Experimental and Theoretical Studies of Ultrasonic Velocity in Ternary Liquid Mixtures of Methyl Benzoate, Cyclohexane and Alcohols at Different Temperatures

UMADEVI MUTHUKRISHNAN^{1,*}, KESAVASAMY RAMASAMY², PALANI RATHINASAMY³ and PRIYA NACHIMUTHU SENNIAPPAN²

¹Department of Physics, SVS College of Engineering, Coimbatore-642 109, India

²Department of Physics, Sri Ramakrishna Engineering College, Coimbatore-641 022, India

³Department of Physics, DDE, Annamalai University, Chidambaram-608 002, India

*Corresponding author: Fax: +91 422 2321332; E-mail: devuma55@gmail.com

Received: 21 October 2013;

Accepted: 16 April 2014;

Published online: 28 July 2014;

AJC-15649

Ultrasonic velocities of ternary liquid mixtures of methyl benzoate and cyclohexane with 1-propanol, 1-butanol, 1-pentanol and 1-hexanol are measured using ultrasonic interferometer at 303, 308 and 313 K over the entire range of composition. Theoretical values of ultrasonic velocity have been evaluated at the three temperatures using Nomoto's relation, ideal mixture relation, impedance relation, Junjie's method, Rao's specific velocity relation and free length theory. Theoretical values are compared with the experimental values and $U_{\text{exp}}^2/U_{\text{imx}}^2$ is evaluated for non-ideality in the mixtures. A good agreement has been found between experimental and theoretical values of ultrasonic velocity. The relative applicability of these theories to the present systems has been checked and discussed. The results are explained in terms of molecular interactions occurring in these ternary liquid mixtures.

Keywords: Ultrasonic velocity, Methyl benzoate, Cyclohexane, Molecular interactions, Theoretical models.

INTRODUCTION

With the growth of scientific knowledge, it became increasingly necessary to specialize in the theoretical and experimental investigation by putting much effort. The material structures and contents may be achieved only if the scientist knows about theoretical background and technological aspect of the materials to be studied. Hence it is decided to evaluate the theoretical parameters of the selected components, when it is exposed to ultrasonic waves¹.

Among the non-spectroscopic methods in the study of molecular interactions, the ultrasonic velocity measurement finds extensive applications owing to its ability of characterizing the physico-chemical behaviour of liquid systems from speed data. The ultrasonic velocity of liquid is fundamentally related to the binding forces between the atoms or the molecules. According to physical concept of liquid model, molecules in the liquid state are so loosely packed as to leave some free space in between them. The intermolecular free space and its dependent properties are related to intermolecular interactions and may reveal the some information regarding the interaction, which may be occurring when the liquids are mixed together²⁻⁴.

Several researchers³⁻¹⁰ carried out ultrasonic investigations on binary and ternary liquid mixtures and compared the experimental results with the theoretical relations of Nomoto¹¹, Van

Dael and Vangeel¹², impedance dependence¹³, Rao's specific velocity¹⁴ and Junjie's¹⁵ equations and the results are explained in terms of molecular interactions. An attempt has been made to compare the merits of relations for the ternary liquid mixtures investigated at different temperatures. The relative applicability of these theories to the present systems have been checked and discussed. Based on the deviation in $U_{\text{exp}}^2/U_{\text{imx}}^2$ and degree of intermolecular interaction or molecular association (α) the molecular interactions are studied. Theoretical values of ultrasonic velocities in ternary liquid mixtures were calculated using different theories and empirical relations. Comparison of theoretical values of ultrasonic velocities with those obtained experimentally in the present ternary liquid mixtures is expected to reveal the nature of interaction between the component molecules in the mixture. Such theoretical study is useful in building the comprehensive theoretical model for the liquid mixtures.

EXPERIMENTAL

All the chemicals used in the present work are AnalaR grade. The purity of the chemicals was ascertained by comparing their density, viscosity and ultrasonic velocity at 303, 308 and 313 K which agrees with the corresponding literature values. The mixtures of methyl benzoate + cyclohexane + 1-propanol (system 1), methyl benzoate + cyclohexane + 1-butanol

(system 2), methyl benzoate + cyclohexane + 1-pentanol (system 3) and methyl benzoate + cyclohexane + 1-hexanol (system 4) were prepared by weight. The mole fraction of the second component, cyclohexane ($x_2 = 0.4$) was kept constant, while the mole fractions of the remaining two (x_1, x_3) were varied from 0.1 to 0.6. The ultrasonic velocity was measured by a single crystal interferometer with a high degree of accuracy operating at a frequency of 3 MHz (model F-05, with digital micrometer) at 303, 308 and 313 K. The viscosity was measured by Ostwald's viscometer. An electronically operated constant temperature water bath is used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature. Densities of the mixtures have been found by relative measurement method.

