



Theoretical evaluation of ultrasonic velocity in certain ternary liquid mixtures at different temperatures and concentrations and their comparison with the experimental values, studied through percentage deviation and interaction parameters

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ABSTRACT

Ultrasonic velocity in ternary mixture containing Benzene group of liquids has been evaluated as function of concentration and temperature, by using certain theoretical models like Nomoto's relation, Impedance dependence relation, Ideal mixture relation, Rao's specific velocity relation and Jungie's method. The experimental values of ultrasonic velocity are compared with the theoretical values. Molecular interaction parameter and the percentage deviation thus calculated indicate the non-ideality in the mixtures.

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Introduction

Ultrasonic velocity measurements have been successfully employed to detect and assess weak and strong molecular interaction in binary and ternary mixtures [1-4]. Such studies led to the determination of acoustic and thermodynamic parameters of liquid mixtures [5-8].

Several researchers [9-12] correlated the experimental results of ultrasonic velocity with theoretical relations obtained from Nomoto's relation [13,14], Vandal and Vangeel IMR relation [15], Impedance relation (IDR) [16], Rao's specific velocity relation [17] and Jungie's relation [18] and analyzed the differences between them.

In this paper, we have compared the experimentally determined ultrasonic velocity with the theoretical values using the above relations at different temperatures and different concentrations for the following ternary mixtures [19-22].

System I → N,N Dimethylformamide + Cyclohexane + Benzene

System II → N,N Dimethylformamide + Cyclohexane +

Chlorobenzene

System III → N,N Dimethylformamide + Cyclohexane +

Nitrobenzene

System IV → N,N Dimethylformamide+Cyclohexane+Pyridine.

Molecular interactions are analyzed based on the percentage deviation of the velocity and the molecular interaction parameter [23].

Experimental Method

The mixtures of various concentrations in mole fraction were prepared by taking analytical reagent grade and spectroscopic reagent grade chemicals with minimum assay of 99.9% and obtained from E.Merck Ltd (India). In all the mixtures, the mole fraction of the second component, cyclohexane ($X_2 = 0.4$), was kept fixed while the mole fractions of the remaining two (X_1 and X_3) were varied from 0.0 to 0.6, so as to have the mixture of different concentration. There is nothing significant in fixing the mole fraction of the second

component at 0.4. The density, viscosity, and ultrasonic velocity were measured as a function of concentration of the ternary liquid mixture at temperature $T = 288$ K, 298 K, 308 K and 318 K.

Velocity measurement

Ultrasonic velocity measurements were made using an ultrasonic interferometer (Model M-84, supplied by M/S Mittal Enterprises, New Delhi), with the accuracy of $\pm 0.1 \text{ m} \cdot \text{s}^{-1}$. The measuring cell of interferometer is a specially designed double-walled vessel with provision for temperature constancy. An electronically operated digital constant temperature bath (Model SSI-03 Spl, supplied by M/S Mittal Enterprises, New Delhi), operating in the temperature range of -10°C to 85°C with an accuracy of $\pm 0.1^\circ\text{C}$ has been used to circulate water through the outer jacket of the double-walled measuring cell containing the experimental liquid.

Density measurement

The densities of the mixture were measured using a 10-ml specific gravity bottle by relative measurement method with an accuracy of $\pm 0.01 \text{ kg} \cdot \text{m}^{-3}$. The specific gravity bottle with the experimental mixture was immersed in the temperature-controlled water bath to maintain required temperatures.

Theory

The following relations are used for the determination of ultrasonic velocity in the ternary liquid mixtures.

Nomoto's relation (U_{NR}):

Nomoto established an empirical formula for ultrasonic velocity in binary liquid mixtures on the assumption of linear dependence of the molar sound velocity on concentration in mole fractions and the additivity of molar volume as

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

Where X_1 , X_2 and X_3 are the mole fractions of the components, R_1 , R_2 and R_3 respective molar sound velocities and V_1 , V_2 and V_3 are the molar volumes respectively.

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Where molar sound velocity

$$R = \frac{M}{\rho} \cdot U^{1/3} = V \cdot U^{1/3}$$

The molar volume

$$V = \frac{M}{\rho}$$

Impedance Dependence Relation (U_{IDR})

The sound speed in the mixture is given by Impedance dependence relation (IDR) as

$$U_{IDR} = \frac{\sum_{i=1}^3 X_i Z_i}{\sum_{i=1}^3 X_i \rho_i} \quad (2)$$

Where ' z_i ' is the acoustic impedance and ' ρ_i ' is the density of the components in the mixture.

