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Excess Molar Volumes and Viscosity Deviations for the Ternary System *N,N*-Dimethylformamide + *N*-Methylformamide + Water and the Binary Subsystems at 298.15 K

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Densities and viscosities at 298.15 K are reported for the binary systems *N,N*-dimethylformamide (DMF) + *N*-methylformamide (NMF), NMF + water, and DMF + water and the ternary system DMF + NMF + water. The excess molar volumes and viscosity deviations of the binary and ternary systems were derived and correlated with the Redlich−Kister and Cibulka equations for the binary systems and the ternary system. The binary data were compared with the values in the literature, and the ternary data were also compared with the predicted values using the binary contribution models of Tsao−Smith, Kohler, Rastogi, and Radojkovič.

Introduction

Studies of the phase equilibrium behavior and the excess properties of liquid mixtures are of great importance for the design of separation processes and theoretical understanding of the nature of molecular interaction.[1] While a large number of phase equilibria and other mixture properties are available for binary mixtures, those for ternary mixtures are spare. The excess properties for ternary mixtures, required to obtain insight into the nature and degree of interactions, are still rare. It is important to test methods of estimating excess properties of ternary mixtures from binary data.[2−4]

The thermodynamic analysis and prediction of phase equilibria and thermophysical properties are useful to solve separation problems. However, when dealing with complex mixtures and new processes, experimental data are still needed for a reliable process design.[5]

In the present work, excess molar volumes (V^E) at 298.15 K for the binary and ternary systems of *N,N*-dimethylformamide (DMF) + *N*-methylformamide (NMF) + water were determined from the densities measured using a digital tube densimeter. DMF and NMF are well-known materials that are used as selective solvents in extraction or extractive distillation processes. Additionally, the viscosities at the same temperature for the same mixtures were measured using an Ubbelhode viscometer. Viscosity deviations ($\Delta\eta$) were determined from the experimental kinematic viscosities. The experimental excess molar volumes and viscosity deviations of the binary and ternary systems were correlated using the Redlich−Kister polynomial[6] and the Cibulka equation.[7]

For the ternary system, the measured excess molar volumes and viscosity deviations were additionally compared with the predicted values using several estimation methods with binary Redlich−Kister parameters.

Table 1. Purities, Densities, and Viscosities of the Pure Components

		ρ/g·cm^{-3} at 298.15 K		η/mPa·s at 298.15 K	
chemicals	GC analysis	present study	literature[8] value	present study	literature[8] value
DMF	99.9	0.9442	0.94406	0.799	0.805
NMF	99.8	0.9988	0.99929	1.681	1.760
water	no impurity	0.9970	0.99705	0.895	0.890

Experimental Section

Materials. DMF and NMF of analytical grade purity supplied from Junsei Chemical Co. and doubly distilled water were used. DMF and NMF were dried using molecular sieves with a pore diameter of 0.4 nm. The purity of the chemicals was checked by gas chromatography and by comparing the density and viscosity with values reported in the literature.[8] The water content of DMF and NMF, determined by Karl Fischer titration (Metrohm 684 KF-coulometer), was less than 50 ppm. The purities, densities, and viscosities of the pure substances are summarized in Table 1.

Apparatus and Procedure. Densities are measured using a digital vibrating glass tube densimeter (Anton Paar, model DMA 48, Graz, Austria). Excess molar volumes (V^E) are calculated from the measured densities of the pure compounds and mixtures. The uncertainty of the density and excess molar volume is less than $\pm 1 \times 10^{-4}$ g·cm^{-3} or $\pm 3 \times 10^{-4}$ cm^3·mol^{-1}. Before each series of measurements, the apparatus was calibrated with doubly distilled water and dried air. Mixture samples were prepared in ca. 20 mL vial by mass using a microbalance (A&D, HA202) with a precision of $\pm 1 \times 10^{-5}$ g, charging the higher boiling component first to minimize vaporization effects. The uncertainty in the mole fraction of the mixture sample is less than $\pm 1 \times 10^{-4}$. The operating procedure has been described elsewhere.[9,10]

