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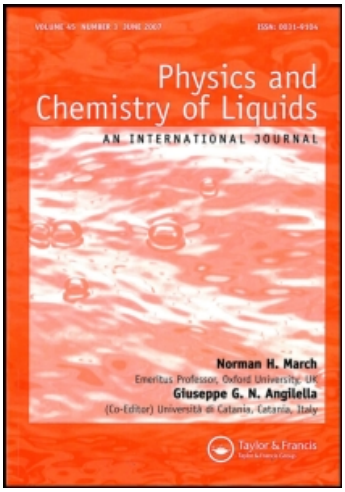

Physics and Chemistry of Liquids
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646857

Ultrasonic velocities and isentropic compressibilities of N-methyl-2-pyrrolidone with ketones and branched alcohols at 303.15 K

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To cite this Article Kumari, P. Gnana , Sekhar, G. Chandra , Ramamohan, K. and Rao, M. V. P.(2006) 'Ultrasonic velocities and isentropic compressibilities of N-methyl-2-pyrrolidone with ketones and branched alcohols at 303.15 K', Physics and Chemistry of Liquids, 44: 6, 701 — 710
To link to this Article: DOI: 10.1080/00319100600814556
URL: http://dx.doi.org/10.1080/00319100600814556

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Physics and Chemistry of Liquids
Vol. 44, No. 6, December 2006, 701–710

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Ultrasonic velocities and isentropic compressibilities of N-methyl-2-pyrrolidone with ketones and branched alcohols at 303.15 K

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(*Received 1 December 2005; in final form 6 March 2006*)

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The ultrasonic sound velocities and densities are measured for the binary mixtures of N-methyl-2-pyrrolidone (NMP) with ketones and branched alcohols at 303.15 K. The ketones include methyl ethyl ketone, methyl propyl ketone, diethyl ketone, methyl isobutyl ketone and cyclohexanone. The branched alcohols include 2-propanol, 2-methyl-1-propanol, 3-methyl-1-butanol, 2-butanol and 2-methyl-2-propanol. The ultrasonic sound velocity data were used to compute isentropic compressibilities (k_s). The deviations in the value of k_s from ideal value were computed. Except cyclohexanone all the binary mixtures formed by NMP with ketones at 303.15 K exhibit negative deviation from ideal behaviour over the entire range of composition. Cyclohexanone with NMP exhibit positive deviation over the entire range of composition. An inversion in the sign of Δk_s from positive to negative was observed for 2-propanol system and negative deviation was observed in four binary mixtures formed by NMP with other branched alcohols at 303.15 K. The ultrasonic sound velocities of these mixtures have been analysed is terms of Free Length Theory (FLT), Collision Factor Theory (CFT) and Nomoto's relation.

Keywords: Ultrasonic velocity; Isentropic compressibility; N-methyl-2-pyrrolidone; Binary mixtures; Derived properties

1. Introduction

In recent years the ultrasonic velocities have been adequately employed in understanding the nature of molecular interactions in binary liquid mixtures. Reddy and Naidu [1,2] studied the molecular interactions of some alcohols in diethyl ketone and isobutyl ketone. Grolier and Benson [3] investigated the binary systems containing ketones. Kehiain and Grolier [4] and Venkateswarlu and Raman [5] analysed the properties of binary mixtures containing ketones in terms of quasi-chemical group

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ISSN 0031-9104 print/ISSN 1029-0451 online © 2006 Taylor & Francis
DOI: 10.1080/00319100600814556

contribution model. Venkatesulu and Rao [6] determined ultrasonic velocities of some binary mixtures containing ketones, Pal *et al.* [7] determined excess volumes and isentropic compressibilities for few binary mixtures. However, no effort appears to have been made to collect the isentropic compressibility data for the binary mixtures of N-methyl-2-pyrrolidone (NMP) with ketones. Hence, new experimental data for isentropic compressibility are reported for the mixtures of NMP with methyl ethyl ketone, methyl propyl ketone, diethyl methyl isobutyl ketone, cyclohexanone, 2-propanol, 2-methyl-1-propanol, 3-methyl-1-butanol, 2-butanol and 2-methyl-2-propanol at 303.15 K. The ultrasonic sound velocities of these mixtures have been analysed in terms of Free Length Theory (FLT) [8–11], Collision Factor Theory (CFT) [8,12,13] and Nomoto's relation [14]. The theoretical aspect has been already discussed in our earlier paper [15].