Ideal mixture relation (U_{IMR}):

Van Deal and Vangeel suggested the following relation for the velocity of sound

$$U_{IMR} = \left(\frac{1}{X_1 M_1 + X_2 M_2 + X_3 M_3} \right)^{1/2} \cdot \left(\frac{X_1}{M_1 U_1^2} + \frac{X_2}{M_2 U_2^2} + \frac{X_3}{M_3 U_3^2} \right)^{1/2} \quad (3)$$

Where, symbols have their usual meanings.

Rao,s specific velocity relation (U_R):

Rao,s specific velocity relation is given as

$$U_R = \sum_{i=1}^3 (X_i r_i \rho)^3 \quad (4)$$

Where, ' X_i ' is the mole fraction, ' ρ ' is the density of mixture and ' r_i ' is the Rao's specific sound velocity, which is given by

$$r_i = \frac{U_i^{1/3}}{\rho_i}$$

Jungie's equation (U_j):

The Jungie's equation is given by

$$U_j = \left[\frac{\sum_{i=1}^3 X_i V_i}{\left(\sum_{i=1}^3 X_i M_i \right)^{1/2}} \right] \left[\sum_{i=1}^3 \frac{X_i V_i}{\rho_i U_i^2} \right]^{-1/2} \quad (5)$$

Where, ' X_i ' is the mole fraction, ' M_i ' is molecular weights, ' V_i ' is the molar mass and ' ρ_i ' is density of constituent components.

Percentage deviation in Velocity :

Percentage deviation can be calculate by using the relation

$$\% \Delta U = \left(\frac{U_{exp} - U_{theo}}{U_{exp}} \right) \times 100 \quad (6)$$

Molecular interaction parameter (α):

Molecular interaction parameter can be calculate by using the relation

$$\alpha = \left(\frac{U_{exp}^2}{U_{IMR}^2} - 1 \right) \quad (7)$$

This depends on molar mass, composition and temperature.

Result And Discussion

The theoretical values of ultrasonic velocity obtained through the expressions given by equations 1 to 5 along with the experimental velocity at different concentrations and at different temperatures are summarized in the table-I. It is observed that the difference between theoretical and experimental values of ultrasonic velocities decreases with increase of temperature although there are slight variations. This is because the various theoretical formulas have not taken molecular interactions in to

account. At higher temperatures the intermolecular interactions becomes weak.

It can be seen from table II, III and IV that the theoretical values of ultrasonic velocity calculated by using various theories show deviation from experimental values through percentage deviation and interaction parameter. The limitations and approximations incorporated in these theories are responsible for these deviations.

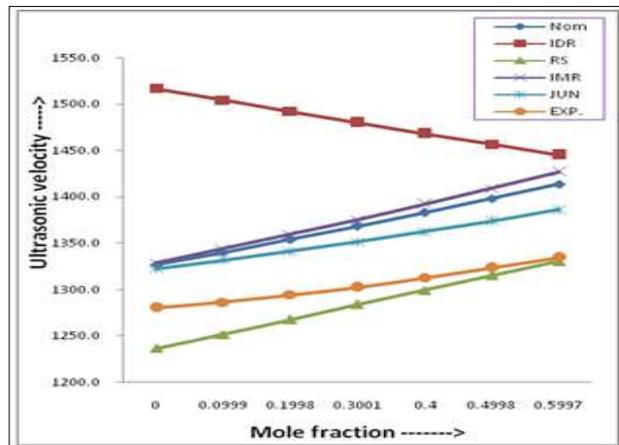


Fig 1. Variation of ultrasonic velocity with Concentration of DMF for system-I at 288K

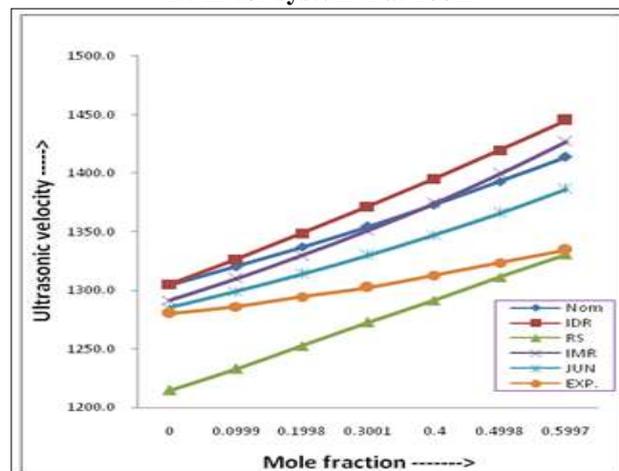


Fig 2. Variation of ultrasonic velocity with Concentration of DMF for system-II at 288K.

