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Short Articles

Densities and Viscosities of *N*,*N*-Dimethylformamide + *N*-Methyl-2-pyrrolidinone and + Dimethyl Sulfoxide in the Temperature Range (303.15 to 353.15) K

Changsheng Yang,* Guanghu He, Yifu He, and Peisheng Ma

Key Laboratory for Green Chemical Technology of State Education Ministry, School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, PRC

Densities and viscosities of the binary mixtures of *N*,*N*-dimethylformamide + *N*-methyl-2-pyrrolidinone and *N*,*N*-dimethylformamide + dimethyl sulfoxide have been measured at different temperatures and atmospheric pressure, over the entire composition range. From these data, excess molar volumes (V^E) were calculated for each of the systems. The densities and viscosities of pure components have been compared with those reported by other authors available in the literature. The ρ and η were fitted to a Redlich−Kister equation, and the coefficients of the Redlich−Kister equation and the estimate of standard deviation are also presented.

Introduction

The physical properties and the thermodynamic behavior of binary mixtures have been studied for many reasons, one of the most important of which is that these properties may provide information about molecular interactions.

This paper is part of our systematic program of research on the measurement of physical and transport properties of binary liquid mixtures containing *N*,*N*-dimethylformamide (DMF), which is an aprotic polar solvent with a large dipole moment and a relatively high dielectric constant. In the pure state, it shows self-association through dipole−dipole interactions.[1] DMF is biologically important and also a very good polar solvent which is widely used in polymer science and pharmaceutical industry.

A detailed search in the literature shows that systematic investigations have been carried out on the excess thermodynamic properties of binary mixtures of DMF, + toluene, + methyl benzoate, + nitrobenzene, + water, + glycerol, + alkanenitriles, + ketones, + ethylbenzene, + chlorobenzene, and so on.[2–7]

For a binary mixture of *N*,*N*-dimethylformamide + dimethyl sulfoxide, Kinart et al.[8] measured densities at 298.15 K. Chauhan et al.[9] measured the viscosity and density at 298.15 K, 308.15 K, and 318.15 K over the entire solvent composition range. Nakamura et al.[10] also determined the density for this system at 298.15 K.

However, there are no reports in the literature on the densities or viscosities of binary mixtures of *N*,*N*-dimethylformamide + *N*-methyl-2-pyrrolidinone in the temperature range (303.15 to 353.15) K. In this work, the densities and viscosities of DMF + *N*-methyl-2-pyrrolidinone (NMP) and dimethyl sulfoxide (DMSO) mixtures were measured at (303.15 to 353.15) K and atmospheric pressure. The results were used to calculate excess molar volumes over the entire mole fraction range. Experimental values were fitted by the Redlich−Kister equation. The standard deviations between the experimental data and values calculated from the Redilch−Kister equation are also presented.

Table 1. Comparison of Experimental and Literature Values of Densities, ρ, and Viscosities, η, for Pure Components

		ρ/g·cm^{-3}		η/mPa·s	
liquid	T/K	exptl	lit.	exptl	lit.
DMF	303.15	0.94009	0.9398[3]	0.759	0.756[3]
			0.9397[4]		0.752[15]
			0.9414[5]		0.765[5]
	313.15	0.93074	0.9302[6]	0.678	0.673[6]
			0.9298[7]		0.664[7]
NMP	303.15	1.02376	1.02347[11]	1.521	1.554[11]
	313.15	1.01508	1.01519[12]	1.319	1.322[12]
			1.0157[13]		1.332[13]
	323.15	1.00630	1.00627[14]	1.174	1.175[14]
			1.00566[11]		1.160[11]
	333.15	0.99689	0.99671[11]	1.033	1.035[11]
			0.9974[13]		1.022[13]
	343.15	0.98805	0.98846[11]	0.914	0.908[11]
			0.98775[12]		0.922[12]
DMSO	303.15	1.09037	1.09027[14]	1.786	1.790[21]
			1.09041[15]		
			1.09042[16]		
	313.15	1.08058	1.08043[17]	1.506	1.516[19]
			1.0806[18]		
	323.15	1.07075	1.0703[19]	1.278	1.294[19]
			1.0698[20]		

Experimental Section

Materials. The chemicals employed were of analytical grade and were purchased from Tianjin Reagent Company. The mass fraction purities, determined by gas chromatography, were DMF > 0.998, DMP > 0.995, and DMSO > 0.995. All chemicals were used without further purification. They were degassed ultrasonically and dried over 4 Å molecular sieves.

