Study of the Solution Properties of Ternary Mixtures of 1,3-Dioxolane (1), Diethyl Ether (2), and *n*-Amyl Alcohol (3) and the Corresponding Binary Mixtures by Density, Viscosity, Refractivity, and Ultrasonic Speed Measurements at 298.15 K[†]

Radhey Shyam Sah, Biswajit Sinha, and Mahendra Nath Roy*

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

The excess molar volume (V^E), viscosity deviation ($\Delta\eta$), deviation in isentropic compressibility (ΔK_S), and excess molar refractivity (ΔR) have been investigated using experimentally measured densities, viscosities, speeds of sound, and refractive indices for the three binary mixtures 1,3-dioxolane (1) + diethyl ether (2), diethyl ether (1) + *n*-amyl alcohol (2), and 1,3-dioxolane (1) + *n*-amyl alcohol (2) and the corresponding ternary mixture 1,3-dioxolane (1) + diethyl ether (2) + *n*-amyl alcohol (3) at 298.15 K. The calculated quantities were further fitted to the Redlich−Kister equation to estimate the binary fitting parameters and standard deviations from the regression lines. The excess or deviation properties were found to be either negative or positive depending on the molecular interactions and the nature of the liquid mixtures.

Introduction

A knowledge of densities, excess volumes, and viscosities of fluids and fluid mixtures is essential for understanding the molecular interactions between unlike molecules and developing new theoretical models as well as for engineering applications in the process industry. Ultrasonic methods have found extensive applications because of their ability to characterize the physicochemical behavior of liquid systems from absorption and velocity data. It is also possible to investigate molecular packing, molecular motion, and various types and extents of intermolecular interactions influenced by the size, shape, and chemical nature of the component molecules and the microscopic structures of the liquids.

Amyl alcohols are used fundamentally for the composition of perfumes and the synthesis of fruit essences. They are also used as solvents for surfaces and lacquer baths, inks for print, and dyes for wool as well as in the chemical production of photographic and pharmaceutical substances. Furthermore, they are an intermediate in the production of amyl acetate and other amyl esters. 1,3-Dioxolane is a versatile solvent that is used in the separation of saturated and unsaturated hydrocarbons and in pharmaceutical synthesis and also serves as a solvent for many polymers.[1,2]

Ethers such as diethyl ether are regarded as ideal potential fuel alternatives or additives, which have good combustion characteristics. The investigation of the thermophysical properties is very important for the increased applications of oxygenated fuels or fuel additives. Considering all of these aspects, we undertook investigations of the thermodynamic and transport properties of binary and ternary mixtures involving 1,3-dioxolane (1,3-DO), diethyl ether (DEE), and *n*-amyl alcohol (AL).

In this paper, we report excess molar volumes (V^E), viscosity deviations ($\Delta\eta$), deviations in isentropic compressibility (ΔK_S), and excess molar refractivities (ΔR) for the three binary mixtures 1,3-DO (1) + DEE (2), 1,3-DO (1) + AL (2), and DEE (1) + AL (2) and the corresponding ternary mixture 1,3-DO (1) + DEE (2) + AL (3) at 298.15 K over the entire range of composition. The excess or deviation properties of the binary mixtures were fitted to the Redlich−Kister polynomial equation to obtain their coefficients and have been interpreted in terms of molecular interactions and structural effects.

Experimental Section

Materials. 1,3-Dioxolane (CAS no. 646-06-0), *n*-amyl alcohol (CAS no. 71-41-0), and diethyl ether (CAS no. 60-29-7) with minimum mass fraction purities of 0.99 used in this study were purchased from S.D.Fine-Chem Ltd. (Mumbai, India). The pure chemicals were stored over activated 4 Å molecular sieves to reduce their water content before use. The purity of each substance was evaluated by comparing experimental values of density, viscosity, speed of sound, and refractive index with those reported in the literature whenever available, as presented in Table 1.