2. Experimental

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All the chemicals were purified by standard procedures described by Riddick and Bunger [16]. The NMP (Merck, >99%) was distilled under reduced pressure. All liquids were stored over freshly activated molecular sieves of type 3A (Union Carbide). The purity of compounds were checked by comparing the measured densities with those reported in literature [17]. The density data of all the components were measured by a bicapillary pycnometer with an accuracy of two parts in 10^5. The density of the pure components are listed in table 1 and are found to be in agreement with the literature data.

Isentropic compressibilities were computed from the measured ultrasonic velocities and densities, evaluated from the measured excess volumes. Ultrasonic velocities were measured by a single crystal ultrasonic interferometer at 4 MHz frequency at 303.15 K. These were accurate to ±0.02%. A Thermostatically controlled, well-stirred water bath with temperature controlled to (303.15 ± 0.01 K) was used for all measurements.

Table 1. Densities (ρ) of the pure components at 303.15 K and speed of sound of pure components.

	ρ (g cm^{-3})		U_{exp} (ms^{-1})
Substance	Experimental	Literature	
N-methyl-2-pyrrolidone	1.02340	1.01910[a]	1551.6
Methyl ethyl ketone	0.79451	0.79452	1167.0
Diethyl ketone	0.80457	0.80461	1198.0
Methyl propyl ketone	0.79654	0.79656	1204.4
Methyl isobutyl ketone	0.79124	0.79127[b]	1168.0
Cyclohexanone	0.93758	0.93761	1392.0
2-Propanol	0.77688	0.77690	1104.6
2-Methyl-1-propanol	0.79434	0.79437	1170.2
3-Methyl-1-butanol	0.80174	0.80179	1218.0
2-Mutanol	0.79890	0.79896	1192.2
2-Methyl-2-propanol	0.77616	0.77620	1124.0

[a] [18] at 298.15 K; [b] [19].

3. Results and discussion

The densities of binary mixtures are calculated from excess volume data, using the relation:

$$\rho = \frac{x_1 M_1 + x_2 M_2}{V + V^{\mathrm{E}}} \tag{1}$$

where x_1, x_2 denote the mole fractions and M_1, M_2 are molar masses of components 1 and 2 respectively, V is the molar volume of ideal mixture and V^{E} the excess volume of the mixture.

Isentropic compressibilities were calculated from sound velocity (U) and density (ρ).

$$k_{\mathrm{s}} = U^{-2}\rho^{-1} \tag{2}$$

Deviation in isentropic compressibility (Δk_{s}) was obtained from the equation.

$$\Delta k_{\mathrm{s}} = k_{\mathrm{s}} - \varphi_1 k_{\mathrm{s1}} - \varphi_2 k_{\mathrm{s2}} \tag{3}$$

where k_{s}, k_{s1} and k_{s2} are the isentropic compressibilities of the mixture and the pure components respectively. ϕ_1 and ϕ_2 are the volume fractions of components 1 and 2.

The experimental data for ten binary mixtures of ketones or branched alcohols with NMP along with theoretical sound velocities are reported in table 2. The variation of Δk_{s} with volume fraction of NMP with ketones and alkanols, is graphically presented in figures 1 and 2 respectively. The dependence of Δk_{s} on volume fraction has been expressed by the polynomial.

$$\Delta k_{\mathrm{s}} = \varphi_1\varphi_2[b_0 + b_1(\varphi_1 - \varphi_2) + b_2(\varphi_1 - \varphi_2)^2] \tag{4}$$

where b_0, b_1 and b_2 are binary constants obtained by the method of least squares and are given in table 4 along with standard deviation. In mixtures of ketones with NMP, negative deviation is observed for four binary mixtures and cyclohexanone exhibit small positive deviation from ideal behaviour. The Δk_{s} values for the mixtures of NMP with branched alcohols 2-methyl-1-propanol, 3-methyl-1-butanol, 2-butanol and 2-methyl-2-propanol are negative over the entire range of composition at 303.15 K. But an inversion in sign from positive to negative was observed for the mixture of 2-propanol with NMP.