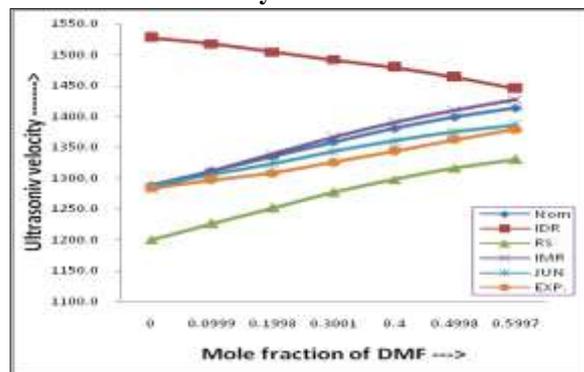


Fig 3. Variation of ultrasonic velocity with Concentration of DMF for system-III at 288K

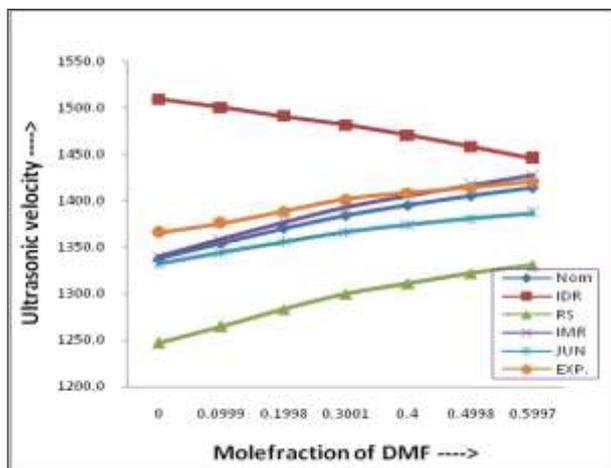
Nomoto's theory proposes that the volume does not change upon mixing. Hence interaction between the components of the liquid mixture has not been taken in to account. In the assumption for the ideal mixture relation, the ratios of specific heats and the volumes are taken to be equal. Here also intermolecular interaction is not taken in to account.

