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Measurement and Correlation of Mutual Solubilities for High-Viscosity Binary Systems: Aniline + Methylcyclohexane, Phenol + Heptane, Phenol + Octane, and Glycerol + 1-Pentanol

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A laser light scattering technique was used for the determination of mutual solubilities at moderate pressure (saturated vapor pressure of a mixture) in four high-viscosity binary systems: aniline + methylcyclohexane, phenol + heptane, phenol + octane, and glycerol + 1-pentanol. The liquid−liquid solubility data were measured up to the upper critical solution temperature. The experimentally determined solubility data were satisfactory correlated by the NRTL equation and Tsuboka−Katayama's modification of the Wilson equation.

Introduction

Liquid−liquid equilibrium (LLE) has an important role in the design and development of separation processes. LLE data are also essential in theoretical studies, such as the applicability of the activity coefficient model. However, the experimental values of the solubilities reported in the literature frequently show discrepancies. In addition, liquid mutual solubility data for high-viscosity binary systems are lacking.

In our recent studies, we have measured the LLE data for the following systems containing viscous components: diethylene glycol + benzene,[1] aniline + hexane,[2] aniline + cyclohexane,[4] aniline + heptane,[8] and phenol + hexane,[8] up to the upper critical solution temperature (UCST) using a laser scattering method developed in our laboratory. As an extension of that work, we made detailed measurements of the four binary solubility curves of aniline + methylcyclohexane, phenol + heptane, phenol + octane, and glycerol + 1-pentanol systems at moderate pressures, and the UCSTs of these systems were determined.

Experimental Section

Apparatus and Procedure. The cloud points of the binary mixtures were measured using a laser scattering technique. Details of the apparatus and its operation have been described elsewhere.[1−8] In this study, a light sensor (NP type silicon capsule module, model 3-400S, Solar Tech, Japan) was used. This sensor is placed away from the incident path of the light beam so that the low-intensity scattered light, which arises at the beginning of turbidity formation, can be detected effectively. A thermoregulator temperature control system (model UT/350, Yokokawa Electric Co. Ltd.) was adopted for the purpose of shortening the measurement time and improving the measurement accuracy. The equilibrium cell was covered with a film heater for heating.

The liquid mole fraction was determined by mass with a Mettler digital balance (model H315 having a sensitivity of 0.1 mg and a maximum load of 1 kg), and the reproducibility of the temperature of the cloud point was within ±0.1 K over the mole fraction range.

Table 1. Experimental Cloud Point Results for the Aniline (1) + Methylcyclohexane (2) System

T/K	x_1^a	T/K	x_1^a	T/K	x_1^a	T/K	x_1^a
292.1_5	0.1011	312.5_6	0.3649	313.7_6	0.5131	312.2_5	0.6600
296.7_7	0.1209	313.1_5	0.3994	313.7_8	0.5174	311.8_2	0.6781
300.9_5	0.1521	313.1_8	0.4088	313.7_6	0.5194	310.5_8	0.7106
304.2_8	0.1725	313.2_8	0.4142	313.7_9	0.5269	309.5_9	0.7309
306.3_3	0.2008	313.4_3	0.4319	313.7_8^b	0.5280^b	308.2_4	0.7513
307.8_9	0.2196	313.5_3	0.4426	313.7_6	0.5382	306.6_7	0.7721
309.4_5	0.2562	313.4_7	0.4500	313.6_6	0.5574	304.5_8	0.7944
310.1_7	0.2696	313.5_8	0.4625	313.6_2	0.5657	302.6_0	0.8116
310.8_8	0.2931	313.6_5	0.4770	313.3_3	0.6034	299.0_8	0.8340
311.6_9	0.3167	313.7_3	0.4973	313.0_8	0.6167	294.1_7	0.8608
312.2_2	0.3412	313.7_5	0.5084	312.8_9	0.6354	290.6_2	0.8747

[a] Mole fraction of aniline. [b] The upper critical solution point.