In the case of NMP, the pyrrolidone ring affords better packing in the liquid state as is evidenced by its larger density and sound velocity compared with the other components. The polar nature of the two components leads to the dipolar attractive forces that act between the electron deficient carbon of ketone and negatively charged oxygen of NMP.

Thus the complex formation between the two component molecules lead to a decrease in the intermolecular distances and increase in the sound velocities, thereby decreasing the compressibility. The study of the data shows that complex formation between the component molecules is maximum at the volume fraction 0.5 of NMP. Thus the negative Δk_{s} values indicate that the contribution made by dipole–dipole

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Table 2. Volume fraction (ϕ_1) of N-methyl-2-pyrrolidone, ρ, U, k_s, Δk_s and predicted sound velocities (U) from various models for the binary mixtures of NMP (**1**) with ketones (**2**) and branched alcohols (**2**) at 303.15 K.

					$U_{theoretical}$ (ms^{-1})		
ϕ_1	ρ ($g\,cm^{-3}$)	U_{exp} (ms^{-1})	k_s (TPa^{-1})	Δk_s (TPa^{-1})	FLT	CFT	Nomoto
N-Methyl-2-pyrrolidone (**1**) + methyl ethyl ketone (**2**)							
0.0000	0.79451	1167	924	–	1167.0	1167.0	1167.0
0.1517	0.83054	1214	816	−28	1205.4	1220.4	1220.5
0.2207	0.84676	1245.2	761	−48	1224.7	1245.2	1245.4
0.2988	0.8652	1282	702	−66	1248.0	1273.8	1274.0
0.426	0.89483	1345.2	617	−85	1289.5	1321.3	1321.5
0.5205	0.91661	1391.4	563	−90	1323.7	1357.4	1357.5
0.5952	0.93369	1421.4	530	−85	1352.9	1386.4	1386.5
0.7078	0.95914	1456	491	−65	1400.7	1431.0	1430.9
0.8167	0.98339	1483.2	462	−38	1452.1	1475.1	1474.7
0.9184	1.00574	1519.2	430	−17	1505.1	1517.2	1516.4
1.0000	1.0234	1551.6	405	–	1551.6	1551.6	1551.6
N-methyl-2-pyrrolidone (**1**) + diethyl ketone (**2**)							
0.0000	0.80457	1198.0	866	–	1198.0	1198.0	1198.0
0.1401	0.83752	1230.2	788	−12	1232.2	1249.4	1243.7
0.2265	0.85708	1252.0	744	−17	1255.6	1280.8	1272.5
0.3711	0.88922	1297.2	668	−26	1298.7	1332.8	1321.7
0.4230	0.90070	1316.2	640	−30	1315.2	1351.2	1339.6
0.5121	0.92045	1347.8	598	−32	1345.1	1382.8	1370.9
0.5666	0.93249	1365.2	574	−30	1364.3	1402.0	1390.2
0.6937	0.96046	1415.2	519	−26	1412.4	1446.4	1436.0
0.7900	0.98130	1452.2	483	−19	1452.1	1476.4	1467.5
0.9040	1.00483	1502.4	440	−9	1503.7	1418.0	1513.9
1.0000	1.02340	1551.6	405	–	1551.6	1551.6	1551.6
N-methyl-2-pyrrolidone (**1**) + methyl propyl ketone (**2**)							
0.0000	0.79654	1202.4	868	–	1202.4	1202.4	1202.4
0.1165	0.82433	1230.6	801	−13	1237.8	1245.1	1239.9
0.1930	0.84258	1254	754	−24	1254.0	1272.8	1265.0
0.3417	0.87777	1308	665	−44	1309.5	1326.0	1314.7
0.4382	0.90032	1344.6	614	−51	1332.3	1360.1	1447.7
0.4930	0.91303	1367.4	585	−54	1351.3	1379.3	1366.6
0.6094	0.93974	1402.6	540	−45	1383.6	1419.6	1407.5
0.7189	0.96415	1428.8	508	−27	1424.4	1457.1	1446.6
0.8475	0.99198	1466.6	468	−7	1483.4	1500.7	1493.5
0.9297	1.00918	1498.2	441	−3	1522.1	1528.2	1524.0
1.0000	1.0234	1551.6	405	–	1551.6	1551.6	1551.6
N-methyl-2-pyrrolidone (**1**) + methyl isobutyl ketone (**2**)							
0.0000	0.79124	1168.0	926	–	1168.0	1168.0	1168.0
0.0946	0.81441	1198.0	855	−21	1194.3	1211.3	1201.1
0.1879	0.83688	1232.4	786	−41	1222.2	1252.5	1234.5
0.2615	0.85450	1259.2	738	−52	1245.1	1283.4	1261.0
0.3961	0.88644	1310.4	656	−63	1290.3	1338.0	1310.9
0.4444	0.89782	1331.6	628	−66	1307.5	1356.9	1329.0
0.6087	0.93620	1385.0	556	−52	1370.1	1418.5	1392.2
0.7236	0.96258	1422.4	513	−36	1418.3	1459.4	1437.5
0.8232	0.98502	1456.0	478	−18	1463.2	1493.6	1477.5
0.8989	1.00174	1491.2	448	−9	1499.6	1518.9	1508.5
1.0000	1.02340	1551.6	405	–	1551.6	1551.6	1551.6