Theory

The following theories/relations are used for the prediction of ultrasonic velocity in the ternary liquid mixtures.

Nomoto's relation (U_{NR})¹¹

$$U_{NR} = \left[\frac{X_1 R_1 + X_2 R_2 + X_3 R_3}{X_1 V_1 + X_2 V_2 + X_3 V_3} \right]^3 \quad (1)$$

where R_i is molar sound velocity and V_i is molar volume. X_i is the mole fraction.

Molar sound velocity

$$R_1 = \frac{m_1}{d_1} U_1^{1/3}; R_2 = \frac{m_2}{d_2} U_2^{1/3}; R_3 = \frac{m_3}{d_3} U_3^{1/3}$$

Molar volume

$$V_1 = \frac{m_1}{d_1}; V_2 = \frac{m_2}{d_2}; V_3 = \frac{m_3}{d_3}$$

m_1, m_2 and m_3 are the molecular weight and d_1, d_2 and d_3 are the density of the component liquid. U_1, U_2 and U_3 are the ultrasonic velocity of the pure liquids.

Van Dael relation (ideal mixtures relation): Van Dael and Vangeel¹² suggested the following relation for the measurement of velocity of sound

$$\frac{1}{X_1 M_1 + X_2 M_2 + X_3 M_3} \cdot \frac{1}{U_{id,mix}^2} = \frac{X_1}{M_1 U_1} + \frac{X_2}{M_2 U_2} + \frac{X_3}{M_3 U_3} \quad (2)$$

where $U_{id,mix}$ is the ideal mixture ultrasonic velocity in liquid mixture. X_i is the mole fraction, M_i is the molar mass and U_i is ultrasonic velocity.

Impedance relation (IMP)¹³: Product of ultrasonic velocity (U) and density (ρ) of the mixture is termed as acoustic impedance (Z) of the mixture. Hence the sound velocity in the mixture can be predicted from the knowledge of acoustic impedance and the density of the pure components.

$$U_{IMP} = \left[\frac{X_1 Z_1 + X_2 Z_2 + X_3 Z_3}{X_1 \rho_1 + X_2 \rho_2 + X_3 \rho_3} \right] \quad (3)$$

where x_i , is the mole fraction, ρ_i the density of the mixture and Z_i is the acoustic impedance.

Junjie's method (JM)¹⁵

$$U_{JM} = \left[\frac{X_1 V_1 + X_2 V_2 + X_3 V_3}{(X_1 m_1 + X_2 m_2 + X_3 m_3)^{1/2}} \right] \cdot \left[\frac{X_1 V_1}{d_1 U_1^2} + \frac{X_2 V_2}{d_2 U_2^2} + \frac{X_3 V_3}{d_3 U_3^2} \right]^{-1/2} \quad (4)$$

X_i is the mole fraction, d_i is the density, U_i is ultrasonic velocity, m_i is the molar mass and V_i is the molar volume.

Rao's specific sound velocity (UR)¹⁴

$$U_R = \left(\sum x_i r_i d \right)^3 \quad (5)$$

where $r_i = \frac{U_i^{1/3}}{d_i}$, x_i is the mole fraction, U_i is the ultrasonic

velocity, d the density of the mixture.

Free length theory (FLT)²⁶

$$U_{FLT} = \frac{k}{L_{f,mix} d_{expt}^{1/2}} \quad (6)$$

where k is the Boltzmann constant, d is the density and $L_{f,mix}$

$$= 2 \left[\frac{V_m - (X_1 V_{01} + X_2 V_{02} + X_3 V_{03})}{X_1 Y_1 + X_2 Y_2 + X_3 Y_3} \right]$$

X_i is the mole fraction

Molar volume at absolute zero is given by

$$V_{01} = V_1 \frac{U_1}{U_\alpha}; V_{02} = V_2 \frac{U_2}{U_\alpha}; V_{03} = V_3 \frac{U_3}{U_\alpha}$$

Surface area per mole

$$Y_i = \frac{2(V_1 - V_{01})}{L_{f_i}}$$

U_i is the ultrasonic velocity

RESULTS AND DISCUSSION

Experimental and computed ultrasonic velocity using eqn. 1-6 and their percentage relative deviation values for the ternary systems at 303, 308 and 313 K are given in Tables 1-8. The predictive abilities of various ultrasonic theories depend upon the strength of the interaction prevailing in a system. These theories generally fail to predict accurately the ultrasonic velocities where strong interactions supposed to exist and the average absolute percentage relative deviation is small in systems where the interactions are less or nil. The theoretically found ultrasonic velocities of liquid mixtures reveal that Nomoto's relation shows better agreement than the other relations.