Materials. Special grade aniline, methylcyclohexane, phenol, heptane, octane, glycerol, and 1-pentanol were supplied by Wako Pure Chemical Industries Ltd., Japan. All chemicals were stored over molecular sieves in order to reduce the water content. The purity of chemicals was established by gas chromatography to be better than 99.91 mol % for aniline, 99.94 mol % for methylcyclohexane, 99.9 mol % for phenol, 99.92 mol % for heptane, 99.95 mol % for octane, 99.9 mol % for glycerol, and 99.92 mol % for 1-pentanol.

Results and Discussion

Aniline (1) + Methylcyclohexane (2). The experimentally determined cloud points for the aniline (1) + methylcyclohexane (2) system are listed in Table 1 and graphically shown in Figure 1. The determined UCST in this study is marked with the sign of ⊗ in this figure. This UCST was interpolated from the experimental cloud points near the UCST. In this system, Angelescu and Zinca[9] have presented the mutual solubilities at 293.15 and 310.9 K, and the data at 298.15 K have been reported by Varteressian and Fenske.[10] However, the solubility curve at all mole fraction range is not considered. In this study, the cloud points were measured in small composition incre-

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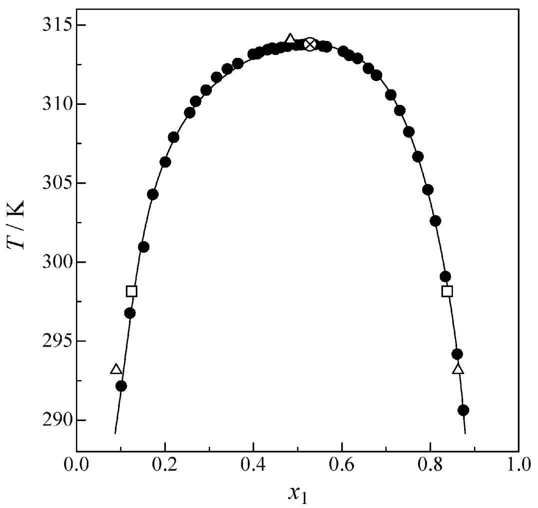

Figure 1. Liquid mutual solubilities for the aniline (1) + methylcyclohexane (2) system: ●, this work; ⊗, UCST; △, Angelescu and Zinca (1941, ref 9); □, Varteressian and Fenske (1937, ref 10); —, T−K−Wilson equation.

Table 2. Experimental Cloud Point Results for the Phenol (1) + Heptane (2) System

T/K	x_1[a]	T/K	x_1[a]	T/K	x_1[a]	T/K	x_1[a]
305.9_5	0.1372	316.6_1	0.2127	326.5_8	0.4497	323.0_1	0.7010
307.0_5	0.1429	319.5_8	0.2512	326.9_0	0.4698	319.5_4	0.7465
311.2_8	0.1693	321.9_9	0.2949	327.3_0[b]	0.5188[b]	317.3_3	0.7700
311.9_3	0.1737	324.2_7	0.3442	327.0_1	0.5563	315.0_1	0.7783
314.2_7	0.1946	324.9_9	0.3773	326.6_3	0.6051	311.2_6	0.7998
314.5_0	0.1981	325.9_2	0.4142	325.0_7	0.6417	305.6_3	0.8297
315.6_7	0.2043	326.0_5	0.4222	324.0_4	0.6712		

[a] Mole fraction of phenol. [b] The upper critical solution point.