(*Continued*)

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Table 2. Continued.

					$U_{\text{theoretical}}$ (ms^{-1})		
ϕ_1	ρ (g cm^{-3})	U_{exp} (ms^{-1})	k_s (TPa^{-1})	Δk_s (TPa^{-1})	FLT	CFT	Nomoto
N-methyl-2-pyrrolidone (**1**) + cyclohexanone (**2**)							
0.1156	0.94792	1392.0	544	6	1363.8	1406.5	1404.3
0.2076	0.95474	1402.0	532	8	1416.2	1422.6	1419.0
0.3050	0.96290	1415.0	518	9	1431.0	1439.3	1434.8
0.4073	0.97145	1426.0	506	11	1447.8	1456.6	1451.4
0.4984	0.97917	1438.0	493	13	1462.0	1471.8	1466.4
0.5729	0.98558	1454.0	479	10	1474.4	1484.0	1478.6
0.7078	0.99722	1480.0	457	8	1497.7	1505.9	1501.1
0.7856	1.00404	1496.0	445	7	1511.6	1518.2	1514.1
0.9104	1.01517	1522.0	425	5	1534.6	1537.8	1535.2
1.0000	1.02340	1551.6	405	–	1551.6	1551.6	1551.6
N-methyl-2-pyrrolidone (**1**) + 2-propanol (**2**)							
0.0000	0.77688	1124.0	1018	–	1124.0	1124.0	1124.0
0.1807	0.82176	1157.2	908	5	1172.2	1188.9	1193.8
0.2458	0.83790	1176.8	861	−2	1192.0	1213.5	1219.8
0.3709	0.86909	1225.6	766	−22	1233.5	1262.4	1271.1
0.4875	0.89801	1277.6	682	−35	1277.5	1310.1	1320.1
0.5586	0.91571	1316.2	630	−43	1306.8	1340.5	1417.1
0.6387	0.93553	1351.0	585	−39	1342.8	1375.8	1385.5
0.7645	0.96646	1416.0	516	−32	1406.0	1433.8	1441.5
0.8463	0.98645	1456.4	477	−21	1452.2	1473.3	1478.8
0.9236	1.00519	1495.8	444	−7	1499.9	1511.1	1514.6
1.0000	1.02340	1551.6	405	–	1551.6	1551.6	1551.6
N-methyl-2-pyrrolidone (**1**) + 2-methyl-1-propanol (**2**)							
0.0000	0.79434	1170.2	919	–	1170.2	1170.2	1170.2
0.1400	0.82728	1216.2	817	−30	1207.0	1242.0	1218.5
0.2265	0.84756	1248.6	756	−46	1231.7	1283.4	1249.5
0.2998	0.86232	1278.0	708	−57	1254.2	1316.8	1276.2
0.4193	0.89463	1326.0	637	−66	1293.6	1367.7	1320.5
0.5237	0.91648	1368.0	583	−67	1331.3	1408.6	1360.0
0.6083	0.93596	1398.2	546	−60	1364.2	1439.2	1392.7
0.6994	0.95676	1432.4	509	−51	1402.3	1469.7	1428.4
0.8102	0.98183	1466.0	473	−29	1452.7	1503.3	1472.1
0.9263	1.00752	1516.2	431	−12	1511.1	1534.2	1519.9
1.0000	1.02340	1551.6	405	–	1551.6	1551.6	1551.6
N-methyl-2-pyrrolidone (**1**) + 3-methyl-1-butanol (**2**)							
0.0000	0.80174	1218.0	840	–	1218.0	1218.0	1218.0
0.0891	0.82207	1242.2	788	−13	1239.8	1248.9	1244.7
0.2493	0.85835	1296.4	693	−39	1282.5	1304.1	1295.2
0.3427	0.87936	1332.0	640	−50	1309.5	1335.8	1325.4
0.3851	0.88887	1348.2	618	−54	1322.2	1350.1	1339.2
0.4937	0.91296	1382.0	573	−52	1356.6	1386.5	1374.9
0.6102	0.93863	1424.0	525	−49	1396.1	1425.1	1414.1
0.6763	0.95306	1442.4	504	−42	1419.9	1446.8	1436.7
0.8549	0.99197	1496.2	450	−18	1489.8	1504.1	1498.8
0.9306	1.00840	1522.4	427	−8	1521.0	1529.4	1525.6
1.0000	1.02340	1551.6	405	–	1551.6	1551.6	1551.6

