J. Chem. Eng. Data **2009,** *54,* 3307–3312 **3307**

Density, Viscosity, Refractive Index, and Ultrasonic Speed of Binary Mixtures of 1,3-Dioxolane with 2-Methoxyethanol, 2-Ethoxyethanol, 2-Butoxyethanol, 2-Propylamine, and Cyclohexylamine

Lovely Sarkar and Mahendra Nath Roy*

Department of Chemistry, University of North Bengal, Darjeeling 734013, India

---

The excess molar volumes (V^E) and viscosity deviations ($\Delta\eta$) have been calculated from experimental density (ρ) and viscosity (η) data of the binary mixtures of 1,3-dioxolane (DO) with 2-methoxyethanol (ME), 2-ethoxyethanol (EE), 2-butoxyethanol (BE), 2-propylamine (PA), and cyclohexylamine (CA) measured over the entire range of composition at 298.15 K. Moreover, deviations in isentropic compressibility (ΔK_S) and molar refraction (ΔR) have been calculated from ultrasonic speed of sounds (u) and refractive indices (n_D) of the mixtures, respectively. The excess or deviation properties were fitted to the Redlich−Kister polynomial equation to derive the adjustable parameters (A_k) and corresponding standard deviations (σ).

Introduction

The mixture thermodynamic properties derived from a measurement of density, viscosity, ultrasonic speed of sound, and refractive index are useful to design engineering processes. The mixing of different solvents gives rise to solutions that generally behave differently. This deviation is expressed by many thermodynamic variables, particularly by excess properties. Excess thermodynamic properties of solvent mixtures, thus, are useful in the study of molecular interactions and arrangements. This work is a part of our program to provide data for the characterization of the molecular interactions between solvents in binary systems.[1,2] The study of binary mixtures of 1,3-dioxolane (DO (1)) with other solvents is of great interest because of its wide use as an important industrial solvent and is miscible with water. On the other hand, alkoxyethanols are of considerable interest for studying the heteroproximity effects of the etheric oxygen on the −O−H bond. 2-Propylamine (PA (5)) and cyclohexylamine (CA (6)) are important in characterizing the associated nature of the liquids in mixtures because of the presence of both a proton donor and a proton acceptor, and they form water insoluble compounds of medical importance.[3]

In the present work, solutions of 2-methoxyethanol (ME (2)), 2-ethoxyethanol (EE (3)), 2-butoxyethanol (BE (4)), PA (5), and CA (6) with DO (1) have been investigated. Such oxygen-containing cyclic ethers, i.e., DO (1), can interact through H-bonding with the hydroxyl (−OH) groups of the alcohols and amine (−NH_2) groups of amines. The experimental data are used to calculate excess molar volumes (V^E), viscosity deviations ($\Delta\eta$), deviations in isentropic compressibility (ΔK_S), and molar refraction (ΔR) of the mixtures.

The excess or deviation properties have been fitted to the Redlich−Kister polynomial equation to obtain their coefficients and standard deviations.

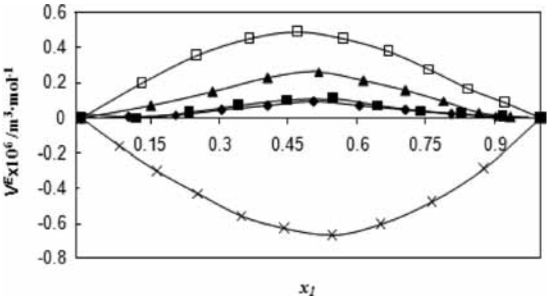

Figure 1. Excess molar volumes, $V^E \cdot 10^6$/m$^3\cdot$mol^{-1}, for binary mixtures of 1,3-dioxolane (1) with: ♦, 2-methoxyethanol (2); ■, 2-ethoxyethanol (3); ▲, 2-butoxyethanol (4); ×, 2-propylamine (5); and □, cyclohexylamine (6) as a function of mole fraction of 1,3-dioxolane (1) at 298.15 K.

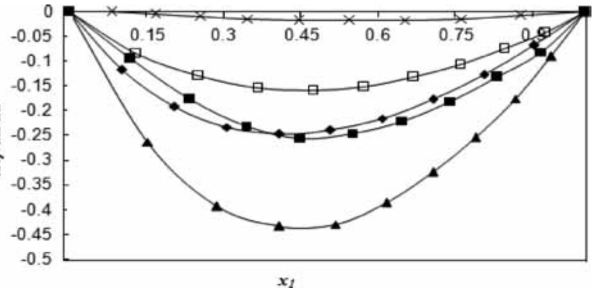

Figure 2. Viscosity deviations, $\Delta\eta$/mPa·s, for binary mixtures of 1,3-dioxolane (1) with: ♦, 2-methoxyethanol (2); ■, 2-ethoxyethanol (3); ▲, 2-butoxyethanol (4); ×, 2-propylamine (5); and □, cyclohexylamine (6) as a function of mole fraction of 1,3-dioxolane (1) at 298.15 K.

Experimental Section

Materials. ME (2), EE (3), and BE (4) were purchased from S. D. Fine Chemicals, AR, India, and purified by standard methods.[4] PA (5) and CA (6) were procured from Merck, India, and were used as purchased. DO was purchased from Aldrich and used without further purification. The pure chemicals were stored over activated 4 Å molecular sieves to reduce water content before use. The mole percent purities for the liquids used as checked by GC (HP6890) using an FID detector were better than 99.8. Thus, the chemicals after purification were 99.8

---

* Corresponding author. Tel.: +91 353 2776381. Fax: +91 353 2699001. E-mail: mahendraroy2002@yahoo.co.in.

