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Density and Viscosity Studies of Binary Mixtures of *N*,*N*-Dimethylformamide with Toluene and Methyl Benzoate at (298.15, 303.15, 308.15, and 313.15) K

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Densities and viscosities of binary mixtures of *N*,*N*-dimethylformamide with toluene and methyl benzoate have been measured over the entire range of composition, at (298.15, 303.15, 308.15, and 313.15) K and at atmospheric pressure. From the experimental data, excess molar volumes (V^E) and deviations in viscosity ($\Delta\eta$) have been calculated. More-negative excess molar volumes for *N*,*N*-dimethylformamide + toluene systems are due to π-electron donor−acceptor-type specific interactions between toluene and *N*,*N*-dimethylformamide molecules. The deviations in viscosity for *N*,*N*-dimethylformamide + toluene systems are very small. The excess molar volumes and deviations in viscosity have been fit to the Redlich−Kister polynomial equation. McAllister's three-body interaction model has also been used to correlate the kinematic viscosities of binary liquid mixtures with mole fractions.

Introduction

Studies on thermodynamic and transport properties are important in understanding the nature of molecular interactions in binary mixtures. *N*,*N*-Dimethylformamide is a versatile solvent used in the separation of saturated and unsaturated hydrocarbons and in pharmaceutical synthesis and serves as a solvent for many polymers. Thermodynamic and transport properties of binary mixtures of *N*,*N*-dimethylformamide with different organic liquids have been studied by many authors.[1−7] The calculated excess quantities from such data have been interpreted in terms of specific and nonspecific interactions taking place between the components of the mixtures. There are few reports[8,9] on the density and viscosity data of binary mixtures of *N*,*N*-dimethylformamide with toluene, but there are no reports on the density and viscosity data of binary mixtures of *N*,*N*-dimethylformamide with methyl benzoate. Therefore, in the present paper we report density and viscosity studies of binary mixtures of *N*,*N*-dimethylformamide with toluene and methyl benzoate over the entire range of composition, at (298.15, 303.15, 308.15, and 313.15) K and at atmospheric pressure.

Experimental Section

Methyl benzoate (PCL, purity >99%) was purified by a suitable procedure[10] involving distillation and drying. Toluene (S. D. Fine Chemicals, analytical reagent, purity >99.5%) and *N*,*N*-dimethylformamide (S. D. Fine Chemicals, analytical reagent, purity >99.5%) were directly used. The purity of the solvents was ascertained by GLC and also by comparing experimental values of densities and viscosities with those reported in the literature (Table 1). Our experimental values of densities and viscosities match very well with those reported in the literature. Binary mixtures were prepared by mixing a known mass of each liquid in an airtight, stoppered glass bottle. The masses were recorded on an Adairdutt balance to an accuracy of $\pm 1 \times 10^{-4}$ g. The uncertainty in the mole fractions was $\pm 1 \times 10^{-4}$. Care was taken to avoid contamination during mixing.

Table 1. Comparison of Experimental Densities (ρ) and Viscosities (η) of Pure Liquids with Literature Values at (298.15, 303.15, 308.15, and 313.15) K

		ρ/g·cm^{-3}		η/mPa·s	
liquid	T/K	exptl	lit	exptl	lit
toluene	298.15	0.8622	0.8622[10]	0.558	0.552[17]
	303.15	0.8576	0.8575[10]	0.526	0.526[16]
	308.15	0.8530		0.496	0.492[10]
	313.15	0.8482	0.8483[17]	0.470	0.471[16]
methyl benzoate	298.15	1.0850	1.0839[21]	1.918	
	303.15	1.0803	1.0790[21]	1.778	1.673[21]
	308.15	1.0756	1.0739[21]	1.663	
	313.15	1.0708	1.0690[21]	1.402	
DMF	298.15	0.9445	0.9445[18]	0.803	0.802[10]
	303.15	0.9398	0.9397[19]	0.756	0.752[7]
	308.15	0.9351	0.9356[19]	0.710	0.707[7]
	313.15	0.9302	0.9298[20]	0.673	0.664[7]

The densities of degassed, pure liquids and binary mixtures were measured using a 15-cm^3 double-arm pycnometer, as reported earlier,[11] in a transparent glass- walled water bath having a thermal stability of ±0.01 K. The pycnometer was calibrated using conductivity water[12] having a conductivity of $<1 \times 10^{-6}\ \Omega^{-1}\ \text{cm}^{-1}$. The uncertainty in the density measurements was within $\pm 1 \times 10^{-4}$ g·cm^{-3}.