Viscosity deviations ($\Delta\eta$) were calculated from the experimental viscosities of the pure and mixture samples. Viscosity (η) of the pure components and mixtures was determined from the kinematic viscosities (ν) measured using an Ubbelhode viscometer with an automatic measur-

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Table 2. Densities, Viscosities, Excess Molar Volumes, and Viscosity Deviations for the Three Binary Systems of DMF, NMF, and Water at 298.15 K

x_1	ρ g·cm^{-3}	η mPa·s	V^E cm^3·mol^{-1}	$\Delta\eta$ mPa·s
DMF (1) + NMF (2)				
0.0497	0.9954	1.618	−0.004	−0.02
0.0997	0.9919	1.524	−0.003	−0.07
0.2002	0.9855	1.373	−0.009	−0.13
0.2999	0.9794	1.274	−0.015	−0.14
0.4001	0.9736	1.171	−0.014	−0.16
0.4999	0.9681	1.085	−0.018	−0.16
0.5999	0.9630	1.006	−0.024	−0.15
0.6990	0.9580	0.947	−0.023	−0.12
0.8001	0.9532	0.897	−0.017	−0.08
0.9001	0.9486	0.855	−0.013	−0.03
0.9500	0.9464	0.840	−0.009	−0.00
NMF (1) + Water (2)				
0.0504	1.0032	1.174	−0.120	0.24
0.1001	1.0089	1.422	−0.250	0.45
0.2000	1.0146	1.793	−0.435	0.74
0.2998	1.0163	2.001	−0.546	0.87
0.4002	1.0151	2.067	−0.575	0.86
0.4996	1.0122	2.040	−0.525	0.75
0.5996	1.0094	1.961	−0.460	0.59
0.6994	1.0061	1.878	−0.348	0.43
0.7999	1.0037	1.794	−0.255	0.27
0.8983	1.0009	1.734	−0.118	0.13
0.9498	0.9997	1.707	−0.053	0.07
DMF (1) + Water (2)				
0.0500	0.9960	1.310	−0.185	0.42
0.1001	0.9968	1.728	−0.406	0.84
0.2000	0.9951	2.346	−0.765	1.47
0.2990	0.9916	2.491	−1.037	1.62
0.3998	0.9844	2.273	−1.124	1.42
0.4997	0.9759	1.878	−1.063	1.03
0.5993	0.9684	1.532	−0.947	0.69
0.6995	0.9607	1.243	−0.726	0.42
0.8000	0.9549	1.045	−0.535	0.23
0.8991	0.9491	0.901	−0.268	0.09
0.9488	0.9469	0.843	−0.161	0.04

ing unit (LAUDA, model PVS1, Germany) and a precision thermostat with an accuracy of 0.01 K. The accuracy of the flowing time measurement is ± 0.01 s. The mole fractions of the mixture samples were the same as the samples used in the density measurements. The samples were transferred to the viscometer cell and kept for more than 30 min to reach the desired temperature in the thermostat. The sample was pumped up through the capillary of the viscometer until the upper part of the measuring cell was filled with the fixed volume of the sample (approximately 12 cm^3). Then the sample flowed down through the capillary, whereby the required flowing time was measured. For every sample, the measurement was repeated five times automatically, and the mean flowing time value was calculated. The uncertainty of ν is less than $\pm 1 \times 10^{-3}$ mm^2·s^{-1} (cSt).

Results and Discussion

Binary Systems. The excess molar volumes for the multicomponent mixtures are calculated from the measured densities of the pure substances and mixtures using eq 1:

$$V^{E}/\text{cm}^3\cdot\text{mol}^{-1} = \frac{\sum_i x_i M_i}{\rho_m} - \sum_i \left(\frac{x_i M_i}{\rho_i}\right) \qquad (1)$$

where x_i, M_i, ρ_i, and ρ_m are the mole fraction, molar mass, pure component density, and mixture density, respectively.