The measurements of ultrasonic velocity in solutions formed by liquid components reveals the degree of deviation from ideality whenever there are interactions among the component molecules. The deviations have been used to gain insight into nature and degree of interactions among the interacting systems. In the case of liquid mixtures the data on composition dependency of ultrasonic velocity help to understand the nature of molecular interactions in terms of some physical parameters.

It can be seen from Tables 1, 3, 5 and 7 that the theoretical values of ultrasonic velocity calculated shows deviation from experimental values. The limitations and approximation

TABLE-1
EXPERIMENTAL AND THEORETICAL VALUES OF VELOCITY (m s^{-1}) FOR SYSTEM I AT DIFFERENT TEMPERATURES

X_1	X_3	U_{EXP}	U_{NR}	U_{IMP}	U_J	U_R	U_{IMR}	U_{FLT}
303 K								
0.5164	0.1396	1278.6	1278.1	1277.2	1279.9	1273.3	1275.5	1277.6
0.4428	0.2107	1246.1	1245.6	1244.7	1247.4	1240.8	1243.0	1245.1
0.3692	0.3028	1231.5	1231.0	1230.1	1232.8	1226.2	1228.4	1230.5
0.2955	0.4297	1218.6	1218.1	1217.2	1219.9	1213.3	1215.5	1217.6
0.1479	0.5046	1190.4	1189.9	1189.0	1191.7	1185.1	1187.3	1189.4
0.0740	0.5805	1174.8	1174.3	1173.4	1176.1	1169.5	1171.7	1173.8
308 K								
0.5164	0.1396	1256.4	1255.9	1255.0	1257.7	1251.2	1253.4	1255.4
0.4428	0.2107	1220.2	1219.7	1218.8	1221.5	1215.0	1217.2	1219.2
0.3692	0.3028	1208.1	1207.6	1206.7	1209.4	1202.9	1205.1	1207.1
0.2955	0.4297	1197.6	1197.1	1196.2	1198.9	1192.4	1194.6	1196.6
0.1479	0.5046	1177.2	1176.7	1175.8	1178.5	1172.0	1174.2	1176.2
0.0740	0.5805	1148.4	1147.9	1147.0	1149.7	1143.2	1145.4	1147.4
313 K								
0.5164	0.1396	1227.1	1226.6	1225.7	1228.4	1221.9	1224.1	1226.1
0.4428	0.2107	1207.1	1206.6	1205.7	1208.4	1201.9	1204.1	1206.1
0.3692	0.3028	1198.8	1198.3	1197.4	1200.1	1193.6	1195.8	1197.8
0.2955	0.4297	1181.4	1180.9	1180.0	1182.7	1176.2	1178.4	1180.4
0.1479	0.5046	1148.4	1147.9	1147.0	1149.7	1143.2	1145.4	1147.4
0.0740	0.5805	1118.2	1117.7	1116.8	1119.5	1113.0	1115.2	1117.2

TABLE-2
PERCENTAGE DEVIATION BETWEEN EXPERIMENTAL AND THEORETICAL VALUES OF VELOCITY (m s^{-1}) FOR SYSTEM I AT DIFFERENT TEMPERATURES

