

Density, ultrasonic velocity, viscosity and their excess parameters of the binary mixtures of N,N-dimethylaniline+1-alkanols (C_3 - C_5), +2-alkanols (C_3 - C_4), +2-methyl-1-propanol, +2-methyl-2-propanol at 303.15 K

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Abstract—The density, ultrasonic velocity of sound and viscosity of binary mixtures of N,N-dimethyl aniline (N,N-DMA) with 1-propanol, +2-propanol, +1-butanol, +2-butanol, +1-pentanol, +2-methyl-1-propanol, +2-methyl-2-propanol were measured at 303.15 K. These experimental data have been used to calculate excess volume V^E , excess ultrasonic speeds u^E , excess intermolecular free length L_f^E , excess acoustic impedance Z^E , excess isentropic compressibility κ_s^E , deviation in viscosity $\Delta\eta$ and excess Gibbs free energy of activation of viscous flow (G^E). The values of L_f^E and κ_s^E are negative over the wide range of composition for all the binary mixtures, while the values of Z^E are positive. These results have been used to discuss the nature of interaction between unlike molecules in terms of hydrogen bonding, dipole-dipole interaction, proton-acceptor interaction and dispersive forces. The viscosity data have been correlated using three equations: Grunberg and Nissan, Katti & Chaudhri and Hind et al. The excess/deviations were fitted by a Redlich-Kister equation and the results were analyzed in terms of specific interactions present in these mixtures.

Key words: Ultrasonic Velocity, Viscosity, Density, Excess Thermodynamic Parameters

INTRODUCTION

Complex formation in liquid mixtures has been extensively studied using optical and ultrasonic technique. The formation of hydrogen bond in solutions and its effect on the physical properties of the mixtures have received much attention. Hydrogen bonding plays an important role in fundamental sciences and in industrial applications. Although many experimental and theoretical studies have been directed towards understanding hydrogen bonding, it remains an area of active research. Knowledge of physico-chemical properties of liquid mixtures formed by two or more components associated through hydrogen bonds is important from theoretical and process design aspects. From the theoretical viewpoint, volumetric properties of these mixtures are important sources of information for the characterization of the interactions between the components and they are also useful for understanding the liquid state theory. In addition, alcohols and amines are widely used in a variety of industrial and consumer applications and hence, the knowledge of their physical properties is also of great importance from a practical point of view. The liquids were chosen in the present investigation on the basis of their industrial importance. N,N-dimethylaniline is used as a reagent for methyl furfural, hydrogen peroxide and as a solvent for manufacture of vanillin, michlers ketones. Alcohols are used as hydraulic fluids in pharmaceutical and cosmetics, in medications for animals, in manufacturing perfumes, paint removers, flavors

and dyestuffs, as defrosting and as an antiseptic agent.

In continuation of our research program to determine the thermodynamic and transport properties of binary liquid mixtures [1-4], we report herein the excess volume V^E , excess ultrasonic sound velocity u^E , excess isentropic compressibility κ_s^E , excess intermolecular free length L_f^E , excess acoustic impedance Z^E , deviation in viscosity $\Delta\eta$ and excess Gibbs free energy of activation of viscous flow (G^E) of the binary systems: N,N-dimethylaniline with homologous series of aliphatic alcohols (1-propanol, 2-propanol, 1-butanol, 2-butanol, 1-pentanol, 2-methyl-1-propanol and 2-methyl-2-propanol). These results have been used to discuss the nature of interaction between unlike molecules in terms of hydrogen bonding, dipole-dipole interaction, proton-acceptor interaction and dispersive forces, and also used to analyze the effect of branching in the alkanol and position of the hydroxyl group in the interaction with the N,N-dimethylaniline. A survey of the literature reveals that these parameters are not reported for the selected systems. N,N-dimethylaniline is stabilized by dipole-dipole interactions, whereas alcohols are self-associated through the hydrogen bonding of their hydroxyl groups, creating multimers of different degrees.

The present work was undertaken to determine the effect of position of -OH group of an alcohol molecule that may influence both the sign and magnitude of various thermodynamic functions when it is mixed with N,N-dimethylaniline. Recently, substantial research work has been reported on the excess properties of methyl isobutyl ketone+alkanols [5], 1,3-Dioxolane+Monoalcohols [6], acetonitrile with alkanols-1 [7], 2-Butanone with Branched alcohols [8], Anisole with Alkanols [9], Methyl Methacrylate with alcohols [10], Tet-

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Table 1. Comparison of experimental and literature values of density ρ viscosity η for pure components at 298.15 K

Components	$\rho \times 10^{-3} / \text{Kg} \cdot \text{m}^{-3}$		$\eta / \text{mPa} \cdot \text{s}$	
	Experimental	Literature	Experimental	Literature
N,N-dimethyl aniline	0.95199	0.95200 ^a	1.280	1.283 ^a
2-Methyl-1-propanol	0.79978	0.79980 ^b	3.334	3.333 ^b
2-Methyl-2-propanol	0.78119	0.78120 ^b	4.437	4.438 ^b
1-Propanol	0.79961	0.79960 ^b	1.947	1.943 ^b
2-Propanol	0.78124	0.78126 ^b	2.044	2.043 ^b
1-Butanol	0.80573	0.80575 ^b	2.568	2.571 ^b
2-Butanol	0.80239	0.80241 ^b	2.996	2.998 ^b
1-Pentanol	0.81112	0.81110 ^c	3.351	3.350 ^c

^aReference [14]^bReference [15]^cReference [16]

rahydrofuran with Methol and o-Cresol [11], Aniline with alcohols [12] while that on N,N-dimethylaniline with alkanols is relatively rare. These results have been fitted to the Redlich-Kister polynomial equation [13] the binary coefficients to estimate the standard deviation between experimental and calculated data. Results have been used to explain the nature of interaction between unlike molecules in terms of hydrogen bonding, dipole-dipole interaction, proton-acceptor interaction and dispersive forces.

EXPERIMENTAL

N,N-dimethylaniline $\geq 99\%$ of purity, 1-propanol $\geq 99\%$ of purity, 2-propanol $\geq 99\%$ of purity, 1-butanol $\geq 99\%$ of purity, 2-butanol $\geq 99\%$ of purity, 1-pentanol $\geq 99\%$ of purity, 2-methyl-1-propanol $\geq 99.4\%$ of purity and 2-methyl-2-propanol $\geq 98\%$ of purity were purchased from Merck (S.D. Fine Chemicals Ltd., India). All the liquids were used after single distillation and the alcohols were stored over type 3 A \times 1.5 mm molecular sieves from Merck. The purities of

the samples were further confirmed by GLC single sharp peaks. The purity of chemicals was checked by comparing the measured densities, which were in good agreement with literature values [14–16] as can be seen in Table 1.