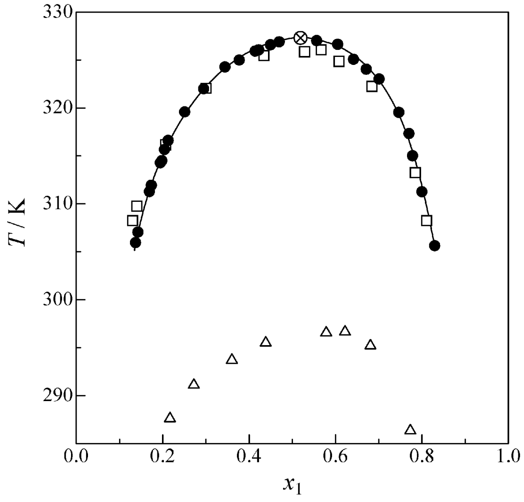

Figure 2. Liquid mutual solubilities for the phenol (1) + heptane (2) system: ●, this work; ⊗, UCST; △, Campetti and Grosso (1913, ref 11); □, Vondráček (1937, ref 12); —, T−K−Wilson equation.

Table 3. Experimental Cloud Point Results for the Phenol (1) + Octane (2) System

T/K	x_1[a]	T/K	x_1[a]	T/K	x_1[a]	T/K	x_1[a]
306.6_7	0.1486	328.3_0	0.4176	328.7_2	0.6493	310.9_9	0.8472
314.6_9	0.1943	329.3_3	0.4935	327.2_8	0.6979	306.1_8	0.8622
323.3_3	0.2960	329.5_3	0.5437	325.2_2	0.7451		
326.0_8	0.3479	329.5_4[b]	0.5457[b]	320.1_6	0.7949		
327.4_6	0.3990	329.3_4	0.5991	316.5_5	0.8191		

[a] Mole fraction of phenol. [b] The upper critical solution point.

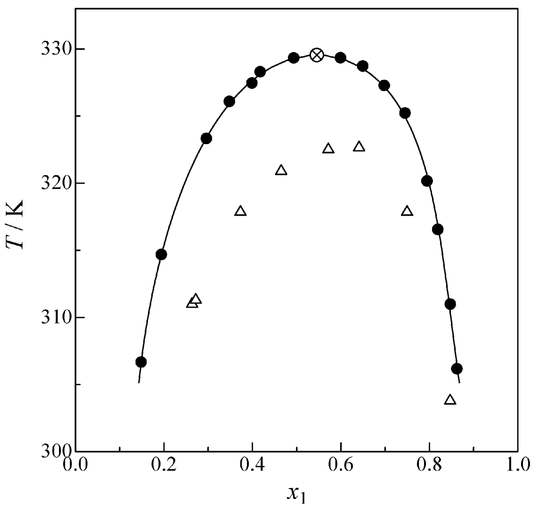

Figure 3. Liquid mutual solubilities for the phenol (1) + octane (2) system: ●, this work; ⊗, UCST; △, Campetti and Grosso (1913, ref 11); —, T−K−Wilson equation.

ments, especially in the vicinity of the UCST. Delicate temperature control was possible using the thermoregulator, which allowed for high clarity in this region.

Phenol (1) + Heptane (2). Table 2 reports the experimental cloud points for the phenol (1) + heptane (2) system, and plots of the cloud points against the mole fraction of phenol, x_1, are shown in Figure 2, along with the literature values.[11,12] Serious differences are apparent between the two literature data sets. Roozeboom[13] has raised doubts about the experimental data of Campetti and Grosso. Vondráček has also pointed out the impure heptane used by Campetti and Grosso, containing naphthenes or aromatic hydrocarbons, as the reason for these differences. Our experimental data showed the same behavior as the data of Vondráček. The determined UCST in this work is 327.3_0 K, while that of Vondráček is 326.0 K, for a difference of about 1.3 K. As we pointed out in our previous work,[8] these literature values were determined visually. Our experimental data seem to be more accurate than the literature data by visual determinations.

Phenol (1) + Octane (2). The experimental solubility data are presented in Table 3. Figure 3 compares our experimental cloud point values for this system with the literature values of Campetti and Grosso.[11] As can be seen, serious discrepancies are also found between our experimentally determined data and the values of Campetti and Grosso. In this system, the intensity of the scattered light was weak, and the variation of voltage is lower than 1 mV. Our experimental apparatus has an advantage in that low-intensity light can be detected efficiently. Therefore, we consider our experimental cloud points to be more accurate than the literature values, as was the case for the phenol (1) + heptane (2) system.