X_1	X_3	$\%U_{\text{NR}}$	$\%U_{\text{IMP}}$	$\%U_J$	$\%U_R$	$\%U_{\text{IMR}}$	$\%U_{\text{FLT}}$
303 K							
0.5164	0.1396	-0.039	-0.109	0.102	-0.415	-0.242	-0.078
0.4428	0.2107	-0.040	-0.112	0.104	-0.425	-0.249	-0.080
0.3692	0.3028	-0.041	-0.114	0.106	-0.430	-0.252	-0.081
0.2955	0.4297	-0.041	-0.115	0.107	-0.435	-0.254	-0.082
0.1479	0.5046	-0.042	-0.118	0.109	-0.445	-0.260	-0.084
0.0740	0.5805	-0.043	-0.119	0.111	-0.451	-0.264	-0.085
308 K							
0.5164	0.1396	-0.040	-0.111	0.103	-0.414	-0.239	-0.080
0.4428	0.2107	-0.041	-0.115	0.107	-0.426	-0.246	-0.082
0.3692	0.3028	-0.041	-0.116	0.108	-0.430	-0.248	-0.083
0.2955	0.4297	-0.042	-0.117	0.109	-0.434	-0.251	-0.084
0.1479	0.5046	-0.042	-0.119	0.110	-0.442	-0.255	-0.085
0.0740	0.5805	-0.044	-0.122	0.113	-0.453	-0.261	-0.087
313 K							
0.5164	0.1396	-0.041	-0.114	0.106	-0.424	-0.244	-0.081
0.4428	0.2107	-0.041	-0.116	0.108	-0.431	-0.249	-0.083
0.3692	0.3028	-0.042	-0.117	0.108	-0.434	-0.250	-0.083
0.2955	0.4297	-0.042	-0.119	0.110	-0.440	-0.254	-0.085
0.1479	0.5046	-0.044	-0.122	0.113	-0.453	-0.261	-0.087
0.0740	0.5805	-0.045	-0.125	0.116	-0.465	-0.268	-0.089

incorporated in these theories are responsible for the deviations of theoretical values from experimental values. In Nomoto's theory, it is supposed that the volume does not change on mixing. But on mixing three liquids, the interaction between the molecules of the liquids takes place because of the presence of various types of forces such as hydrogen bonding, dipole-dipole, dispersive forces, charge transfer and dipole-induced dipole interactions. The deviations of experimental values from theoretical values calculated using Van Dael and Vangeel equation might be due to the compressibility of the component liquids in the present mixture. The deviations of experimental values and values calculated from impedance relation and Rao's

relation imply non-additivity of acoustic impedance and Rao's velocity in the liquid mixture. Large deviations are observed in case of Junjie's relation. Thus, the observed deviation of theoretical values of velocity from the experimental values shows that the molecular interactions are taking place¹⁶⁻¹⁸ between the unlike molecules in the liquid mixture. On increasing the temperature, the ultrasonic velocity values decrease in the ternary liquid mixture. This is probably due to the fact that the thermal energy activates the molecule, which would increase the rate of association of unlike molecules¹⁹. Similar kinds of results were obtained by earlier workers²⁰⁻²⁵. The ratio $U_{\text{exp}}^2/U_{\text{IMR}}^2$ is used to measure the non-ideality in

liquid mixtures, especially in those cases where the properties other than sound velocity are not known.

It is observed from the Tables 1, 3, 5 and 7 that Nomoto's relation predicts a good agreement of sound velocity with the experimental values in ternary mixtures all the systems. The ideal mixture relation (IMR) yields a fair estimation of sound velocity for system III and system IV whereas a poor agreement for the other system. Impedance dependence value closely follows the experimental value in system III. It also estimates the sound velocity values in systems I & II to a certain degree of accuracy. The collision factor model of sound velocity

confirms well with the experiment in ternary system of system I. It fails to confirm with the values of system III whereas the system (I, II, IV) is better compared to the above said systems. Thus from the comparison of experimental velocity values with theoretical models, it is clear that the collision factor theory is best suited for system I and II and impedance dependence relation for system IV. The percentage of deviation of the theoretical ultrasonic velocity values from the experimental values are shown in Tables 2, 4, 6 and 8. We infer from Tables 2, 4, 6 and 8 that for all the systems, the theoretical ultrasonic velocity values based on Nomoto relation and free length

