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Study of Inter Molecular Interactions in Industrially useful Binary Liquid Mixtures of N-Methyl-2-Pyrrolidone with the Help of Scaled Particle Theory (SPT)

D Bala Karuna Kumar[1,2*], Y Subba Reddy[1], P Sugandha Kumar[1], G Srinivasa Rao[2] and C Rambabu[3]

[1]Department of Chemistry, Andhra Loyola College, Vijayawada, Andhra Pradesh, India
[2]Department of Physics, Andhra Loyola College, Vijayawada, Andhra Pradesh, India
[3]Department of Chemistry, Acharya Nagarjuna University, Nagarjuna Nagar, Andhra Pradesh, India

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ABSTRACT

The ultrasonic speed (U) studies are carried out using single crystal variable path fixed frequency (2 MHz) ultrasonic interferometer on some aliphatic amines (Propylamine, PA, Butylamine, BA, dipropylamine, DPA) in N-methyl-2-pyrrolidone (NMP) at temperatures of 303.15, 308.15, 313.15 and 318.15 K. To examine the intermolecular interactions in these liquid mixtures ultrasonic speeds are theoretically evaluated based on scaled particle theory and compared with the experimentally measured values. An attempt is made to study the temperature effect on the shapes of the interacting molecules in the binary liquid mixture using scaled particle theory in the temperature range 303.15-318.15 K.

Keywords: Ultrasonic speed; Scaled particle theory; Molecular interactions

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INTRODUCTION

Liquid mixtures are indispensable in almost all industries and all biological sciences. There exists an imperative need to understand these systems and to be able to predict their behavior from the molecular point of view. The use of measurable macroscopic or global quantities to probe into the microscopic, or the local properties of the system has become an essential area of research. The types of properties probed by these tools are local composition, local change of order, or structure etc. The traditional characterization and study of the properties of liquid mixtures by means of the global excess thermodynamic functions has become handy as it provides richer and more detailed information on the immediate environment of molecules in the mixture [1-3]. This paper is a continuation of our systematic studies on the thermodynamic properties of industrially useful binary liquid mixtures of N-methyi-2-Pyrrolidone (NMP) [4-9]. In this paper we report the molecular interactions in the liquid mixtures of NMP + Propylamine (PA), + Butylamine (BA), + Dipropylamine (DPA) using Scaled Particle Theory (SPT).

EXPERIMENTAL SECTION

The commercially available pure solvents were used in the present investigation. NMP (Merck >99%), propylamine (>99%), butylamine (>99%) and dipropylamine (>99%) procured from S.D Fine chemicals (India) were purified using standard procedures [10,11] like fractional distillation and distillation under reduced pressure and only the middle fractions are collected. The ultrasonic speed of pure components and their mixtures were measured by single crystal variable path fixed frequency interferometer provided by Mittal Enterprises, New Delhi (Model-F 05). The

measurements of ultrasonic speed were taken at a fixed frequency of 2 MHz. The calibration of ultrasonic interferometer was done by measuring the velocity in AR grade benzene and carbon tetrachloride. The expanded uncertainty in the measurement of ultrasonic speed was found to be 0.8 ms^{-1}. Temperature control for the measurement of density and ultrasonic speed was achieved by using a microprocessor assisted circulating water bath, (supplied by Mac, New Delhi) regulated to ±0.01 K, using a proportional temperature controller.