Dynamic viscosities were measured using an Ubbelohde suspended-level viscometer,[11] calibrated with conductivity water.[12] An electronic digital stopwatch with an accuracy of ±0.01 s was used for flow-time measurements. At least three repetitions of each data point obtained were reproducible to ±0.05 s, and the results were averaged. Because all flow times were >300 s, kinetic energy corrections were not applied. To evaluate the viscometer constant, the length of the capillary of the viscometer (l) term is to be corrected as $l' = l + 0.5\ r$, with r being the radius of the viscometer capillary. Because l is much larger (50 to 60 mm) than r (r = 0.5 mm), $l = l'$ and hence end effects in viscometers are

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Table 2. Density (ρ), Viscosity (η), Excess Molar Volume (V^E), and Deviations in Viscosity ($\Delta\eta$) for DMF (1) + Toluene (2) and DMF (1) + Methyl Benzoate (2) at (298.15, 303.15, 308.15, and 313.15) K

x_1	$10^3 \rho$ / kg·m^{-3}	η / mPa·s	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s	x_1	$10^3 \rho$ / kg·m^{-3}	η / mPa·s	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s
DMF (1) + Toluene (2) 298.15 K									
0.0000	0.8622	0.558	0.000	0.000	0.6570	0.9150	0.723	−0.476	0.004
0.1224	0.8713	0.584	−0.184	−0.004	0.7413	0.9222	0.742	−0.411	0.002
0.2414	0.8805	0.612	−0.327	−0.005	0.8336	0.9300	0.764	−0.291	0.001
0.3485	0.8891	0.639	−0.426	−0.004	0.9129	0.9359	0.781	−0.084	−0.001
0.4031	0.8936	0.654	−0.465	−0.003	1.0000	0.9445	0.803	0.000	0.000
0.5545	0.9063	0.699	−0.508	0.005					
DMF (1) + Toluene (2) 303.15 K									
0.0000	0.8576	0.526	0.000	0.000	0.6570	0.9104	0.680	−0.487	0.003
0.1224	0.8667	0.550	−0.187	−0.004	0.7413	0.9176	0.698	−0.423	0.001
0.2414	0.8759	0.576	−0.333	−0.005	0.8336	0.9254	0.718	−0.301	0.000
0.3485	0.8845	0.601	−0.434	−0.005	0.9129	0.9313	0.735	−0.092	−0.001
0.4031	0.8890	0.615	−0.474	−0.004	1.0000	0.9398	0.756	0.000	0.000
0.5545	0.9018	0.657	−0.529	−0.003					
DMF (1) + Toluene (2) 308.15 K									
0.0000	0.8530	0.496	0.000	0.000	0.6570	0.9058	0.638	−0.498	0.001
0.1224	0.8621	0.518	−0.190	0.004	0.7413	0.9130	0.655	−0.434	0.002
0.2414	0.8713	0.541	−0.339	−0.007	0.8336	0.9208	0.674	−0.312	0.000
0.3485	0.8799	0.565	−0.442	0.006	0.9129	0.9268	0.690	−0.110	−0.001
0.4031	0.8844	0.578	−0.483	0.004	1.0000	0.9351	0.710	0.000	0.000
0.5545	0.8972	0.617	−0.540	0.002					
DMF (1) + Toluene (2) 313.15 K									