TABLE-3
EXPERIMENTAL AND THEORETICAL VALUES OF VELOCITY (m s^{-1}) FOR SYSTEM II AT DIFFERENT TEMPERATURES

X_1	X_3	U_{EXP}	U_{NR}	U_{IMP}	U_{J}	U_{R}	U_{IMR}	U_{FLT}
303 K								
0.5164	0.1396	1361.6	1361.1	1360.2	1362.9	1356.5	1358.6	1360.6
0.4428	0.2107	1295.6	1295.1	1294.2	1296.9	1290.5	1292.6	1294.6
0.3692	0.3028	1256.1	1255.6	1254.7	1257.4	1251.0	1253.1	1255.1
0.2955	0.4297	1224.3	1223.8	1222.9	1225.6	1219.2	1221.3	1223.3
0.1479	0.5046	1187.7	1187.2	1186.3	1189.0	1182.6	1184.7	1186.7
0.0740	0.5805	1046.2	1045.7	1044.8	1047.5	1041.1	1043.2	1045.2
308 K								
0.5164	0.1396	1338.2	1337.7	1336.8	1339.5	1333.1	1335.2	1337.2
0.4428	0.2107	1244.1	1243.6	1242.7	1245.4	1239.0	1241.1	1243.1
0.3692	0.3028	1194.9	1194.4	1193.5	1196.2	1189.8	1191.9	1193.9
0.2955	0.4297	1193.5	1193.0	1192.1	1194.8	1188.4	1190.5	1192.5
0.1479	0.5046	1172.2	1171.7	1170.8	1173.5	1167.1	1169.2	1171.2
0.0740	0.5805	1010.9	1010.4	1009.5	1012.2	1005.8	1007.9	1009.9
313 K								
0.5164	0.1396	1316.1	1315.6	1314.7	1317.4	1311.0	1313.1	1315.1
0.4428	0.2107	1226.1	1225.6	1224.7	1227.4	1221.0	1223.1	1225.1
0.3692	0.3028	1184.1	1183.6	1182.7	1185.4	1179.0	1181.1	1183.1
0.2955	0.4297	1178.2	1177.7	1176.8	1179.5	1173.1	1175.2	1177.2
0.1479	0.5046	1127.8	1127.3	1126.4	1129.1	1122.7	1124.8	1126.8
0.0740	0.5805	986.3	985.8	984.9	987.6	981.2	983.3	985.3

TABLE-4
PERCENTAGE DEVIATION BETWEEN EXPERIMENTAL AND THEORETICAL VALUES OF VELOCITY (m s^{-1}) FOR SYSTEM II AT DIFFERENT TEMPERATURES

X_1	X_3	$\%U_{\text{NR}}$	$\%U_{\text{IMP}}$	$\%U_{\text{J}}$	$\%U_{\text{R}}$	$\%U_{\text{IMR}}$	$\%U_{\text{FLT}}$
303 K							
0.5164	0.1396	-0.037	-0.103	0.095	-0.375	-0.220	0.073
0.4428	0.2107	-0.039	-0.108	0.100	-0.394	-0.232	0.077
0.3692	0.3028	-0.040	-0.111	0.103	-0.406	-0.239	0.080
0.2955	0.4297	-0.041	-0.114	0.106	-0.417	-0.245	0.082
0.1479	0.5046	-0.042	-0.118	0.109	-0.429	-0.253	0.084
0.0740	0.5805	-0.048	-0.134	0.124	-0.487	-0.287	0.096
308 K							
0.5164	0.1396	-0.037	-0.105	0.097	-0.381	-0.224	-0.075
0.4428	0.2107	-0.040	-0.113	0.104	-0.410	-0.241	-0.080
0.3692	0.3028	-0.042	-0.117	0.109	-0.427	-0.251	-0.084
0.2955	0.4297	-0.042	-0.117	0.109	-0.427	-0.251	-0.084
0.1479	0.5046	-0.043	-0.119	0.111	-0.435	-0.256	-0.085
0.0740	0.5805	-0.049	-0.138	0.129	-0.505	-0.297	-0.099
313 K							
0.5164	0.1396	-0.038	-0.106	0.099	-0.388	-0.228	-0.076
0.4428	0.2107	-0.041	-0.114	0.106	-0.416	-0.245	-0.082
0.3692	0.3028	-0.042	-0.118	0.110	-0.431	-0.253	-0.084
0.2955	0.4297	-0.042	-0.119	0.110	-0.433	-0.255	-0.085
0.1479	0.5046	-0.044	-0.124	0.115	-0.452	-0.266	-0.089
0.0740	0.5805	-0.051	-0.142	0.132	-0.517	-0.304	-0.101

TABLE-5
EXPERIMENTAL AND THEORETICAL VALUES OF VELOCITIES (m s^{-1}) OF
VELOCITY (m s^{-1}) FOR SYSTEM III AT DIFFERENT TEMPERATURES