10.1021/je900240s CCC: $40.75

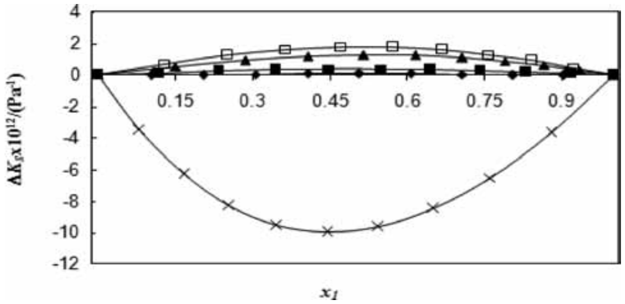

Figure 3. Deviations in isentropic compressibility, $\Delta K_S \cdot 10^{12}/\mathrm{Pa}^{-1}$, for binary mixtures of 1,3-dioxolane (1) with: ♦, 2-methoxyethanol (2); ■, 2-ethoxyethanol (3); ▲, 2-butoxyethanol (4); ×, 2-propylamine (5); and □, cyclohexylamine (6) as a function of mole fraction of 1,3-dioxolane (1) at 298.15 K.

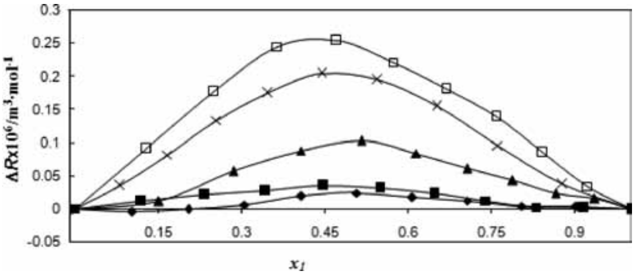

Figure 4. Deviations in molar refraction, $\Delta R \cdot 10^6/\mathrm{m}^3 \cdot \mathrm{mol}^{-1}$, for binary mixtures of 1,3-dioxolane (1) with: ♦, 2-methoxyethanol(2); ■, 2-ethoxyethanol (3); ▲, 2-butoxyethanol (4); ×, 2-propylamine (5); and □, cyclohexylamine (6) as a function of mole fraction of 1,3-dioxolane (1) at 298.15 K.

% pure, and their purity was ascertained by comparing experimental values of densities and viscosities with those reported in the literature[5-15] as presented in Table 1.

Apparatus and Procedures. Densities were measured with an Ostwald-Sprengel-type pycnometer having a bulb volume of about 25 cm^3 and an internal diameter of the capillary of about 0.1 cm. The pycnometer was calibrated at 298.15 K with doubly distilled water. The pycnometer with experimental liquid was equilibrated in a glass-walled thermostatted water bath maintained at ± 0.01 K of the desired temperature. The pycnometer was then removed from the thermostat, properly dried, and weighed in an electronic balance with a precision of ± 0.01 mg. Adequate precautions were taken to avoid evaporation losses during the time of measurements. An average of triplicate measurement was taken into account. The uncertainty of density values is within $\pm 3 \cdot 10^{-4}\ \mathrm{g \cdot cm^{-3}}$. The details of the methods and measurement techniques had been described earlier.[16]

Solvent viscosities were measured by means of a suspended Ubbelohde-type viscometer, calibrated at 298.15 K with doubly distilled water and purified methanol using density and viscosity values from the literature.[17,18] A thoroughly cleaned and perfectly dried viscometer filled with experimental liquid was placed vertically in the glass-walled thermostat maintained to ± 0.01 K. After attainment of thermal equilibrium, efflux times of flow were recorded with a stopwatch correct to ± 0.1 s. At least three repetitions of each data reproducible to ± 0.1 s were taken to average the flow times. The uncertainty of viscosity values is ± 0.003 mPa·s. The details of the methods and measurement techniques had been described earlier.[19]

Ultrasonic speeds of sound were determined by a multifrequency ultrasonic interferometer (Mittal Enterprise, New Delhi) working at 1 MHz, calibrated with water, methanol, and benzene at 298.15 K. The precision of ultrasonic speed measurements was $\pm 0.2\ \mathrm{m \cdot s^{-1}}$. The temperature stability was maintained within ± 0.01 K by circulating thermostatted water around the cell with a circulating pump. The details of the methods and techniques have been described elsewhere.[20]

Refractive indices (n_D) were measured with a Abbe refractometer. The values of refractive index were obtained using sodium D light. The temperature was controlled to ± 0.01 K with circulating thermostat water to a jacketed sample vessel. Calibration was performed periodically using double-distilled water. The sample mixtures were directly injected into the prism assembly of the instrument using an airtight hypodermic syringe, and an average of four measurements was taken for each mixture. The uncertainty of the refractive index measurement was estimated to be ± 0.0002 units.

Results and Discussion

The experimental densities ρ, viscosities η, excess molar volumes V^E, and viscosity deviations $\Delta\eta$ for the binary mixtures of DO (1)[5-7] with ME (2),[8-12] EE (3),[9,13,14] BE (4),[9,14,15] PA (5),[9] and CA (6)[9] along with the coreresponding mole fractions of DO (1), x_1, at 298.15 K are given in Table 2. The excess molar volume V^E was calculated using the equation[21,22]

$$V^{E} = \sum_{i=1}^{2} x_i M_i (1/\rho - 1/\rho_i) \qquad (1)$$

where ρ is the density of the mixture and M_i, x_i, and ρ_i, are the molecular weight, mole fraction, and density of the *i*th component in the mixture, respectively.