Apparatus and Procedures. Densities (ρ) were measured with an Ostwald−Sprengel-type pycnometer having a bulb volume of about 25 cm^3 and a capillary internal diameter of about 0.1 cm. The measurements were done in a thermostat bath controlled to ± 0.01 K. Viscosity (η) was measured by means of a suspended Ubbelohde-type viscometer calibrated at 298.15 K with triply distilled water and purified methanol using density and viscosity values from the literature. The flow times were accurate to ± 0.1 s, and the uncertainty in the viscosity measurements was $\pm\ 2\cdot10^{-4}$ mPa·s. The mixtures were prepared by mixing known volumes of the pure liquids in airtight stopper bottles, and each solution thus prepared was distributed into three receptacles to perform all of the measurements in triplicate, with the aim of determining possible dispersion of the obtained results. Adequate precautions were taken to minimize evaporation losses during the actual measurements. The reproducibility of the mole fraction was within ± 0.0002. The mass measurements were done on a Mettler AG-285 electronic balance with a precision of ± 0.01 mg. The precision of the density measurements was $\pm\ 3\cdot10^{-4}\ g\cdot cm^{-3}$.

Ultrasonic speeds of sound (u) were determined using a multifrequency ultrasonic interferometer (M-81, Mittal Enterprises, New Delhi, India) working at 1 MHz that had been calibrated with triply distilled and purified water, methanol, and benzene at 303.15 K. The precision of the ultrasonic speed measurements was $\pm\ 0.2\ m\cdot s^{-1}$. The details of the methods and techniques have been described in earlier papers.[3−5]

[†] Part of the “Sir John S. Rowlinson Festschrift”.

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

10.1021/je100594u

Table 1. Densities (ρ), Viscosities (η), Speeds of Sound (u), and Refractive Indices (n_D) of the Pure Liquids at T = 298.15 K

	$10^{-3}\cdot\rho$/kg·m^{-3}		η/mPa·s		u/m·s^{-1}		n_D	
liquid	exptl	lit	exptl	lit	exptl	lit	exptl	lit
1,3-DO	1.0587	1.0586[14]	0.589	0.5886[14]	1339.4	1338.8[15]	1.3985	1.398[16]
AL	0.8112	0.8110[17]	3.504	3.510[18]	1271.6	1274.1[19]	1.4100	1.409[20]
DEE	0.7083	0.7083[21]	0.247	–	982.3	983[21]	1.3515	–

Table 2. Densities (ρ), Viscosities (η), Excess Molar Volumes (V^E), and Viscosity Deviations ($\Delta\eta$) for Binary Mixtures of 1,3-Dioxolane, Diethyl Ether, and *n*-Amyl Alcohol at T = 298.15 K

x_1	$10^{-3}\cdot\rho$ kg·m^{-3}	η mPa·s	$10^6\cdot V^E$ m^3·mol^{-1}	$\Delta\eta$ mPa·s
1,3-Dioxolane (1) + Diethyl Ether (2)				
0.0000	0.7083	0.247	0.000	0.000
0.1000	0.7364	0.266	−0.533	−0.016
0.2000	0.7656	0.284	−0.905	−0.031
0.3001	0.7963	0.306	−1.176	−0.044
0.4001	0.8290	0.331	−1.381	−0.053
0.5001	0.8634	0.360	−1.480	−0.058
0.6001	0.8997	0.394	−1.480	−0.058
0.7001	0.9382	0.435	−1.400	−0.052
0.8000	0.9770	0.478	−1.072	−0.043
0.9000	1.0184	0.531	−0.694	−0.025
1.0000	1.0587	0.589	0.000	0.002
1,3-Dioxolane (1) + *n*-Amyl Alcohol (2)				
0.0000	0.8112	3.504	0.000	0.000
0.1168	0.8329	2.505	−0.287	−0.659
0.2293	0.8545	1.885	−0.414	−0.951
0.3377	0.8766	1.440	−0.460	−1.080
0.4424	0.8992	1.063	−0.440	−1.152
0.5434	0.9225	0.906	−0.377	−1.014
0.6409	0.9468	0.756	−0.290	−0.880
0.7352	0.9725	0.647	−0.213	−0.714
0.8264	0.9993	0.570	−0.117	−0.525
0.9146	1.0282	0.541	−0.058	−0.297
1.0000	1.0587	0.589	0.000	0.000
Diethyl Ether (1) + *n*-Amyl Alcohol (2)				
0.0000	0.8112	3.504	0.000	0.000
0.1167	0.8052	2.651	−0.750	−0.384
0.2292	0.7975	1.966	−1.250	−0.714
0.3377	0.7889	1.491	−1.575	−0.847
0.4423	0.7797	1.123	−1.788	−0.884
0.5433	0.7704	0.829	−1.950	−0.859
0.6409	0.7595	0.620	−1.860	−0.761
0.7352	0.7479	0.481	−1.638	−0.602
0.8263	0.7361	0.382	−1.345	−0.413
0.9146	0.7230	0.314	−0.820	−0.203
1.0000	0.7083	0.247	0.000	0.000