In this study, we considered the change in behavior of LLE for phenol + hydrocarbon systems according to the increase of the carbon number of the hydrocarbon. Figure 4 illustrates the plots of the LLE for our experimental data and the two literature data sets, respectively. The UCST, T_c, and the mole fraction of phenol at UCST and $x_{1,\mathrm{UCST}}$ are also shown in this figure. As can be seen, our experimental data show that the behavior of the mutual solubility for three phenol + hydrocarbon systems is almost the same to about 0.3 mole fraction of phenol. Furthermore, our determined UCST rises as the carbon number of hydrocarbons increases, and the $x_{1,\mathrm{UCST}}$ moves toward the phenol-

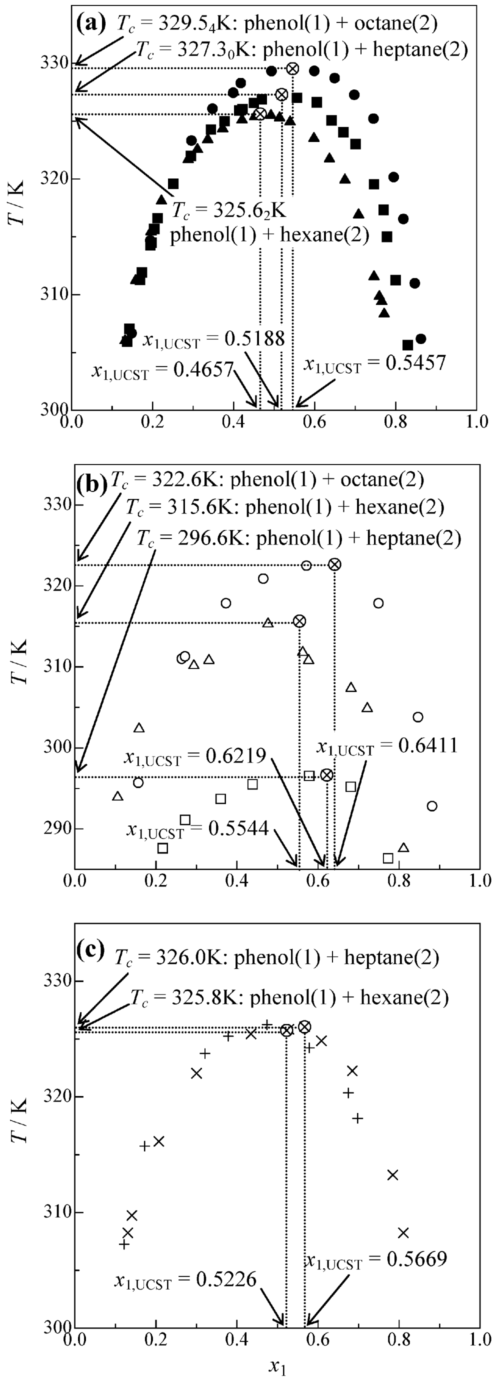

Figure 4. Changes in the behavior of LLE for three phenol + hydrocarbon systems. (a) Our measurements: ▲, phenol (1) + hexane (2) (ref 8); ■, phenol (1) + heptane (2) (this work); ●, phenol (1) + octane (2) (this work). (b) Campetti and Grosso (1913, ref 11): △, phenol (1) + hexane (2); □, phenol (1) + heptane (2); ○, phenol (1) + octane (2). (c) Vondráček (1937, ref 12): +, phenol (1) + hexane (2); ×, phenol (1) + heptane (2).

rich side. This tendency is the same as that for the methanol + hydrocarbon systems measured in our laboratory.[6,7] On the other hand, the solubility curve of Campetti and Grosso is different from our data, although the $x_{1,\mathrm{UCST}}$ values show the same trend. The behavior of Vondráček's LLE data is similar to that of our data, although the magnitude of change with the increase in carbon number is smaller than that of our data.