Binary mixtures of N,N-dimethylaniline with 1-propanol, 2-propanol, 1-butanol, 2-butanol, 1-pentanol, 2-methyl-1-propanol and 2-methyl-2-propanol were prepared in glass bottles with air tight stoppers and adequate precaution was taken to minimize evaporation losses to weights of solutions measured by using Acculab ALC-210.4 digital electronic balance with a precision of ± 0.1 mg. The uncertainty in solution composition expressed in mole fraction was found to be less than 1×10^{-4} . After mixing, the bubble-free homogeneous sample was transferred into the U-tube of the densimeter through a syringe. The density measurements were performed with a Rudolph Research Analytical digital densimeter (DDH-2911 Model), equipped with a built-in solid-state thermostat and a resident program with accuracy of temperature of $303.15 \text{ K} \pm 0.02 \text{ K}$. The estimated accuracy in the density measurement is $\pm 2 \times 10^{-5} \text{ Kg} \cdot \text{m}^{-3}$.

Table 2. Calculated values of density (ρ), ultrasonic sound velocity (u), excess ultrasonic sound velocity (u^E), excess molar volume (V^E), excess intermolecular free length (L_f^E) and excess acoustic impedance (Z^E) of the binary mixtures at 303.15 K

N,N-dimethyl aniline (1)+2-methyl-2-propanol (2)							
x_1	$\rho \times 10^{-3} / \text{Kg} \cdot \text{m}^{-3}$	$u \text{ m} \cdot \text{sec}^{-1}$	$u^E \text{ m} \cdot \text{sec}^{-1}$	$V^E \text{ m}^3 \cdot \text{mol}^{-1}$	$\kappa_s^E \times 10^{-12} \text{ Pa}^{-1}$	$L_f^E \times 10^{-9} \text{ m}$	$Z^E \times 10^{-3} \text{ kgm}^{-2} \text{ s}^{-1}$
0.0000	0.77629	1061.3	0.000	0.00000	0.000	0.000	0.000
0.0534	0.78917	1084.3	0.694	−0.10368	−3.057	−0.965	0.970
0.1054	0.80110	1106.9	1.574	−0.17945	−5.515	−1.740	1.969
0.1689	0.81494	1134.4	2.550	−0.24587	−7.843	−2.475	3.013
0.2256	0.82670	1159.0	3.466	−0.28737	−9.398	−2.965	3.933
0.2958	0.84054	1189.2	4.344	−0.31865	−10.68	−3.370	4.802
0.3612	0.85282	1217.2	5.026	−0.33641	−11.34	−3.577	5.501
0.4502	0.86867	1254.8	5.451	−0.34722	−11.48	−3.623	6.038
0.5246	0.88124	1285.9	5.474	−0.34903	−11.05	−3.486	6.234
0.6589	0.90246	1341.2	4.677	−0.33240	−9.195	−2.901	5.838
0.7421	0.91469	1375.0	3.724	−0.30437	−7.468	−2.356	5.091
0.8046	0.92339	1400.2	2.818	−0.26679	−5.924	−1.869	4.215
0.8645	0.93133	1424.3	1.898	−0.21424	−4.271	−1.348	3.171
0.9472	0.94159	1457.6	0.654	−0.10294	−1.744	−0.550	1.362
1.0000	0.94767	1479.0	0.000	0.00000	0.000	0.000	0.000

Table 2. Continued

N,N-dimethyl aniline (1)+2-methyl-1-propanol (2)							
x_1	$\rho \times 10^{-3} / \text{Kg} \cdot \text{m}^{-3}$	$u \text{ m} \cdot \text{sec}^{-1}$	$u^E \text{ m} \cdot \text{sec}^{-1}$	$V^E \text{ m}^3 \cdot \text{mol}^{-1}$	$\kappa_s^E \times 10^{-12} \text{ Pa}^{-1}$	$L_f^E \times 10^{-9} \text{ m}$	$Z^E \times 10^{-3} \text{ kgm}^{-2} \text{ s}^{-1}$
0.0000	0.79417	1075.2	0.000	0.00000	0.000	0.000	0.000
0.0545	0.80616	1098.3	1.093	-0.08914	-2.779	-0.877	1.664
0.1056	0.81685	1119.9	2.059	-0.15512	-4.901	-1.546	3.060
0.1875	0.83300	1154.5	3.588	-0.23432	-7.475	-2.358	5.111
0.2546	0.84541	1182.7	4.693	-0.27856	-8.907	-2.810	6.527
0.3526	0.86234	1223.4	5.820	-0.31501	-10.05	-3.169	7.972
0.4278	0.87449	1254.3	6.354	-0.32724	-10.28	-3.242	8.670
0.5047	0.88623	1285.4	6.402	-0.32775	-10.00	-3.156	8.838
0.5869	0.89807	1318.3	6.110	-0.31563	-9.236	-2.914	8.582
0.6514	0.90688	1343.8	5.565	-0.29704	-8.330	-2.628	7.994
0.7348	0.91767	1376.5	4.588	-0.25932	-6.820	-2.152	6.822
0.8025	0.92596	1402.7	3.450	-0.21754	-5.336	-1.684	5.413
0.8621	0.93290	1425.7	2.384	-0.16858	-3.872	-1.221	3.961
0.9285	0.94023	1451.4	1.272	-0.09714	-2.090	-0.659	2.207
1.0000	0.94767	1479.0	0.000	0.00000	0.000	0.000	0.000
N,N-dimethyl aniline (1)+2-propanol (2)							
x_1	$\rho \times 10^{-3} / \text{Kg} \cdot \text{m}^{-3}$	$u \text{ m} \cdot \text{sec}^{-1}$	$u^E \text{ m} \cdot \text{sec}^{-1}$	$V^E \text{ m}^3 \cdot \text{mol}^{-1}$	$\kappa_s^E \times 10^{-12} \text{ Pa}^{-1}$	$L_f^E \times 10^{-9} \text{ m}$	$Z^E \times 10^{-3} \text{ kgm}^{-2} \text{ s}^{-1}$
0.0000	0.77799	1112.2	0.000	0.00000	0.000	0.000	0.000
0.0564	0.79400	1134.2	1.312	-0.07401	-2.867	-0.905	5.026
0.1136	0.80901	1156.4	2.532	-0.13582	-5.153	-1.626	9.332
0.1989	0.82936	1189.3	4.143	-0.20497	-7.592	-2.395	14.40
0.2689	0.84446	1216.0	5.167	-0.24304	-8.855	-2.794	17.37
0.3245	0.85562	1237.3	6.073	-0.27043	-9.503	-2.998	19.34
0.3767	0.86535	1256.8	6.426	-0.27862	-9.779	-3.085	20.26
0.4258	0.87396	1274.9	6.517	-0.27875	-9.809	-3.095	20.56
0.4878	0.88422	1297.8	6.675	-0.27768	-9.608	-3.031	20.64
0.5655	0.89610	1326.1	6.475	-0.26258	-8.970	-2.830	19.75
0.6305	0.90531	1349.5	6.033	-0.24194	-8.157	-2.573	18.28
0.7385	0.91931	1387.9	4.818	-0.19218	-6.327	-1.996	14.56
0.8187	0.92877	1416.1	3.601	-0.14335	-4.642	-1.464	10.86
0.9243	0.94017	1452.8	1.567	-0.06591	-2.060	-0.650	4.875
1.0000	0.94767	1479.0	0.000	0.00000	0.000	0.000	0.000
N,N-dimethyl aniline (1)+1-propanol (2)							
x_1	$\rho \times 10^{-3} / \text{Kg} \cdot \text{m}^{-3}$	$u \text{ m} \cdot \text{sec}^{-1}$	$u^E \text{ m} \cdot \text{sec}^{-1}$	$V^E \text{ m}^3 \cdot \text{mol}^{-1}$	$\kappa_s^E \times 10^{-12} \text{ Pa}^{-1}$	$L_f^E \times 10^{-9} \text{ m}$	$Z^E \times 10^{-3} \text{ kgm}^{-2} \text{ s}^{-1}$
0.0000	0.79577	1194.8	0.000	0.00000	0.000	0.000	0.000
0.0503	0.80883	1210.6	1.505	-0.07401	-1.666	-0.526	5.707
0.1049	0.82198	1227.5	2.887	-0.13582	-3.113	-0.982	10.90
0.1825	0.83900	1251.2	4.534	-0.20497	-4.632	-1.461	16.70
0.2575	0.85380	1273.7	5.719	-0.24304	-5.587	-1.763	20.61
0.3423	0.86884	1298.6	6.518	-0.27043	-6.158	-1.943	23.17
0.3929	0.87706	1313.3	6.838	-0.27862	-6.289	-1.984	23.93
0.4121	0.88004	1318.8	6.881	-0.27875	-6.297	-1.987	24.03
0.4703	0.88866	1335.4	6.941	-0.27768	-6.214	-1.961	23.91
0.5523	0.89983	1358.5	6.736	-0.26258	-5.836	-1.841	22.65
0.6229	0.90867	1378.1	6.272	-0.24194	-5.297	-1.671	20.64
0.7221	0.92005	1405.2	5.179	-0.19218	-4.253	-1.342	16.53
0.8096	0.92931	1428.8	3.912	-0.14335	-3.105	-0.980	12.03
0.9023	0.93848	1453.4	2.166	-0.06591	-1.684	-0.531	6.428
1.0000	0.94767	1479.0	0.000	0.00000	0.000	0.000	0.000