Refractive indices (n_D) were measured with the help of an Abbe refractometer. The accuracy of the refractive index measurements was ± 0.0002 units. The refractometer was calibrated twice using distilled and deionized water, and the calibration was checked after every few measurements.

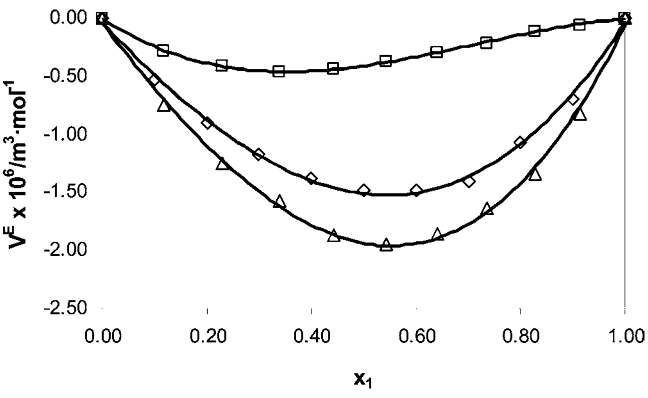


Figure 1. Excess molar volumes $V^E\cdot10^6$/m^3·mol^{-1} as functions of mole fraction (x_1) for the three binary subsystems at T = 298.15 K: □, 1,3-dioxolane (1) + *n*-amyl alcohol (2); ◇, 1,3-dioxolane (1) + diethyl ether (2); △, diethyl ether (1) + *n*-amyl alcohol (2).

Results and Discussion

We calculated excess molar volumes (V^E), viscosity deviations ($\Delta\eta$), deviations in isentropic compressibility (ΔK_S), and excess molar refractivities (ΔR) at 298.15 K for binary mixtures of diethyl ether, dichloromethane, and *n*-amyl alcohol over the whole composition range. The variations of the excess properties over the entire range of composition for the binary mixtures are depicted in Figures 1−4.

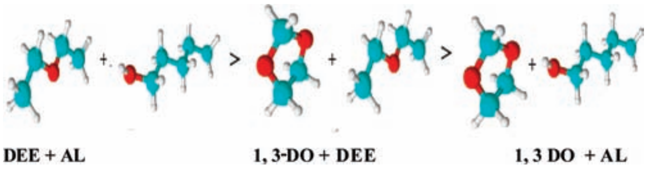
Excess Molar Volume. Density (ρ) values were used to calculate the excess molar volumes (V^E) for binary (n = 2) and ternary (n = 3) mixtures using the following equation:[6]

$$V^E = \sum_{i=1}^{n} x_i M_i\left(\frac{1}{\rho} - \frac{1}{\rho_i}\right) \qquad (1)$$

where ρ is the density of the mixture and M_i, x_i, and ρ_i are the molar mass, mole fraction, and density, respectively, of the ith component in the mixture.

V^E is the result of contributions from several opposing effects. These may be divided arbitrarily into three types, namely, physical, chemical, and structural. Physical contributions, which are nonspecific interactions between the real species present in the mixture, make a positive contribution to V^E. Chemical or specific intermolecular interactions result in a volume decrease, resulting in a negative contribution to V^E. Structural contributions are mostly negative and arise from several effects, especially from interstitial accommodation and changes in free volume.[7]

The values of V^E were found to be negative for all of the binary mixtures, as reported in Table 2 and depicted graphically in Figure 1; the order of the negative deviations is schematically depicted below:



These phenomena result from differences in the energies of interaction between the molecules and packing effects. Disruption of the ordered structure of the pure component during