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Table 2. Continued.

ϕ_1	ρ (g cm^{-3})	U_{exp} (ms^{-1})	k_s (TPa^{-1})	Δk_s (TPa^{-1})	$U_{theoretical}$ (ms^{-1}) FLT	CFT	Nomoto
N-methyl-2-pyrrolidone (**1**) + 2-butanol (**2**)							
0.0000	0.79890	1192.2	880	–	1192.2	1192.2	1192.2
0.1896	0.84261	1236.2	776	−14	1237.6	1255.6	1254.6
0.2571	0.85811	1258.2	736	−22	1255.8	1278.7	1277.7
0.3188	0.87228	1278.0	701	−27	1273.3	1300.0	1299.0
0.4924	0.91175	1336.2	614	−32	1328.3	1361.3	1360.3
0.5844	0.93235	1372.2	569	−33	1361.4	1394.5	1393.6
0.6454	0.94584	1396.4	542	−32	1385.2	1416.9	1415.9
0.7574	0.97060	1442.2	495	−25	1431.6	1458.6	1457.5
0.8151	0.98330	1463.6	474	−18	1457.8	1480.4	1479.3
0.9175	1.00555	1514.2	433	−11	1507.6	1519.5	1518.4
1.0000	1.02340	1551.6	405	–	1551.6	1551.6	1551.6
N-methyl-2-pyrrolidone (**1**) + 2-methyl-2-propanol (**2**)							
0.0000	0.77616	1104.6	1055	–	1104.6	1104.6	1104.6
0.1135	0.80450	1144.2	949	−32	1135.3	1152.2	1149.5
0.2449	0.83745	1194.8	836	−60	1175.4	1208.4	1203.9
0.3246	0.85745	1230.6	770	−74	1202.4	1242.9	1237.8
0.4636	0.89220	1292.4	671	−83	1255.1	1304.1	1298.3
0.5438	0.91211	1328.0	621	−80	1289.2	1340.0	1334.2
0.6392	0.93557	1368.8	570	−69	1333.7	1383.2	1377.6
0.7274	0.95718	1406.4	528	−54	1379.1	1423.6	1418.6
0.8422	0.98506	1456.4	478	−29	1445.4	1476.1	1473.2
0.9248	1.00508	1502.2	440	−13	1498.5	1515.8	1513.3
1.0000	1.02340	1551.6	405	–	1551.6	1551.6	1551.6