Table 2. Continued

N,N-dimethyl aniline (1)+2-butanol (2)							
x_1	$\rho \times 10^{-3} / \text{Kg} \cdot \text{m}^{-3}$	$u \text{ m} \cdot \text{sec}^{-1}$	$u^E \text{ m} \cdot \text{sec}^{-1}$	$V^E \text{ m}^3 \cdot \text{mol}^{-1}$	$\kappa_s^E \times 10^{-12} \text{ Pa}^{-1}$	$L_f^E \times 10^{-9} \text{ m}$	$Z^E \times 10^{-3} \text{ kgm}^{-2} \text{ s}^{-1}$
0.0000	0.79855	1152.2	0.000	0.00000	0.000	0.000	0.000
0.0452	0.80814	1169.0	2.029	-0.05375	-1.696	-0.535	2.862
0.0845	0.81618	1183.3	3.485	-0.09476	-2.931	-0.925	5.008
0.1321	0.82555	1200.3	4.930	-0.13592	-4.163	-1.313	7.210
0.1893	0.83633	1220.3	6.237	-0.17667	-5.309	-1.675	9.333
0.2452	0.84637	1239.4	7.069	-0.20566	-6.111	-1.928	10.83
0.2756	0.85163	1249.7	7.434	-0.21651	-6.440	-2.032	11.49
0.3065	0.85686	1260.0	7.636	-0.22676	-6.692	-2.111	11.97
0.3655	0.86646	1279.7	8.055	-0.23535	-7.008	-2.211	12.73
0.4292	0.87634	1300.6	8.137	-0.23575	-7.088	-2.236	13.01
0.4906	0.88541	1320.7	8.172	-0.22710	-6.966	-2.198	13.04
0.6021	0.90085	1356.7	7.734	-0.19253	-6.270	-1.978	12.17
0.7323	0.91742	1398.0	6.484	-0.13189	-4.806	-1.516	9.851
0.8911	0.93588	1447.0	3.589	-0.04775	-2.227	-0.703	5.051
1.0000	0.94767	1479.0	0.000	0.00000	0.000	0.000	0.000
N,N-dimethyl aniline (1)+1-butanol (2)							
x_1	$\rho \times 10^{-3} / \text{Kg} \cdot \text{m}^{-3}$	$u \text{ m} \cdot \text{sec}^{-1}$	$u^E \text{ m} \cdot \text{sec}^{-1}$	$V^E \text{ m}^3 \cdot \text{mol}^{-1}$	$\kappa_s^E \times 10^{-12} \text{ Pa}^{-1}$	$L_f^E \times 10^{-9} \text{ m}$	$Z^E \times 10^{-3} \text{ kgm}^{-2} \text{ s}^{-1}$
0.0000	0.80201	1235.6	0.000	0.00000	0.000	0.000	0.000
0.0489	0.81201	1250.1	2.598	-0.03783	-1.231	-0.389	4.050
0.0742	0.81704	1257.3	3.640	-0.05697	-1.765	-0.557	5.831
0.1249	0.82682	1271.4	5.399	-0.09239	-2.674	-0.844	8.966
0.1802	0.83704	1286.2	6.739	-0.12524	-3.430	-1.082	11.64
0.2362	0.84692	1300.7	7.609	-0.15015	-3.982	-1.256	13.63
0.265	0.85181	1308.1	7.999	-0.15785	-4.203	-1.326	14.47
0.2944	0.85667	1315.5	8.243	-0.16185	-4.375	-1.380	15.09
0.3549	0.86629	1330.5	8.517	-0.16061	-4.597	-1.450	15.90
0.4190	0.87593	1346.2	8.615	-0.14318	-4.667	-1.473	16.16
0.4854	0.88536	1362.4	8.654	-0.10896	-4.592	-1.449	15.93
0.5905	0.89927	1387.7	8.372	-0.02916	-4.184	-1.320	14.47
0.7423	0.91774	1423.5	7.224	0.09228	-3.082	-0.972	10.62
0.9099	0.93710	1460.9	3.830	0.11199	-1.252	-0.395	4.404
1.0000	0.94767	1479.0	0.000	0.00000	0.000	0.000	0.000
N,N-dimethyl aniline (1)+1-pentanol (2)							
x_1	$\rho \times 10^{-3} / \text{Kg} \cdot \text{m}^{-3}$	$u \text{ m} \cdot \text{sec}^{-1}$	$u^E \text{ m} \cdot \text{sec}^{-1}$	$V^E \text{ m}^3 \cdot \text{mol}^{-1}$	$\kappa_s^E \times 10^{-12} \text{ Pa}^{-1}$	$L_f^E \times 10^{-9} \text{ m}$	$Z^E \times 10^{-3} \text{ kgm}^{-2} \text{ s}^{-1}$
0.0000	0.80761	1245.2	0.000	0.00000	0.000	0.000	0.000
0.0799	0.82098	1269.0	5.119	-0.05888	-1.693	-0.534	4.550
0.1210	0.82761	1280.3	6.810	-0.07397	-2.318	-0.731	6.041
0.2034	0.84041	1301.9	9.145	-0.07398	-3.224	-1.017	7.954
0.2784	0.85155	1320.5	10.21	-0.04579	-3.710	-1.170	8.598
0.3584	0.86297	1339.6	10.61	0.00563	-3.953	-1.247	8.484
0.4284	0.87263	1356.0	10.64	0.06202	-3.990	-1.259	8.018
0.5047	0.88286	1373.6	10.40	0.12968	-3.868	-1.220	7.215
0.5722	0.89173	1389.2	10.22	0.18516	-3.658	-1.154	6.582
0.6406	0.90059	1404.8	9.828	0.23217	-3.338	-1.053	5.856
0.7103	0.90956	1420.5	9.232	0.26134	-2.914	-0.919	5.138
0.7810	0.91865	1436.1	8.302	0.26549	-2.383	-0.752	4.386
0.8168	0.92328	1443.8	7.632	0.25395	-2.074	-0.654	3.969
0.9058	0.93494	1461.9	4.924	0.17677	-1.171	-0.369	2.485
1.0000	0.94767	1479.0	0.000	0.00000	0.000	0.000	0.000