Table I. Experimental and Theoretical values of ultrasonic velocity

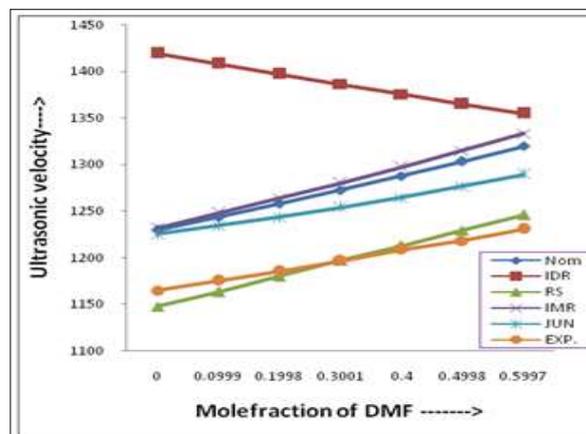
Mole fraction		U_{EXP} ($10^{-10} N^{-1} m^2$)				U_{NOM}				U_{IDR}				U_{RS}				U_{IMR}				U_J			
X_1	X_3	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K	288 K	298 K	308 K	318 K
SYSTEM – I (DMF + Cyclohexane + benzene)																									
0.0000	0.6000	1280.4	1247.7	1220.1	1165.1	1326.8	1276.1	1230.0	1185.9	1516.8	1465.8	1419.4	1371.9	1236.7	1189.9	1147.9	1107.2	1328.4	1277.7	1232.1	1187.9	1322.7	1271.9	1225.2	1181.4
0.0999	0.4999	1286.2	1252.8	1226.1	1175.4	1340.2	1289.9	1243.7	1200.0	1504.3	1454.2	1408.1	1361.6	1251.5	1205.3	1163.3	1122.9	1344.0	1293.6	1247.9	1203.9	1331.6	1281.1	1234.2	1190.6
0.1998	0.4001	1294.4	1258.2	1233.3	1185.3	1353.9	1304.2	1257.9	1214.5	1492.1	1442.8	1397.1	1351.6	1267.5	1221.8	1179.9	1139.8	1359.5	1309.7	1263.8	1220.0	1341.2	1290.9	1243.7	1200.4
0.3001	0.3000	1302.3	1267.4	1241.4	1196.4	1368.2	1318.9	1272.7	1229.6	1480.1	1431.5	1386.3	1341.7	1284.0	1239.0	1197.1	1157.3	1375.4	1326.0	1280.1	1236.6	1351.4	1301.5	1253.9	1210.8
0.4000	0.1999	1312.7	1280.9	1250.8	1208.5	1382.9	1334.2	1287.9	1245.1	1468.2	1420.4	1375.6	1331.9	1299.2	1254.7	1212.9	1173.5	1392.2	1343.4	1297.3	1254.1	1362.2	1312.7	1264.8	1222.0
0.4998	0.1001	1323.5	1287.7	1259.5	1218.5	1398.0	1349.9	1303.5	1261.2	1456.6	1409.6	1365.1	1322.3	1315.2	1271.4	1229.6	1190.6	1409.1	1361.0	1314.8	1272.0	1373.9	1324.7	1276.5	1234.0
0.5997	0.0000	1334.5	1298.8	1270.2	1230.9	1413.7	1366.2	1319.8	1277.8	1445.0	1398.8	1354.7	1312.8	1330.5	1287.4	1245.8	1207.1	1426.9	1379.4	1333.1	1290.7	1386.3	1337.6	1289.1	1246.9
SYSTEM – II (DMF + Cyclohexane + Chlorobenzene)																									
0.0000	0.6000	1280.4	1242.0	1206.0	1166.4	1304.9	1260.3	1218.2	1178.8	1305.3	1261.6	1221.1	1182.3	1214.6	1173.6	1135.1	1098.8	1291.5	1246.8	1204.1	1164.6	1285.3	1240.2	1197.0	1157.9
0.0999	0.4999	1291.1	1254.8	1218.3	1178.1	1320.4	1275.4	1232.7	1193.0	1326.6	1282.6	1241.6	1202.3	1232.9	1191.6	1152.5	1115.9	1310.1	1265.0	1221.8	1182.0	1299.1	1253.5	1209.5	1170.0
0.1998	0.4001	1304.6	1269.6	1233.3	1190.6	1336.9	1291.4	1248.1	1207.9	1348.7	1304.2	1262.7	1222.9	1252.4	1210.7	1171.2	1134.1	1329.7	1284.2	1240.6	1200.3	1313.8	1267.8	1223.0	1183.1
0.3001	0.3000	1320.1	1287.3	1246.9	1205.4	1354.4	1308.5	1264.5	1223.9	1371.6	1326.7	1284.6	1244.3	1272.7	1230.6	1190.5	1153.0	1350.8	1305.0	1260.8	1220.1	1329.9	1283.2	1237.6	1197.2
0.4000	0.1999	1336.8	1303.5	1263.0	1220.6	1373.0	1326.6	1281.8	1240.7	1395.2	1349.9	1307.1	1266.4	1291.5	1249.1	1208.5	1170.6	1374.2	1327.9	1283.0	1241.8	1347.1	1299.9	1253.3	1212.4
0.4998	0.1001	1355.0	1321.9	1280.1	1237.6	1392.7	1345.7	1300.2	1258.7	1419.7	1373.9	1330.5	1289.1	1311.3	1268.5	1227.4	1189.1	1399.3	1352.4	1306.9	1265.1	1365.9	1317.9	1270.4	1228.9
0.5997	0.0000	1377.2	1350.1	1300.5	1256.4	1413.7	1366.2	1319.8	1277.8	1445.0	1398.8	1354.7	1312.8	1330.5	1287.4	1245.8	1207.1	1426.9	1379.4	1333.1	1290.7	1386.3	1337.6	1289.1	1246.9
SYSTEM – III (DMF + Cyclohexane + Nitrobenzene)																									
0.0000	0.6000	1284.2	1245.9	1210.2	1170.2	1288.5	1245.6	1204.1	1163.1	1528.0	1477.7	1430.0	1377.7	1200.1	1160.2	1121.5	1083.6	1281.2	1237.6	1195.0	1154.0	1287.2	1244.1	1202.1	1161.0
0.0999	0.4999	1297.1	1259.8	1221.3	1185.3	1312.2	1269.2	1225.3	1185.9	1517.6	1465.5	1417.6	1368.7	1226.7	1186.8	1146.3	1109.7	1311.4	1267.7	1223.2	1183.4	1306.1	1262.7	1218.2	1178.6
0.1998	0.4001	1308.8	1275.1	1239.5	1199.5	1335.1	1292.6	1249.4	1208.0	1504.5	1454.9	1407.7	1360.8	1252.0	1212.7	1172.9	1134.7	1339.3	1296.1	1252.3	1210.7	1324.1	1281.2	1237.1	1195.6
0.3001	0.3000	1325.6	1294.2	1252.2	1210.7	1358.8	1315.8	1269.8	1227.6	1492.1	1442.4	1395.9	1350.2	1277.2	1237.5	1195.5	1156.6	1366.4	1322.9	1276.7	1234.3	1343.2	1299.6	1252.3	1210.1
0.4000	0.1999	1343.6	1310.0	1273.6	1229.0	1380.4	1335.4	1290.3	1247.5	1479.6	1430.6	1384.9	1340.7	1298.0	1256.8	1215.9	1176.5	1390.8	1345.5	1300.3	1257.2	1360.4	1314.6	1268.0	1225.1
0.4998	0.1001	1362.8	1332.5	1287.0	1242.6	1399.0	1353.0	1306.4	1263.8	1463.9	1416.1	1370.8	1327.7	1316.5	1274.6	1232.5	1193.3	1410.8	1364.8	1318.2	1275.2	1375.0	1328.1	1279.6	1236.9
0.5997	0.0000	1379.5	1352.5	1313.1	1261.7	1413.7	1366.2	1319.8	1277.8	1445.0	1398.8	1354.7	1312.8	1330.5	1287.4	1245.8	1207.1	1426.9	1379.4	1333.1	1290.7	1386.3	1337.6	1289.1	1246.9
SYSTEM – IV (DMF + Cyclohexane + Pyridine)																									
0.0000	0.6000	1365.3	1320.2	1287.1	1247.2	1337.4	1289.9	1248.5	1207.3	1508.9	1458.3	1411.8	1364.8	1246.9	1203.3	1165.9	1128.0	1339.5	1292.1	1251.0	1209.7	1331.8	1283.9	1241.2	1199.8
0.0999	0.4999	1375.5	1331.2	1292.7	1251.3	1353.7	1306.6	1261.9	1219.8	1500.3	1450.4	1405.1	1359.6	1264.7	1221.6	1181.0	1142.3	1358.5	1311.4	1266.9	1224.7	1343.9	1296.1	1250.3	1208.3
0.1998	0.4001	1388.1	1337.6	1298.8	1255.0	1369.7	1320.0	1274.9	1232.0	1490.9	1441.0	1397.2	1352.4	1282.7	1237.3	1196.4	1156.8	1376.4	1326.8	1281.8	1238.6	1355.7	1305.6	1259.3	1216.4
0.3001	0.3000	1401.5	1347.5	1303.9	1258.9	1384.0	1333.9	1287.0	1244.0	1481.1	1432.1	1388.1	1343.6	1299.3	1253.4	1211.0	1171.3	1392.4	1342.2	1295.5	1252.1	1366.3	1315.5	1267.4	1224.4
0.4000	0.1999	1408.1	1352.1	1309.8	1267.1	1394.8	1345.2	1298.7	1256.5	1470.1	1421.4	1377.7	1334.3	1310.6	1265.3	1223.4	1184.4	1405.1	1355.4	1309.1	1266.5	1373.7	1323.3	1275.2	1232.9
0.4998	0.1001	1413.5	1360.2	1315.6	1272.5	1404.6	1356.3	1309.6	1267.5	1458.1	1411.0	1366.9	1324.1	1321.4	1277.4	1235.4	1196.6	1416.3	1368.0	1321.4	1278.9	1380.3	1331.0	1282.4	1240.2
0.5997	0.0000	1420.1	1369.0	1324.8	1282.5	1413.7	1366.2	1319.8	1277.8	1445.0	1398.8	1354.7	1312.8	1330.5	1287.4	1245.8	1207.1	1426.9	1379.4	1333.1	1290.7	1386.3	1337.6	1289.1	1246.9