Table 3. Densities (ρ), Viscosities (η), Excess Molar Volumes (V^E), and Viscosity Deviations ($\Delta\eta$) for Ternary Mixtures of 1,3-Dioxolane (1) + Diethyl Ether (2) + *n*-Amyl Alcohol (3) at T = 298.15 K

x_1	x_2	$10^{-3}\cdot\rho$ kg·m^{-3}	η mPa·s	$10^6\cdot V^E$ m^3·mol^{-1}	$\Delta\eta$ mPa·s
0.5001	0.4999	0.8634	0.360	−1.480	−0.058
0.4574	0.4572	0.8602	0.431	−1.595	−0.121
0.4132	0.4131	0.8567	0.480	−1.668	−0.222
0.3676	0.3675	0.8525	0.552	−1.680	−0.322
0.3205	0.3204	0.8477	0.651	−1.608	−0.421
0.2717	0.2716	0.8424	0.759	−1.476	−0.543
0.2212	0.2211	0.8367	0.966	−1.293	−0.607
0.1689	0.1688	0.8307	1.243	−1.046	−0.654
0.1146	0.1146	0.8246	1.682	−0.770	−0.609
0.0584	0.0584	0.8181	2.304	−0.424	−0.476
0.0000	0.0000	0.8112	3.404	0.000	0.000

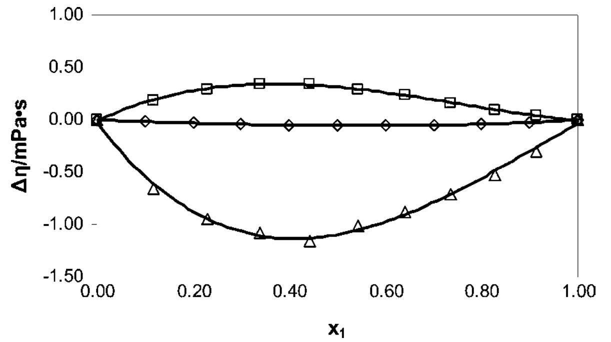


Figure 2. Viscosity deviations $\Delta\eta$/m·Pa s as functions of mole fraction (x_1) for the three binary subsystems at T = 298.15 K: □, 1,3-dioxolane (1) + *n*-amyl alcohol (2); ◇, 1,3-dioxolane (1) + diethyl ether (2); △, diethyl ether (1) + *n*-amyl alcohol (2).

Table 4. Ultrasonic Speeds (u), Isentropic Compressibilities (K_S), Deviations in Isentropic Compressibility (ΔK_S), Refractive Indices (n_D), and Excess Molar Refractivities (ΔR) for Binary Mixtures of 1,3-Dioxolane, Diethyl Ether, and *n*-Amyl Alcohol at T = 298.15 K

	u	$10^{12}\cdot K_S$	$10^{12}\cdot\Delta K_S$		$10^{-6}\cdot\Delta R$
x_1	$\mathrm{m\cdot s^{-1}}$	$\mathrm{Pa^{-1}}$	$\mathrm{Pa^{-1}}$	n_D	$\mathrm{m^3\cdot mol^{-1}}$
1,3-Dioxolane (1) + Diethyl Ether (2)					
0.0000	982.3	1463.2	0.00	1.3515	0.0000
0.1000	1003.9	1347.5	−22.0	1.3568	−0.0390
0.2000	1033.4	1223.1	−52.7	1.3604	−0.0781
0.3001	1067.4	1102.1	−80.0	1.3649	−0.1054
0.4001	1108.5	981.7	−106.7	1.3696	−0.1268
0.5001	1152.8	871.6	−123.2	1.3743	−0.1460
0.6001	1191.4	783.1	−118.0	1.3791	−0.1520
0.7001	1230.9	703.5	−104.0	1.3842	−0.1320
0.8000	1265.3	639.3	−74.5	1.3893	−0.1000
0.9000	1302.4	578.9	−41.3	1.3956	−0.0530
1.0000	1339.4	526.5	0.0	1.3985	0.0000
1,3-Dioxolane (1) + *n*-Amyl Alcohol (2)					
0.0000	1271.6	762.4	0.0	1.4100	0.0000
0.1168	1278.8	734.2	−0.7	1.4085	−0.1038
0.2293	1287.5	705.9	−2.4	1.4072	−0.1510
0.3377	1296.8	678.3	−4.4	1.4060	−0.1715
0.4424	1305.0	653.0	−5.0	1.4050	−0.1616
0.5434	1313.4	628.4	−5.8	1.4040	−0.1365
0.6409	1321.5	604.8	−6.4	1.4030	−0.1054
0.7352	1327.1	583.9	−5.1	1.4022	−0.0730
0.8264	1332.7	563.4	−4.1	1.4010	−0.0415
0.9146	1337.0	544.1	−2.5	1.4000	−0.0121
1.0000	1339.5	526.5	−0.0	1.3985	0.0000
Diethyl Ether (1) + *n*-Amyl Alcohol (2)					
0.0000	1271.6	762.4	0.0	1.4100	0.0000
0.1167	1241.8	803.5	−40.7	1.4043	−0.1830
0.2292	1216.7	846.3	−76.7	1.3977	−0.2680
0.3377	1193.9	889.3	−109.7	1.3922	−0.2740
0.4423	1172.2	933.3	−139.0	1.3874	−0.2480
0.5433	1141.6	996.6	−146.5	1.3825	−0.2054
0.6409	1110.7	1068.2	−143.3	1.3774	−0.1430
0.7352	1077.3	1152.1	−125.5	1.3724	−0.0720
0.8263	1046.3	1240.8	−100.7	1.3664	−0.0350
0.9146	1011.2	1350.8	−52.5	1.3601	−0.0070
1.0000	982.3	1463.2	0.0	1.3515	0.0000