Proper calibration at 303.15 K temperature was achieved with doubly distilled, deionized water and with air as standards. A multi-frequency ultrasonic interferometer (M-82 Model, Mittal Enterprise, New Delhi, India) operated at 2 MHz, was used to measure the ultrasonic velocities of binary liquid mixtures (with an accuracy of $\pm 0.2\%$) at 303.15 K. A thermostatically controlled, well-stirred circulated water bath with temperature controlled to 303.15 ± 0.02 K was used for all the ultrasonic sound velocity measurements. The viscosities of pure and liquid mixtures were determined by a suspended Ubbelohde-type viscometer described earlier [17]. The estimated uncertainty in viscosity is ± 0.005 mPa·s.

RESULTS AND DISCUSSION

The experimental values of density ρ , viscosity η and ultrasonic

velocity u of pure liquids and their mixtures as function of mole fraction of N,N-dimethylaniline at 303.15 K are given in Tables 2 and 3. The derived parameters such as κ_s , L_f , and Z were calculated using the following relations:

$$\kappa_s = u^{-2} \rho^{-1} \quad (1)$$

$$L_f = K/u (\rho)^{1/2} \quad (2)$$

$$Z = u \rho \quad (3)$$

where ρ is the density and u is the ultrasonic speed of solutions. K is a temperature-dependent constant [18]. The deviations in excess functions from ideality provide a relatively better tool to assess the strength of interaction between the component molecules of the binary mixtures. V^E , κ_s^E , $\Delta\eta$ and u^E were calculated from experimental data using the following relevant expressions:

Table 3. Experimental values of viscosity (η), deviation in viscosities ($\Delta\eta$), excess Gibbs free energy of activation of viscous flow (G^{*E}), Grunberg - Nissan interaction parameters (d), Katti-Chaudhri interaction parameters (W_{vis}/RT) and Hind interaction parameters (H_{12}) at 303.15 K

N,N-dimethyl aniline (1)+2-methyl-2-propanol (2)						
x_1	η mPa·s	$\Delta\eta$ mPa·s	G^{*E} J·mol ⁻¹	d'	W_{vis}/RT	H_{12}
0.0000	3.378	0.000	0.000			
0.0534	3.315	0.055	0.043	0.323	0.335	2.817
0.1054	3.242	0.097	0.079	0.324	0.336	2.786
0.1689	3.141	0.136	0.120	0.328	0.340	2.758
0.2256	3.041	0.161	0.151	0.332	0.345	2.734
0.2958	2.907	0.182	0.185	0.339	0.352	2.710
0.3612	2.775	0.194	0.210	0.349	0.362	2.695
0.4502	2.587	0.202	0.237	0.368	0.381	2.683
0.5246	2.423	0.202	0.252	0.388	0.401	2.680
0.6589	2.112	0.188	0.256	0.440	0.452	2.692
0.7421	1.908	0.167	0.240	0.486	0.498	2.711
0.8046	1.747	0.144	0.215	0.531	0.542	2.732
0.8645	1.584	0.113	0.175	0.585	0.595	2.757
0.9472	1.340	0.052	0.086	0.677	0.687	2.790
1.0000	1.172	0.000	0.000			
N,N-dimethyl aniline (1)+2-methyl-1-propanol (2)						
x_1	η mPa·s	$\Delta\eta$ mPa·s	G^{*E} J·mol ⁻¹	d'	W_{vis}/RT	H_{12}
0.0000	2.845	0.000	0.000			
0.0545	2.804	0.050	0.039	0.285	0.301	2.495
0.1056	2.755	0.087	0.071	0.282	0.298	2.467
0.1875	2.663	0.132	0.115	0.285	0.301	2.440
0.2546	2.576	0.157	0.146	0.289	0.305	2.422
0.3526	2.434	0.179	0.181	0.298	0.314	2.400
0.4278	2.316	0.187	0.200	0.308	0.324	2.389
0.5047	2.189	0.188	0.213	0.322	0.338	2.385
0.5869	2.044	0.181	0.217	0.340	0.356	2.381
0.6514	1.926	0.171	0.214	0.358	0.374	2.384
0.7348	1.767	0.151	0.199	0.390	0.406	2.396
0.8025	1.628	0.126	0.174	0.420	0.435	2.404
0.8621	1.501	0.098	0.141	0.457	0.472	2.421
0.9285	1.345	0.053	0.083	0.485	0.501	2.410
1.0000	1.172	0.000	0.000			

