Densities, Viscosities, and Speeds of Sound for Diethylene Glycol Dimethyl Ether + Methyl Acetate

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Densities, speeds of sound, and viscosities of binary mixtures of diethylene glycol dimethyl ether + methyl acetate have been measured in the temperature interval of 298.15–313.15 K. From these data excess molar volume, viscosity, and speed of sound deviations have been derived. The results are fitted to the Redlich–Kister polynomial relation and are presented as a function of composition.

Introduction

The study of molecular interactions in binary organic mixtures has been actively investigated in our laboratories (1–5). Knowledge of mixing properties of binary mixtures has relevance in theoretical and applied areas of research because such results are needed for design processes in chemical and petrochemical industries.

Diethylene glycol dimethyl ether (bis(2-methoxyethyl) ether or dimethyl diglycol) finds application as a medium in polymerization reactions. However, esters are used as plasticizing agents in the polymer processing industries. Therefore, an understanding of the mixing properties of binary mixtures of these compounds has value in the polymer industry. A search of the literature suggests that the physical properties of these mixtures have not been studied. As a contribution in this area, we now present the experimental data of densities, ρ, viscosities, η, and speeds of sound, u, of binary mixtures of diethylene glycol dimethyl ether with methyl acetate. From these data the mixing properties have been calculated.

Experimental Section

Materials. Diethylene glycol dimethyl ether (BDH, England) was used directly, and methyl acetate (Sisco, India) was purified by the recommended methods (6, 7). The purity of methyl acetate was ascertained by the constancy of its boiling temperature during the final distillation and also by comparing its density and refractive index at 298.15 K which agreed reasonably with the literature values (see Table I) (8, 9). The gas chromatographic tests using a flame ionization detector (Nucon Series 5700/5765 with fused silica columns) having a sensitivity better than 10^{-8} g of fatty acid/μL solvent showed a purity of >99 mol %.

Mixtures were prepared by mixing the appropriate volumes of liquids in specially designed ground-glass air-tight bottles and weighed in a single-pan Mettler balance, AE-240 (Switzerland), to an accuracy of ±0.01 mg. Preferential evaporation losses of solvents from the mixtures were kept to a minimum as evidenced by a repeated measurement of the physical properties over an interval of 2–3 days, during which time no changes in the physical properties were observed. The possible uncertainty in the mole fractions is around ±0.0001. Stock solutions of the mixture were kept in air-tight glass-stoppered bottles and then transferred carefully to the viscometer, pycnometer, and ultrasonic cell in a controlled atmosphere, taking extreme care to avoid contamination with external air or other impurities.

Table I. Comparison of Densities ρ and Refractive Indexes n_D of Liquids with the Literature at 298.15 K

	ρ/(g cm^{-3})		n_D	
liquid	obsd	lit.	obsd	lit.
diethylene glycol dimethyl ether	0.9396	0.9384[a]	1.4070	1.4058[a]
methyl acetate	0.9213	0.9274[b]	1.3587	1.3588[c]

[a] Reference 7. [b] Reference 8. [c] Reference 9.

Measurements. Densities of pure liquids and their binary mixtures in the composition range 0.1–0.9 at 0.1 mole fraction increments were measured with a pycnometer having a bulb volume of about 10 cm^3 and a capillary with an internal diameter of 1 mm. The procedural details were given earlier (1–5). The densities at 298.15, 303.15, 308.15, and 313.15 K are considered significant to four figures. An average of triplicate measurements was taken into account, and these were reproducible within ±0.0001 g cm^{-3}.

Viscosities were measured with a Cannon Fenske viscometer (size 75) ASTM D 445, supplied by the Industrial Research Glassware Ltd., New Jersey. An electronic stopwatch with a precision of ±0.01 s was used for flow time measurements. Triplicate measurements of flow times were reproducible within ±0.01 s. The calibration of the viscometer and the experimental details are the same as given earlier (1–5). The viscosities are accurate to ±0.001 mPa s. The viscosity of methyl acetate at 303.15 K, 0.366 mPa s, compares well with the literature (8) value of 0.361 mPa s.

The speeds of sound were measured with a variable-path single-crystal interferometer (Mittal Enterprises, New Delhi, Model M-84) as described in earlier publications (1–5). The speed of sound data are accurate to ±2 ms^{-1}. Experimental data of ρ, η, and u at different temperatures are given in Table II.

Results and Discussion

The experimental values of densities are used to calculate the excess molar volume, V^E, of the mixture as

$$V^E/(\text{cm}^3\ \text{mol}^{-1}) = V_m - V_1x_1 - V_2x_2 \qquad (1)$$

where V_m is the molar volume of the mixture, V_1 and V_2 are the molar volumes of the components 1 and 2, respectively, and x_1 and x_2 refer to mole fractions of components 1 and 2.

