the activation of the molecules by thermal energy.

Figs. 4-6 show the average molecular interaction parameter ( $\alpha_{avg}$ ) values of the studied three binary mixtures at 303.15-318.15 K and it supports the above that  $\alpha_{JOU}$  followed by  $\alpha_{NOM}$ ,  $\alpha_{IMP}$  and  $\alpha_{RAO}$  are in good agreement with the

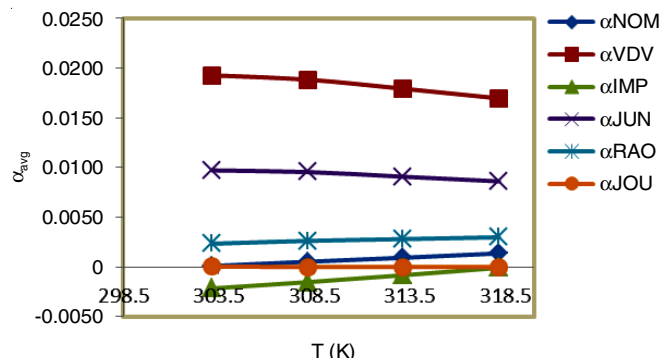


Fig. 4. Average interaction parameter ' $\alpha_{avg}$ ' values of propiophenone + *o*-xylene binary mixture at 303.15 to 318.15 K

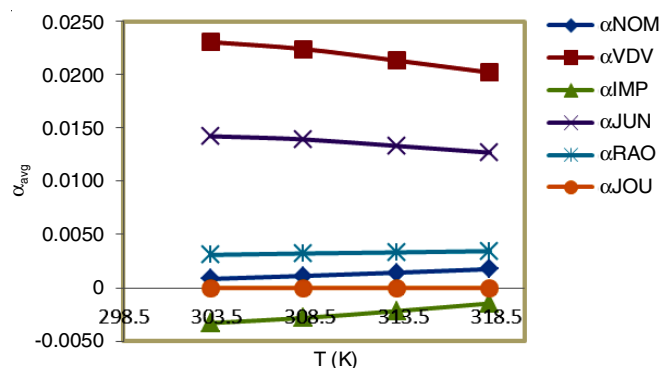


Fig. 5. Average interaction parameter ' $\alpha_{avg}$ ' values of propiophenone + *m*-xylene binary mixture at 303.15 to 318.15 K

experimental data. The algebraic order of ' $\alpha_{avg}$ ' values is  $\alpha_{JOU} < \alpha_{NOM} < \alpha_{IMP} < \alpha_{RAO} < \alpha_{JUN} < \alpha_{VDV}$  for all the three binary mixtures.

Validity of the applied theories is checked by using chi square test for the goodness of fit. If the chi square values are closer to zero, then there is no significant difference between

TABLE-4  
RELATIVE PERCENTAGE ERROR AND CHI SQUARE VALUES OF BINARY MIXTURES  
PROPIOPHENONE WITH *o*-XYLENE, *m*-XYLENE AND *p*-XYLENE AT 303.15-318.15 K