Theory

The scaled-particle theory (SPT) offers a powerful conceptual and computational frame work within which molecular order and thermodynamic properties can be examined [12]. SPT links the microscopic parameters viz., radius, surface area and hard core volume of a molecule with the macroscopic parameters like ultrasonic speed. The ultrasonic speeds of pure liquids and their binary liquid mixtures can be estimated theoretically based on some, statistical, empirical and semi empirical models like Junjee, Nomoto, Free length theory (FLT), Vandeel Vangeal and Collision factor theory (CFT). However, all these models have a common drawback that, the shapes of the participating species have not been taken into consideration while estimating ultrasonic speed. On the other hand, in scaled particle theory [13] participating components are considered to have different shapes (like sphere, cube, tetrahedral, disc A, disc B, disc C and disc D) and when the shape of the participating components match the actual shape of the species, then the theoretical ultrasonic speeds estimated based on this model will give values close to the experimental values. Although, the chemical structure of a molecule is known, no definite shape has been attached to it in liquid state [14]. Recently, Ghosh et al. [14] have used the scaled particle theory for the binary mixtures of 1,1,1-trichloroethane with 1-alkanols by considering only three different shapes namely spherical, cubical and tetrahedral. However, Kalidoss et al. [15] have considered seven shapes (sphere, cube, tetrahedron, disc A, disc B, disc C and disc D) for each component while estimating the ultrasonic speeds of CCl_4 + benzene, CCl_4 + propanol, using SPT. Characteristic parameters while assigning different shapes to the molecules are given in table below. In the present investigation, theoretical ultrasonic speeds for the three binary mixtures of NMP at different temperatures are estimated using SPT and compared with experimental speeds. Chi-square fit determines the closeness between experimental and theoretical ultrasonic speeds when the assumed shape matches the actual shape of the components [16].

According to the scaled particle theory the equation of state of fluid is given by:

$$P/\rho_N K_B T = \frac{1+\alpha+\alpha^2}{(1-\alpha)^2} \quad \text{..... (1)}$$

Where $\alpha = V_H \rho_N$, ρ_N is number density and V_H is hard core volume, the other quantities have their usual meaning.

SPT for mixtures of hard convex (not necessarily spherical) molecules gives the equation for mixture as follows [14,15]:

$$\frac{P}{\rho_N K_B T} = \frac{1}{(1-V\rho_N)^2} + \frac{AB\rho_N}{(1-V\rho_N)^2} + \frac{B^2 C\rho_N^2}{(1-V\rho_N)^3} \quad \text{..... (2)}$$

Where $A = \sum x_i R_i$, $B = \sum x_i S_i$, $C = \sum x_i R_i^2$, $V = \sum x_i V_{Hi}$

x_i is the mole fraction of i^{th} species, R_i, S_i and V_{Hi} are mean radius of curvature, surface area and volume, respectively, of a molecule of species 'i'.

Relating the above equation with the equation: $\gamma(dp/d\rho)_T = u^2$ where u is the ultrasonic speed, and γ is the ratio of specific heats, we get:

$$\frac{Mu^2}{\gamma RT} = \frac{1}{(1-V\rho_N)^2} + 2AB\frac{\rho_N}{(1-V\rho_N)^3} + B^2 C\frac{\rho_N^2}{(1-V\rho_N)^4} \quad \text{..... (3)}$$

Eq. (3) is used to evaluate the ultrasonic speeds in the binary mixtures. For the case of pure liquids the above equation is modified by introducing the dimensionless shape parameter, $X = RS/V_H$ and $\lambda = V_H \rho_N$

$$\frac{Mu^2}{\gamma RT} = \frac{[1+(X-1)\lambda]^2}{(1-\lambda)^4} \quad \text{..... (4)}$$

Solution for the above equation is obtained as:

$$\lambda = K - \sqrt{K^2 + L - 1}$$

Where $K = 1 + L(X-1)/2$ and $L = \sqrt{\gamma RT/Mu^2}$

Mean radius and the surface area of a molecule can be written as $R = YV_H^{1/3}$ and $S = ZR^2$ where Y and Z are the parameters related to shape of the molecule.

RESULTS AND DISCUSSION

When the molecule present in the liquid mixture is assigned different shapes as described in Table 1, then the corresponding values of X, Y and Z (also known as shape parameters) can be calculated as shown in Table 2. The corresponding evaluated 'λ' values for all the three mixtures at all the four temperatures under study are given in Table 3.