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Table II. Experimental Densities ρ, Viscosities η, Speeds of Sound *u*, and Excess Molar Volumes V^E of Diethylene Glycol Dimethyl Ether (1) + Methyl Acetate (2) at Different Temperatures

x_1	ρ/(g cm^{-3})	η/(mPa s)	u/(m s^{-1})	V^E/(cm^3 mol^{-1})
		298.15 K		
0.0000	0.9213	0.385	1163	
0.1000	0.9248	0.441	1182	−0.046
0.1980	0.9277	0.498	1202	−0.078
0.2969	0.9302	0.559	1218	−0.109
0.4015	0.9323	0.621	1232	−0.122
0.4988	0.9342	0.683	1243	−0.142
0.5951	0.9357	0.741	1254	−0.147
0.6954	0.9370	0.801	1264	−0.135
0.7941	0.9380	0.861	1273	−0.100
0.8973	0.9387	0.922	1279	−0.038
1.0000	0.9396	0.973	1288	
		303.15 K		
0.0000	0.9144	0.366	1141	
0.1000	0.9184	0.418	1160	−0.060
0.1980	0.9216	0.470	1180	−0.109
0.2969	0.9243	0.526	1196	−0.130
0.4015	0.9266	0.582	1211	−0.136
0.4988	0.9285	0.639	1223	−0.148
0.5951	0.9301	0.691	1234	−0.139
0.6954	0.9316	0.746	1242	−0.134
0.7941	0.9326	0.799	1251	−0.087
0.8973	0.9336	0.855	1258	−0.033
1.0000	0.9346	0.904	1265	
		308.15 K		
0.0000	0.9077	0.348	1118	
0.1000	0.9120	0.396	1138	−0.073
0.1980	0.9155	0.445	1158	−0.121
0.2969	0.9183	0.496	1176	−0.144
0.4015	0.9207	0.548	1191	−0.140
0.4988	0.9228	0.600	1202	−0.153
0.5951	0.9247	0.646	1213	−0.165
0.6954	0.9263	0.696	1222	−0.147
0.7941	0.9275	0.745	1229	−0.110
0.8973	0.9284	0.795	1237	−0.041
1.0000	0.9295	0.839	1246	
		313.15 K		
0.0000	0.9004	0.331	1096	
0.1000	0.9058	0.376	1116	−0.142
0.1980	0.9093	0.418	1137	−0.164
0.2969	0.9124	0.469	1155	−0.190
0.4015	0.9149	0.516	1171	−0.166
0.4988	0.9173	0.564	1181	−0.192
0.5951	0.9191	0.607	1193	−0.177
0.6954	0.9208	0.652	1202	−0.160
0.7941	0.9221	0.697	1209	−0.098
0.8973	0.9233	0.741	1217	−0.047
1.0000	0.9244	0.781	1225	

The deviations of the viscosity and speed of sound from a mole fraction average are given by

$$\Delta Y = Y_m - Y_1 x_1 - Y_2 x_2 \qquad (2)$$

where ΔY refers to either $\Delta\eta$ or Δu and the same meaning applies to Y_1 and Y_2. The computed values of V^E are included in Table II. The results of V^E, $\Delta\eta$, and Δu are presented in Figure 1. The values of V^E are negative while those of Δu are positive over the entire composition scale of the mixture. However, $\Delta\eta$ values vary sigmiodally from negative to positive values.

Each set of the derived quantities Y^E ($=V^E$, $\Delta\eta$, and Δu) is fitted to the Redlich–Kister polynomial relation

$$\Delta Y \text{ or } V^E = x_1 x_2 \sum_{i=0}^{3} a_i (x_2 - x_1)^i \qquad (3)$$

to estimate the regression coefficients, a_i, and standard errors, σ. These values are summarized in Table III. A third-order fit in almost all cases reproduced insignificant differences between the calculated and observed excess quantities.

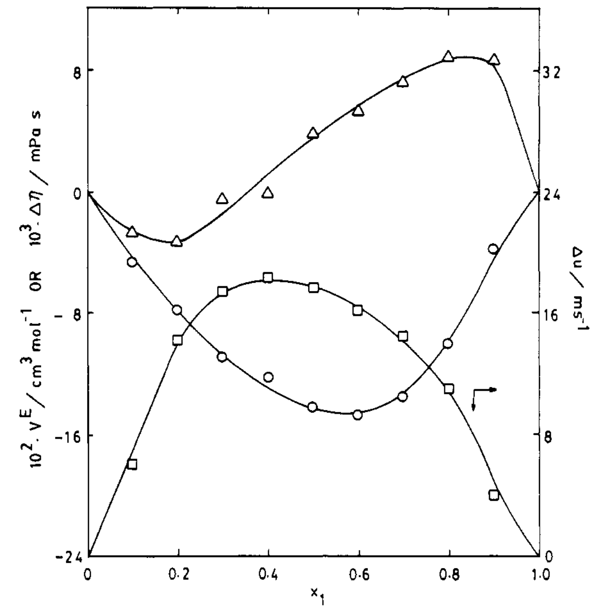

Figure 1. Dependence of excess volumes, changes in viscosities, and speeds of sound on the mole fraction of the mixture at 298.15 K: (○), V^E, (△) $\Delta\eta$, (□) Δu.

Table III. Estimated Parameters and Standard Errors of Equation 3

quantity	T/K	a_0	a_1	a_2	a_3	σ
V^E/(cm^3 mol^{-1})	298.15	−0.575	0.250	0.110	−0.472	0.005
	303.15	−0.595	0.051	0.025	−0.422	0.008
	308.15	−0.640	0.161	−0.121	−0.645	0.009
	313.15	−0.730	0.043	−0.387	−1.137	0.012
$\Delta\eta$/(mPa s)	298.15	0.012	−0.044	0.023	−0.053	0.001
	303.15	0.014	−0.033	0.005	−0.039	0.001
	308.15	0.020	−0.027	0.005	−0.041	0.001
	313.15	0.030	−0.031	−0.001	−0.026	0.002
Δu/(m s^{-1})	298.15	73.451	20.883	−5.462	−7.870	1.231
	303.15	79.576	21.230	−16.266	−15.880	0.789
	308.15	86.218	36.359	−31.905	−25.097	0.603
	313.15	89.110	38.296	−22.971	−26.669	1.026

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