The excess molar volume for the five binary systems has been depicted in Figure 1 as a function of mole fraction of DO (1) at 298.15 K. It is seen that the values of V^E (see Table 2) for all the experimental binary mixtures are positive except PA (5), and the largest values of V^E were in the mole fraction range $x_1 = 0.45$ to 0.6. The positive values of V^E for the five binary systems decrease in the following order:

Table 1. Comparison of Density ρ, Viscosity η, Speed of Sound u, and Refractive Index n_D with Literature Data at 298.15 K

	$\rho \cdot 10^{-3}/\mathrm{kg \cdot m^{-3}}$		$\eta/\mathrm{mPa \cdot s}$		$u/\mathrm{m \cdot s^{-1}}$		n_D	
pure solvent	expt	lit	expt	lit	expt	lit	expt	lit
1,3-dioxolane	1.0572	1.05862[6]	0.588	0.592[7]	1338.2	1338.8[5]	1.3980	1.3983[7]
2-methoxyethanol	0.9603	0.96002[8] 0.9603[9]	1.562	1.540[9] 1.5414[12]	1340.2	1340.2[9] 1341.9[10]	1.4002	1.4002[11]
2-ethoxyethanol	0.9256	0.9250[9] 0.9256[13]	1.850	1.850[9,13]	1302.8	1302.8[9] 1302.51[14]	1.4051	–
2-butoxyethanol	0.8966	0.8965[9] 0.8966[15]	2.795	2.792[9] 2.795[15]	1303.4	1303.2[9] 1305.84[14]	1.4176	–
2-propylamine	0.6815	0.6815[9]	0.278	0.278[9]	1075.6	1075.6[9]	1.3759	–
cyclohexylamine	0.8668	0.8668[9]	1.753	1.753[9]	1416.4	1416.4[9]	1.4565	–

$$\text{DO (1)} + \text{CA (6)} > \text{DO (1)} + \text{BE (4)} > \text{DO (1)} + \text{EE (3)} > \text{DO (1)} + \text{ME (2)} > \text{DO (1)} + \text{PA (5)}$$

A perusal of Table 2 shows that the values of V^E generally increase as the chain length of the alkyl group of the alkoxyethanol increases. This indicates dominance of disruption forces for the binary systems containing alkoxyethanols in DO (1). As the +I effect of the alkyl group increases, the disruption forces become more effective in characterizing these binary systems. This may be due the increased +I effect and size of the alkyl group, thereby decreasing the ether−ether interactions in DO (1). A similar result was also reported by us earlier.[9] On the contrary, the amine systems behaved rather in an opposite manner, while the V^E values were negative for the PA (5) system and those for CA (6) were positive. This implies dominance of specific interaction[21] for the component molecules of DO (1)

Table 2. Values of Density ρ, Viscosity η, Excess Molar Volume V^E, and Viscosity Deviation for the Binary Mixtures of DO (1) at 298.15 K

x_1	$\rho \cdot 10^{-3}$ / $\text{kg} \cdot \text{m}^{-3}$	η / $\text{mPa} \cdot \text{s}$	$V^E \cdot 10^6$ / $\text{m}^3 \cdot \text{mol}^{-1}$	$\Delta\eta$ / $\text{mPa} \cdot \text{s}$
		DO (1) + ME (2)		
0.0000	0.9600	1.562	0.0000	0.000
0.1024	0.9691	1.345	0.0064	−0.118
0.2043	0.9780	1.171	0.0181	−0.193
0.3057	0.9869	1.032	0.0429	−0.233
0.4065	0.9959	0.920	0.0720	−0.247
0.5067	1.0052	0.829	0.0906	−0.239
0.6064	1.0152	0.756	0.0712	−0.215
0.7056	1.0255	0.698	0.0451	−0.177
0.8043	1.0360	0.652	0.0201	−0.127
0.9024	1.0465	0.616	0.0094	−0.068
1.0000	1.0572	0.588	0.0000	0.000
		DO (1) + EE (3)		
0.0000	0.9250	1.850	0.0000	0.000
0.1191	0.9367	1.605	0.0016	−0.095
0.2332	0.9484	1.379	0.0313	−0.177
0.3427	0.9603	1.187	0.0691	−0.230
0.4478	0.9725	1.030	0.1052	−0.255
0.5488	0.9854	0.912	0.1075	−0.245
0.6460	0.9991	0.814	0.0744	−0.221
0.7395	1.0132	0.737	0.0410	−0.180
0.8295	1.0275	0.671	0.0230	−0.133
0.9163	1.0422	0.613	0.0071	−0.081
1.0000	1.0572	0.588	0.0000	0.000
		DO (1) + BE (4)		
0.0000	0.8966	2.795	0.0000	0.000
0.1506	0.9099	2.200	0.0719	−0.263
0.2851	0.9235	1.773	0.1475	−0.393
0.4061	0.9374	1.465	0.2295	−0.434
0.5154	0.9521	1.229	0.2633	−0.429
0.6147	0.9681	1.053	0.2134	−0.386
0.7053	0.9848	0.916	0.1544	−0.323
0.7882	1.0021	0.801	0.0977	−0.254
0.8645	1.0202	0.712	0.0332	−0.176
0.9349	1.0385	0.642	0.0072	−0.090
1.0000	1.0572	0.588	0.0000	0.000
		DO (1) + PA (5)		
0.0000	0.6815	0.278	0.0000	0.000
0.0814	0.7079	0.303	−0.1569	−0.001
0.1663	0.7363	0.324	−0.3032	−0.005
0.2548	0.7668	0.346	−0.4284	−0.011
0.3472	0.7999	0.372	−0.5551	−0.014
0.4438	0.8354	0.398	−0.6308	−0.018
0.5448	0.8737	0.429	−0.6676	−0.018
0.6506	0.9145	0.463	−0.6076	−0.017
0.7614	0.9584	0.500	−0.4791	−0.014
0.8778	1.0060	0.543	−0.2884	−0.007
1.0000	1.0572	0.588	0.0000	0.000
		DO (1) + CA (6)		
0.0000	0.8668	1.753	0.0000	0.000
0.1295	0.8811	1.517	0.1971	−0.085
0.2508	0.8961	1.331	0.3565	−0.130
0.3646	0.9121	1.174	0.4535	−0.155
0.4716	0.9292	1.044	0.4920	−0.160
0.5724	0.9478	0.935	0.4483	−0.152
0.6676	0.9675	0.843	0.3785	−0.132
0.7575	0.9884	0.763	0.2811	−0.107
0.8427	1.0105	0.696	0.1695	−0.076
0.9234	1.0333	0.634	0.0843	−0.043
1.0000	1.0572	0.588	0.0000	0.000