Table 1: Molecular assignment for different shapes

Shape	Size	R	S	V_H
Sphere	Radius=b	b	$4pb^2$	$4pb^2/3$
Cube	Side=a	3a/4	$6a^2$	a^3
Tetrahedron	Side=a	$(3a \arctan\sqrt{2})/2p$	$\sqrt{3}\ a^2$	$(\sqrt{2}/12)\ a^3$
Discs	**radius=b and depth=l**			
Disc A	l=b	(p +1)b/4	$4pb^2$	$p\ b^3$
Disc B	l=b/4	(p+0.25)b/4	$5pb^2/2$	$p\ b^3/4$
Disc C	l=b/2	(p +0.50)b/4	$3pb^2$	$p\ b^3/2$
Disc D	l=b/10	(p +0.10)b/4	$11pb^2/5$	$p\ b^3/10$

Table 2: Shape parameters

Shape	X	Y	Z
Sphere	3	0.6204	12.5664
Cube	4.5	0.75	10.6666
Tetrahedron	6.7035	0.9303	8.3247
Disc A	4.1416	0.707	11.7218
Disc B	8.479	0.919	10.9244
Disc C	5.4624	0.7832	11.3712
Disc D	17.8274	1.192	10.5253

Table 3: Values of 'λ' obtained from equation (1)

Shape	NMP	PA	BAM	DPA	NMP	PA	BAM	DPA
	303.15 K				**308.15 K**			
Sphere	0.5336	0.4263	0.4559	0.4828	0.5288	0.4203	0.4504	0.4768
Cube	0.4718	0.3642	0.3934	0.4201	0.4669	0.3585	0.3879	0.4141
Tetrahedron	0.4083	0.3035	0.3313	0.3572	0.4034	0.2981	0.3261	0.3513
Disc A	0.4848	0.377	0.4063	0.4332	0.4799	0.3712	0.4008	0.4271
Disc B	0.3703	0.2687	0.2952	0.3202	0.3654	0.2636	0.2902	0.3146
Disc C	0.4412	0.3345	0.3631	0.3895	0.4362	0.3289	0.3577	0.3836
Disc D	0.2545	0.1706	0.1915	0.2118	0.2502	0.1667	0.1875	0.2072
	313.15 K				**318.15 K**			
Sphere	0.5241	0.4148	0.4449	0.4711	0.5196	0.4087	0.439	0.4656
Cube	0.462	0.3531	0.3825	0.4084	0.4575	0.3472	0.3767	0.4029
Tetrahedron	0.3985	0.293	0.3208	0.3458	0.394	0.2875	0.3153	0.3405
Disc A	0.475	0.3658	0.3954	0.4214	0.4704	0.3598	0.3895	0.4159
Disc B	0.3606	0.2588	0.2852	0.3092	0.3561	0.2536	0.2799	0.3041
Disc C	0.4313	0.3236	0.3524	0.378	0.4267	0.3179	0.3467	0.3725
Disc D	0.2461	0.163	0.1835	0.2028	0.2422	0.159	0.1794	0.1986

The theoretical speeds based on SPT are computed for the NMP + X (X= PA or BA or DPA) binary mixtures by considering different shapes, viz., sphere, cube, tetrahedron, disc A, disc B, disc C and disc D for both the participating components. At each temperature for NMP+X binary mixture 7 × 7 combinations of the different shapes are possible. Of these 49 combinations the best-fit combination of shapes of the participating component molecules is arrived at, using Chi-square test (χ^2). At 303.15 K, the theoretical ultrasonic speeds calculated using SPT for the NMP+PA mixture is found to have lowest χ^2 value when NMP assumes cube shape. Therefore in Table 4, the theoretical and experimental ultrasonic speeds for only short listed seven (out of 7 × 7) shape combinations with cube shape for NMP and other possible shapes (viz., sphere, cube, tetrahedron, disc A, disc B, disc C and disc D) for PA are presented.