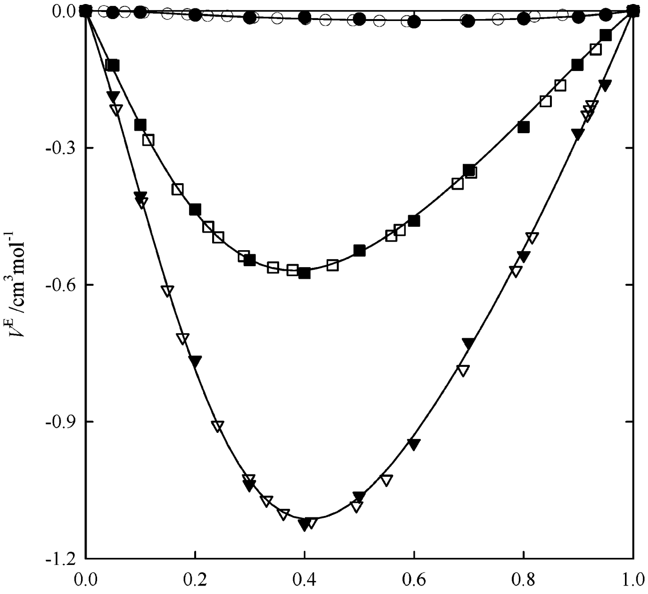

Figure 1. Excess molar volumes (V^E) for the three binary systems of DMF, NMF, and water at 298.15 K: ●, DMF + NMF; ○, DMF + NMF from García et al.;[8] ■, NMF + water; □, NMF + water from García et al.;[11] ▼, DMF + water; □, DMF + water from García et al.[11] Solid curves were calculated from the Redlich−Kister equation.

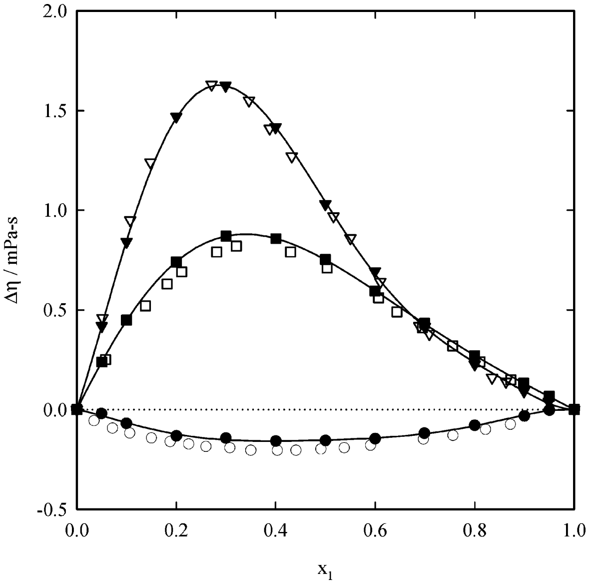

Figure 2. Viscosity deviations ($\Delta\eta$) for the three binary systems of DMF, NMF, and water at 298.15 K: ●, DMF + NMF; ○, DMF + NMF from García et al.;[8] ■, NMF + water; □, NMF + water from García et al.;[11] ▼, DMF + water; □, DMF + water from García et al.[11] Solid curves were calculated from the Redlich−Kister equation.

The experimental excess molar volumes at 298.15 K for the binary systems of DMF (1) + NMF (2), NMF (1) + water (2), and DMF (1) + water (2) are listed in Table 2 and shown in Figure 1. The system DMF (1) + NMF (2) shows very small deviations from ideal behavior, probably because the molecular structures and characteristics of the two components are very similar. In contrast, the systems NMF (1) + water (2) and DMF (1) + water (2) show relatively large deviations from ideal mixing caused by the strong interactions between NMF and water or DMF and water. This means that in this case the breaking of