Table 3. Values of Speeds of Sound u, Isentropic Compressibilty K_S, and Deviations in Isentropic Compressibilty ΔK_S, at 298.15 K

x_1	u / $\text{m} \cdot \text{s}^{-1}$	$K_S \cdot 10^{12}$ / Pa^{-1}	$\Delta K_S \cdot 10^{12}$ / Pa^{-1}
	DO (1) + ME (2)		
0.0000	1340.2	579.95	0.00
0.1024	1339.9	574.76	0.02
0.2043	1339.6	569.78	0.04
0.3057	1339.3	564.90	0.08
0.4065	1339.0	560.05	0.12
0.5067	1338.8	555.03	0.13
0.6064	1338.7	549.64	0.11
0.7056	1338.6	544.21	0.08
0.8043	1338.5	538.77	0.05
0.9024	1338.3	533.52	0.03
1.0000	1338.2	528.20	0.00
	DO (1) + EE (3)		
0.0000	1302.8	636.95	0.00
0.1191	1306.6	625.34	0.13
0.2332	1310.3	614.14	0.26
0.3427	1313.9	603.21	0.35
0.4478	1317.6	592.30	0.41
0.5488	1321.3	581.28	0.40
0.6460	1325.0	570.11	0.34
0.7395	1328.5	559.22	0.27
0.8295	1331.9	548.62	0.19
0.9163	1335.1	538.30	0.10
1.0000	1338.2	528.20	0.00
	DO (1) + BE (4)		
0.0000	1303.4	656.52	0.00
0.1506	1307.6	642.77	0.56
0.2851	1311.7	629.35	0.94
0.4061	1315.5	616.44	1.20
0.5154	1319.2	603.53	1.31
0.6147	1322.9	590.24	1.26
0.7053	1326.3	577.26	1.12
0.7882	1329.5	564.56	0.92
0.8645	1332.5	552.05	0.65
0.9349	1335.5	539.89	0.33
1.0000	1338.2	528.20	0.00
	DO (1) + PA (5)		
0.0000	1075.6	1268.33	0.00
0.0814	1097.3	1173.21	−3.49
0.1663	1120.0	1082.70	−6.25
0.2548	1143.6	997.17	−8.26
0.3472	1168.1	916.23	−9.51
0.4438	1193.6	840.21	−9.96
0.5448	1220.2	768.73	−9.63
0.6506	1247.9	702.19	−8.46
0.7614	1276.7	640.14	−6.46
0.8778	1306.8	582.08	−3.65
1.0000	1338.3	528.12	0.00
	DO (1) + CA (6)		
0.0000	1416.4	575.05	0.00
0.1295	1403.3	576.33	0.73
0.2508	1391.8	576.09	1.28
0.3646	1381.5	574.45	1.65
0.4716	1372.7	571.14	1.82
0.5724	1365.3	566.01	1.78
0.6676	1358.7	559.89	1.61
0.7575	1352.9	552.76	1.32
0.8427	1347.7	544.85	0.93
0.9234	1342.7	536.80	0.50
1.0000	1338.2	528.20	0.00

Table 4. Values of Experimental Refractive Index n_D, Experimental Molar Refractivity R_{exp} by the Lorentz−Lorenz Equation (Equation 5), and Deviation in Molar Refractivity ΔR at 298.15 K