Apparatus and Procedure. The densities of the pure components and their mixtures were measured with a high precision vibrating-tube digital density meter. The uncertainty in density measurements was $\pm\, 5 \cdot 10^{-5}$ g·cm^{-3}. Density measurements were reproducible to $\pm\, 3 \cdot 10^{-5}$ g·cm^{-3}.

The liquid mixtures were prepared by mass using a BP210s balance accurate to within ± 0.1 mg. The average uncertainty

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* To whom correspondence should be addressed. E-mail: yangchangsheng@tju.edu.cn.

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Table 2. Densities ρ, Viscosities η, and Excess and Molar Volumes V^E for the Binary Mixtures at Different Temperatures

x_1	ρ $\mathrm{g\cdot cm^{-3}}$	η $\mathrm{mPa\cdot s}$	V^E $\mathrm{cm^3\cdot mol^{-1}}$	x_1	ρ $\mathrm{g\cdot cm^{-3}}$	η $\mathrm{mPa\cdot s}$	V^E $\mathrm{cm^3\cdot mol^{-1}}$	x_1	ρ $\mathrm{g\cdot cm^{-3}}$	η $\mathrm{mPa\cdot s}$	V^E $\mathrm{cm^3\cdot mol^{-1}}$	x_1	ρ $\mathrm{g\cdot cm^{-3}}$	η $\mathrm{mPa\cdot s}$	V^E $\mathrm{cm^3\cdot mol^{-1}}$
(x_1)DMF + $(1 - x_1)$NMP								(x_1)DMF + $(1 - x_1)$DMSO							
$T = 303.15$ K								$T = 303.15$ K							
0.0000	1.02376	1.521	0.000	0.5950	0.97895	1.018	−0.042	0.0000	1.09037	1.786	0.000	0.6001	0.99694	1.025	0.025
0.1003	1.01738	1.417	−0.046	0.7000	0.96961	0.943	−0.035	0.1000	1.07407	1.617	0.009	0.7001	0.98231	0.952	0.025
0.2001	1.01028	1.328	−0.048	0.8000	0.96030	0.872	−0.029	0.2000	1.05813	1.466	0.012	0.8000	0.96798	0.877	0.020
0.2997	1.00295	1.249	−0.054	0.9003	0.95041	0.809	−0.015	0.3000	1.04235	1.331	0.022	0.9000	0.95396	0.814	0.007
0.4000	0.99519	1.170	−0.054	1.0000	0.94009	0.759	0.000	0.4001	1.02675	1.228	0.038	1.0000	0.94009	0.759	0.000
0.5007	0.98701	1.085	−0.052					0.5002	1.01189	1.109	0.036				
$T = 313.15$ K								$T = 313.15$ K							
0.0000	1.01508	1.319	0.000	0.5950	0.96984	0.891	−0.039	0.0000	1.08058	1.506	0.000	0.6001	0.98743	0.900	0.023
0.1003	1.00862	1.231	−0.046	0.7000	0.96043	0.832	−0.033	0.1000	1.06434	1.365	0.007	0.7001	0.97287	0.839	0.020
0.2001	1.00148	1.152	−0.049	0.8000	0.95101	0.775	−0.023	0.2000	1.04844	1.248	0.010	0.8000	0.95856	0.775	0.017
0.2997	0.99403	1.085	−0.050	0.9003	0.94111	0.721	−0.019	0.3000	1.03275	1.141	0.017	0.9000	0.94459	0.724	0.005
0.4000	0.98624	1.020	−0.055	1.0000	0.93074	0.678	0.000	0.4001	1.01714	1.057	0.037	1.0000	0.93074	0.678	0.000
0.5007	0.97802	0.948	−0.053					0.5002	1.00234	0.969	0.033				
$T = 323.15$ K								$T = 323.15$ K							
0.0000	1.00630	1.174	0.000	0.5950	0.96068	0.794	−0.043	0.0000	1.07075	1.278	0.000	0.6001	0.97785	0.792	0.017