Table II: Percentage of deviation at different concentration and at temperature 288 K

Mole fraction		Percentage of deviation				
X_1	X_3	$\% \Delta U_{NOM}$	$\% \Delta U_{IDR}$	$\% \Delta U_{RS}$	$\% \Delta U_{IMR}$	$\% \Delta U_J$
SYSTEM – I (DMF + Cyclohexane + benzene)						
0.0000	0.6000	0.00	0.00	0.00	0.00	0.00
0.0999	0.4999	-1.91	-1.94	5.14	-0.87	-0.38
0.1998	0.4001	-2.27	-2.75	4.51	-1.47	-0.62
0.3001	0.3000	-2.48	-3.38	4.00	-1.92	-0.71
0.4000	0.1999	-2.60	-3.90	3.59	-2.33	-0.74
0.4998	0.1001	-2.71	-4.37	3.39	-2.80	-0.77
0.5997	0.0000	-2.78	-4.77	3.23	-3.27	-0.80
SYSTEM – II (DMF + Cyclohexane + Chlorobenzene)						
0.0000	0.6000	-1.91	-1.94	5.14	-0.87	-0.38
0.0999	0.4999	-2.27	-2.75	4.51	-1.47	-0.62
0.1998	0.4001	-2.48	-3.38	4.00	-1.92	-0.71
0.3001	0.3000	-2.60	-3.90	3.59	-2.33	-0.74
0.4000	0.1999	-2.71	-4.37	3.39	-2.80	-0.77
0.4998	0.1001	-2.78	-4.77	3.23	-3.27	-0.80
0.5997	0.0000	-2.65	-4.93	3.39	-3.61	-0.66
SYSTEM – III (DMF + Cyclohexane + Nitrobenzene)						
0.0000	0.6000	-0.34	-18.99	6.55	0.23	-0.23
0.0999	0.4999	-1.16	-17.00	5.43	-1.10	-0.69
0.1998	0.4001	-2.01	-14.95	4.34	-2.33	-1.17
0.3001	0.3000	-2.50	-12.56	3.65	-3.08	-1.33
0.4000	0.1999	-2.74	-10.12	3.39	-3.51	-1.25
0.4998	0.1001	-2.65	-7.42	3.40	-3.52	-0.89
0.5997	0.0000	-2.48	-4.75	3.55	-3.43	-0.50
SYSTEM – IV (DMF + Cyclohexane + Pyridine)						
0.0000	0.6000	2.04	-10.52	8.67	1.89	2.45
0.0999	0.4999	1.58	-9.07	8.06	1.24	2.30
0.1998	0.4001	1.33	-7.41	7.59	0.84	2.33
0.3001	0.3000	1.25	-5.68	7.29	0.65	2.51
0.4000	0.1999	0.94	-4.41	6.92	0.21	2.44
0.4998	0.1001	0.63	-3.16	6.51	-0.20	2.35
0.5997	0.0000	0.45	-1.76	6.31	-0.48	2.38