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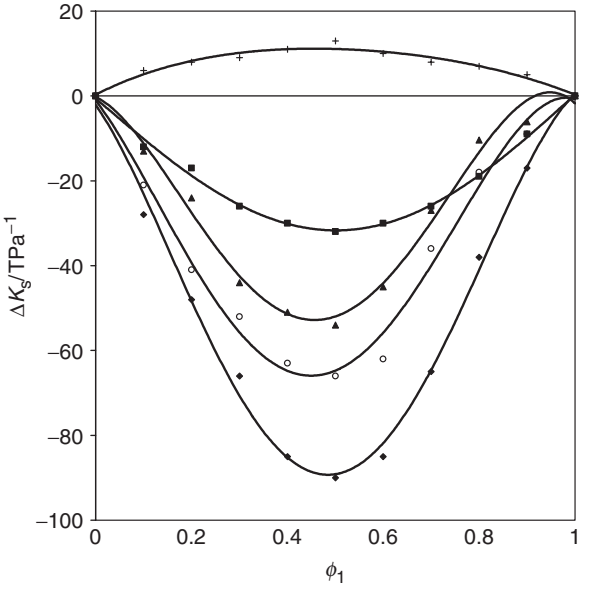

Figure 1. Excess isentropic compressibilities (k_S^E) as a function of volume fraction (ϕ_1) for N-methyl-2-pyrrolidone **(1)** + cyclohexanone**(2)** (+); +diethyl ketone **(2)** (■); + methyl propyl ketone **(2)** (▲) + methyl isobutyl ketone **(2)** (○) and + methyl ethyl ketone **(2)** (◆) at 303.15 K.

interactions between unlike moelcules of the components are predominant. The relatively small Δk_s values in the system NMP with cyclohexanone are due to the fact that the free length is not affected much.

The negative Δk_s for the binary mixture of NMP with methyl ethyl ketone are higher than that compared with the mixture of NMP with diethyl ketone and methyl isobutyl ketone. The lower Δk_s values of the mixtures of NMP with diethyl ketone and methyl isobutyl ketone are probably due to the larger steric hindrance of the isobutyl chain on the carbonyl group in methyl isobutyl ketone molecules. The negative values of Δk_s at equimolar mixtures are found to vary in the following order:

Cyclohexanone > Diethyl ketone > Methyl propyl ketone
> Methyl isobutyl ketone > Methyl ethyl ketone.

The negative Δk_s for the binary mixtures of NMP with 2-propanol, 2-methyl-1-propanol, 3-methyl-1-butanol, 2-butanol and 2-methyl-2-propanol can be explained in terms of contributions made by the following factors: (i) the break-up of hydrogen bonds in branched alcohols with NMP. (ii) The formation of new species acting as an adduct between the branched alcohols and NMP. The dissociation of alcohol leads to an increase in Δk_s and the formation of an adduct leads to decrease in Δk_s. The brief, the former and latter effects contribute to positive and negative deviation in isentropic compressibilities. The observed Δk_s values are the resultant of the aforesaid effects.

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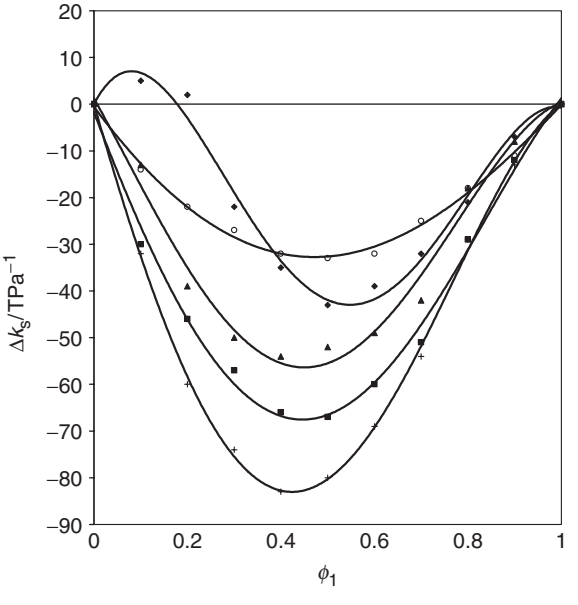

Figure 2. Excess isentropic compressibilities (k_S^E) as a function of volume fraction (ϕ_1) for N-methyl-2-pyrrolidone **(1)** + 2-propanol **(2)** (◆); +2-butanol **(2)** (○); +3-methyl-1-butanol **(2)** (▲) + 2-methyl-1-propanol **(2)** (■) and +2-methyl-2-propanol **(2)** (+) at 303.15 K.