0.1003	0.99980	1.097	−0.049	0.7000	0.95117	0.742	−0.034	0.1000	1.05458	1.170	0.004	0.7001	0.96336	0.740	0.012
0.2001	0.99254	1.024	−0.048	0.8000	0.94168	0.692	−0.025	0.2000	1.03870	1.075	0.007	0.8000	0.94903	0.690	0.014
0.2997	0.98506	0.967	−0.053	0.9003	0.93172	0.646	−0.017	0.3000	1.02304	0.990	0.014	0.9000	0.93509	0.647	0.002
0.4000	0.97717	0.908	−0.054	1.0000	0.92125	0.608	0.000	0.4001	1.00750	0.922	0.031	1.0000	0.92125	0.608	0.000
0.5007	0.96891	0.846	−0.056					0.5002	0.99271	0.850	0.028				
$T = 333.15$ K								$T = 333.15$ K							
0.0000	0.99689	1.033	0.000	0.5950	0.95090	0.711	−0.047	0.0000	1.06026	1.104	0.000	0.6001	0.96764	0.706	0.012
0.1003	0.99033	0.970	−0.051	0.7000	0.94131	0.664	−0.037	0.1000	1.04411	1.015	0.004	0.7001	0.95316	0.661	0.010
0.2001	0.98305	0.910	−0.054	0.8000	0.93174	0.623	−0.026	0.2000	1.02831	0.938	0.004	0.8000	0.93890	0.621	0.010
0.2997	0.97546	0.857	−0.055	0.9003	0.92172	0.583	−0.018	0.3000	1.01268	0.868	0.011	0.9000	0.92498	0.583	0.000
0.4000	0.96751	0.811	−0.057	1.0000	0.91117	0.552	0.000	0.4001	0.99714	0.818	0.032	1.0000	0.91117	0.552	0.000
0.5007	0.95917	0.754	−0.058					0.5002	0.98246	0.753	0.023				
$T = 343.15$ K								$T = 343.15$ K							
0.0000	0.98805	0.914	0.000	0.5950	0.94159	0.639	−0.047	0.0000	1.05030	0.962	0.000	0.6001	0.95792	0.635	0.006
0.1003	0.98141	0.860	−0.051	0.7000	0.93192	0.601	−0.037	0.1000	1.03424	0.895	−0.001	0.7001	0.94347	0.597	0.004
0.2001	0.97402	0.811	−0.051	0.8000	0.92225	0.565	−0.025	0.2000	1.01848	0.829	−0.003	0.8000	0.92922	0.561	0.006
0.2997	0.96638	0.767	−0.055	0.9003	0.91214	0.531	−0.016	0.3000	1.00288	0.771	0.004	0.9000	0.91532	0.530	−0.001
0.4000	0.95837	0.722	−0.059	1.0000	0.90154	0.501	0.000	0.4001	0.98738	0.726	0.024	1.0000	0.90154	0.501	0.000
0.5007	0.94993	0.679	−0.058					0.5002	0.97271	0.674	0.017				
$T = 353.15$ K								$T = 353.15$ K							
0.0000	0.97884	0.808	0.000	0.5950	0.93194	0.581	−0.051	0.0000	1.04003	0.850	0.000	0.6001	0.94783	0.575	0.001
0.1003	0.97212	0.769	−0.052	0.7000	0.92219	0.547	−0.041	0.1000	1.02403	0.793	−0.005	0.7001	0.93343	0.541	−0.002
0.2001	0.96469	0.729	−0.056	0.8000	0.91242	0.514	−0.027	0.2000	1.00831	0.739	−0.008	0.8000	0.91920	0.512	0.001
0.2997	0.95698	0.691	−0.060	0.9003	0.90224	0.483	−0.018	0.3000	0.99273	0.691	−0.002	0.9000	0.90531	0.483	−0.004
0.4000	0.94889	0.653	−0.064	1.0000	0.89154	0.459	0.000	0.4001	0.97726	0.654	0.018	1.0000	0.89154	0.459	0.000
0.5007	0.94036	0.614	−0.062					0.5002	0.96263	0.611	0.009				

in the mole fraction of the mixtures was estimated to be less than ± 0.0001. The molar excess volumes were calculated from composition−density data with an uncertainty better than ±