Table 4: Experimental and theoretical (obtained using SPT) ultrasonic speeds with behavioral shapes having least χ^2- fit for the binary mixture NMP + PA at 303.15 K

x_1	U_{exp}	U (Cu-Sp)	U (Cu-Cu)	U (Cu -Te)	U (Cu-disc A)	U (Cu-disc B)	U (Cu-disc C)	U (Cu-disc D)
0	1193.1	1193.1	1193.1	1193.1	1193.1	1193.1	1193.1	1193.1
0.0873	1226.4	1225.9	1221.7	1219.6	1222.4	1217.3	1220.1	1214.6
0.1771	1260.2	1259.6	1251.9	1248.1	1253.3	1243.8	1249	1238.7
0.2695	1294.7	1294	1283.8	1278.6	1285.6	1272.7	1279.8	1265.5
0.3646	1330	1329.2	1317.3	1311.1	1319.5	1304.1	1312.6	1295.3
0.4626	1366.1	1365.1	1352.4	1345.6	1354.7	1338.1	1347.3	1328.3
0.5636	1402.8	1401.6	1389.1	1382.4	1391.4	1374.8	1384.1	1364.7
0.6676	1439.5	1438.7	1427.5	1421.3	1429.6	1414.4	1423	1404.9
0.7749	1476.6	1476.2	1467.6	1462.7	1469.2	1457.1	1464	1449.3
0.8857	1514.3	1514.3	1509.3	1506.4	1510.3	1503.2	1507.2	1498.4
1	1552.8	1552.8	1552.8	1552.8	1552.8	1552.8	1552.8	1552.8
	χ^2	0.0036	0.7269	1.6294	0.4999	3.0624	1.3674	5.6274

It is clear that at 303.15 K, when NMP + PA binary mixture takes the shapes of disc-A + sphere, the ultrasonic speed estimated based on SPT is found to be very close to the experimental values. Further, it is found that change of temperature has effect on the shape of the participating molecules in this system (Table 5).

Table 5: Experimental and theoretical (obtained using SPT) ultrasonic speeds with behavioral shape having least χ^2- fit for the binary mixture NMP + PA at different temperatures

x_1	303.15 K		308.15 K		313.15 K		318.15 K	
	U_{exp}	$U_{(disc\ A\text{-}Sp}$	U_{exp}	$U_{(disc\ D\text{-}disc\ D)}$	U_{exp}	$U_{(Te\text{-}disc\ C)}$	U_{exp}	$U_{(Cu\text{-}Sp)}$
0	1193.1	1193.1	1170.6	1170.6	1150.6	1150.6	1128.4	1128.4
0.0873	1226.4	1225.9	1204.3	1204.6	1184.8	1185	1163.6	1162.6
0.1771	1260.2	1259.6	1238.4	1239.1	1219.2	1219.7	1198.5	1197.1
0.2695	1294.7	1294	1273.1	1274	1253.8	1254.6	1233.2	1231.9
0.3646	1330	1329.2	1308.4	1309.3	1288.9	1289.7	1268.5	1266.8
0.4626	1366.1	1365.1	1344.4	1345	1324.3	1324.8	1303.8	1301.9
0.5636	1402.8	1401.6	1380.9	1380.9	1359.8	1360.1	1338.9	1337.1
0.6676	1439.5	1438.7	1416.8	1417	1394.8	1395.3	1373.6	1372.3
0.7749	1476.6	1476.2	1453	1453.4	1430.1	1430.6	1408.6	1407.5
0.8857	1514.3	1514.3	1489.4	1489.8	1465.7	1465.8	1443.5	1442.8
1	1552.8	1552.8	1526.4	1526.4	1501	1501	1478	1478
	χ^2	0.0036		0.0023		0.002		0.0137

In the binary mixtures of NMP+BA and NMP+DPA (Tables 6 and 7), the theoretical ultrasonic speeds match with the experimental values, when the participating molecules take the shapes of Tetrahedron + disc-A and Sphere + cube, respectively at 303.15 K. The shape of the participating molecules with rise in temperature in the binary mixtures NMP+BA and NMP + DPA are shown in Tables 8 and 9. From these tables it is clear that increase in temperature effects the assumed shapes of the constituent molecules in all the liquid mixtures under investigation.