Table 4. Experimental Cloud Point Results for the Glycerol (1) + 1-Pentanol (2) System

T/K	x_1[a]	T/K	x_1[a]	T/K	x_1[a]	T/K	x_1[a]
289.9$_7$	0.2046	330.3$_8$	0.4613	337.6$_2$[b]	0.7032[b]	330.9$_7$	0.8497
301.0$_6$	0.2526	334.1$_0$	0.5208	337.4$_7$	0.7246	327.4$_9$	0.8720
307.7$_1$	0.2850	335.3$_2$	0.5431	337.1$_5$	0.7678	320.0$_0$	0.8959
316.1$_6$	0.3357	336.2$_7$	0.5901	334.9$_6$	0.8081	312.8$_1$	0.9124
322.2$_6$	0.3823	337.1$_3$	0.6298	334.3$_9$	0.8202		
326.7$_3$	0.4169	337.6$_3$	0.6918	333.8$_1$	0.8291		

[a] Mole fraction of glycerol. [b] The upper critical solution point.

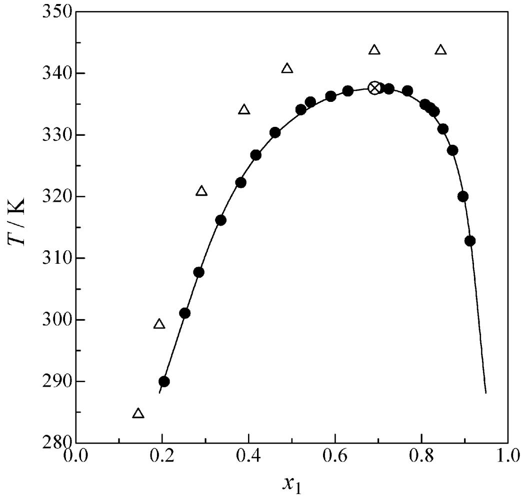

Figure 5. Liquid mutual solubilities for the glycerol (1) + 1-pentanol (2) system: ●, this work; ⊗, UCST; △, Mel'nikova and Zhuravlev (1965, ref 14); —, T−K−Wilson equation.

Glycerol (1) + 1-Pentanol (2). Table 4 and Figure 5 show our experimental cloud point values for this system with the literature values of Mel'nikova et al.[14] Data in the glycerol-rich region are not available in the literature. It is thought that the determination of cloud points in this region was difficult since the viscosity of glycerol is very high. In this work, the cloud points were measured in the glycerol-rich region in detail. As shown in Figure 5, serious discrepancies exist between the experimental and literature values. For this system, the scattered light is low in intensity, much like the phenol (1) + octane (2) system. Therefore, our experimental technique for this system provides more reliable mutual solubility data than those determined visually by Mel'nikova et al.

Data Reduction

The experimental cloud point data were correlated with the NRTL equation[15] and Tsuboka−Katayama's modification of the Wilson equation (T−K−Wilson).[16]

NRTL equation:

$$\ln \gamma_i = x_j^2\left[\tau_{ji}\left(\frac{G_{ji}}{x_i + G_{ji}x_j}\right)^2 + \frac{\tau_{ij}G_{ij}}{(G_{ij}x_i + x_j)^2}\right] \tag{1}$$

$$G_{ij} = \exp(-\alpha_{ij}\tau_{ij}) \tag{2}$$

$$\tau_{ij} = \frac{g_{ij} - g_{jj}}{RT} \tag{3}$$

In eqs 1−3, x_i is the mole fraction of component i, γ_i is the activity coefficient of component i, α_{ij} is the nonrandomness parameter of the NRTL equation, $g_{ij} - g_{jj}$ is the binary interaction parameter, T is the absolute temperature, and R is the gas constant. In this study, we adopted the constant value $\alpha_{12} = 0.4$ for the four systems, so that the behavior of $g_{ij} - g_{jj}$ versus temperature is linear in nature.