T (K)	Average relative deviation error (%)						Chi square values ( $\chi^2$ )						Interaction parameter ( $\alpha_{avg}$ )					
	$U_{NOM}$	$U_{VDV}$	$U_{IMP}$	$U_{JUN}$	$U_{RAO}$	$U_{JOU}$	$U_{NOM}$	$U_{VDV}$	$U_{IMP}$	$U_{JUN}$	$U_{RAO}$	$U_{JOU}$	$\alpha_{NOM}$	$\alpha_{VDV}$	$\alpha_{IMP}$	$\alpha_{JUN}$	$\alpha_{RAO}$	$\alpha_{JOU}$
PPH + <i>o</i> -xylene																		
318.15	0.0676	0.8336	-0.0047	0.4266	0.1484	-0.0001	0.0086	1.3888	0.0003	0.3596	0.0426	0.0000	0.0014	0.0170	-0.0001	0.0086	0.0030	0.0000
313.15	0.0459	0.8800	-0.0418	0.4492	0.1393	0.0000	0.0040	1.5713	0.0038	0.4052	0.0383	0.0000	0.0009	0.0179	-0.0008	0.0091	0.0028	0.0000
308.15	0.0261	0.9237	-0.0767	0.4723	0.1305	0.0000	0.0013	1.7577	0.0121	0.4556	0.0345	0.0000	0.0005	0.0188	-0.0015	0.0095	0.0026	0.0000
303.15	0.0049	0.9445	-0.1084	0.4805	0.1162	0.0009	0.0001	1.8646	0.0236	0.4800	0.0284	0.0000	0.0001	0.0193	-0.0022	0.0097	0.0023	0.0000
PPH + <i>m</i> -xylene																		
318.15	0.0879	0.9907	-0.0730	0.6269	0.1720	0.0000	0.0145	1.9469	0.0112	0.7728	0.0568	0.0000	0.0018	0.0202	-0.0015	0.0127	0.0035	0.0000
313.15	0.0561	1.0446	-0.1088	0.6576	0.1661	0.0000	0.0098	2.1986	0.0240	0.8645	0.0542	0.0000	0.0014	0.0213	-0.0022	0.0133	0.0033	0.0000
308.15	0.0420	1.0960	-0.1414	0.6872	0.1610	0.0000	0.0063	2.4572	0.0402	0.9592	0.0522	0.0000	0.0011	0.0224	-0.0028	0.0139	0.0032	0.0000
303.15	0.0420	1.1273	-0.1654	0.7015	0.1544	0.0000	0.0037	2.6330	0.0550	1.0130	0.0490	0.0000	0.0008	0.0231	-0.0033	0.0142	0.0031	0.0000
PPH + <i>p</i> -xylene																		
318.15	0.1112	1.0669	-0.0772	0.7055	0.1985	0.0000	0.0229	2.2466	0.0131	0.9731	0.0747	0.0000	0.0022	0.0218	-0.0015	0.0143	0.0040	0.0000
313.15	0.0950	1.1238	-0.1163	0.7428	0.1920	0.0000	0.0170	2.5313	0.0282	1.0961	0.0712	0.0000	0.0019	0.0230	-0.0023	0.0151	0.0039	0.0000
308.15	0.0761	1.1754	-0.1555	0.7740	0.1828	0.0000	0.0113	2.8128	0.0491	1.2101	0.0662	0.0000	0.0015	0.0241	-0.0031	0.0157	0.0037	0.0000
303.15	0.0597	1.2029	-0.1831	0.7873	0.1728	0.0000	0.0072	2.9859	0.0677	1.2701	0.0605	0.0000	0.0012	0.0247	-0.0036	0.0160	0.0035	0.0000

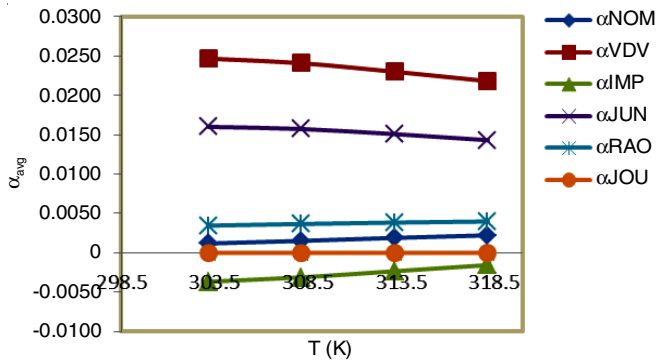


Fig. 6. Average interaction parameter ' $\alpha_{avg}$ ' values of propiophenone + *p*-xylene binary mixture at 303.15 to 318.15 K

the experimental and the calculated values, therefore those binary mixtures are in agreement with goodness of fit (null hypothesis), if the chi square values are deviating away from

zero it comes under alternative hypothesis. The chi square values of  $U_{JOU}$  followed by  $U_{NOM}$ ,  $U_{IMP}$  and  $U_{RAO}$  are closer to zero and are in good agreement under the studied binary mixtures. Overall, Jouyban-Acree's relation values are in best agreement with the experimental values. Chi square values are in the order of PPH + *p*-xylene > PPH + *m*-xylene > PPH + *o*-xylene.

**Conclusion**

In this paper, the evaluation of ultrasonic velocity studies in binary liquid mixture containing propiophenone and isomeric xylenes using Nomoto ( $U_{NOM}$ ), impedance ( $U_{IMP}$ ), Van-Dael and Vangeel ( $U_{VDV}$ ), Junjie ( $U_{JUN}$ ), Jouyban-Acree's relation ( $U_{JOU}$ ) and Rao's specific velocity ( $U_{RAO}$ ) models were done and they are compared with the experimental data of ultrasonic values at 303.15 to 318.15 K. From the results, the U values of Jouyban-Acree followed by Nomoto's relation, Impedance relation and Rao's specific velocity relations are in good

agreement with the experimental data. Larger deviations in the  $U_{VDV}$  and  $U_{JUN}$  relations indicate the molecular interactions between the component molecules. The relative percentage error of ultrasonic velocity is attributed to the presence of molecular interactions in the studied system. Further, the positive values of molecular interaction parameter ( $\alpha$ ) in the present systems at all the temperatures except for  $\alpha_{VDV}$  clearly indicate the existence of good association and show the strength of interaction between the component molecules of the liquid mixture and the non-ideal behaviour. Chi square test for the goodness of fit indicates  $U_{JOU}$ ,  $U_{NOM}$ ,  $U_{IMP}$  and  $U_{RAO}$  relations are in good agreement.

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#### CONFLICT OF INTEREST

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

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