**Fig 4. Variation of ultrasonic velocity with Concentration of DMF for system-IV at 288K**

Deviation of U_{IMR} from U_{EXP} may be due to the compressibility of the component liquids in the mixture. It may be noted that increase in adiabatic compressibility suggest minimum interaction between molecules.

**Fig 5. Variation of ultrasonic velocity with Concentration of DMF for system-I at 308K**

Deviations observed in impedance dependent velocity (U_{IDR}) and Rao's velocity, imply non-additivity of acoustic impedance and Rao's velocity in the liquid mixture respectively. Acoustic impedance changes with temperature as well as concentration, hence large deviations are observed in case of

impedance dependent velocity.

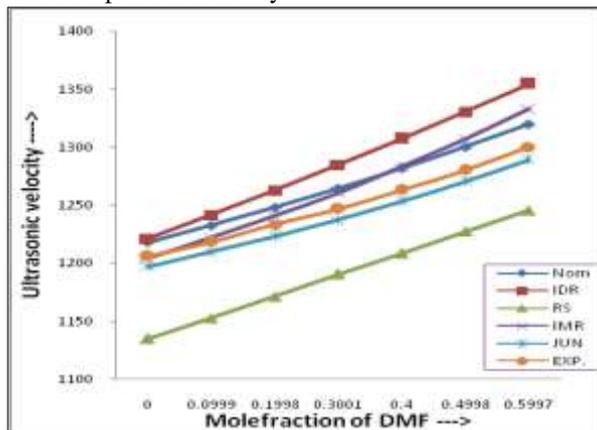


Fig 6. Variation of ultrasonic velocity with Concentration of DMF for system-II at 308K

On mixing two or more liquids the interaction between the molecules takes place because of the various types of forces such as dispersion forces, charge transfer, hydrogen bonding, dipole-dipole and dipole-induced dipole interactions. This shows that the intermolecular interaction potential for a system of liquid mixture will require for its full description the knowledge of the relative strengths of the various interactions in like and unlike molecules. Such a comprehensive expression for the intermolecular potential including all these factors has not yet been developed.

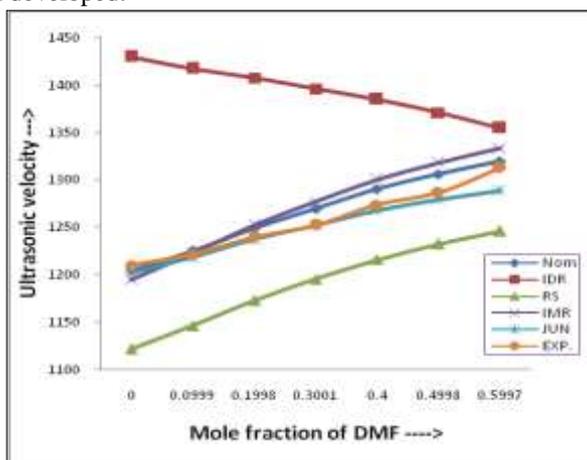


Fig 7. Variation of ultrasonic velocity with Concentration of DMF for system-III at 308K

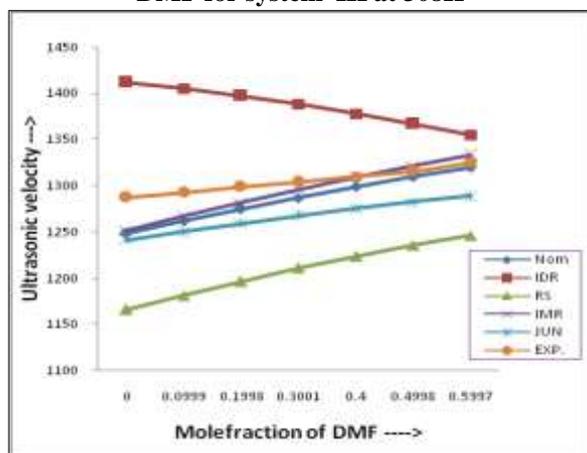


Fig 8. Variation of ultrasonic velocity with Concentration of DMF for system-IV at 308K