Table 3. Continued

N,N-dimethyl aniline (1)+2-propanol (2)						
x_1	η mPa·s	$\Delta\eta$ mPa·s	G^{*E} J·mol ⁻¹	d'	W_{vis}/RT	H_{12}
0.0000	1.806	0.000	0.000			
0.0564	1.727	-0.043	-0.015	-0.166	-0.109	1.082
0.1136	1.655	-0.079	-0.028	-0.165	-0.109	1.096
0.1989	1.560	-0.120	-0.044	-0.165	-0.110	1.112
0.2689	1.492	-0.144	-0.055	-0.165	-0.112	1.124
0.3245	1.439	-0.161	-0.066	-0.172	-0.120	1.121
0.3767	1.401	-0.166	-0.069	-0.168	-0.117	1.135
0.4258	1.369	-0.167	-0.070	-0.165	-0.114	1.147
0.4878	1.330	-0.167	-0.072	-0.165	-0.115	1.155
0.5655	1.288	-0.159	-0.072	-0.165	-0.116	1.164
0.6305	1.259	-0.147	-0.068	-0.164	-0.116	1.172
0.7385	1.220	-0.118	-0.057	-0.164	-0.117	1.184
0.8187	1.199	-0.088	-0.044	-0.163	-0.117	1.192
0.9243	1.180	-0.040	-0.021	-0.161	-0.116	1.203
1.0000	1.172	0.000	0.000			
N,N-dimethyl aniline (1)+1-propanol (2)						
x_1	η mPa·s	$\Delta\eta$ mPa·s	G^{*E} J·mol ⁻¹	d'	W_{vis}/RT	H_{12}
0.0000	1.735	0.000	0.000			
0.0503	1.685	-0.022	-0.003	-0.086	-0.022	1.226
0.1049	1.625	-0.051	-0.012	-0.113	-0.049	1.182
0.1825	1.552	-0.080	-0.020	-0.116	-0.055	1.184
0.2575	1.482	-0.108	-0.033	-0.129	-0.068	1.171
0.3423	1.413	-0.129	-0.044	-0.137	-0.078	1.166
0.3929	1.377	-0.137	-0.049	-0.140	-0.082	1.166
0.4121	1.365	-0.138	-0.050	-0.140	-0.082	1.168
0.4703	1.328	-0.142	-0.055	-0.144	-0.088	1.168
0.5523	1.290	-0.134	-0.052	-0.140	-0.084	1.182
0.6229	1.265	-0.119	-0.046	-0.132	-0.077	1.199
0.7221	1.242	-0.086	-0.028	-0.110	-0.056	1.238
0.8096	1.225	-0.054	-0.013	-0.086	-0.032	1.277
0.9023	1.198	-0.029	-0.006	-0.081	-0.028	1.289
1.0000	1.172	0.000	0.000			

$$V^E = [x_1 M_1 + x_2 M_2] / \rho_m - [x_1 M_1 / \rho_1 + x_2 M_2 / \rho_2] \quad (4)$$

$$\kappa_s^E = \kappa_s - [x_1 \kappa_{s1} + x_2 \kappa_{s2}] \quad (5)$$

$$\Delta\eta = \eta - [x_1 \eta_1 + x_2 \eta_2] \quad (6)$$

$$u^E = u - [x_1 u_1 + x_2 u_2] \quad (7)$$

M_1 , M_2 , κ_{s1} , κ_{s2} , η_1 , η_2 , u_1 , u_2 , ρ_1 , ρ_2 , ρ_m , κ_s , η , and u represent molecular weight, isentropic compressibility, viscosity, ultrasonic sound velocity and density data of component 1, component 2 and mixture, respectively. The variation of V^E , κ_s^E , $\Delta\eta$ and u^E with mole fraction was fitted to the Redlich-Kister equation [13] of the type,

$$Y^E = x_1 x_2 [a_0 + a_1 (x_1 - x_2) + a_2 (x_1 - x_2)^2] \quad (8)$$

where Y^E is V^E or u^E or κ_s^E or $\Delta\eta$. The values of a_0 , a_1 and a_2 are the coefficients of the polynomial equation and were obtained by method of least squares and are given in Table 4 along with standard deviation values at 303.15 K. The standard deviations are calculated by using the equation

$$\sigma(Y^E) = [\sum (Y_{obs}^E - Y_{cal}^E)^2 / (n - m)]^{1/2} \quad (9)$$

where n is the total number of experimental points and m is the number of coefficients.

The excess Gibbs free energy of activation of viscous flow (G^{*E}) is obtained by the equation

$$G^{*E} = RT [\ln \eta V - (x_1 \ln \eta_1 V_1 + x_2 \ln \eta_2 V_2)] \quad (10)$$

where V_1 , V_2 and V are the molar volumes of the component 1 and component 2 and molar volume of the mixture, respectively, R and T have their usual meanings.

Grunberg and Nissan [19] proposed the following equation for the measurement of viscosity of liquid mixtures:

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 d' \quad (11)$$

where d is a parameter proportional to interchange energy, which

Table 3. Continued

N,N-dimethyl, aniline (1)+2-butanol (2)						
x_1	η mPa·s	$\Delta\eta$ mPa·s	G^{*E} J·mol ⁻¹	d'	W_{vis}/RT	H_{12}
0.0000	2.742	0.000	0.000			
0.0452	2.686	0.015	0.021	0.179	0.197	2.130
0.0845	2.638	0.028	0.040	0.186	0.205	2.142
0.1321	2.582	0.047	0.062	0.197	0.216	2.163
0.1893	2.512	0.067	0.087	0.207	0.226	2.175
0.2452	2.442	0.085	0.110	0.217	0.235	2.186
0.2756	2.402	0.092	0.120	0.221	0.240	2.189
0.3065	2.362	0.101	0.131	0.227	0.246	2.195
0.3655	2.279	0.110	0.148	0.235	0.254	2.196
0.4292	2.183	0.114	0.161	0.242	0.261	2.191
0.4906	2.086	0.114	0.168	0.249	0.268	2.185
0.6021	1.893	0.096	0.165	0.256	0.274	2.158
0.7323	1.65	0.057	0.134	0.253	0.272	2.104
0.8911	1.353	0.010	0.060	0.228	0.247	2.008
1.0000	1.172	0.000	0.000			
N,N-dimethyl aniline (1)+1-butanol (2)						
x_1	η mPa·s	$\Delta\eta$ mPa·s	G^{*E} J·mol ⁻¹	d'	W_{vis}/RT	H_{12}
0.0000	2.226	0.000	0.000			
0.0489	2.200	0.025	0.024	0.183	0.204	1.973
0.0742	2.187	0.039	0.036	0.189	0.210	1.984
0.1249	2.156	0.061	0.058	0.191	0.212	1.981
0.1802	2.120	0.083	0.080	0.196	0.217	1.983
0.2362	2.079	0.102	0.100	0.200	0.220	1.981
0.2650	2.056	0.109	0.109	0.201	0.222	1.979
0.2944	2.032	0.116	0.117	0.204	0.224	1.978
0.3549	1.978	0.126	0.131	0.207	0.228	1.974
0.4190	1.915	0.130	0.142	0.211	0.231	1.967
0.4854	1.844	0.129	0.148	0.214	0.235	1.958
0.5905	1.722	0.118	0.147	0.219	0.241	1.943
0.7423	1.526	0.082	0.119	0.223	0.247	1.914
0.9099	1.295	0.028	0.051	0.222	0.248	1.870
1.0000	1.172	0.000	0.000			

reflects the non-ideality of the system.

Katti and Chaudhri [20] proposed the following equation:

$$\ln \eta V = x_1 \ln V_1 \eta_1 + x_2 \ln V_2 \eta_2 + x_1 x_2 W_{vis}/RT \quad (12)$$

where W_{vis}/RT is an interaction term.

Hind et al. [21] suggested an equation for the viscosity of binary liquid mixtures as

$$\eta = x_1^2 \eta_1 + x_2^2 \eta_2 + 2x_1 x_2 H_{12} \quad (13)$$

where H_{12} is Hind interaction parameter.