x_1	n_D	$R_{exp} \cdot 10^6$ $m^3 \cdot mol^{-1}$	$\Delta R \cdot 10^6$ $m^3 \cdot mol^{-1}$
		DO (1) + ME (2)	
0.0000	1.4002	19.2257	0.0000
0.1024	1.4000	18.9850	−0.0038
0.2043	1.3998	18.7529	−0.0001
0.3057	1.3996	18.5253	0.0069
0.4065	1.3995	18.3043	0.0191
0.5067	1.3993	18.0782	0.0249
0.6064	1.3990	17.8402	0.0176
0.7056	1.3988	17.6060	0.0128
0.8043	1.3985	17.3694	0.0047
0.9024	1.3983	17.1418	0.0040
1.0000	1.3980	16.9120	0.0000
		DO (1) + EE (3)	
0.0000	1.4051	23.8844	0.0000
0.1191	1.4047	23.0659	0.0120
0.2332	1.4041	22.2795	0.0211
0.3427	1.4034	21.5231	0.0282
0.4478	1.4027	20.7978	0.0356
0.5488	1.4019	20.0893	0.0314
0.6460	1.4012	19.4040	0.0238
0.7395	1.4004	18.7411	0.0128
0.8295	1.3995	18.1034	0.0026
0.9163	1.3988	17.4974	0.0018
1.0000	1.3980	16.9120	0.0000
		DO (1) + BE (4)	
0.0000	1.4176	33.1887	0.0000
0.1506	1.4158	30.7489	0.0115
0.2851	1.4145	28.6062	0.0580
0.4061	1.4129	26.6672	0.0885
0.5154	1.4112	24.9029	0.1032
0.6147	1.4092	23.2676	0.0842
0.7053	1.4071	21.7707	0.0619
0.7882	1.4050	20.4033	0.0439
0.8645	1.4028	19.1407	0.0232
0.9349	1.4006	17.9870	0.0154
1.0000	1.3980	16.9120	0.0000
		DO (1) + PA (5)	
0.0000	1.3759	19.8976	0.0000
0.0814	1.3789	19.6895	0.0350
0.1663	1.3823	19.4833	0.0822
0.2548	1.3859	19.2707	0.1339
0.3472	1.3895	19.0357	0.1748
0.4438	1.3928	18.7767	0.2041
0.5448	1.3952	18.4658	0.1948
0.6506	1.3965	18.1099	0.1548
0.7614	1.3971	17.7204	0.0961
0.8778	1.3975	17.3146	0.0377
1.0000	1.3980	16.9120	0.0000
		DO (1) + CA (6)	
0.0000	1.4565	31.1328	0.0000
0.1295	1.4522	29.3826	0.0913
0.2508	1.4479	27.7434	0.1771
0.3646	1.4435	26.1933	0.2454
0.4716	1.4381	24.6802	0.2539
0.5724	1.4321	23.2144	0.2216
0.6676	1.4258	21.8193	0.1803
0.7575	1.4194	20.4996	0.1390
0.8427	1.4125	19.2341	0.0852
0.9234	1.4052	18.0356	0.0343
1.0000	1.3980	16.9120	0.0000

and PA (5) most probably through H-bonding between the ethereal oxygen of DO (1) and hydrogen of the amine group of PA (5). However, large positive values for the CA (6) system may be attributed to the presence of the bulky cyclohexyl ring in its molecule.

The deviations in viscosities $\Delta\eta$ were calculated as[22]

$$\Delta\eta = \eta - \sum_{i=1}^{2} (x_i \eta_i) \qquad (2)$$

where η is the dynamic viscosity of the mixture and x_i and η_i are the mole fraction and viscosity of the *i*th component in the mixture, respectively.

As far as $\Delta\eta$ values were concerned, the studied binary mixtures exhibit negative values over the entire range of composition at 298.15 K as depicted in Figure 2 as a function of mole fraction of DO (1). The alkoxyethanol systems are characterized by negative $\Delta\eta$ values in the order:

$$\text{DO (1) + ME (2)} < \text{DO (1) + EE (3)} < \text{DO (1) + BE (4)}$$

and for the amine systems, the order of negative $\Delta\eta$ values is as follows

$$\text{DO (1) + PA (5)} < \text{DO (1) + CA (6)}$$

Generally it is mentioned in some works[23−26] that where V^E values are positive for a system the $\Delta\eta$ values are negative, and vice versa, though there may be discrepancies of such sign reversal for V^E and $\Delta\eta$ values. Our systems follow this sign reversal.[9,26] In fact, some authors[27,28] have pointed out that when one of the compounds of a binary mixture is strongly associated the deviations of viscosity are usually negative. Also, the negative V^E values indicate specific interaction and its positive value dominance of disruption forces. In addition, positive $\Delta\eta$ values indicate specific interaction, and its negative values indicate dominance of disruption forces. Thus, the order of $\Delta\eta$ values (Figure 2) supports our earlier results obtained from V^E values. Table 3 gives the experimental ultrasonic speed of sounds u, isentropic compressibility K_S, and deviation in isentropic compressibility for the binary mixtures of DO (1) with ME (2), EE (3), BE (4), PA (5), and CA (6) along with the coreresponding mole fractions of DO (1), x_1.

Isentropic compressibilities, K_S, and the deviations in isentropic compressibilities, ΔK_S, were calculated from the experimental densities, ρ, and speeds of sound, u, using the following equations[29−31]

Table 5. Redlich−Kister Coefficients A_k and Standard Deviations σ for the Binary Mixtures