formation of the mixture leads to a positive effect on excess volume, while formation of order in the mixture leads to a negative contribution. The negative V^E values indicate the specific interactions, such as intermolecular hydrogen bonding and interstitial accommodation of the mixing components because of the difference in molar volumes.

The ρ and V^E values for the ternary mixture 1,3-DO (1) + DEE (2) + AL (3) are reported in Table 3. The V^E values are negative in the low mole fraction region for AL, suggesting a specific intermolecular interaction between the two components 1,3-DO (1) and DEE (2), but as the mole fraction of AL in the ternary mixture increases, the magnitude of V^E decreases, suggesting that dispersion forces now begin to operate. Thus, it can be said that the addition of AL to a binary mixture of 1,3-DO (1) and DEE (2) causes a decrease in the intermolecular interactions between the mixture components.

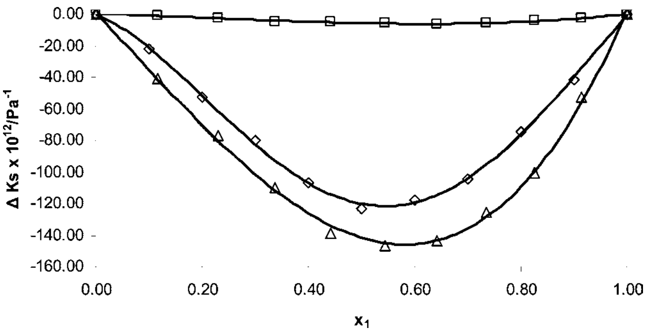


Figure 3. Deviations in isentropic compressibility $\Delta K_s\cdot10^{12}/\mathrm{Pa^{-1}}$ as functions of mole fraction (x_1) for the three binary subsystems at T = 298.15 K: □, 1,3-dioxolane (1) + *n*-amyl alcohol (2); ◇, 1,3-dioxolane (1) + diethyl ether (2); △, diethyl ether (1) + *n*-amyl alcohol (2).

Table 5. Ultrasonic Speeds (u), Isentropic Compressibilities (K_S), Deviations in Isentropic Compressibility (ΔK_S), Refractive Indices (n_D), and Excess Molar Refractivities (ΔR) for Ternary Mixtures of 1,3-Dioxolane (1) + Diethyl Ether (2) + *n*-Amyl Alcohol (3) at T = 298.15 K