Table 3. Densities, Viscosities, Excess Molar Volumes, and Viscosity Deviations for the Ternary System of DMF (1) + NMF (2) + Water (3) at 298.15 K

x_1	x_2	ρ/$g\cdot cm^{-3}$	η/mPa·s	V^E/$cm^3\cdot mol^{-1}$	$\Delta\eta$/mPa·s
0.0600	0.5400	1.0035	1.995	−0.471	0.68
0.0500	0.4503	1.0078	2.129	−0.579	0.89
0.0402	0.3599	1.0111	2.190	−0.616	1.02
0.0299	0.2696	1.0119	2.151	−0.549	1.05
0.0201	0.1801	1.0112	1.946	−0.436	0.91
0.1795	0.4214	0.9940	1.906	−0.553	0.70
0.1498	0.3499	0.9994	2.116	−0.676	0.96
0.1198	0.2800	1.0028	2.238	−0.670	1.13
0.0901	0.2097	1.0058	2.263	−0.625	1.21
0.0300	0.0701	1.0032	1.591	−0.255	0.64
0.2996	0.2992	0.9859	1.843	−0.668	0.74
0.2499	0.2500	0.9921	2.075	−0.790	1.01
0.1997	0.1995	0.9975	2.293	−0.817	1.26
0.1499	0.1497	1.0006	2.355	−0.715	1.36
0.1000	0.1000	1.0023	2.163	−0.545	1.20
0.3410	0.1461	0.9855	2.085	−0.882	1.11
0.2798	0.1200	0.9919	2.333	−0.938	1.37
0.2100	0.0901	0.9963	2.468	−0.827	1.52
0.0699	0.0301	0.9985	1.727	−0.318	0.81
0.3601	0.0401	0.9860	2.367	−1.028	1.48
0.2682	0.0300	0.9925	2.587	−0.941	1.69
0.1797	0.0201	0.9963	2.430	−0.713	1.54
0.0400	0.5960	1.0042	1.940	−0.422	0.58
0.0300	0.6978	1.0029	1.858	−0.334	0.42
0.0201	0.7990	1.0011	1.783	−0.209	0.26
0.1500	0.5005	0.9954	1.815	−0.486	0.54
0.1201	0.5997	0.9960	1.759	−0.379	0.40
0.0901	0.6993	0.9964	1.726	−0.263	0.29
0.0304	0.9006	0.9980	1.705	−0.083	0.11
0.2996	0.3995	0.9840	1.615	−0.510	0.43
0.2500	0.4992	0.9860	1.583	−0.386	0.32
0.1996	0.5988	0.9884	1.572	−0.287	0.23
0.1501	0.7014	0.9910	1.579	−0.207	0.15
0.0998	0.8010	0.9936	1.613	−0.130	0.10
0.4192	0.4018	0.9743	1.344	−0.332	0.17
0.3492	0.4997	0.9781	1.375	−0.256	0.12
0.2796	0.5990	0.9821	1.413	−0.199	0.07
0.2102	0.6996	0.9860	1.467	−0.137	0.04
0.0699	0.8996	0.9944	1.624	−0.039	0.03
0.5343	0.3995	0.9667	1.126	−0.144	−0.03
0.4497	0.4994	0.9712	1.184	−0.103	−0.06
0.3594	0.6001	0.9762	1.255	−0.075	−0.08
0.2698	0.7000	0.9815	1.346	−0.057	−0.07
0.1798	0.7998	0.9869	1.458	−0.031	−0.05
0.3919	0.0588	0.9831	2.177	−1.006	1.27
0.5000	0.0501	0.9746	1.834	−0.947	0.95
0.5934	0.0503	0.9674	1.523	−0.807	0.65
0.6991	0.0299	0.9607	1.264	−0.674	0.41
0.7993	0.0195	0.9545	1.078	−0.466	0.24
0.4000	0.1802	0.9800	1.804	−0.794	0.81
0.5002	0.1502	0.9724	1.563	−0.723	0.60
0.5997	0.1204	0.9657	1.345	−0.623	0.41
0.7001	0.0900	0.9597	1.164	−0.509	0.27
0.9006	0.0301	0.9490	0.908	−0.198	0.08
0.3996	0.2999	0.9778	1.547	−0.576	0.45
0.5005	0.2502	0.9708	1.373	−0.514	0.33
0.5995	0.2002	0.9647	1.217	−0.449	0.22
0.6994	0.1501	0.9591	1.087	−0.364	0.14
0.7999	0.1000	0.9539	0.975	−0.271	0.08
0.5001	0.3501	0.9694	1.215	−0.302	0.09
0.5893	0.2748	0.9647	1.146	−0.311	0.09
0.7008	0.2103	0.9586	1.021	−0.229	0.03
0.8994	0.0700	0.9488	0.881	−0.094	0.02
0.6000	0.3600	0.9631	1.043	−0.098	−0.08
0.7005	0.2701	0.9581	0.973	−0.084	−0.07
0.7991	0.1798	0.9533	0.917	−0.067	-0.04