Table 6: Experimental and theoretical (obtained using SPT) ultrasonic speeds with behavioral shapes having least χ^2- fit for the binary mixture NMP + BAM at 303.15 K

x_1	U_{exp}	U (Te-Sp)	U (Te-Cu)	U (Te -Te)	U (Te-disc A)	U (Te-disc B)	U (Te-disc C)	U (Te-disc D)
0	1226	1226	1226	1226	1226	1226	1226	1226
0.1033	1261.4	1267	1259.4	1254.1	1261.3	1253.1	1257.6	1248.6
0.2059	1295.7	1305.9	1292.6	1283.3	1296	1281.4	1289.4	1273.2
0.3077	1329.3	1342.7	1325.8	1313.5	1330.1	1310.9	1321.5	1299.9
0.4087	1362.6	1377.7	1358.7	1344.7	1363.6	1341.7	1353.9	1328.6
0.5091	1395.4	1410.8	1391.6	1377	1396.5	1373.6	1386.5	1359.6
0.6087	1427.7	1442.2	1424.2	1410.2	1428.8	1406.9	1419.3	1392.9
0.7076	1459.5	1472	1456.6	1444.4	1460.6	1441.4	1452.3	1428.8
0.8057	1491	1500.3	1488.9	1479.5	1491.8	1477.2	1485.6	1467.2
0.9032	1522	1527.2	1521	1515.7	1522.6	1514.3	1519.1	1508.5
1	1552.8	1552.8	1552.8	1552.8	1552.8	1552.8	1552.8	1552.8
	χ^2	0.9069	0.0594	1.312	0.0044	1.8228	0.3099	4.9305

Table 7: Experimental and theoretical (obtained using SPT) ultrasonic speeds with behavioral shape having least χ^2- fit for the binary mixture NMP + BAM at different temperatures

x_1	303.15 K		308.15 K		313.15 K		318.15 K	
	U_{exp}	$U_{(disc\ A\text{-}Sp)}$	U_{exp}	$U_{(disc\ D\text{-}disc\ D)}$	U_{exp}	$U_{(Te\text{-}disc\ C)}$	U_{exp}	$U_{(Cu\text{-}Sp)}$
0	1226	1226	1204.3	1204.3	1182.7	1182.7	1159.6	1159.6
0.1033	1261.4	1261.3	1239.4	1239	1217.7	1216.9	1196.4	1194.6
0.2059	1295.7	1296	1273.5	1273.1	1251.2	1250.6	1230.6	1228.9
0.3077	1329.3	1330.1	1306.9	1306.7	1284.6	1283.8	1263.8	1262.5
0.4087	1362.6	1363.6	1339.7	1339.6	1317.6	1316.5	1297.5	1295.4
0.5091	1395.4	1396.5	1372.2	1372.1	1349.5	1348.7	1328.4	1327.6
0.6087	1427.7	1428.8	1404	1403.9	1380.5	1380.3	1359.6	1359.1
0.7076	1459.5	1460.6	1435.3	1435.3	1411.5	1411.4	1390.1	1389.9
0.8057	1491	1491.8	1466.2	1466.2	1442	1441.8	1419.8	1419.9
0.9032	1522	1522.6	1496.4	1496.5	1471.9	1471.7	1449.5	1449.3
1	1552.8	1552.8	1526.4	1526.4	1501	1501	1478	1478
	χ^2	0.0044		0.0003		0.0027		0.0104