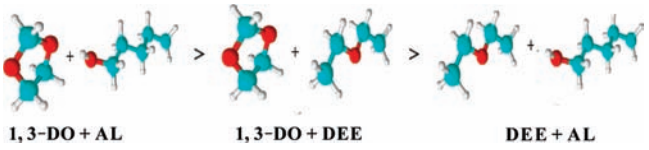
		u	$10^{12}\cdot K_S$	$10^{12}\cdot\Delta K_S$		$10^{-6}\cdot\Delta R$
x_1	x_2	$\mathrm{m\cdot s^{-1}}$	$\mathrm{Pa^{-1}}$	$\mathrm{Pa^{-1}}$	n_D	$\mathrm{m^3\cdot mol^{-1}}$
0.5001	0.4999	1149.2	877.0	−83.7	1.3750	−0.1081
0.4574	0.4572	1165.9	858.4	−88.7	1.3775	0.0710
0.4132	0.4131	1183.4	843.3	−92.7	1.3805	0.1540
0.3676	0.3675	1196.6	828.9	−89.0	1.3850	0.2608
0.3205	0.3204	1207.6	815.9	−80.5	1.3875	0.3340
0.2717	0.2716	1213.9	805.6	−64.5	1.3925	0.3829
0.2212	0.2211	1224.6	796.9	−53.2	1.3945	0.3600
0.1689	0.1688	1235.8	788.3	−41.1	1.3980	0.3049
0.1146	0.1146	1246.7	780.3	−27.6	1.4005	0.1720
0.0584	0.0584	1258.2	772.2	−13.4	1.4035	0.0140
0.0000	0.0000	1271.6	762.4	0.00	1.4100	0.0000

Viscosity Deviation. The measured η values for binary systems are listed in Table 2 and depicted graphically in Figure 2. The viscosity deviations ($\Delta\eta$) from a linear dependence for binary (n = 2) and ternary (n = 3) mixtures can be calculated as[13]

$$\Delta\eta = \eta - \sum_{i=1}^{n} x_i\eta_i \tag{2}$$

where η is the viscosity of the mixture and x_i and η_i are the mole fraction and viscosity, respectively, of pure component i.

The values of $\Delta\eta$ over entire range of mole fraction for the binary mixtures follow the trend which is schematically shown below:



The trend in the deviations of the $\Delta\eta$ values was found to be the opposite of that in the V^E values for all three binary mixtures, in agreement with the view proposed by Brocos et al.[8] The values of $\Delta\eta$ for the ternary mixture are listed in Table 3. For

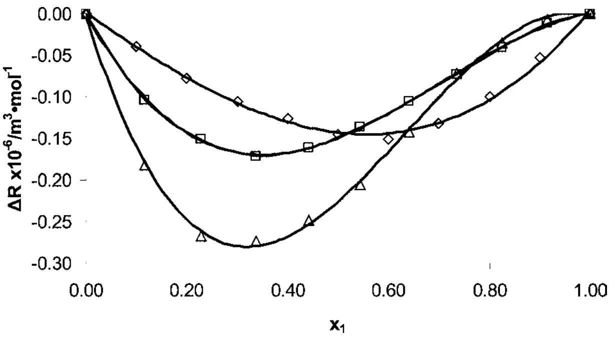


Figure 4. Excess molar refractivities $\Delta R \cdot 10^{-6}/\text{m}^3 \cdot \text{mol}^{-1}$ as functions of mole fraction (x_1) for the for the three binary subsystems at $T = 298.15$ K: □, 1,3-dioxolane (1) + *n*-amyl alcohol (2); ◇, 1,3-dioxolane (1) + diethyl ether (2); △, diethyl ether (1) and *n*-amyl alcohol (2).

Table 6. Redlich−Kister Coefficients (a_n) and Standard Deviations (σ) for the Binary Mixtures at T = 298.15 K