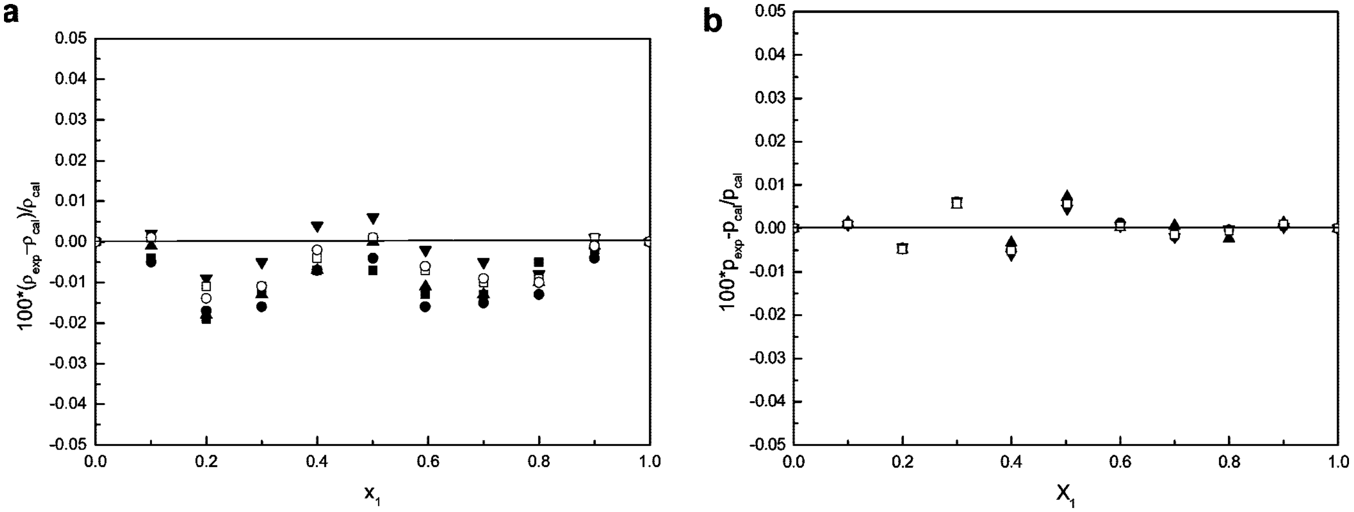

Figure 1. Relative deviation of the densities $(\rho_{\mathrm{exptl}} - \rho_{\mathrm{calcd}})/\rho_{\mathrm{exptl}}$ between the experimental densities ρ_{exptl} and the calculation values ρ_{calcd} obtained with the correlation of eq 2 for the two systems (a) DMF (1) + NMP (2) and (b) DMF (1) + DMSO (2) at different temperatures. ■, 303.15 K; ●, 313.15 K; ▲, 323.15 K; ▼, 333.15 K; □, 343.15 K; ○, 353.15 K.