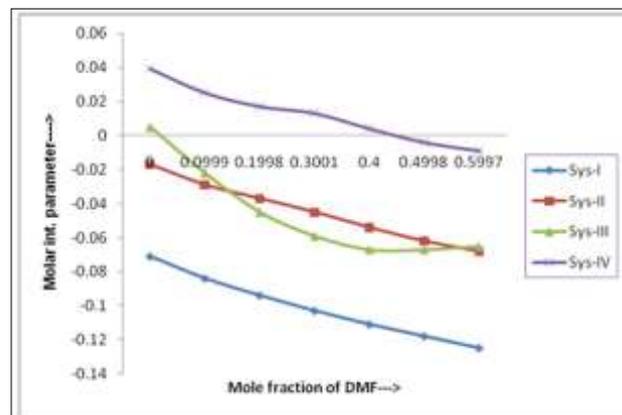


Fig 9. Variation of Molar interaction parameter with concentration of DMF at 288K

Percentage deviations of the ultrasonic velocity are both negative and positive. Positive deviations are attributed to molecular association (stronger interactions) and complex formation whereas negative deviations indicate molecular dissociations (weak interaction). Since the interaction in all the four mixtures are weak, the percentage deviations are found to be negative in all the relations i.e. Nomoto, IDR, IMR, Rao's specific velocity relation, Junge's relation. However it is different in case of the mixture containing pyridine. This may be due to the fact that pyridine molecules are spherical in shape and monomers, leading to strong intra-molecular interaction. However this deviation shows a decline as concentration of pyridine decreases and DMF increases, indicating weak intermolecular interaction.

Percentage deviation observed in impedance relation is due to the non-additivity of acoustic impedance. It is negative for all the mixtures, indicating weak interaction. For system-I, system-III and system-IV negativity decreases with decrease of concentrations of benzene, nitrobenzene and pyridine (increase of concentration of DMF) implying that the interaction increases due to dipole-induced dipole (intermolecular) interaction. It has already been observed [5] that acoustic impedance in system-I, III and IV decreases with increase in concentration of DMF.

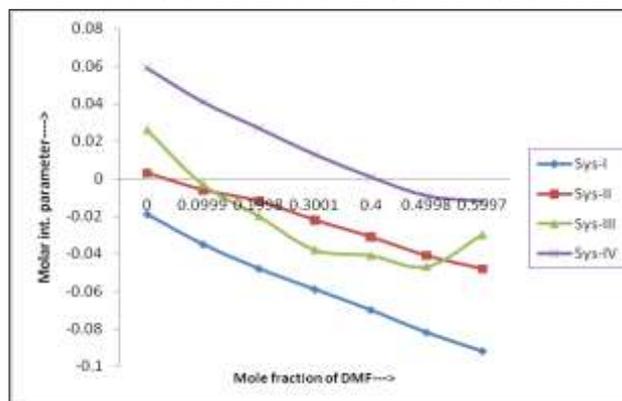


Fig 10. Variation of Molar interaction parameter with concentration of DMF at 308K

In system-II the percentage deviation increases with decrease in concentration of chlorobenzene (or increase in concentration of DMF). It has also been observed [19] that acoustic impedance increases with (increase in concentration of DMF) decrease in concentration of chlorobenzene.

If interaction parameter is positive, it represents strong interaction between mixing molecules. Larger value of ' α ' indicates larger deviation from ideality.

Table III. Percentage of deviation at different concentration and at temperature 308 K