excess property	A_0	A_1	A_2	σ
		DO (1) + ME (2)		
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	0.319	0.025	−0.491	0.008
$\Delta\eta$/mPa·s	−1.964	0.310	−0.109	0.001
$\Delta K_S \cdot 10^{12}/Pa^{-1}$	0.504	−0.031	−0.396	0.009
$\Delta R \cdot 10^6/m^3 \cdot mol^{-1}$	0.084	0.034	−0.173	0.003
		DO (1) + EE (3)		
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	0.404	0.033	−0.666	0.009
$\Delta\eta$/mPa·s	−1.000	0.130	0.272	0.006
$\Delta K_S \cdot 10^{12}/Pa^{-1}$	1.635	−0.172	−1.005	0.005
$\Delta R \cdot 10^6/m^3 \cdot mol^{-1}$	0.131	−0.059	−0.136	0.002
		DO (1) + BE (4)		
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	0.992	−0.063	−1.172	0.011
$\Delta\eta$/mPa·s	−1.722	0.443	0.009	0.003
$\Delta K_S \cdot 10^{12}/Pa^{-1}$	5.161	0.874	−0.535	0.011
$\Delta R \cdot 10^6/m^3 \cdot mol^{-1}$	0.410	−0.005	−0.600	0.009
		DO (1) + PA (5)		
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	−2.610	−0.378	0.446	0.009
$\Delta\eta$/mPa·s	−0.074	−0.022	0.044	0.001
$\Delta K_S \cdot 10^{12}/Pa^{-1}$	−39.490	7.842	−0.778	0.007
$\Delta R \cdot 10^6/m^3 \cdot mol^{-1}$	0.799	−0.143	−0.664	0.003
		DO (1) + CA (6)		
$V^E \cdot 10^6/m^3 \cdot mol^{-1}$	1.934	−0.350	−0.993	0.006
$\Delta\eta$/mPa·s	−0.632	0.114	−0.052	0.002
$\Delta K_S \cdot 10^{12}/Pa^{-1}$	7.299	0.289	−1.765	0.015
$\Delta R \cdot 10^6/m^3 \cdot mol^{-1}$	0.920	−0.187	−0.574	0.011

$$K_S = 1/u^2\rho \tag{3}$$

$$\Delta K_S = K_S - \sum_{i=1}^{2}(x_i K_{S,i}) \tag{4}$$

where u is the speed of sound of the mixture and x_i and $K_{S,i}$ are the mole fraction and isentropic compressibility of the ith component in the mixture, respectively.

The variations of the deviation in isentropic compressibilty, ΔK_S, against mole fraction of DO (1), x_1, are represented in Figure 3. The values are positive for all the investigated binary systems except the DO (1) + PA (5) system over the entire range of mole fraction. The largest negative values are observed for the system DO (1) + PA (5), while for the system DO (1) + ME (2) the ΔK_S values are close to zero. The algebraic values of ΔK_S of binary mixtures fall in the order:

DO (1) + CA (6) > DO (1) + BE (4) > DO (1) + EE (3) > DO (1) + ME (2) > DO (1) + PA (5)

Table 4 represents the experimental refractive index, molar refractivity by the Lorentz−Lorenz equation, and deviation in molar refractivity at 298.15 K. The molar refractivity, R, was calculated using the Lorentz−Lorenz equation[32]

$$R = (n_D^2 - 1/n_D^2 + 2)V \tag{5}$$

where n_D and V are the refractive indices and the molar volume, respectively; and ΔR is the molar refraction changes of mixing, which were obtained from

$$\Delta R = R - \sum_{i=1}^{2}(x_i R_i) \tag{6}$$

where R is the molar refraction of the mixture and x_i and R_i are the mole fraction and molar refraction of the ith component in the mixture, respectively. The dependence of molar refraction (ΔR) on mole fraction (x_1) of DO (1) at 298.15 K is depicted in Figure 4. It is observed that for all the mixtures ΔR is positive for the entire range of composition; however, in the case of the DO (1) + PA (5) system, the ΔR values are negative up to $x_1 = 0.2$, but beyond that, ΔR becomes positive. Such values may be due to the electronic perturbation of the individual molecules during mixing and therefore depend very much on the nature of the mixing components. The positive ΔR values vary in the order:

DO (1) + CA (6) > DO (1) + PA (5) > DO (1) + BE (4) > DO (1) + EE (3) > DO (1) + ME (2)

Redlich−Kister Polynomial Equation. For each mixture, the excess or deviation properties (V^E, $\Delta\eta$, ΔK_S, and ΔR) were fitted to the Redlich−Kister polynomial equation[33] of the type

$$Y^E_{ij} = x_i x_j \sum_{k=1}^{2} A_k(x_i - x_j)^k \tag{8}$$

where Y^E_{ij} refers to an excess property (V^E, $\Delta\eta$, ΔK_S, ΔR) for each $i-j$ binary pair; and x_i is the mole fraction of the ith component; and A_k represents the coefficients.

The calculated values of A_k along with the standard deviations (σ) are listed in Table 5. The values of coefficients A_k of eq 8 and the corresponding standard deviations (σ) obtained by the method of least-squares with equal weights assigned to each point are calculated. The standard deviations (σ) were calculated as

$$\sigma = [(Y^E_{\text{exp}} - Y^E_{\text{cal}})^2/(n - m)]^{1/2} \tag{9}$$

where n represents the number of measurements and m the number of coefficients. The values of coefficients (A_k) were determined by a multiple-regression analysis based on the least-squares method and summarized along with the standard deviations between the experimental and fitted values of V^E, $\Delta\eta$, ΔK_S, and ΔR. Finally, it can be concluded that the expressions used for interpolating the experimental data measured in this work gave good results, as the σ values for the V^E, $\Delta\eta$, ΔK_S, and ΔR data fitted to the Redlich−Kister polynomial equation were in the range from 0.006 to 0.011, 0.001 to 0.006, 0.005 to 0.015, and 0.002 to 0.011, respectively. These results indicate that the derived V^E, $\Delta\eta$, ΔK_S, and ΔR are quite systematic and are a function of composition of the binary mixtures.