The sign and magnitude of u^E play an important role in describing molecular rearrangements as a result of molecular interactions occurring among the component molecules in the mixtures. An examination of data in the Table 2 suggests that the excess ultrasonic velocity (u^E) is positive in all the binary systems over the entire range of composition. Positive deviations indicate the increasing strength of interaction between component molecules of binary liquid mix-

tures [22]. If strong interactions arise among the components of a mixture, leading to the formation of molecular aggregates and more compact structures, then sound will travel faster through the mixture by means of longitudinal waves and hence the speed of sound deviations with respect to a linear behavior will be positive. While if the structure-breaking factor in the mixture predominates, resulting in expansion, then the speed of sound through the mixture will be slower, resulting into negative deviation in speed of sound [23]. The positive values of κ_s and L_f are observed with the opposite trend of Z values.

An examination of data in the Table 2 suggests that the excess isentropic compressibility (κ_s^E) and excess intermolecular free length (L_f^E) are negative in all the binary systems over the entire range of composition. According to Sri Devi et al. [24], the negative excess values are due to the closely packed molecules, which accounts for the existence of strong molecular interaction, whereas positive excess values causes weak interaction between unlike molecules. The sign

Table 3. Continued

N,N-dimethyl aniline (1)+1-pentanol (2)						
x_1	η mPa·s	$\Delta\eta$ mPa·s	G^{*E} J·mol ⁻¹	d'	W_{vis}/RT	H_{12}
0.0000	2.835	0.000	0.000			
0.0799	2.750	0.047	0.044	0.237	0.239	2.329
0.1210	2.704	0.070	0.066	0.243	0.246	2.333
0.2034	2.605	0.108	0.105	0.254	0.258	2.337
0.2784	2.506	0.134	0.136	0.265	0.269	2.337
0.3584	2.392	0.153	0.163	0.277	0.282	2.336
0.4284	2.284	0.161	0.182	0.287	0.294	2.333
0.5047	2.158	0.162	0.193	0.300	0.307	2.328
0.5722	2.039	0.155	0.197	0.312	0.320	2.321
0.6406	1.913	0.143	0.194	0.325	0.334	2.314
0.7103	1.777	0.123	0.180	0.338	0.348	2.302
0.7810	1.635	0.098	0.157	0.354	0.364	2.292
0.8168	1.562	0.085	0.141	0.364	0.375	2.288
0.9058	1.374	0.045	0.085	0.385	0.398	2.269
1.0000	1.172	0.000	0.000			

Table 4. Coefficients of Redlich-Kister equation and standard deviation values at 303.15 K

Binary mixtures	Functions	a_0	a_1	a_2	σ
N,N-DMA+2-methyl-2-propanol	V^E (m ³ ·mol ⁻¹)	-1.394	-0.001	-0.822	0.001
	u^E (msec ⁻¹)	22.281	-0.730	-10.667	0.042
	κ_s^E (Pa ⁻¹)	-44.991	14.227	-3.442	0.015
	$\Delta\eta$ (mPa·s)	0.811	-0.025	0.318	0.001
N,N-DMA+2-methyl-1-propanol	V^E (m ³ ·mol ⁻¹)	-1.312	0.131	-0.370	0.001
	u^E (msec ⁻¹)	25.701	-1.532	-7.684	0.035
	κ_s^E (Pa ⁻¹)	-40.126	12.575	-3.037	0.015
	$\Delta\eta$ (mPa·s)	0.755	-0.067	0.181	0.002
N,N-DMA+2-propanol	V^E (m ³ ·mol ⁻¹)	-1.101	0.258	-0.078	0.001
	u^E (msec ⁻¹)	26.56	-1.359	-4.034	0.017
	κ_s^E (Pa ⁻¹)	-37.947	13.516	-4.591	0.065
	$\Delta\eta$ (mPa·s)	-0.663	0.132	-0.031	0.001
N,N-DMA+1-propanol	V^E (m ³ ·mol ⁻¹)	-0.711	0.746	0.306	0.001
	u^E (msec ⁻¹)	27.702	-4.029	0.150	0.021
	κ_s^E (Pa ⁻¹)	-24.392	8.765	-2.971	0.019
	$\Delta\eta$ (mPa·s)	-0.552	0.132	0.230	0.003
N,N-DMA+2-butanol	V^E (m ³ ·mol ⁻¹)	-0.905	0.461	0.091	0.003
	u^E (msec ⁻¹)	32.548	-4.481	12.832	0.036
	κ_s^E (Pa ⁻¹)	-27.578	8.881	-4.132	0.029
	$\Delta\eta$ (mPa·s)	0.451	-0.184	-0.331	0.001
N,N-DMA+1-butanol	V^E (m ³ ·mol ⁻¹)	-0.395	1.340	0.980	0.001
	u^E (msec ⁻¹)	34.396	-3.331	22.251	0.031
	κ_s^E (Pa ⁻¹)	-18.107	6.013	-3.341	0.022
	$\Delta\eta$ (mPa·s)	0.517	-0.137	-0.096	0.001
N,N-DMA+1-pentanol	V^E (m ³ ·mol ⁻¹)	0.498	1.740	0.231	0.001
	u^E (msec ⁻¹)	41.858	-5.953	31.634	0.032
	κ_s^E (Pa ⁻¹)	-15.332	5.332	-3.841	0.032
	$\Delta\eta$ (mPa·s)	0.649	-0.078	-0.087	0.001

of excess isentropic compressibility (κ_s^E) and excess intermolecular free length (L_f^E) play a vital role in assessing the compactness due

to molecular interaction in liquid mixtures through hydrogen-bonding, charge transfer, dipole-dipole interactions and dipole-induced

dipole interactions, interstitial accommodation and orientational ordering [25], leading to more compact structure making negative excess isentropic compressibility and excess intermolecular free length values. Hence, negative values of excess isentropic compressibility (κ_s^E) and excess intermolecular free length (L_f^E) in the present investigation suggest that dipole-dipole interactions are prevailing through H-bonding in the liquid mixtures [26].

An examination of data in the Table 2 suggests that the excess acoustic impedance Z^E is positive in all the binary systems over the entire range of composition. Trends in Z^E values (Table 2) reinforce our earlier view that there is the possibility of specific intermolecular reactions in all mixtures. Positive values of Z^E in all mole fraction concentration further hint to the possibility of the presence of strong attractive forces between the reacting components of the mixture.

The algebraic negative κ_s^E values of N,N-dimethylaniline with alcohols fall in the order:

1-pentanol < 1-butanol < 2-butanol < 1-propanol < 2-propanol < 2-methyl-1-propanol < 2-methyl-2-propanol

The V^E is the resultant contribution from several opposing effects, namely chemical, physical and structural. The chemical or specific interactions result in volume contraction, leading to negative excess volume and these include charge-transfer complexes, dipole-dipole and dipole-induced dipole interactions, proton-acceptor interactions and H-bonding between unlike molecules finally leading to the more effective packing in the mixture than in the pure liquids. The physical interactions or nonspecific interactions are weak and hence contribute positive V^E , while the structural contributions are mostly negative and arise from several effects such as interstitial accommodation and geometrical fitting of one component into another due to the differences in the molar volume and free volume between components. The negative V^E values of the investigated systems assume heteroassociates that are forming complexes in the alcohol+amine mixtures have stronger O-H...N bonds than O-H...O bonds. This can be explained qualitatively by the fact that the free electron pair around the N atoms with less S and more P character has a higher polarizability and acts as a good proton acceptor for the donor -OH groups of the alcohols, which are more efficient than the -OH group itself.