X_1	X_3	U_{EXP}	U_{NR}	U_{IMP}	U_J	U_R	U_{IMR}	U_{FLT}
303 K								
0.5164	0.1396	1356.7	1356.2	1355.3	1358.0	1351.6	1353.7	1355.7
0.4428	0.2107	1326.1	1325.6	1324.7	1327.4	1321.0	1323.1	1325.1
0.3692	0.3028	1296.4	1295.9	1295.0	1297.7	1291.3	1293.4	1295.4
0.2955	0.4297	1260.9	1260.4	1259.5	1262.2	1255.8	1257.9	1259.9
0.1479	0.5046	1226.3	1225.8	1224.9	1227.6	1221.2	1223.3	1225.3
0.0740	0.5805	1214.1	1213.6	1212.7	1215.4	1209.0	1211.1	1213.1
308 K								
0.5164	0.1396	1315.3	1314.8	1313.9	1316.6	1310.2	1312.3	1314.3
0.4428	0.2107	1290.1	1289.6	1288.7	1291.4	1285.0	1287.1	1289.1
0.3692	0.3028	1253.7	1253.2	1252.3	1255.0	1248.6	1250.7	1252.7
0.2955	0.4297	1234.2	1233.7	1232.8	1235.5	1229.1	1231.2	1233.2
0.1479	0.5046	1212.1	1211.6	1210.7	1213.4	1207.0	1209.1	1211.1
0.0740	0.5805	1205.3	1204.8	1203.9	1206.6	1200.2	1202.3	1204.3
313 K								
0.5164	0.1396	1301.2	1300.7	1299.8	1302.5	1296.1	1298.2	1300.2
0.4428	0.2107	1281.1	1280.6	1279.7	1282.4	1276.0	1278.1	1280.1
0.3692	0.3028	1224.4	1223.9	1223.0	1225.7	1219.3	1221.4	1223.4
0.2955	0.4297	1217.4	1216.9	1216.0	1218.7	1212.3	1214.4	1216.4
0.1479	0.5046	1188.2	1187.7	1186.8	1189.5	1183.1	1185.2	1187.2
0.0740	0.5805	1200.2	1199.7	1198.8	1201.5	1195.1	1197.2	1199.2

TABLE-6
PERCENTAGE DEVIATION BETWEEN EXPERIMENTAL AND THEORETICAL VALUES OF
VELOCITY (m s^{-1}) FOR SYSTEM III AT DIFFERENT TEMPERATURES

X_1	X_3	$\%U_{\text{NR}}$	$\%U_{\text{IMP}}$	$\%U_J$	$\%U_R$	$\%U_{\text{IMR}}$	$\%U_{\text{FLT}}$
303 K							
0.5164	0.1396	-0.037	-0.103	0.096	-0.376	-0.221	-0.074
0.4428	0.2107	-0.038	-0.106	0.098	-0.385	-0.226	-0.075
0.3692	0.3028	-0.039	-0.108	0.100	-0.393	-0.231	-0.077
0.2955	0.4297	-0.040	-0.111	0.103	-0.404	-0.238	-0.079
0.1479	0.5046	-0.041	-0.114	0.106	-0.416	-0.245	-0.082
0.0740	0.5805	-0.041	-0.115	0.107	-0.420	-0.247	-0.082
308 K							
0.5164	0.1396	-0.038	-0.106	0.099	-0.388	-0.228	-0.076
0.4428	0.2107	-0.039	-0.109	0.101	-0.395	-0.233	-0.078
0.3692	0.3028	-0.040	-0.112	0.104	-0.407	-0.239	-0.080
0.2955	0.4297	-0.041	-0.113	0.105	-0.413	-0.243	-0.081
0.1479	0.5046	-0.041	-0.116	0.107	-0.421	-0.248	-0.083
0.0740	0.5805	-0.041	-0.116	0.108	-0.423	-0.249	-0.083
313 K							
0.5164	0.1396	-0.038	-0.108	0.100	-0.392	-0.231	-0.077
0.4428	0.2107	-0.039	-0.109	0.101	-0.398	-0.234	-0.078
0.3692	0.3028	-0.041	-0.114	0.106	-0.417	-0.245	-0.082
0.2955	0.4297	-0.041	-0.115	0.107	-0.419	-0.246	-0.082
0.1479	0.5046	-0.042	-0.118	0.109	-0.429	-0.252	-0.084
0.0740	0.5805	-0.042	-0.117	0.108	-0.425	-0.250	-0.083

theory gives less percentage of deviation than that of other theories. On the whole, all the theoretical models fairly predict that the ultrasonic velocities are reasonably close to the experimental values for the three ternary mixtures reported in this work, thus showing the validity of these theoretical models.

Percentage deviations of the ultrasonic velocity are both negative and positive. Such deviations indicate the non ideal behaviour of liquid mixtures. It is used as an important tool to measure the non-ideality in the mixtures, especially in those cases where the properties other than sound velocity are not known.