Mole fraction		Percentage of deviation				
X_1	X_3	% ΔU_{NOM}	% ΔU_{IDR}	% ΔU_{RS}	% ΔU_{IMR}	% ΔU_J
SYSTEM – I (DMF + Cyclohexane + benzene)						
0.0000	0.6000	-0.81	-16.33	5.92	-0.98	-0.42
0.0999	0.4999	-1.44	-14.85	5.12	-1.78	-0.66
0.1998	0.4001	-2.00	-13.28	4.33	-2.47	-0.84
0.3001	0.3000	-2.52	-11.67	3.57	-3.11	-1.01
0.4000	0.1999	-2.96	-9.97	3.03	-3.72	-1.12
0.4998	0.1001	-3.50	-8.38	2.37	-4.39	-1.35
0.5997	0.0000	-3.90	-6.65	1.92	-4.95	-1.48
SYSTEM – II (DMF + Cyclohexane + Chlorobenzene)						
0.0000	0.6000	-1.01	-1.26	5.88	0.16	0.75
0.0999	0.4999	-1.18	-1.91	5.40	-0.29	0.72
0.1998	0.4001	-1.20	-2.38	5.04	-0.59	0.84
0.3001	0.3000	-1.41	-3.02	4.52	-1.11	0.75
0.4000	0.1999	-1.49	-3.49	4.32	-1.59	0.76
0.4998	0.1001	-1.57	-3.94	4.12	-2.09	0.76
0.5997	0.0000	-1.48	-4.17	4.21	-2.51	0.88
SYSTEM – III (DMF + Cyclohexane + Nitrobenzene)						
0.0000	0.6000	0.51	-18.16	7.33	1.26	0.67
0.0999	0.4999	-0.33	-16.07	6.14	-0.16	0.25
0.1998	0.4001	-0.80	-13.57	5.37	-1.03	0.20
0.3001	0.3000	-1.40	-11.47	4.53	-1.96	-0.01
0.4000	0.1999	-1.31	-8.74	4.53	-2.09	0.44
0.4998	0.1001	-1.50	-6.51	4.23	-2.42	0.58
0.5997	0.0000	-0.51	-3.17	5.13	-1.52	1.83
SYSTEM – IV (DMF + Cyclohexane + Pyridine)						
0.0000	0.6000	3.00	-9.69	9.41	2.80	3.57
0.0999	0.4999	2.38	-8.69	8.64	2.00	3.28
0.1998	0.4001	1.84	-7.57	7.88	1.31	3.04
0.3001	0.3000	1.29	-6.46	7.12	0.64	2.80
0.4000	0.1999	0.85	-5.18	6.60	0.05	2.64
0.4998	0.1001	0.45	-3.90	6.09	-0.44	2.52
0.5997	0.0000	0.38	-2.26	5.97	-0.63	2.70

Table – IV: Molecular interaction parameter at different concentration at temperatures 288K and 308 K

Mole fraction		Temperature	
X ₁	X ₃	288 K	308 K
SYSTEM – I (DMF + Cyclohexane + benzene)			
0.0000	0.6000	-0.071	-0.019
0.0999	0.4999	-0.084	-0.035
0.1998	0.4001	-0.094	-0.048
0.3001	0.3000	-0.103	-0.059
0.4000	0.1999	-0.111	-0.070
0.4998	0.1001	-0.118	-0.082
0.5997	0.0000	-0.125	-0.092
SYSTEM – II (DMF + Cyclohexane + Chlorobenzene)			
0.0000	0.6000	-0.017	0.003
0.0999	0.4999	-0.029	-0.006
0.1998	0.4001	-0.037	-0.012
0.3001	0.3000	-0.045	-0.022
0.4000	0.1999	-0.054	-0.031
0.4998	0.1001	-0.062	-0.041
0.5997	0.0000	-0.068	-0.048
SYSTEM – III (DMF + Cyclohexane + Nitrobenzene)			
0.0000	0.6000	0.005	0.026
0.0999	0.4999	-0.022	-0.003
0.1998	0.4001	-0.045	-0.020
0.3001	0.3000	-0.059	-0.038
0.4000	0.1999	-0.067	-0.041
0.4998	0.1001	-0.067	-0.047
0.5997	0.0000	-0.065	-0.030
SYSTEM – IV (DMF + Cyclohexane + Pyridine)			
0.0000	0.6000	0.039	0.059
0.0999	0.4999	0.025	0.041
0.1998	0.4001	0.017	0.027
0.3001	0.3000	0.013	0.013
0.4000	0.1999	0.004	0.001
0.4998	0.1001	-0.004	-0.009
0.5997	0.0000	-0.009	-0.012

In the present case ' α ' is usually negative, indicating weak interaction between the molecules of the mixture. In case of system-IV however ' α ' is slightly positive. It is larger at higher concentration of pyridine and decreases with lowering of concentration of pyridine, finally becoming negative. At larger concentration of pyridine strong intermolecular interaction leads to positive value of ' α '. When pyridine concentration decreases the weak intermolecular interaction between pyridine and DMF molecules yields negative value of ' α '. Nature of variation of ' α ' is same at low as well as high temperature (table-IV), however ' α ' has higher magnitude at higher temperature indicating larger deviation from ideality.

In spite of being empirical, the Junje's relation performs well for system-I, II and III at all temperatures whereas the Ideal mixing relation and Nomoto's relation perform well for system-IV (table-II&III).

Conclusion:

Ultrasonic velocity predicted using Nomoto's, IMR, IDR, Junje's and Rao's relations were compared with experimentally measured velocity values at different concentrations and temperatures for the systems I, II, III and IV. The observed deviations in theoretical values of velocity from the experimental values are attributed to the presence of intermolecular interaction in the systems.

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