An examination of data in the Table 2 suggests that the excess volume data for the binary systems 1-Alkanols (C_3 - C_4), 2-Alkanols (C_3 - C_4), 2-methyl-1-propanol, 2-methyl-2-propanol are negative over the entire composition range at 303.15 K, whereas an inversion in sign for the binary mixture containing N,N-dimethyl aniline with 1-pentanol was observed.

The negative V^E values imply that volume contraction takes place upon mixing N,N-dimethylaniline with alkanols due to hydrogen bond formation between dissimilar molecules. It is well known that alkanols are strongly self associated with degree of association depending on such variables as chain length, the position of the -OH group, temperature and dilution by other substances. The strength of bonding in alkanols is expected to decrease with increase in their chain length and molar volume, implying that higher alkanols have smaller proton-donor capacity. N,N-dimethylaniline favors dipole-dipole interactions due to its high dipole moment.

The decrease in negative V^E with increasing chain length of alkanols indicates that weak dipole-dipole interactions are prevailing in higher

alkanols owing to their decrease in polarizability with increasing chain length of alkanols.

The more negative V^E values for N,N-dimethylaniline+2-Alkanols when composed to N,N-dimethylaniline+1-Alkanols may be due to the fact that in 2-Alkanols the presence of $-CH_3$ groups at the α -carbon atom increases the electron density at the oxygen atom of hydroxyl group to a greater extent than in 1-alkanols, which has one alkyl group on the α -carbon atom, resulting in stronger interaction (H-bonding) in former. More negative V^E values for N,N-dimethylaniline+2-methyl-2-propanol mixture (Fig. 1) when compared with of N,N-dimethylaniline+2-methyl-1-propanol may be ascribed to the fact that, in 2-methyl-2-propanol, the presence of three $-CH_3$ groups at the α -carbon atom increases the electron density at the oxygen atom to a greater extent than that in 2-methyl-1-propanol, which has two $-CH_3$ groups on the α -carbon atom, resulting in stronger interaction in the N,N-dimethylaniline+2-methyl-2-propanol mixture. Hence, it may be concluded that the interaction

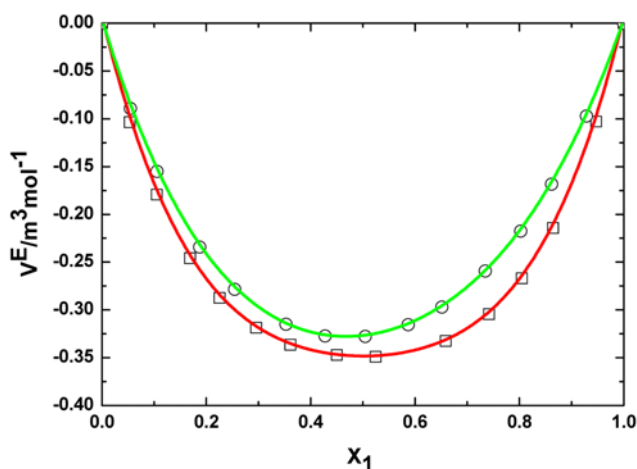


Fig. 1. Variation of excess molar volume (V^E) with mole fraction (x_1) of N,N-DMA in the binary liquid mixtures of N,N-DMA with 2-methyl-2-propanol (\square), 2-methyl-1-propanol (\circ) at 303.15 K.

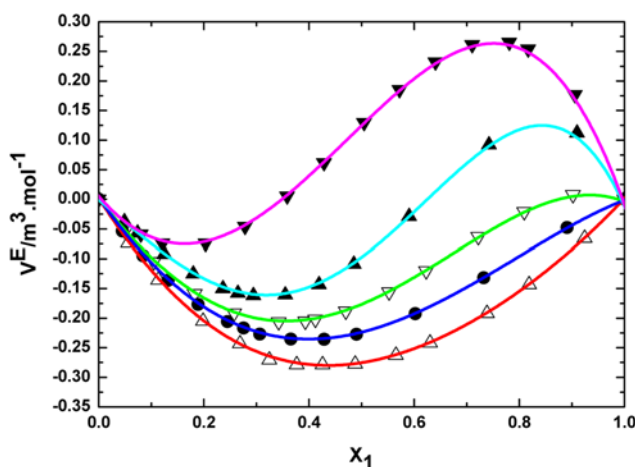


Fig. 2. Variation of excess molar volume (V^E) with mole fraction (x_1) of N,N-DMA in the binary liquid mixtures of N,N-DMA with 2-propanol (Δ), 1-propanol (∇), 2-butanol (\bullet), 1-butanol (\blacktriangle), 1-pentanol (\blacktriangledown) at 303.15 K.

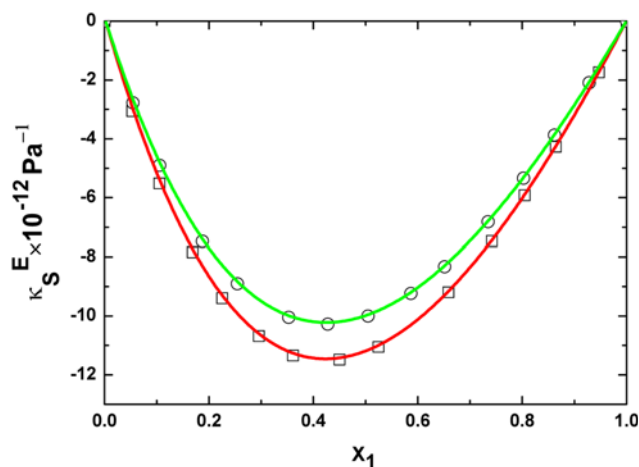


Fig. 3. Deviation of excess isentropic compressibility (κ_S^E) with mole fraction (x_1) of N,N-DMA in the binary liquid mixtures of N,N-DMA with 2-methyl-2-propanol (□), 2-methyl-1-propanol (○) at 303.15 K.