Conclusion

Ultrasonic velocities are calculated experimentally and the results are compared with the values calculated from various theories and relations. Ultrasonic method is a powerful probe for characterizing the physio-chemical properties and existence of molecular interaction in the liquid mixtures. In addition, ultrasonic velocity and the percentage deviation provide evidence of confirmation. It is concluded that the strong molecular interaction exists between the mixing components which may be due to the various types of forces such as hydrogen bonding, dipole-dipole, dispersive forces. Further the strength

TABLE-7
EXPERIMENTAL AND THEORETICAL VALUES OF VELOCITY (m s^{-1}) FOR SYSTEM IV AT DIFFERENT TEMPERATURES

X_1	X_3	U_{EXP}	U_{NR}	U_{IMP}	U_{J}	U_{R}	U_{IMR}	U_{FLT}
303 K								
0.5164	0.1396	1387.1	1386.6	1385.7	1388.4	1382.0	1384.1	1386.1
0.4428	0.2107	1353.5	1353.0	1352.1	1354.8	1348.4	1350.5	1352.5
0.3692	0.3028	1311.8	1311.3	1310.4	1313.1	1306.7	1308.8	1310.8
0.2955	0.4297	1297.4	1296.9	1296.0	1298.7	1292.3	1294.4	1296.4
0.1479	0.5046	1254.1	1253.6	1252.7	1255.4	1249.0	1251.1	1253.1
0.0740	0.5805	1229.1	1228.6	1227.7	1230.4	1224.0	1226.1	1228.1
308 K								
0.5164	0.1396	1348.4	1347.9	1347.0	1349.7	1343.3	1345.4	1347.4
0.4428	0.2107	1317.2	1316.7	1315.8	1318.5	1312.1	1314.2	1316.2
0.3692	0.3028	1265.7	1265.2	1264.3	1267.0	1260.6	1262.7	1264.7
0.2955	0.4297	1245.9	1245.4	1244.5	1247.2	1240.8	1242.9	1244.9
0.1479	0.5046	1230.3	1229.8	1228.9	1231.6	1225.2	1227.3	1229.3
0.0740	0.5805	1187.7	1187.2	1186.3	1189.0	1182.6	1184.7	1186.7
313 K								
0.5164	0.1396	1313.1	1312.6	1311.7	1314.4	1308.0	1310.1	1312.1
0.4428	0.2107	1297.4	1296.9	1296.0	1298.7	1292.3	1294.4	1296.4
0.3692	0.3028	1248.3	1247.8	1246.9	1249.6	1243.2	1245.3	1247.3
0.2955	0.4297	1234.5	1234.0	1233.1	1235.8	1229.4	1231.5	1233.5
0.1479	0.5046	1200.3	1199.8	1198.9	1201.6	1195.2	1197.3	1199.3
0.0740	0.5805	1166.5	1166.0	1165.1	1167.8	1161.4	1163.5	1165.5

TABLE-8
PERCENTAGE DEVIATION BETWEEN EXPERIMENTAL AND THEORETICAL VALUES OF VELOCITY (ms^{-1}) FOR SYSTEM IV AT DIFFERENT TEMPERATURES

X_1	X_3	$\%U_{\text{NR}}$	$\%U_{\text{IMP}}$	$\%U_{\text{J}}$	$\%U_{\text{R}}$	$\%U_{\text{IMR}}$	$\%U_{\text{FLT}}$
303 K							
0.5164	0.1396	-0.036	-0.101	0.094	-0.368	-0.216	-0.072
0.4428	0.2107	-0.037	-0.103	0.096	-0.377	-0.222	-0.074
0.3692	0.3028	-0.038	-0.107	0.099	-0.389	-0.229	-0.076
0.2955	0.4297	-0.039	-0.108	0.100	-0.393	-0.231	-0.077
0.1479	0.5046	-0.040	-0.112	0.104	-0.407	-0.239	-0.080
0.0740	0.5805	-0.041	-0.114	0.106	-0.415	-0.244	-0.081
308 K							
0.5164	0.1396	-0.037	-0.104	0.096	-0.378	-0.222	-0.074
0.4428	0.2107	-0.038	-0.106	0.099	-0.387	-0.228	-0.076
0.3692	0.3028	-0.040	-0.111	0.103	-0.403	-0.237	-0.079
0.2955	0.4297	-0.040	-0.112	0.104	-0.409	-0.241	-0.080
0.1479	0.5046	-0.041	-0.114	0.106	-0.415	-0.244	-0.081
0.0740	0.5805	-0.042	-0.118	0.109	-0.429	-0.253	-0.084
313 K							
0.5164	0.1396	-0.038	-0.107	0.099	-0.388	-0.228	-0.076
0.4428	0.2107	-0.039	-0.108	0.100	-0.393	-0.231	-0.077
0.3692	0.3028	-0.040	-0.112	0.104	-0.409	-0.240	-0.080
0.2955	0.4297	-0.041	-0.113	0.105	-0.413	-0.243	-0.081
0.1479	0.5046	-0.042	-0.117	0.108	-0.425	-0.250	-0.083
0.0740	0.5805	-0.043	-0.120	0.111	-0.437	-0.257	-0.086