Conclusion

In this work, density, viscosity, ultrasonic speed of sound, and refractive index have been measured for the binary mixtures of 1,3-dioxolane with 2-methoxyethanol, 2-ethoxyethanol, 2-butoxyethanol, 2-propylamine, and cyclohexylamine. Excess molar volume, viscosity deviation, deviation in isentropic compressibility, and molar refraction of the mixtures have also been measured.

Literature Cited

(1) Roy, M. N.; Das, M. Thermodynamic and Transport Properties of Binary Mixtures of Dimethyl Sulfoxide with *t*-Butyl Alcohol, Butyl Acetate, 2-Butanone and Butyl Amine at Different Temperatures. *Russ. J. Phys. Chem.* **2006**, *80*, 163–171.

(2) Roy, M. N.; Sinha, A.; Sinha, B. Excess Molar Volumes, Viscosity Deviations and Isentropic Compressibility of Binary Mixtures Containing 1,3-Dioxolane and Monoalcohols at 303.15 K. *J. Solution Chem.* **2005**, *34* (11), 1311–1325.

(3) Subha, M. C. S.; Swamy, G. N.; Bai, M. E.; Krishna Rao, K. S. V. Excess Volume and Viscosity of Ethoxy Ethanol with *n*-Butylamine, *sec*-Butylamine, *tert*-Butylamine, *n*-Hexylamine, *n*-Octylamine and Cyclohexylamine. *Indian J. Chem.* **2004**, *43A*, 1876–1881.

(4) Riddick, J. A.; Bunger, W. B.; Sakano, T. K. *Organic Solvents. Techniques of Chemistry*, 4th ed.; John Wiley and Sons: New York, 1986; Vol. 11.

(5) Lafuente, C.; Giner, B.; Villares, A.; Gascon, I.; Cea, P. Speeds of Sound and Isentropic Compressibilities of Binary Mixtures Containing Cyclic Ethers and Haloalkanes at 298.15 and 313.15 K. *Int. J. Thermophys.* **2004**, *25* (6), 1735–1746.

(6) Gascon, I.; Mainar, A. M.; Royo, F. M.; Urieta, J. S. Experimental viscosities and viscosity predictions of the ternary mixture (Cyclohexane + 1,3-dioxolane + 2-butanol) at 298.15 and 313.15 K. *J. Chem. Eng. Data* **2000**, *45*, 751–755.

(7) Ottani, S.; Vitalini, D.; Comelli, F.; Castellari, C. Densities, Viscosities, and Refractive Indices of Poly(ethyleneglycol) 200 and 400 + Cyclic Ethers at 303.15 K. *J. Chem. Eng. Data* **2002**, *47*, 1197–1204.

(8) Douhéret, G.; Pal, A.; Davis, M. I. Ultrasonic speeds and isentropic functions of (a 2-alkoxyethanol + water) at 298.15 K. *J. Chem. Thermodyn.* **1990**, *22*, 99–108.

(9) Sinha, B.; Dakua, V. K.; Roy, M. N. Excess Molar Volumes, Viscosity Deviations and ultrasonic speeds of sound of Binary Mixtures of 2-butanone with some alkoxyethanols and amines at 298.15 K. *J. Mol. Liq.* **2007**, *136*, 128–137.

(10) Nishimoto, N.; Tabata, S.; Tamura, K.; Murakami, S. Thermodynamic properties of the mixture of methoxyethanol and cyclohexane: Measurements at the temperatures 293.15, 298.15 and 303.15 K above and below UCST. *Fluid Phase Equilib.* **1987**, *136*, 235–247.

(11) Aminabhavi, T. M.; Raikar, S. K.; Balundgi, R. H. Volumetric, Acoustic, Optical, and Viscometric Properties of Binary Mixtures of 2-Methoxyethanol with Aliphatic Alcohols (Cl-C8). *Ind. Eng. Chem. Res.* **1993**, *32*, 931–936.

(12) Muhuri, P. K.; Hazra, D. K. Density and Viscosity of Propylene Carbonate + 2-Methoxyethanol at 298.15, 308.15, and 318.15 K. *J. Chem. Eng. Data* **1995**, *40*, 582.

(13) Jhonson, I.; Kalidoss, M.; Srinivasamoorthy, R. Density, Viscosity, and Speed of Sound in the Ternary Mixtures of 2-Ethoxyethanol + *N*,*N*-Dimethylformamide + *N*,*N*-Dimethylacetamide and 2-Ethoxy-

ethanol + Dimethyl Sulfoxide + *N*,*N*-Dimethylacetamide at 308.15 K. *J. Chem. Eng. Data* **2002**, *47*, 1388.

(14) Tamura, K.; Osaki, A.; Murakami, S.; Laurent, B.; Grolier, J.-P. E. Thermodynamic properties of x (2-alkoxyethanol) + (1-x) n-octane: densities at 298.15 and 303.15 K, and speeds of sound at 298.15 K. *Fluid Phase Equilib.* **2000**, *173*, 285–296.

(15) Shastry, N. V.; Patel, S. R. Densities, viscosities, sound speeds and excess properties of binary mixtures of methyl methacrylate with alkoxyethanols and 1-alchohols at 298.15 and 308.15 K. *Int. J. Thermophys.* **2000**, *21*, 1153–1174.

(16) Roy, M. N.; Hazra, D. K. Densities and Viscosities of the binary aqueous mixture of tetrahydrofuran and 1, 2 dimethoxyethane at 298, 308 and 318 K. *Indian J. Chem. Technol.* **1994**, *1*, 93–97.