0.0000	0.8482	0.470	0.000	0.000	0.6570	0.9011	0.604	−0.520	0.001
0.1224	0.8573	0.490	−0.914	−0.005	0.7413	0.9082	0.619	−0.446	−0.001
0.2414	0.8665	0.512	−0.346	−0.007	0.8336	0.9160	0.638	−0.323	−0.001
0.3485	0.8751	0.534	−0.451	−0.007	0.9129	0.9220	0.653	−0.119	−0.002
0.4031	0.8796	0.547	−0.493	−0.005	1.0000	0.9302	0.673	0.000	0.000
0.5545	0.8924	0.584	−0.551	0.001					
DMF (1) + Methyl Benzoate (2) 298.15 K									
0.0000	1.0850	1.918	0.000	0.000	0.6218	1.0159	1.141	−0.154	−0.084
0.1213	1.0744	1.740	−0.047	−0.043	0.7468	0.9956	1.020	−0.113	−0.065
0.1914	1.0678	1.644	0.076	−0.061	0.8028	0.9855	0.969	−0.086	−0.054
0.2976	1.0570	1.507	−0.116	−0.079	0.9311	0.9599	0.860	−0.038	−0.020
0.3961	1.0460	1.388	−0.149	−0.088	1.0000	0.9445	0.803	0.000	0.000
0.5284	1.0294	1.239	−0.176	−0.090					
DMF (1) + Methyl Benzoate (2) 303.15 K									
0.0000	1.0803	1.778	0.000	0.000	0.6218	1.0113	1.078	−0.167	−0.064
0.1213	1.0698	1.622	−0.059	−0.032	0.7468	0.9909	0.963	−0.116	−0.052
0.1914	1.0632	1.536	−0.089	−0.046	0.8028	0.9809	0.915	−0.097	−0.042
0.2976	1.0524	1.413	−0.129	−0.061	0.9311	0.9522	0.810	−0.030	−0.016
0.3961	1.0414	1.306	−0.162	−0.067	1.0000	0.9398	0.756	0.000	0.000
0.5284	1.0248	1.169	−0.190	−0.069					
DMF (1) + Methyl Benzoate (2) 308.15 K									
0.0000	1.0756	1.663	0.000	0.000	0.6218	1.0067	1.022	−0.180	−0.048
0.1213	1.0651	1.524	−0.067	−0.023	0.7468	0.9863	0.913	−0.128	−0.038
0.1914	1.0586	1.447	−0.102	−0.033	0.8028	0.9762	0.867	−0.100	−0.031
0.2976	1.0478	1.334	−0.143	−0.045	0.9311	0.9506	0.764	−0.040	−0.012
0.3961	1.0368	1.235	−0.176	−0.050	1.0000	0.9351	0.751	0.000	0.000
0.5284	1.0202	1.107	−0.203	−0.052					
DMF (1) + Methyl Benzoate (2) 313.15 K									
0.0000	1.0708	1.402	0.000	0.000	0.6218	1.0019	0.936	−0.189	−0.013
0.1213	1.0603	1.305	−0.063	−0.009	0.7468	0.9815	0.848	−0.138	−0.010
0.1914	1.0538	1.252	−0.105	−0.010	0.8028	0.9714	0.809	−0.110	−0.008
0.2976	1.0431	1.173	−0.159	−0.012	0.9311	0.9457	0.721	−0.040	−0.002
0.3961	1.0321	1.100	−0.194	−0.013	1.0000	0.9302	0.673	0.000	0.000
0.5284	1.0154	1.003	−0.212	−0.014					

negligible. The dynamic viscosity (η) of the liquids was calculated[11] using

$$\frac{\eta}{\eta_0} = \frac{\rho t}{\rho_0 t_0} \tag{1}$$

where ρ, ρ_0; t, t_0; and η, η_0 refer to the density, flow time, and viscosity of liquids and water, respectively. The uncertainty in the measurement of viscosity was ±0.003 mPa·s. All of the measurements were made at atmospheric pressure and at (298.15, 303.15, 308.15, and 313.15) K.