hydrogen bondings, and specific dipole−dipole interactions of the nitrogen compounds are less important than the interactions between different molecules. The measured data were compared with the values in the literature[8,11] and showed good agreements. The literature values are represented in Figure 1 together with the experimental data. The binary excess volumes were correlated with a five-parameter Redlich−Kister polynomial equation:

$$V^{E}_{12}/\mathrm{cm^3\cdot mol^{-1}} = x_1x_2\sum_{i=1}^{5}A_i(x_1 - x_2)^{i-1} \qquad (2)$$

where A_i values are the adjustable parameters.

The standard deviation of fit (σ_{st}) is defined as

$$\sigma_{st}/\mathrm{cm^3\cdot mol^{-1}} = \left[\frac{\sum_i (V^{E}_{cal} - V^{E}_{exp})_i^2}{(N - n)}\right]^{1/2} \qquad (3)$$

where N is the number of experimental data and n is the number of the fitted parameters.

In Figure 1, the continuous lines represent the correlated values according to the Redlich−Kister polynomial. As can be seen, the calculated values agree very well with the experimental data.

The viscosity (η) was determined from the experimental density (ρ) and the kinematic viscosity (ν) (see eq 4). The viscosity deviations ($\Delta\eta$) for the binary and multicomponent systems were calculated from the viscosity of the pure substances (η_i) and mixtures (η_m) following eq 5. The experimental $\Delta\eta$ values for the binary systems were correlated with a four-parameter Redlich−Kister polynomial equation:

$$\eta/\mathrm{mPa\cdot s} = \rho\nu \qquad (4)$$

$$\Delta\eta/\mathrm{mPa\cdot s} = \eta_m - \sum_{i=1}^{n}x_i\eta_i \qquad (5)$$

$$\Delta\eta_{12}/\mathrm{mPa\cdot s} = x_1x_2\sum_{i=1}^{4}A_i(x_1 - x_2)^{i-1} \qquad (6)$$

The experimental viscosity deviations for the binary systems of DMF (1) + NMF (2), NMF (1) + water (2), and DMF (1) + water (2) at 298.15 K are listed in Table 2 and shown in Figure 2. The system DMF (1) + NMF (2) shows small negative values. In contrast, the systems DMF (1) + water (2) and NMF (1) + water (2) show relatively large positive values as shown in Figure 2. The measured binary $\Delta\eta$ data were compared with the values in the literature[8,11] and also showed good agreement. The literature values were plotted in Figure 2 together with the experimental data. The binary $\Delta\eta$ were also correlated well with Redlich−Kister polynomials. Their standard deviations according to eq 3 are 0.010, 0.005, and 0.051 mPa·s for the different binary systems.

Ternary System. The excess molar volumes for the ternary system of DMF (1) + NMF (2) + water (3) measured at 298.15 K are listed in Table 3. The measured ternary V^E data show negative deviations from ideal behavior over the whole composition range. The excess molar properties for the ternary systems were correlated with the Cibulka equation[7] as a modification of the Ra-

Table 4. Fitted Redlich−Kister Parameters and Cibulka Parameters with the Standard Deviations for V^E and $\Delta\eta$ of DMF (1) + NMF (2) + Water (3) at 298.15 K

	system	A_1	A_2	A_3	A_4	A_5	σ_{st}
V^E	1 + 2	−0.0795	−0.0283	−0.0052	−0.0546	0.0093	0.002[a]
	2 + 3	−2.1162	1.2348	−0.1851	−0.4598	0.5398	0.005[a]
	1 + 3	−4.2600	1.9922	0.1142	−1.7065	1.0397	0.009[a]
	1 + 2 + 3	5.1849	−3.7233	−1.3734			0.013[a]
$\Delta\eta$	1 + 2	−0.6404	0.1568	0.1232	0.1670		0.010[b]
	2 + 3	3.0215	−2.7664	0.3345	0.8924		0.005[b]
	1 + 3	4.3620	−7.8642	1.8511	4.0871		0.051[b]
	1 + 2 + 3	−4.5644	3.0561	−1.3487			0.057[b]

[a] The unit is cm^3·mol^{-1}. [b] The unit is mPa·s.