(17) Dean, J. A. *Lange's handbook of chemistry*, 11th ed.; McGraw-Hill Book Company: New York, 1973.

(18) Chatterjee, A.; Das, B. Electrical Conductances of Tetrabutylammonium Bromide, Sodium Tetraphenylborate, and Sodium Bromide in Methanol (1) + Water (2) Mixtures at (298.15, 308.15, and 318.15) K. *J. Chem. Eng. Data* **2006**, *51*, 1352–1355.

(19) Das, B.; Roy, M. N.; Hazra, D. K. Densities and Viscosities of the binary aqueous mixtures of tetrahydrofuran and 1,2 dimethoxyethane at 298, 308 and 318 K. *Indian J. Chem. Technol.* **1994**, *1*, 93–97.

(20) Ali, A.; Nain, A. K.; Sharma, V. K.; Ahmad, S. Ultrasonic studies in binary liquid mixtures. *Indian J. Phys.* **2001**, *758* (6), 519–525.

(21) Gurung, B. B.; Roy, M. N. Studies of Densities, Viscosity Deviations, and Isentropic Compressibilities of Ternary Liquid Mixtures of Water and Ethane-1,2-diol with some monoalcohols at various temperatures. *J. Phys. Chem. Liq.* **2007**, *45*, 331–343.

(22) Nayak, J. N.; Aralaguppi, M. I.; Aminabhavi, T. M. Density, Viscosity, Refractive Index, and Speed of Sound in the Binary Mixtures of 1,4-Dioxane + Ethyl acetoactate, + Diethyl oxalate, + Diethyl pthalate, or + Dioctyl pthalate at 298.15, 303.15, and 308.15 K. *J. Chem. Eng. Data* **2003**, *48*, 1489–1494.

(23) Ansón, A.; Garriga, R.; Martínez, S.; Pérez, P.; Gracia, M. Densities and Viscosities of Binary Mixtures of 1-Chlorobutane with Butanol Isomers at Several Temperatures. *J. Chem. Eng. Data* **2005**, *50*, 677–682.

(24) Chen, H. W.; Wen, C. C.; Tu, C. H. Excess Molar Volumes, Viscosities, and Refractive Indexes for Binary Mixtures of 1-Chlorobutane with Four Alcohols at *T*) (288.15, 298.15, and 308.15) K. *J. Chem. Eng. Data* **2004**, *49*, 347–351.

(25) Nikam, P. S.; Mahale, T. R.; Hasan, M. Density and Viscosity of Binary Mixtures of Ethyl Acetate with Methanol, Ethanol, Propan-1-ol, Propan-2-ol, Butan-1-ol, 2-Methylpropan-1-ol, and 2-Methyl-propan-2-ol at (298.15, 303.15, and 308.15) K. *J. Chem. Eng. Data* **1996**, *41*, 1055–1058.

(26) Wankhede, N. M.; Lande, K.; Arbad, B. R. Excess Molar Volumes and Viscosity Deviations of Binary Mixtures of 2,4,6-Trimethyl-1,3,5-trioxane + Ethanol, 1-Propanol, and 1-Butanol at (298.15, 303.15, and 308.15) K. *J. Chem. Eng. Data* **2005**, *50*, 969–972.

(27) Meyer, R.; Meyer, M.; Metzger, J.; Peneloux, A. EÄ Tude Thermodynamique Des mélanges Binaires. *J. Chim. Phys. Chim. Biol.* **1971**, *68*, 406–412.

(28) Nigam, R. K.; Mahl, B. S. Strength of Interactions in Binary Mixtures of Methylene Chloride with Benzene, Toluene & Xylenes; Chloroform with Acetone, Ether & Dioxane; Benzene with Acetone, *n*-Pentane & *n*-Hexane; & of Cyclohexane with *n*-Pentane. *Indian J. Chem.* **1971**, *9*, 1255–1258.

(29) Artigas, H.; Lafuente, C.; Lopez, M. C.; Royo, F. M.; Urieta, J. S. Calorimetric Behaviour of Primary Bromobutanes with Isomeric Butanols. *Zeit. Phys. Chem.* **2001**, *215*, 933–942.

(30) Gascon, I.; Martin, S.; Cea, P.; Lopez, M. C.; Royo, F. M. Density and Speed of Sound for Binary Mixtures of a Cyclic Ether with a Butanol Isomer. *J. Solution Chem.* **2002**, *31*, 905–915.

(31) Kimura, F.; Treszczanowicz, A.; Halpin, C.; Benson, G. Excess Volumes and Ultrasonic Speeds for (Di-n-Propyl Ether + n-Heptane). *J. Chem. Thermodyn.* **1983**, *15*, 503–510.

(32) Alberto, Arce.; Eladio, P.; Eva, R.; Ana, S. Molar Volume, Refractive Index, and Isentropic Compressibility at 298.15 K for 1-Butanol + Ethanol + 2-Methoxy-2-methylpropane. *J. Chem. Eng. Data* **1999**, *44*, 291–295.

(33) Redlich, O.; Kister, A. T. Algebraic Representation of Thermodynamic Properties and the Classification of Solutions. *Ind. Eng. Chem.* **1948**, *40*, 345–348.

Received for review March 6, 2009. Accepted June 24, 2009. One of the authors is grateful to UGC Research Fellowship in Science for meritorious student ref. UGC letter No. F.4 −1 /2006 (×1 plan /BSR) under **SAP** for sanctioning a Junior Research Fellowship and providing financial aid in order to continue this research work.

JE900240S