Density, Viscosity, and Speed of Sound in the Ternary Mixtures of 2-Ethoxyethanol + *N*,*N*-Dimethylformamide + *N*,*N*-Dimethylacetamide and 2-Ethoxyethanol + Dimethyl Sulfoxide + *N*,*N*-Dimethylacetamide at 308.15 K

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Experimental values of density, viscosity, and speed of sound of the ternary mixtures of 2-ethoxyethanol (1) + *N*,*N*-dimethylformamide (2) + *N*,*N*-dimethylacetamide (3) and 2-ethoxyethanol (1) + dimethyl sulfoxide (2) + *N*,*N*-dimethylacetamide (3) at 308.15 K and at 18 different concentrations are reported. From the data, the excess volume and the deviations in adiabatic compressibility, viscosity, and speed of sound for the ternary mixtures over the additive values were calculated. The deviations were fitted using the Redlich−Kister equation.

1. Introduction

It is not without reason that multicomponent liquid systems have attracted the attention of researchers in the past decade. Such systems find industrial applications such as heat transfer, fluid flow, and so forth. The present work is a continuation of our previous study[1,2] of physicochemical properties of nonaqueous binary and ternary liquid mixtures. The liquids were selected on the basis of their industrial use. 2-Ethoxyethanol, *N*,*N*-dimethylformamide, *N*,*N*-dimethylacetamide, and dimethyl sulfoxide are important liquids which find a variety of applications such as as solvents for lacquers, oils, and resins, as antifreeze for explosives, and so forth. To the best of our knowledge, the properties of mixtures of these liquids have not been reported earlier.

2. Experimental Details

2.1. Materials. Spectroscopic and HPLC grade (purities of 99.5% or better) 2-ethoxyethanol, dimethylformamide, dimethylacetamide and dimethyl sulfoxide were from Merck (India), and no further purification was done. The liquids were stored over molecular sieves. The experimental values of density, viscosity, and speed of sound of the mixtures were determined immediately after mixing. Experimental results of density, viscosity, and ultrasonic sound of the pure liquids at 298.15 K are compared with published data in Table 1.

2.2. Measurements. Experimental details about the preparation of mixtures and the measurements of mass, density, speed of sound, and viscosity of pure liquids and mixtures are the same as those described previously.[3] The ternary mixtures were prepared by mass, by mixing calculated volumes of liquid components in airtight glass bottles. In all the property measurements, an INSREF thermostat was used at a constant digital temperature display accurate to ±0.01 K. All the measurements were carried out at 308.15 K. For all the mixtures and pure solvents, triplicate measurements were performed and the average of these values was considered in all calculations. The mass measurements (±0.01 mg) were made using an electronic balance. The accuracy of the density measurements was 0.0001 g.cm^{-3}, and the mole fraction values were reproducible within 0.0001. A set of 18 compositions was prepared for each system, and their physical properties were measured on the same day. A 10 mL specific gravity bottle and an Oswald viscometer were used for the determination of density and viscosity, respectively. An electronic digital stopwatch with a readability of ±0.01 s was used for flow time measurements. The measured viscosity values are accurate to ±0.001 mPa·s, and the entire viscometer with a tight fitting stopper was held within the constant-temperature bath to prevent convective flow. Speed of sound was determined using a constant frequency (2 MHz) variable path ultrasonic interferometer (model F-81, Mittal Enterprises, New Delhi) with an accuracy of ±1 m·s^{-1} that was calibrated using water and benzene. Calibration of the viscometer was done as outlined in ref 4.

Table 1. Density, ρ, Viscosity, η, and Ultrasonic Speed of Pure Liquids at 298.15 K

	ρ/g·cm^{-3}		η/mPa·s		u/m·s^{-1}	
liquid	expt	ref 8	expt	ref 9	expt	ref 10
2-ethoxyethanol	0.9256	0.9258	1.851	1.850	1308	1300
N,*N*-dimethylformamide	0.9447	0.9442	0.802	0.802	1465	1462
N,*N*-dimethylacetamide	0.9376	0.9372	0.937	0.937	1462	1458
dimethyl sulfoxide	1.0962	1.0958	1.997	1.996	1493	1488

3. Results and Discussion

Deviations of thermodynamical parameters from their ideal (or additive) values give information regarding molecular interactions existing in the mixture. The excess volumes were calculated from

$$V^{E} = V_{m} - (x_1 V_1^{\circ} + x_2 V_2^{\circ} + x_3 V_3^{\circ}) \qquad (1)$$

where V_i° is the molar volume of the i-th pure liquid. Deviations of the other properties were calculated from

$$\Delta X = X_{m} - (x_1 X_1^{\circ} + x_2 X_2^{\circ} + x_3 X_3^{\circ}) \qquad (2)$$