excess property	a_0	a_1	a_2	a_3	σ
1,3-Dioxolane (1) + Diethyl Ether (2)					
$10^6 \cdot V^E/\text{m}^3 \cdot \text{mol}^{-1}$	−5.915	−1.079	−1.113		0.021
$\Delta\eta/\text{mPa} \cdot \text{s}$	−0.230	−0.056			0.001
$10^{12} \cdot \Delta K_S/\text{Pa}^{-1}$	−480.02	−127.24	222.66		1.81
$10^{-6} \cdot \Delta R/\text{m}^3 \cdot \text{mol}^{-1}$	−0.5842	−0.2040	0.0988	0.1994	0.0024
1,3-Dioxolane (1) + *n*-Amyl Alcohol (2)					
$10^6 \cdot V^E/\text{m}^3 \cdot \text{mol}^{-1}$	−1.623	1.286	−0.196		0.005
$\Delta\eta/\text{mPa} \cdot \text{s}$	−3.717	2.154	−0.732	0.356	0.013
$10^{12} \cdot \Delta K_S/\text{Pa}^{-1}$	−22.97	−13.39	6.97		0.27
$10^{-6} \cdot \Delta R/\text{m}^3 \cdot \text{mol}^{-1}$	−0.5995	0.4953			0.0019
Diethyl Ether (1) + *n*-Amyl Alcohol (2)					
$10^6 \cdot V^E/\text{m}^3 \cdot \text{mol}^{-1}$	−7.480	−1.698	−1.748		0.029
$\Delta\eta/\text{mPa} \cdot \text{s}$	−3.548	0.820	0.352		0.014
$10^{12} \cdot \Delta K_S/\text{Pa}^{-1}$	−574.02	−204.92	69.38		2.30
$10^{-6} \cdot \Delta R/\text{m}^3 \cdot \text{mol}^{-1}$	−0.8943	1.0672	−0.0883		0.0048

the ternary mixture, the viscosity deviations are negative over the entire range of composition. These can be interpreted qualitatively by considering the effect of the intermolecular interactions and the shapes of the components.

Deviation in Isentropic Compressibility. Isentropic compressibility (K_S) and deviations in isentropic compressibility (ΔK_S) for binary ($n = 2$) and ternary ($n = 3$) mixtures were calculated using the following relations:

$$K_S = (u^2\rho)^{-1} \tag{4}$$

$$\Delta K_S = K_S - \sum_{i=1}^{n} x_i K_{S,i} \tag{5}$$

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

It is evident from Table 4 and Figure 3 that the ΔK_S values for the binary mixtures are negative over the entire composition range and that order of negative deviation is as follows: DEE (1) + AL (2) > 1,3-DO (1) + DEE (2) > 1,3-DO (1) + AL (2). These results can be explained in terms of molecular interactions[9,10] between unlike molecules. It appears from the sign and magnitude of ΔK_S that specific interactions between the mixture components exist. These results are in excellent agreement with those for V^E discussed earlier.

The values of ΔK_S for the ternary mixture (Table 5) are negative over the entire composition range, which suggests that specific interactions exist between the mixture components.[11]

Excess Molar Refractivity. The molar refractivity $[R]$ gives more information than n_D about the mixture phenomenon because it takes into account electronic perturbations of the molecular orbitals during the liquid mixture process; in addition, $[R]$ is directly related to the dispersion forces, It can be evaluated from the Lorentz−Lorenz relation:[12]

$$[R] = \left(\frac{n_D^{\,2} - 1}{n_D^{\,2} + 2}\right)\left(\frac{M}{\rho}\right) \tag{6}$$

where $[R]$, n_D, and M are the molar refractivity, refractive index, and molar mass of the mixture, respectively. The excess molar refractivity was calculated from the following relation:

$$\Delta R = [R] - (x_1[R]_1 + x_2[R]_2) \tag{7}$$

The values of n_D and ΔR for the binary mixtures are presented in Table 4, and the ΔR values are depicted graphically in Figure 4. The data for ternary mixtures are reported in Table 5.

The excess properties (V^E, $\Delta\eta$, ΔK_S, ΔR) for the binary mixtures were fitted to the Redlich−Kister polynomial equation,[13]

$$Y^E = x_1 x_2 \sum_{n=0}^{K} a_n (x_1 - x_2)^n \tag{8}$$

where Y^E denotes an excess property. The coefficients a_n were obtained by fitting eq 8 to the experimental results using a least-squares regression method. In each case, the optimal number of coefficients, K, was ascertained from an approximation of the variation in the standard deviation. The fitted values of the coefficients along with the standard deviations are summarized for all of the mixtures in Table 6. The standard deviation was calculated using the equation

$$\sigma = \left[\frac{\sum (Y^E_{\text{exptl}} - Y^E_{\text{calcd}})^2}{(N - m)}\right]^{1/2} \tag{9}$$

where N is the number of data points and m is the number of coefficients.

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Received for review May 30, 2010. Accepted August 13, 2010. The authors are grateful to the Departmental Special Assistance Scheme under the University Grants Commission, New Delhi (540/6/DRS/2007, SAP-2) for financial support.

JE100594U