Table 5. Parameters L_i, M_i, and N_i of Equation 8 for Calculating Liquid Molar Volumes of Pure Components

	L_i	M_i	N_i
component	$m^3 \cdot mol^{-1}$	$m^3 \cdot mol^{-1} \cdot K^{-1}$	$m^3 \cdot mol^{-1} \cdot K^{-2}$
aniline	7.41450×10^{-5}	3.88072×10^{-8}	6.54293×10^{-11}
methylcyclohexane	8.97323×10^{-5}	-5.29018×10^{-9}	2.31873×10^{-10}
phenol	8.05245×10^{-5}	-1.75440×10^{-8}	1.41377×10^{-10}
heptane	-1.15533×10^{-4}	1.49912×10^{-6}	-2.07500×10^{-9}
octane	1.23931×10^{-4}	-1.33265×10^{-7}	5.32960×10^{-10}
glycerol	6.70753×10^{-5}	5.00015×10^{-9}	5.20358×10^{-11}
1-pentanol	9.52112×10^{-5}	-7.22510×10^{-9}	1.75587×10^{-10}

T−K−Wilson equation:

$$\ln \gamma_i = \ln \frac{x_i + (v_j^{\mathrm{L}}/v_i^{\mathrm{L}})x_j}{x_i + \Lambda_{ij}x_j} + (\beta - \beta_v)x_j \tag{4}$$

$$\beta = \frac{\Lambda_{ij}}{x_i + \Lambda_{ij}x_j} - \frac{\Lambda_{ji}}{\Lambda_{ji}x_i + x_j} \tag{5}$$

$$\beta_v = \frac{v_j^{\mathrm{L}}/v_i^{\mathrm{L}}}{x_i + (v_j^{\mathrm{L}}/v_i^{\mathrm{L}})x_j} - \frac{v_i^{\mathrm{L}}/v_j^{\mathrm{L}}}{(v_i^{\mathrm{L}}/v_j^{\mathrm{L}})x_i + x_j} \tag{6}$$

$$\Lambda_{ij} = \frac{v_j^{\mathrm{L}}}{v_i^{\mathrm{L}}} \exp\left(-\frac{\lambda_{ij} - \lambda_{ii}}{RT}\right) \tag{7}$$

where v_i^{L} is the liquid molar volume of the pure component i, and $\lambda_{ij} - \lambda_{ii}$ is the binary interaction parameter of the T−K−Wilson equation. The value of v_i^{L} was calculated as a function of temperature in the following form, as in previous studies:[6,8]

$$v_i^{\mathrm{L}} = L_i + M_iT + N_iT^2 \tag{8}$$

The parameters L_i, M_i, and N_i of aniline, phenol, heptane, and octane were taken from previous papers,[6,8] and those of methylcyclohexane, glycerol, and 1-pentanol were determined from the v_i^{L} data in the compilation of Timmermans.[17] These parameters are listed in Table 5.

We used the following equations to express the temperature dependencies of the binary interaction parameters in eqs 3 and 7:

NRTL equation:

$$g_{12} - g_{22} = A_{12} + B_{12}(T_c - T) + C_{12}(T_c - T)^2 + D_{12}(T_c - T)^3 \tag{9a}$$

$$g_{21} - g_{11} = A_{21} + B_{21}(T_c - T) + C_{21}(T_c - T)^2 + D_{21}(T_c - T)^3 \tag{9b}$$

T−K−Wilson equation:

$$\lambda_{12} - \lambda_{11} = A_{12} + B_{12}(T_c - T) + C_{12}(T_c - T)^2 + D_{12}(T_c - T)^3 \tag{10a}$$

$$\lambda_{21} - \lambda_{22} = A_{21} + B_{21}(T_c - T) + C_{21}(T_c - T)^2 + D_{21}(T_c - T)^3 \tag{10b}$$

where T_c is the UCST. The coefficients of eqs 9a−10b were estimated by a previously reported method.[1] The LLE compositions necessary for the reduction of the data were obtained by smoothing of the experimental cloud points.