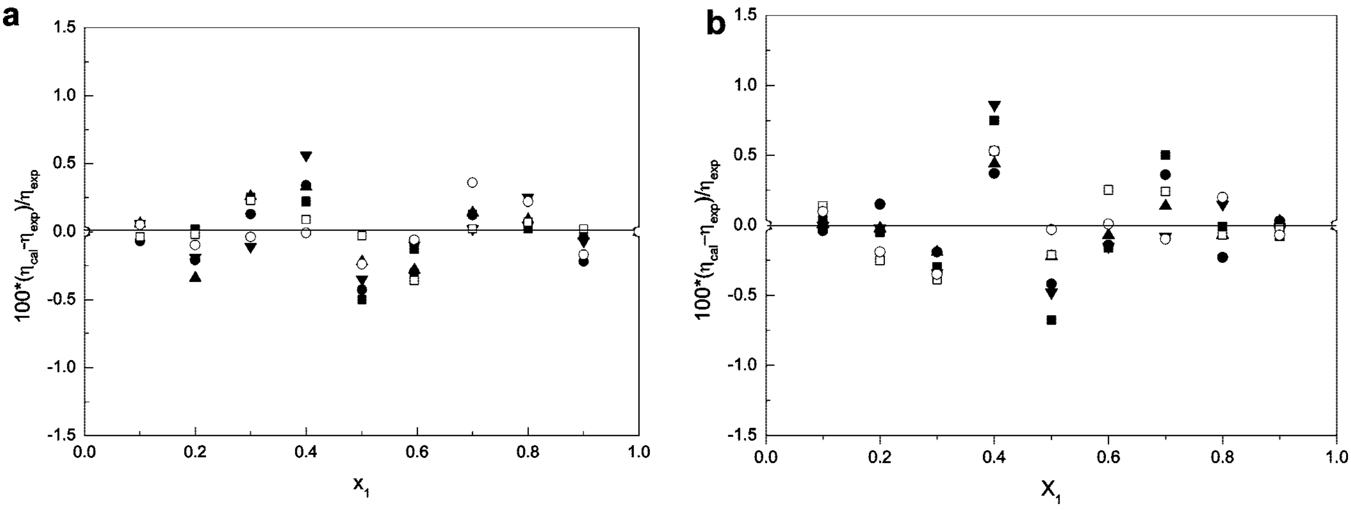

Figure 2. Relative deviation of viscosity deviations $(\eta_{\text{exptl}} - \eta_{\text{calcd}})/\eta_{\text{exptl}}$ between the experimental values of viscosity η_{exptl} and the calculation values η_{calcd} obtained by the correlation of eq 3 for the two systems (a) DMF (1) + NMP (2) and (b) DMF (1) + DMSO (2) at different temperatures *T*. ■, 303.15 K; ●, 313.15 K; ▲, 323.15 K; ▼, 333.15 K; □, 343.15 K; ○, 353.15 K.

Table 3. Coefficients of the Redlich−Kister Equation and Standard Deviation for Density and Viscosity of Mixtures

T/K	property	A_0	A_1	A_2	A_3	σ
		(x_1)DMF + $(1 - x_1)$NMP				
303.15	ρ/g·cm^{-3}	−0.185	0.043	−0.219	0.252	0.0299
	η/mPa·s	−0.196	0.006	−0.153		0.0125
313.15	ρ/g·cm^{-3}	−0.174	0.095	−0.251	0.129	0.0369
	η/mPa·s	−0.184	0.024	−0.097		0.0124
323.15	ρ/g·cm^{-3}	−0.184	0.053	−0.241	0.257	0.0404
	η/mPa·s	−0.171	0.018	−0.073		0.0130
333.15	ρ/g·cm^{-3}	−0.194	0.067	−0.257	0.241	0.0349
	η/mPa·s	−0.142	−0.009	−0.055		0.0114
343.15	ρ/g·cm^{-3}	−0.198	0.053	−0.230	0.289	0.0390
	η/mPa·s	−0.112	0.006	−0.029		0.0054
353.15	ρ/g·cm^{-3}	−0.218	0.079	−0.227	0.234	0.0369
	η/mPa·s	−0.071	−0.039	−0.014		0.0070
		(x_1)DMF + $(1 - x_1)$DMSO				
303.15	ρ/g·cm^{-3}	0.132	0.006	−0.077		0.0230
	η/mPa·s	−0.623	0.138	−0.016		0.0238
313.15	ρ/g·cm^{-3}	0.120	0.002	−0.093		0.0261
	η/mPa·s	−0.475	0.143	−0.079		0.0140
323.15	ρ/g·cm^{-3}	0.098	−0.001	−0.109		0.0258
	η/mPa·s	−0.364	0.090	−0.033		0.0088
333.15	ρ/g·cm^{-3}	0.084	−0.015	−0.104		0.0305
	η/mPa·s	−0.285	0.068	−0.057		0.0149
343.15	ρ/g·cm^{-3}	0.056	0.008	−0.115		0.0307
	η/mPa·s	−0.224	0.036	0.011		0.0125
353.15	ρ/g·cm^{-3}	0.031	0.012	−0.135		0.0292
	η/mPa·s	−0.173	0.027	−0.020		0.0095

0.002 cm^3·mol^{-1}. All relative atomic masses were based on the IUPAC relative atomic mass table.