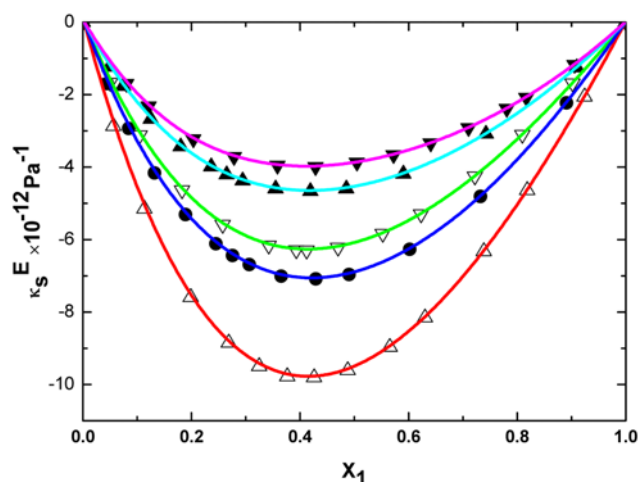


Fig. 4. Deviation of excess isentropic compressibility (κ_S^E) with mole fraction (x_1) of N,N-DMA in the binary liquid mixtures of N,N-DMA with 2-propanol (△), 1-propanol (▽), 2-butanol (●), 1-butanol (▲), 1-pentanol (▼) at 303.15 K.

between N,N-dimethylaniline and alcohols under study increases when a hydroxyl group is attached to a carbon atom with a greater number of $-\text{CH}_3$ groups, i.e., an increase in the number of $-\text{CH}_3$ groups at the α -carbon atom of alkanols. Similar results have been reported for N-methyl acetamide with aromatic hydrocarbon [27] phenyl acetonitrile with aliphatic alcohol [28] and aniline with 1-alkanols [12,29].

The algebraic negative V^E values of N,N-dimethylaniline with Alkanols fall in the order:

1-pentanol < 1-butanol < 2-butanol < 1-propanol < 2-propanol < 2-methyl-1-propanol < 2-methyl-2-propanol.

As number of $-\text{CH}_3$ groups at the α -carbon atom increases, negative V^E value increases due to increase in positive inductive effect than that in 2-methyl-1-propanol which has two $-\text{CH}_3$ groups on the β -carbon atom.

Table 3 shows that the values of η , G^E , W_{vis}/RT and d' are positive

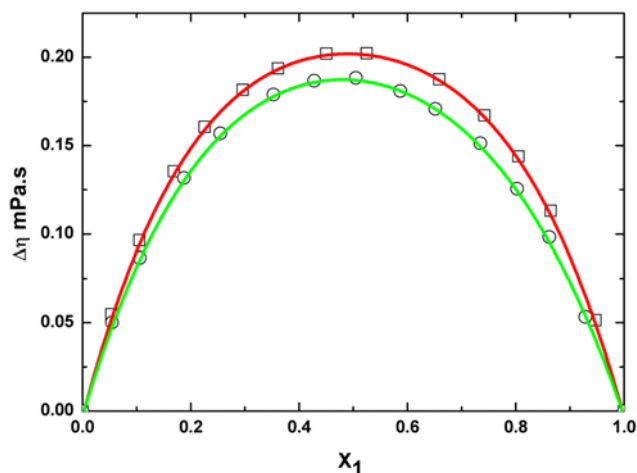


Fig. 5. Deviation of viscosity ($\Delta\eta$) with mole fraction (x_1) of N,N-DMA in the binary liquid mixtures of N,N-DMA with 2-methyl-2-propanol (□), 2-methyl-1-propanol (○) at 303.15 K.

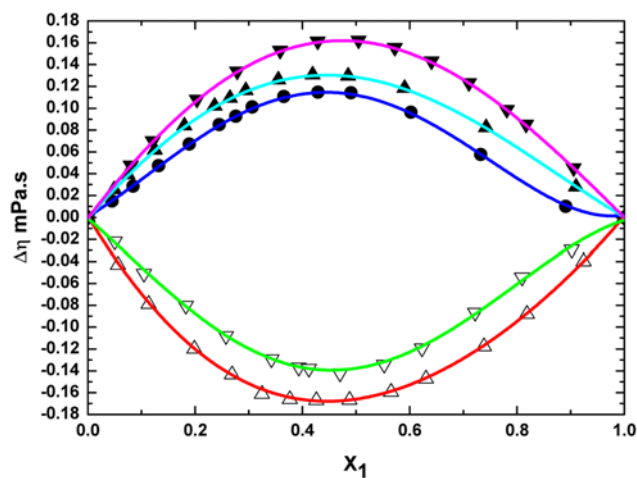


Fig. 6. Deviation of viscosity ($\Delta\eta$) with mole fraction (x_1) of N,N-DMA in the binary liquid mixtures of N,N-DMA with 2-propanol (△), 1-propanol (▽), 2-butanol (●), 1-butanol (▲), 1-pentanol (▼) at 303.15 K.

for N,N-dimethylaniline with 1-butanol, +2-butanol, +1-pentanol, +2-methyl-1-propanol, 2-methyl-2-propanol and negative for the 1-propanol and 2-propanol systems. The sign and magnitude of excess viscosity depend on the combined effect of factors such as molecular size, shape and intermolecular forces [30]. The positive values of $\Delta\eta$ indicate the presence of a specific interaction such as the formation of hydrogen bonding of the type $\text{O-H}\cdots\text{N}$. The negative values of $\Delta\eta$ in the systems of N,N-dimethylaniline with 1-propanol, +2-propanol suggest that mutual loss of specific interactions in like molecules outweigh the specific interactions between unlike molecules [31,32]. Negative deviation in viscosity may also occur where dispersion forces are dominant, particularly, for the systems having different molecular sizes [32,33]. According to Reed et al. [34], the positive deviation in G^E may be attributed to specific interactions like hydrogen bonding and charge transfer, whereas the negative deviations may be ascribed to dispersion forces with systems. Ali

et al. [35] attribute the increasing positive values of G^E in liquid mixtures to hydrogen bond formation between unlike molecules. In the present investigation, the positive G^E obtained in the binary systems shows formation of H-bonding between unlike molecules. The interaction parameter d' in the Grunberg and Nissan equation is a measure of the strength of interaction between the mixing components. In the present investigation, positive values of d' indicate the presence of strong interactions and negative values of d' indicate the presence of weak interactions between the component molecules. Further, Kalra et al. [36] suggested that the negative d' values arise due to strong hydrogen bonds with in the associated component molecules of alcohol when mixed with N,N-dimethylaniline.

CONCLUSIONS

The densities, viscosities and speed of sound at 303.15 K over the entire range of composition of N,N-dimethylaniline with 1-propanol, +2-propanol, +1-butanol, +2-butanol, +1-pentanol, +2-methyl-1-propanol and 2-methyl-2-propanol have been measured. From these measured physical property data, excess molar volumes, deviation in viscosity, deviation in ultrasonic speeds and deviations in isentropic compressibility have been calculated and correlated by a Redlich-Kister type polynomial equation to derive the coefficients and standard deviation. These results have been used to discuss the nature of interaction between unlike molecules in terms of hydrogen bonding, dipole-dipole interaction, proton-acceptor interaction and dispersive forces. Interestingly, the interaction of tertiary alcohols is stronger than that of secondary alcohols in mixtures with N,N-dimethyl aniline. Also, it is clear that the magnitude depends on the chain length of the alcohol and position of the hydroxyl group in the alcohol molecules.

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