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Table 3. Molar volume (V) molar volume at absolute zero (V_0), available volume (V_a), free length (L_f,) surface area (Y), collision factor (S), molecular radius (R_m) and molecular sound velocity (R) of pure components at 303.15 K.

	$cm^3\,mol^{-1}$			A^0	m^2		A^0	
Component	V	V_0	V_a	L_f	Y	S	r_m	R
N-methyl-2-pyrrolidone	96.867	82.017	14.850	0.4020	73.881	2.0746	2.6587	1121.1
Methyl ethyl ketone	90.758	70.653	20.105	0.6066	66.286	1.6860	2.5353	955.5
Diethyl ketone	107.057	84.783	22.274	0.5872	75.864	1.6895	2.7004	1136.9
Methyl propyl ketone	108.130	85.633	22.497	0.5880	76.521	1.6802	2.7177	1149.8
Methyl isobutylketone	126.589	100.899	25.689	0.6067	84.597	1.6365	2.8618	1333.1
Cyclohexanone	104.680	85.943	18.737	0.4702	79.700	1.8521	2.7289	1167.1
2-Propanol	77.357	58.923	18.434	0.6369	57.887	1.7183	2.3590	804.1
2-Methyl-1-propanol	93.314	79.837	13.477	0.6050	66.2771	1.7341	2.5373	983.1
3-Methyl-1-butanol	109.950	88.082	21.868	0.5786	75.589	1.7463	2.7096	1173.9
2-Butanol	92.781	72.249	20.532	0.5921	69.347	1.7483	2.5428	983.5
2-Methyl-2-propanol	95.499	72.608	22.891	0.6484	70.608	1.5910	2.5812	986.9

Table 4. Least square parameters of equation (4) and standard deviation σ (Δk_s) at 303.15 K.

Binary mixture	b_0 (TPa^{-1})	b_1 (TPa^{-1})	b_2 (TPa^{-1})	σ (Δk_s) (TPa^{-1})
N-methyl-2-pyrrolidone +				
Methyl ethyl ketone	−354.2	−19.5	229.6	2
Diethyl ketone	−122.6	−10.9	40.9	1
Methyl propyl ketone	−209.7	69.3	248.2	2
Methyl isobutyl ketone	−250.0	100.3	124.6	1
Cyclohexanone	41.6	−2.4	23.6	1
2-Propanol	−152.5	−152.8	233.4	2
2-Methyl-1-propanol	−266.7	41.9	88.5	1
3-Methyl-1-butanol	−218.3	24.4	106.2	1
2-Butanol	−130.5	−28.7	30.2	1
2-Methyl-2-propanol	−326.7	70.9	114.2	1

The algebraic values of Δk_s for the binary mixtures of NMP with branched alcohols fall in the following order.

$$\text{2-Propanol} > \text{2-Butanol} > \text{3-Methyl-1-butanol} > \text{2-Methyl-1-propanol} > \text{2-Methyl-2-propanol}.$$

The analysis of the experimental and predicted sound velocities given in table 2 indicate that the FLT, CFT and Nomoto models give a rough estimate of sound velocities in all binary mixtures and FLT satisfactorily estimates sound velocity when compared to CFT and Nomoto models. The surface area (Y), collision factor (S) and molecular sound velocity (R) of the pure components used in the FLT, CFT and Nomoto's relation are calculated using experimental sound velocities and densities. The quantities critical temperature (T_c) and surface tension (σ) are taken from the literature [16]. The properties of pure components required to predict sound velocities data in terms of aforesaid theories are given in table 3.

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Acknowledgement

PGK is extremely thankful to UGC, New Delhi for giving Teacher fellowship.

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