The viscosities of pure liquids and the mixtures were measured at atmospheric pressure and at different temperatures using an Ubbelohde suspended level viscometer. The uncertainty of viscosity results was within ± 0.003 mPa·s.

The details of the methods and techniques used to determine densities and viscosities have been described previously.[8] Densities and viscosities of pure compounds are reported in Table 1, together with the corresponding literature data.

Results and Discussion

Excess molar volumes were calculated from our measurements according to the following equations[2]

$$V^{\mathrm{E}} = \frac{x_1M_1 + x_2M_2}{\rho} - \frac{x_1M_1}{\rho_1} - \frac{x_2M_2}{\rho_2} \tag{1}$$

where x_1 and x_2 are mole fractions; M_1 and M_2 are the molar masses; and ρ_1 and ρ_2 are the densities of pure components 1 and 2, respectively. Quantities without subscripts refer to the mixture.

The densities are correlated by the following equation

$$\rho = \frac{M}{V_1 + V_2 + V^{\mathrm{E}}} = \frac{x_1M_1 + x_2M_2}{V_1 + V_2 + x_1(1 - x_1)\sum_{i=0}^{n} A_i(2x_1 - 1)^i} \tag{2}$$

The viscosities are correlated with the eq[8]

$$\eta = x_1\eta_1 + (1 - x_1)\eta_2 + x_1(1 - x_1)\sum_{i=0}^{n} A_i(2x_1 - 1)^i \tag{3}$$

where η is the viscosity of mixtures and η_1 and η_2 are the viscosity of components 1 and 2, respectively. A_i is the adjustable parameter, and x_1 is the mole fraction of component 1.

In each case, the optimum number of coefficients A_i was determined from an examination of the variation of the standard deviation[17]

$$\sigma(Y) = \left[\sum (Y_{\text{calcd}} - Y_{\text{exptl}})^2/(n - m)\right]^{\frac{1}{2}} \tag{4}$$

where n is the total number of experimental values and m is the number of parameters.

The densities, viscosities, and excess molar volume data are presented in Table 2. Table 3 lists the values of the parameters A_i together with the standard deviations.

The relative deviations of the densities and the viscosity were calculated by the following equation[21]

$$\text{RD }\% = \frac{(y_{\text{exptl}} - y_{\text{calcd}})}{y_{\text{calcd}}} \cdot 100 \tag{5}$$

where $y = \rho$ or η; y_{exptl} is the experimental value; and y_{calcd} are the calculated values obtained with eqs 2 and 3, respectively.

The average absolute deviations were calculated by

$$\text{AAD }\% = \frac{\sum \left|(y_{\text{exptl}} - y_{\text{calcd}}) \cdot 100/y_{\text{exptl}}\right|}{n} = \frac{\sum |\Delta y|}{n} \tag{6}$$

The relative deviation of the densities and the viscosities at different mole fractions and temperatures for the two binary systems of DMF with NMP and DMSO are shown in Figure 1 and Figure 2, respectively. It can be seen from the figures that the relative deviation values of the densities for the two binary systems are within ± 0.05 %, and the relative deviation values of the viscosity deviations for the two binary systems are within ± 0.15 %.

Conclusion

Densities and viscosities for the DMF with NMP and DMSO binary systems have been experimentally determined at (303.15 to 353.15) K and normal atmospheric pressure, over the entire mole fraction range. The densities and viscosities were correlated by the Redlich−Kister equation. The relative deviation values of the densities for the two binary systems are within ± 0.05 %, and the relative deviation values of the viscosity deviations for the two binary systems are within ± 0.15 %.

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