The estimated parameters of eqs 9a−10b and the average deviations between the correlated and experimental mutual solubility data are given in Table 6. The difference of the average deviation $|\Delta x_1|_{\mathrm{av}}$ between the two equations is within 0.0007 mole fraction for all four systems. The correlated results by the T−K−Wilson equation are graphically shown in Figures 1−3 and Figure 5. Figure 6 also shows plots of $\delta = T_{\mathrm{exptl}} - T_{\mathrm{calct}}$ versus x_1 for the two equations for each system. The values of δ for the two equations are small for all liquid mole fractions. These

Table 6. Determined Parameters and Deviations between Calculated and Experimental Cloud Points for the Four Binary Systems Using the NRTL and T−K−Wilson Equations

	NRTL		T−K−Wilson	
	$ij = 12$	$ij = 21$	$ij = 12$	$ij = 21$
Aniline (1) + Methylcyclohexane (2) System				
$A_{ij}/J \cdot mol^{-1}$	4.343147×10^3	4.017253×10^3	6.971729×10^3	5.395475×10^3
$B_{ij}/J \cdot mol^{-1} \cdot K^{-1}$	3.174599×10^1	1.567151×10^2	2.556231×10^2	3.329083×10^1
$C_{ij}/J \cdot mol^{-1} \cdot K^{-2}$	9.998120×10^{-1}	-7.103803	-1.283457×10^1	2.260994
$D_{ij}/J \cdot mol^{-1} \cdot K^{-3}$	-3.412528×10^{-2}	1.441789×10^{-1}	2.614165×10^{-1}	-7.052446×10^{-2}
$\lvert \Delta x_1 \rvert_{\mathrm{av}}$[a]	0.0065		0.0072	
Phenol (1) + Heptane (2) System				
$A_{ij}/J \cdot mol^{-1}$	4.476982×10^3	4.249684×10^3	6.778376×10^3	3.583919×10^3
$B_{ij}/J \cdot mol^{-1} \cdot K^{-1}$	3.729624×10^1	6.175039×10^1	8.544784×10^1	5.097953×10^1
$C_{ij}/J \cdot mol^{-1} \cdot K^{-2}$	-1.242309	-4.152565×10^{-3}	4.795651×10^{-1}	-1.665480
$D_{ij}/J \cdot mol^{-1} \cdot K^{-3}$	2.625796×10^{-2}	-2.349639×10^{-2}	-5.276466×10^{-2}	4.049993×10^{-2}
$\lvert \Delta x_1 \rvert_{\mathrm{av}}$[a]	0.0054		0.0054	
Phenol (1) + Octane (2) System				
$A_{ij}/J \cdot mol^{-1}$	4.663718×10^3	4.098099×10^3	6.319255×10^3	2.954215×10^3
$B_{ij}/J \cdot mol^{-1} \cdot K^{-1}$	6.683217×10^1	6.367846×10^1	6.930240×10^1	9.189108×10^1
$C_{ij}/J \cdot mol^{-1} \cdot K^{-2}$	-3.141191	-1.162430×10^{-1}	1.256067	-4.448862
$D_{ij}/J \cdot mol^{-1} \cdot K^{-3}$	6.510830×10^{-2}	-2.375176×10^{-2}	-6.970832×10^{-2}	9.783603×10^{-2}
$\lvert \Delta x_1 \rvert_{\mathrm{av}}$[a]	0.0033		0.0034	
Glycerol (1) + 1-Pentanol (2) System				
$A_{ij}/J \cdot mol^{-1}$	5.842429×10^3	2.652667×10^3	5.358761×10^3	7.250545×10^3
$B_{ij}/J \cdot mol^{-1} \cdot K^{-1}$	3.943988×10^1	7.304410×10^1	5.961458×10^1	5.659340×10^1
$C_{ij}/J \cdot mol^{-1} \cdot K^{-2}$	-1.139288	-1.408668	-1.220574	-1.660783
$D_{ij}/J \cdot mol^{-1} \cdot K^{-3}$	1.270232×10^{-2}	1.375795×10^{-2}	1.265825×10^{-2}	1.816155×10^{-2}
$\lvert \Delta x_1 \rvert_{\mathrm{av}}$[a]	0.0058		0.0057	

[a] $|\Delta x_1|_{\mathrm{av}} = \sum_{k=1}^{\mathrm{NDP}} |x_{1,\mathrm{exptl}} - x_{1,\mathrm{calct}}|_k/\mathrm{NDP}$, where NDP is the number of data points.