of interaction tends to be weaker with rise in temperature due to weak intermolecular forces and thermal dispersive forces. It is understood from the results that out of all the theories and the relations discussed above, the Nomoto's relation provides the best results for all the systems.

REFERENCES

- J. Thennarasu and G. Meenakshi, *Int. Interdiscipl. Res. J.*, **1**, 43 (2011).
- N. Santhi, P. L. Sabarathinam, J. Madhumitha, G. Alamelumangai and M. Emayavaramban, *Int. Lett. Chem. Phy. Astron.*, **2**, 18 (2013).
- A. Ali, Abida, A.K. Nain and S. Hyder, *J. Pure Appl. Ultrason.*, **23**, 73 (2001).
- P.S. Naidu and K. Ravindra Prasad, *J. Pure Appl. Ultrason*, **24**, 1824 (2002).
- C.H. Srinivasu, K. Narendra and C.H. Kalpana, *E-J. Chem.*, **8**, 977 (2011).
- A. Pal and H. Kumar, *Indian J. Phys.*, **78**, 1319 (2004).
- G.V.R. Rao, A.V. Sarma, J.S.R. Krishna and C. Rambabu, *J. Indian Pure Appl. Phys.*, **43**, 345 (2005).
- D. Saravana Kumar and D. Krishna Rao, *J. Indian Pure Appl. Phys.*, **45**, 210 (2007).
- R. Uvarani and S. Punitha, *E-J. Chem.*, **6**(s1), 235 (2009).
- S. Parveen, D. Shukla, S. Singh, K.P. Singh, M. Gupta and J.P. Shukla, *Appl. Acoust.*, **70**, 507 (2009).

11. O. Nomoto, *J. Phys. Soc. Jpn.*, **13**, 1528 (1958).
12. W. Van Dael and E. Vangeel, Proc Int Conf on calorimetry and thermodynamics, Warasa, p. 555 (1955).
13. S. Baluja and P.H. Parsania, *Asian J. Chem.*, **7**, 417 (1995).
14. V.D. Gokhale and N.N. Bhagavan, *Indian J. Pure Appl. Ultrason.*, **11**, 21 (1989).
15. Z. Junjie, *J. China Univ. Sci. Technol.*, **14**, 298 (1984).
16. G.V. Rama Rao, P.B. Sandhya Sri, A. Viswanatha Sarma, C. Rambabu and J. Indian, *Pure Appl. Phys.*, **45**, 135 (2007).
17. K. Saravanaku, R. Baskaran and T.R. Kubendran, *J. Appl. Sci.*, **10**, 1616 (2010).
18. S. Anuradha, S. Prema and K. Rajagopal, *J. Pure Appl. Ultrason.*, **27**, 49 (2005).
19. N. Santhi, P.L. Sabarathinam, M.E. C. Gopi and C. Manivannan, *E-J. Chem.*, **7**, 648 (2010).
20. M. Rastogi, A. Awasthi, M. Gupta and J.P. Shukla, *J. Indian Pure Appl. Phys.*, **40**, 256 (2002).
21. P. Vasantharani, S. Muthu Shailaja, A.N. Kannappan and R. Ezhil Pavai, *J. Appl. Sci.*, **8**, 2329 (2008).
22. S. Babu, S.V.K. Sastry, H.S. Tiong and S.S. Sastry, *E-J. Chem.*, **9**, 2309 (2012).
23. G.R. Bedare, V.D. Bhandakkar and B.M. Suryavanshi, *J. Chem. Pharm. Res.*, **4**, 1028 (2012).
24. A. Ali and A.K. Nain, *J. Pure Appl. Ultrason.*, **22**, 10 (2000).
25. A. Ali, Abida, S. Hyder and A.K. Nain, *Collect. Czech. Chem. Commun.*, **67**, 1125 (2002).