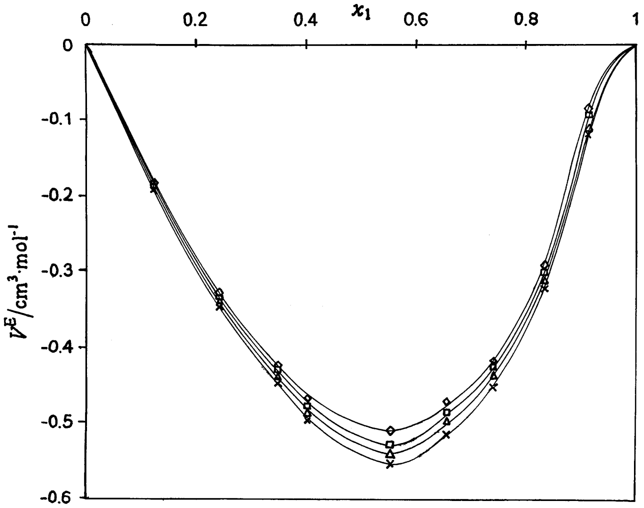

Figure 1. Excess molar volumes (eq 4), V^E, for (x_1) DMF + (1 − x_1) toluene at various temperatures: ◇, 298.15; □, 303.15; △, 308.15; and ×, 313.15 K.

Results and Discussion

Table 2 lists experimental values of densities (ρ) and viscosities (η) of binary mixtures of DMF with toluene and methyl benzoate at (298.15, 303.15, 308.15, and 313.15) K. The ρ values have been used to calculate the excess molar volumes (V^E) using the following equation:

$$V^E/\text{cm}^3\cdot\text{mol}^{-1} = \frac{x_1M_1 + x_2M_2}{\rho_{\text{mix}}} - \frac{x_1M_1}{\rho_1} - \frac{x_2M_2}{\rho_2} \quad (2)$$

where x_1, x_2; M_1, M_2; and ρ_1, ρ_2 are mole fractions, molecular weights, and densities of pure components 1 and 2, respectively, and ρ_{mix} is the density of the binary mixture.

The viscosity deviations ($\Delta\eta$) were calculated by using the equation

$$\Delta\eta/\text{mPa}\cdot\text{s} = \eta_{\text{mix}} - x_1\eta_1 - x_2\eta_2 \quad (3)$$

where η_{mix} is the viscosity of the binary mixture and η_1, η_2 are viscosities of components 1 and 2, respectively. The variation of V^E with the mole fraction of DMF (x_1) for DMF + toluene systems at (298.15, 303.15, 308.15, and 313.15) K is represented in Figure 1. Ramadevi et al.[9] reported the excess molar volumes for DMF + toluene binary mixtures at 303.15 K and 313.15 K. At equimolar concentrations and at 303.15 K, the V^E values obtained in the present investigation and those reported by Ramadevi et al.[9] are −0.529 and −0.384 cm.^{3}·mol^{-1} respectively, because $\Delta\eta$ values for DMF + toluene systems are very small and hence are not represented graphically. Similar $\Delta\eta$ results for DMF + toluene systems were reported by Ramadevi et al.[9] The variations of V^E and $\Delta\eta$ with the mole fraction of DMF (x_1) for DMF + methyl benzoate systems at (298.15, 303.15, 308.15, and 313.15) K are represented in Figures 2 and 3, respectively. The V^E values are negative over the entire range of composition and at all temperatures for binary mixtures of DMF with toluene. For binary mixtures of DMF with methyl benzoate, the V^E and $\Delta\eta$ values are negative over the entire range of composition and at all temperatures.

Treszczanowicz et al.[13] suggested that 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, contribute a positive term to V^E. The chemical or specific intermolecular interactions result in a volume decrease. This effect contributes negative values to V^E. The structural contributions are mostly negative and arise from several effects, especially from interstitial accommodation and changes in the free volume. In other words, structural contributions arising from the geometrical fitting (interstitially accommodated) of one component into other due to the differences in the free volume and molar volume between components lead to negative contribution to V^E.