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Table 2. Experimental Values of Density, ρ, Viscosity, η, and Ultrasonic Speed, u, of 2-Ethoxyethanol (1) + N,N-Dimethylformamide (2) + N,N-Dimethylacetamide (3) at 308.15 K

x_1	x_2	ρ $\mathrm{g \cdot cm^{-3}}$	η $\mathrm{mPa \cdot s}$	u $\mathrm{m \cdot s^{-1}}$
0.3993	0.6006	0.9281	1.342	1373
0.3994	0.4980	0.9268	1.611	1383
0.3974	0.4018	0.9256	1.489	1384
0.4014	0.3010	0.9251	1.307	1377
0.3973	0.2003	0.9245	2.229	1388
0.3973	0.0986	0.9232	1.246	1394
0.4043	0.0000	0.9243	1.216	1385
0.6008	0.3991	0.9244	2.644	1348
0.4989	0.4010	0.9247	1.845	1368
0.3010	0.3999	0.9264	1.663	1402
0.2033	0.4026	0.9274	1.155	1419
0.0967	0.4011	0.9283	0.940	1433
0.0000	0.3996	0.9303	0.916	1443
0.5979	0.0000	0.9221	1.710	1362
0.4966	0.1021	0.9233	2.750	1379
0.3009	0.2985	0.9262	2.770	1391
0.0980	0.5000	0.9298	1.226	1423
0.0000	0.5984	0.9315	1.227	1445

Table 3. Experimental Values of Density, ρ, Viscosity, η, and Ultrasonic Speed, u, of 2-Ethoxyethanol (1) + Dimethyl Sulfoxide (2) + N,N-Dimethylacetamide (3) at 308.15 K

x_1	x_2	ρ $\mathrm{g \cdot cm^{-3}}$	η $\mathrm{mPa \cdot s}$	u $\mathrm{m \cdot s^{-1}}$
0.3996	0.6002	1.0105	1.991	1383
0.3973	0.5019	0.9954	1.809	1404
0.4064	0.3953	0.9783	2.524	1402
0.4028	0.3002	0.9666	2.466	1407
0.3962	0.2007	0.9521	1.797	1401
0.3943	0.0996	0.9387	1.514	1388
0.3970	0.0000	0.9285	1.154	1384
0.5990	0.4009	0.9791	2.304	1364
0.4994	0.4027	0.9800	2.077	1384
0.3022	0.3999	0.9805	1.519	1411
0.2044	0.3988	0.9820	1.355	1429
0.0968	0.4006	0.9833	1.347	1445
0.0000	0.3989	0.9868	1.343	1460
0.5992	0.0000	0.9235	1.678	1355
0.5034	0.0917	0.9356	2.059	1370
0.2972	0.3008	0.9668	1.739	1410
0.0996	0.4990	0.9998	3.002	1455
0.0000	0.5831	1.0234	2.787	1479

Table 4. Excess Values of Volume, V^E, Adiabatic Compressibility, $\beta_a{}^E$, Viscosity, η^E, and Ultrasonic Speed, u^E, of 2-Ethoxyethanol (1) + N,N-Dimethylformamide (2) + N,N-Dimethylacetamide (3) at 308.15 K

x_1	x_2	V^E $\mathrm{cm^3 \cdot mol^{-1}}$	$10^{10}\Delta\beta_a$ $\mathrm{m^2 \cdot N^{-1}}$	$\Delta\eta$ $\mathrm{mPa \cdot s}$	Δu $\mathrm{m \cdot s^{-1}}$
0.3993	0.6006	−0.14	−0.17	0.31	2
0.3994	0.4980	−0.09	−0.25	0.57	14
0.3974	0.4018	−0.03	−0.25	0.45	16
0.4014	0.3010	−0.04	−0.20	0.26	11
0.3973	0.2003	−0.04	−0.29	1.18	23
0.3973	0.0986	0.04	−0.33	0.19	31
0.4043	0.0000	−0.14	−0.28	0.15	24
0.6008	0.3991	−0.15	−0.24	1.47	10
0.4989	0.4010	−0.06	−0.27	0.74	15
0.3010	0.3999	0.01	−0.26	0.68	20
0.2033	0.4026	0.03	−0.25	0.24	23
0.0967	0.4011	0.07	−0.20	0.09	21
0.0000	0.3996	−0.01	−0.13	0.13	17
0.5979	0.0000	−0.14	−0.34	0.52	29
0.4966	0.1021	−0.09	−0.35	1.63	30
0.3009	0.2985	−0.04	−0.18	1.79	10
0.0980	0.5000	−0.01	−0.12	0.38	10
0.0000	0.5984	0.01	−0.12	0.45	16