Table 8: Experimental and theoretical (obtained using SPT) ultrasonic speeds with behavioral shapes Having Least χ^2- fit for the binary mixture NMP+ DPA at 303.15 K

x_1	U_{exp}	U (Sp-Sp)	U (Sp -Cu)	U (Sp -Te)	U (Sp-disc A)	U (Sp-disc B)	U (Sp-disc C)	U (Sp-disc D)
0	1175	1175	1175	1175	1175	1175	1175	1175
0.137	1213.1	1209.8	1204.9	1202.5	1205.6	1199.5	1202.8	1196.3
0.2631	1245.8	1245.2	1236.2	1231.9	1237.6	1226.2	1232.4	1220.2
0.3797	1279.1	1281.1	1269.1	1263.3	1271	1255.4	1263.9	1247
0.4878	1309.7	1317.8	1303.8	1296.8	1305.9	1287.3	1297.5	1276.9
0.5882	1338.9	1355.1	1340.2	1332.6	1342.5	1322.2	1333.4	1310.5
0.6818	1372.7	1393.1	1378.4	1371	1380.7	1360.3	1371.7	1348.1
0.7692	1410.2	1431.8	1418.7	1411.9	1420.8	1402.1	1412.6	1390.3
0.8511	1447.7	1471.4	1461.1	1455.8	1462.8	1447.7	1456.3	1437.9
0.9278	1494.3	1511.7	1505.8	1502.6	1506.7	1497.8	1502.9	1491.7
1	1552.8	1552.8	1552.8	1552.8	1552.8	1552.8	1552.8	1552.8
	χ^2	1.4788	0.5232	0.6967	0.5599	1.6546	0.6537	3.7821

Table 9: Experimental and theoretical (obtained using SPT) ultrasonic speeds with behavioral shape having least x^2- fit for the binary mixture NMP + DPA at different temperatures

	303.15 K		308.15 K		313.15 K		318.15 K	
x_1	U_{exp}	$U_{(disc\ A\text{-}Sp)}$	U_{exp}	$U_{(disc\ D\text{-}disc\ D)}$	U_{exp}	$U_{(Te\text{-}\ disc\ C)}$	U_{exp}	$U_{(Cu\text{-}Sp)}$
0	1175	1175	1150.6	1150.6	1128.3	1128.3	1107.4	1107.4
0.137	1213.1	1204.9	1187.5	1180.3	1165.3	1157.8	1144.7	1136.7
0.2631	1245.8	1236.2	1221.8	1211.5	1199	1188.8	1177.6	1167.5
0.3797	1279.1	1269.1	1254.2	1244.2	1230.8	1221.3	1209.1	1199.8
0.4878	1309.7	1303.8	1284.5	1278.6	1261	1255.4	1239.4	1233.7
0.5882	1338.9	1340.2	1314.7	1314.8	1290.1	1291.3	1268.5	1269.4
0.6818	1372.7	1378.4	1347.5	1352.9	1324.7	1329.1	1301.5	1307
0.7692	1410.2	1418.7	1384.5	1393	1361.2	1368.8	1338.6	1346.5
0.8511	1447.7	1461.1	1423.1	1435.2	1401.5	1410.6	1380.3	1388.1
0.9278	1494.3	1505.8	1469.3	1479.6	1445.3	1454.7	1425.2	1431.9
1	1552.8	1552.8	1526.4	1526.4	1501	1501	1478	1478
	χ^2	0.5232		0.4858		0.4124		0.3857

CONCLUSION

The experimental values of ultrasonic speed, for the binary mixtures of N-methyl-2-pyrrolidone (NMP) with propylamine (PA), butylamine (BA) and dipropylamine (DPA) at 303.15, 308.15, 313.15 and 318.15 K and at atmospheric pressure over the entire composition range of NMP have been used to study the intermolecular interactions in the mixtures. Theoretical ultrasonic speeds for the three binary mixtures are estimated using SPT and compared with experimental speeds. The shapes of the participating molecules are analyzed using SPT in the temperature range 303.15-318.15 K in all the three binary systems, and it was found that due to the presence of varying intermolecular interactions in the liquid mixtures the shapes of the constituent molecules change at any

given temperature under study. Also, increase in temperature affects the assumed shapes of the constituent molecules in all the liquid mixtures under investigation.

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