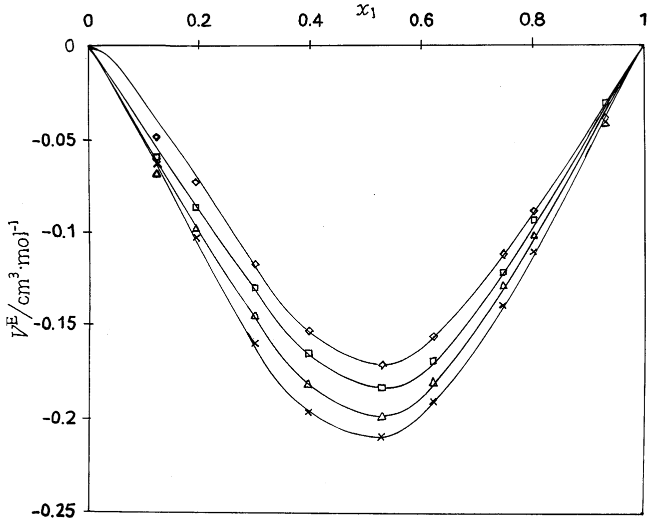

Figure 2. Excess molar volumes (eq 4), V^E, for (x_1) DMF + (1 − x_1) methyl benzoate at various temperatures: ◇, 298.15; □, 303.15; △, 308.15; and ×, 313.15 K.

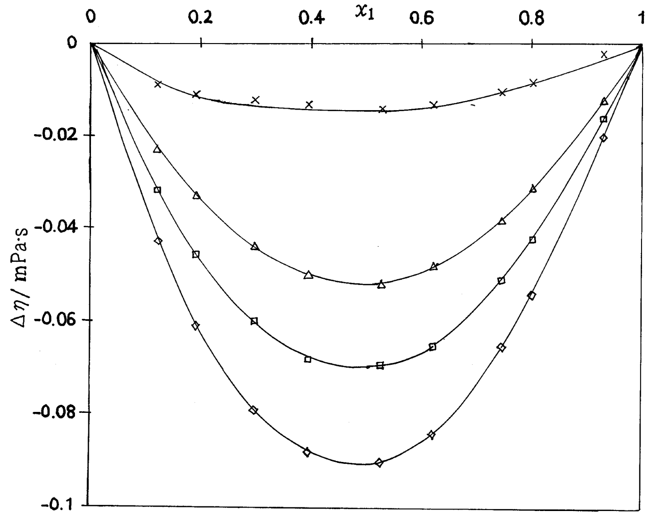

Figure 3. Deviations in viscosity (eq 4), $\Delta\eta$, for (x_1) DMF + (1 − x_1) methyl benzoate at various temperatures: ◇, 298.15; □, 303.15; △, 308.15; and ×, 313.15 K.

The negative excess molar volumes for DMF + toluene systems indicate a π-electron donor−acceptor-type specific interaction between toluene and DMF molecules. The V^E values become more negative at higher temperature, suggesting an increase in interaction between DMF and toluene molecules. The apparent intermolecular interactions in binary mixtures of *N*,*N*-dimethylformamide with methyl benzoate may be classified as weak dipolar induced-type forces resulting from the polarization of methyl benzoate molecules by the dipoles of the surrounding *N*,*N*-dimethylformamide molecules. This effect can be superimposed upon dipole−dipole interactions and leads to a

Table 3. Parameters and Standard Deviations (σ) of Equations 4 and 5 for DMF (1) + Toluene (2) and DMF (1) + Methyl Benzoate (2)

property	T/K	a_0	a_1	a_2	a_3	a_4	a_5	a_6	σ
				DMF (1) + Toluene (2)					
V^E/cm^3·mol^{-1}	298.15	−2.045	−0.294	1.246	−1.069	−6.225	2.773	8.866	0.003
	303.15	−2.117	−0.381	1.669	−0.637	−7.767	2.136	10.319	0.002
	308.15	−2.156	−0.412	1.529	−0.539	−7.036	1.870	9.226	0.002
	313.15	−2.201	−0.493	1.366	−0.293	−6.437	1.572	8.616	0.004
				DMF (1) + Methyl Benzoate (2)					
V^E/cm^3·mol^{-1}	298.15	−0.685	−0.068	0.707	0.002	−0.674			0.003
	303.15	−0.732	−0.100	0.566	0.224	−0.393			0.004
	308.15	−0.793	−0.044	0.663	−0.113	−0.643			0.003
	313.15	−0.840	0.016	0.501	−0.085	−0.207			0.002
				DMF (1) + Methyl Benzoate (2)					
$\Delta\eta$/mPa·s	298.15	−0.363	0.026	−0.003	0.048				0.000
	303.15	−0.279	0.015	−0.001	0.026				0.001
	308.15	−0.209	0.012	0.009	0.006				0.000
	313.15	−0.054	−0.009	−0.013	0.063				0.000

slight increase in the attraction, giving less-negative V^E values. From Figures 2 and 3, it is clear that the V^E values become more negative and $\Delta\eta$ values become less negative at higher temperatures for *N*,*N*-dimethylformamide + methyl benzoate systems. These observations suggest that the specific interactions arising from induced dipole−dipole-type forces tend to increase with increasing mixture temperature.