Table 5. Excess Values of Volume, V^E, Adiabatic Compressibility, $\beta_a{}^E$, Viscosity, η^E, and Ultrasonic Speed, u^E, of 2-Ethoxyethanol (1) + Dimethyl Sulfoxide (2) + N,N-Dimethylacetamide (3) at 308.15 K

x_1	x_2	V^E $\mathrm{cm^3 \cdot mol^{-1}}$	$10^{10}\Delta\beta_a$ $\mathrm{m^2 \cdot N^{-1}}$	$\Delta\eta$ $\mathrm{mPa \cdot s}$	Δu $\mathrm{m \cdot s^{-1}}$
0.3996	0.6002	−0.44	−0.28	0.42	4
0.3973	0.5019	−0.47	−0.44	0.32	20
0.4064	0.3953	−0.37	−0.44	1.12	24
0.4028	0.3002	−0.52	−0.48	1.15	33
0.3962	0.2007	−0.41	−0.42	0.57	30
0.3943	0.0996	0.36	−0.31	0.37	21
0.3970	0.0000	−0.57	−0.29	0.09	22
0.5990	0.4009	−0.63	−0.44	0.77	14
0.4994	0.4027	−0.55	−0.45	0.61	20
0.3022	0.3999	−0.36	−0.35	0.18	18
0.2044	0.3988	−0.38	−0.33	0.08	22
0.0968	0.4006	−0.32	−0.28	0.14	22
0.0000	0.3989	−0.52	−0.24	0.20	23
0.5992	0.0000	−0.28	−0.29	0.49	22
0.5034	0.0917	−0.28	−0.30	0.85	20
0.2972	0.3008	−0.39	−0.34	0.49	20
0.0996	0.4990	−0.46	−0.33	1.71	28
0.0000	0.5831	−1.12	−0.34	1.49	34

To calculate the deviations of adiabatic compressibility, volume fractions (ϕ_i) were used instead of mole fractions. These two are related to one another as

$$\phi_i = \frac{x_i V_i^\circ}{\sum x_i V_i^\circ} \tag{3}$$

Adiabatic compressibility, β_a, was estimated from the speed of sound, u, and density, ρ, using $\beta_a = 1/u^2\rho$.

The calculated deviations of the properties were fitted using a Redlich−Kister polynomial of the type[5]

$$\Delta A_{123} = \Delta A_{12} + \Delta A_{23} + \Delta A_{31} + x_1 x_2 x_3 [C_0 + C_1(x_1 - x_2) + C_2(x_1 - x_2)^2] \tag{4}$$

where ΔA_{ij} is the deviation of the property A of the binary mixture formed by the i-th and j-th liquids. The Redlich−Kister coefficients, C_i's, were determined using a least-squares fit. The standard deviations, σ, of the fitted values were obtained using

$$\sigma = \left[\frac{\sum(\Delta A - \Delta A_{\text{calc}})^2}{M - n}\right]^{1/2} \tag{5}$$

where M and n are the number of experimental points and coefficients used in eq 4, respectively.

Experimental values of the density, viscosity, and speed of sound of the two ternary mixtures are reported in Tables 2 and 3. Excess values of volume and deviations in adiabatic compressibility, viscosity, and speed of sound are given in Tables 4 and 5. Redlich−Kister coefficients and the standard deviations for both the liquid systems are given in Table 6. The mole fraction of one of the liquids was kept constant at ≈0.4, and those of the remaining two liquids were varied such that the sum of their mole fractions was ≈0.6.

For the system 2-ethoxyethanol + dimethylformamide + dimethylacetamide, V^E is negative except for a few ternary mixtures (Table 4). The negative values of excess volume suggest specific hydrogen bond type interactions between the hydroxyl group of 2-ethoxyethanol and the

Table 6. Redlich−Kister Coefficients and the Standard Deviations

	A_0	A_1	A_2	σ
2-Ethoxyethanol (1) + *N*,*N*-Dimethylformamide (2) + *N*,*N*-Dimethylacetamide (3)				
V^E/cm^3·mol^{-1}	−0.16	−4.34	−94.19	0.008
$10^{11}\Delta\beta_a$/m^2·N^{-1}	−6.36	9.41	−351.99	0.015
$\Delta\eta$/mPa·s	20.24	−84.33	765.68	0.060
2-Ethoxyethanol (1) + Dimethyl Sulfoxide (2) + *N*,*N*-Dimethylacetamide (3)				
V^E/cm^3·mol^{-1}	−12.23	−18.69	−325.36	0.042
$10^{11}\Delta\beta_a$/m^2·N^{-1}	−11.31	−16.73	−310.42	0.021
$\Delta\eta$/mPa·s	19.64	8.58	335.99	0.065

amide groups in dimethylformamide and dimethylacetamide. In both the ternary mixtures, $\Delta\beta_a$ values are negative for the entire mole fraction range, further supporting the specific interactions in these mixtures.

In the system 2-ethoxyethanol + dimethyl sulfoxide + dimethylacetamide, the trend of the excess values (Table 5) is the same as that of the other ternary system, the only differences being the negative values are more negative in the second case and the hydrogen bond type interaction is between the hydroxyl group of 2-ethoxyethanol and the sulfoxide group of dimethyl sulfoxide.

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