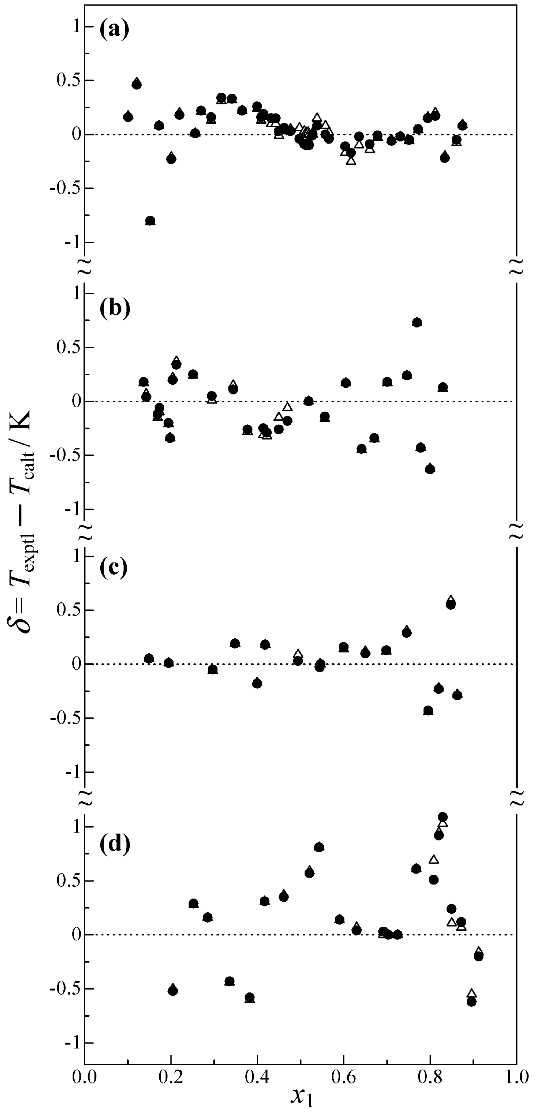

Figure 6. Plots of the difference between experimental and calculated temperature $T_{\text{exptl}} - T_{\text{calct}}$ vs x_1 for the two equations for the (a) aniline (1) + methylcyclohexane (2), (b) phenol (1) + heptane (2), (c) phenol (1) + octane (2), and (d) glycerol (1) + 1-pentanol (2) systems: ●, NRTL equation; △, T−K−Wilson equation.

results show that both the NRTL and T−K−Wilson equations give almost good accuracy in the correlation of LLE for these four systems.

Conclusion

The determination of mutual solubilities was made by using a laser light scattering technique on four high-viscosity binary systems: aniline + methylcyclohexane, phenol + heptane, and phenol + octane, and glycerol + 1-pentanol. LLE data were taken for temperatures up to the UCST. (The determined UCST is $T_c = 313.7_8$ K and $x_{1,\text{UCST}} = 0.5280$ for aniline + methylcyclohexane, $T_c = 327.3_0$ K and $x_{1,\text{UCST}} = 0.5188$ for phenol + heptane, $T_c = 329.5_4$ K and $x_{1,\text{UCST}} = 0.5457$ for phenol + octane, and $T_c = 337.6_2$ K and $x_{1,\text{UCST}} = 0.7032$ for glycerol + 1-pentanol.) Experimental mutual solubility data were satisfactorily correlated with the NRTL and T−K−Wilson equations.

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