The results of V^E and $\Delta\eta$ are fit to the Redlich−Kister equation[14]

$$Y = x_1x_2\sum a_i(x_1 - x_2)^i \tag{4}$$

where Y refers to V^E/cm^3·mol^{-1} or $\Delta\eta$/mPa·s and x_1 and x_2 are the mole fractions of components 1 and 2, respectively. Coefficients a_i were obtained by fitting eq 4 to experimental results using a least-squares regression method. In each case, the optimum number of coefficients is ascertained from an examination of the variation in standard deviation (σ).

σ was calculated using

$$\sigma(Y) = \left(\frac{\sum(Y_{\text{exptl}} - Y_{\text{calcd}})^2}{n - m}\right)^{1/2} \tag{5}$$

where n is the number of data points and m is the number of coefficients. The calculated values of a_i along with standard deviations (σ) are given in Table 3.

Kinematic viscosities (ν) of the binary liquid mixtures are obtained indirectly. Kinematic viscosity values of the binary liquid mixtures are calculated from their dynamic viscosities and densities. McAllister's three-body interaction model[15] has been used to correlate the kinematic viscosities of binary liquid mixtures.

$$\ln \nu = {x_1}^3 \ln \nu_1 + {x_2}^3 \ln \nu_2 + 3{x_1}^2x_2 \ln \nu_{12} + 3{x_2}^2x_1 \ln \nu_{21} - \ln\left(x_1 + \frac{x_2M_2}{M_1}\right) + 3{x_1}^2x_2 \ln\left(\frac{2}{3} + \frac{M_2}{3M_1}\right) + 3{x_2}^2x_1 \ln\left(\frac{1}{3} + \frac{2M_2}{3M_1}\right) + {x_2}^3 \ln\left(\frac{M_2}{M_1}\right) \tag{6}$$

where ν_{12} and ν_{21} are model parameters.

The percentage standard deviation was calculated by using the following equation.

$$\sigma\% = \left(\frac{\sum(100\ (\nu_{\text{exptl}} - \nu_{\text{calcd}})/\nu_{\text{exptl}})^2}{n - m}\right)^{1/2} \tag{7}$$

Table 4. Parameters of McAllister's Model and Standard Percentage Deviation for Kinematic Viscosities at Various Temperatures

system	T/K	ν_{12}	ν_{21}	σ%
DMF (1) + toluene (2)	298.15	0.810	0.711	0.33
	303.15	0.765	0.672	0.33
	308.15	0.720	0.635	0.35
	313.15	0.685	0.601	0.42
DMF (1) + methyl benzoate (2)	298.15	1.124	1.408	0.10
	303.15	1.070	1.332	0.05
	308.15	1.028	1.268	0.12
	313.15	0.963	1.119	0.11

where n represents the number of experimental points and m represents the number of coefficients. Table 4 includes the different parameters and percentage standard deviations. From Table 4, it is clear that McAllister's three-body interaction model is suitable for correlating the kinematic viscosities of the binary mixtures studied.

Therefore, it can be concluded that the negative excess molar volumes for DMF + toluene systems are attributed to a π-electron donor−acceptor-type specific interaction. The less-negative values of V^E and $\Delta\eta$ for binary mixtures of *N*,*N*-dimethylformamide with methyl benzoate are due to the weak specific interactions arising from induced dipole−dipole-type forces. For both systems, the specific interactions increase with increasing temperature.

Note Added after ASAP Posting. This article was released ASAP 1/21/2005. Changes were made to Table 2, equation 3, and reference 3. The paper was reposted 2/9/2005.

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