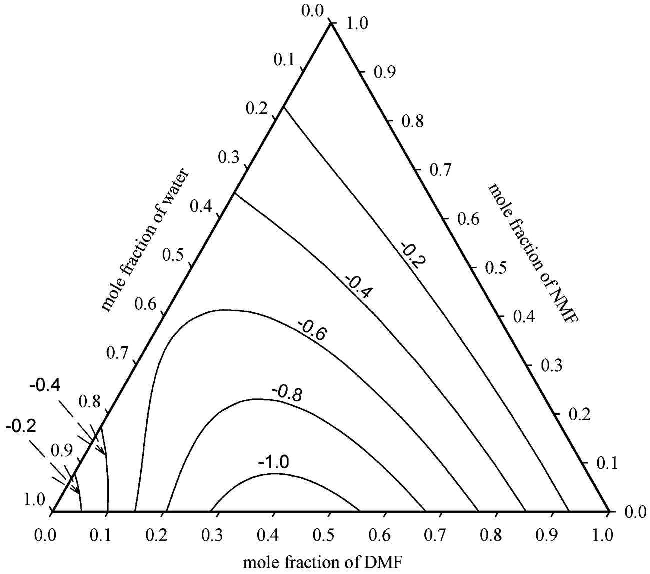

Figure 3. Constant lines of the excess molar volumes (cm^3·mol^{-1}) for the ternary system of DMF (1) + NMF (2) + water (3) at 298.15 K.

dojkovič equation[12] (see eq 11):

$$M^E_{123} = M^E_{12*} + M^E_{23*} + M^E_{13*} + x_1x_2x_3(A_1 + A_2x_1 + A_3x_2) \quad (7)$$

where M^E_{12*}, M^E_{23*}, and M^E_{13*} represent the excess properties, calculated from binary Redlich−Kister parameters and x_1, x_2, and x_3 are the mole fractions in the ternary mixture.

The fitted parameters of the Redlich−Kister and Cibulka equations are listed together with the standard deviations of the binary and ternary systems in Table 4. With a standard deviation of 0.013 cm^3·mol^{-1}, the correlated values are in good agreement with the experimental data. The correlation results are given in Figure 3. The solid lines in Figure 3 represent constant excess molar volumes of the ternary systems calculated by the Cibulka equation.

The viscosity deviations ($\Delta\eta$) for the ternary system were determined using the same procedure starting with $\Delta\eta$ for the binary systems. The experimental viscosity deviations for DMF (1) + NMF (2) + water (3) are listed in Table 3 together with the results of the excess molar volumes. With the exception of low water concentration, the measured ternary $\Delta\eta$ values show positive deviations. The measured $\Delta\eta$ values were compared with the correlated values using the Cibulka equation (eq 6). The Cibulka equation provided good results for the correlation of $\Delta\eta$ with a standard deviation of 0.057 mPa·s. The results are plotted in Figure 4. The fitted parameters and the standard deviations for the Redlich−Kister and Cibulka equations are listed in Table 4 together with those for the excess molar volumes for the same binary and ternary systems.

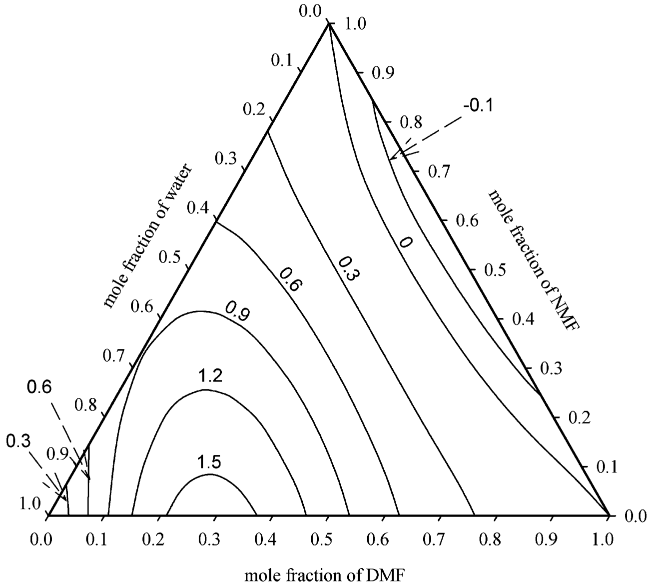

Figure 4. Constant lines of the viscosity deviations (mPa·s) for the ternary system of DMF (1) + NMF (2) + water (3) at 298.15 K.

Table 5. Standard Deviations[a] for the Estimation Results of the Excess Molar Volumes and Viscosity Deviations for the Ternary System of DMF (1) + NMF (2) + Water (3) at 298.15 K

	model equation			
	Tsao−Smith	Kohler	Rastogi	Radojkovič
V^E	0.297[b]	0.273[b]	0.272[b]	0.073[b]
$\Delta\eta$	0.481[c]	0.346[c]	0.355[c]	0.096[c]

[a] Calculated using eq 3. [b] The unit is cm^3·mol^{-1}. [c] The unit is mPa·s.

Prediction of the Ternary Data Using Binary Parameters. Excess molar volumes and viscosity deviations for the ternary system were calculated using the four conventional prediction models of Tsao−Smith,[13] Kohler,[14] Rastogi et al.,[15] and Radojkovič et al.[12] The expressions for these models are as follows:

Tsao−Smith equation:

$$M^E_{123} = \frac{x_2M^E_{12}}{1 - x_1} + \frac{x_3M^E_{13}}{1 - x_1} + (1 - x_1)M^E_{23} \quad (8)$$

where M^E_{ij} is the binary contribution of the ternary property at x^0_i and x^0_j:

$$(x^0_i = 1 - x^0_j = x_i/(x_i + x_j))$$

Kohler equation:

$$M^E_{123} = (x_1 + x_2)^2M^E_{12} + (x_1 + x_3)M^E_{13} + (x_2 + x_3)M^E_{23} \quad (9)$$

Rastogi equation:

$$M^E_{123} = \frac{1}{2}[(x_1 + x_2)M^E_{12} + (x_1 + x_3)M^E_{13} + (x_2 + x_3)M^E_{23}] \quad (10)$$

Radojkovič equation: $M^E_{123} = M^E_{12*} + M^E_{23*} + M^E_{13*}$ (11)

In the Radojkovič equation, the binary contributions M^E_{ij*} are evaluated directly using the ternary mole fractions.

The standard deviations between the experimental ternary data and the estimated values for the four prediction models are listed in Table 5. The Radojkovič equation provided the best results with standard deviations of 0.073 $cm^3 \cdot mol^{-1}$ and 0.096 mPa·s for V^E and $\Delta\eta$, respectively.

Conclusions

Excess molar volumes (V^E) and viscosity deviations ($\Delta\eta$) at 298.15 K were calculated for the binary systems of DMF (1) + NMF (2), NMF (1) + water (2), and DMF (1) + water (2) and also for the ternary system of DMF (1) + NMF (2) + water (3) from the measured density and kinematic viscosity. The binary and ternary V^E values show negative deviations from ideal behavior over the whole composition range. For $\Delta\eta$ in a wide composition range, positive deviations except in the DMF + NMF rich region are obtained. The binary $V^E$and $\Delta\eta$ data were correlated reliably with the Redlich−Kister equation, while the Cibulka equation was applied successfully for the ternary system. The experimental ternary V^E and $\Delta\eta$ results were compared with the predicted values using binary contribution models. The Radojkovič equation provided the best results.

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