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Three-Liquid-Phases Equilibria for the Quaternary System Water + Formamide + Cyclohexane + Diethylene Glycol Monobutyl Ether near the Tricritical Point

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The three-liquid-phases equilibria data of the quaternary system water (1) + formamide (2) + cyclohexane (3) + diethylene glycol monobutyl ether (4) in the tricritical region are reported. It has been found that the mass fraction of formamide in (formamide + water) controls the size of the three-phase region and the approach to the tricritical point. The experimental data have been analyzed to obtain the tricritical temperature and the tricritical composition.

Introduction

The phase behaviors of multicomponent systems including nonionic surfactants have attracted increasing attention for both practical and theoretical reasons. The practical reasons result from the applications of homogeneous microemulsions and fluids containing aqueous, surfactant, and oil phases with similar densities and low interfacial tensions. The theoretical reasons result from the interest in the tricritical phenomena. Nonionic surfactants of the homologous series of *n*-alkylpolyglycol ethers $H(CH_2)_i(OCH_2CH_2)_jOH$, abbreviated as C_iE_j hereafter, were widely used in the studies of phase behavior and critical phenomena for binary, ternary, and quaternary systems. However, relatively few measurements of three-liquid-phase equilibrium exist for systems composed of four liquid components including nonionic surfactants C_iE_j, although such equilibrium data for non-C_iE_j systems have been reported in the literature.[1−5] In this paper, we report equilibrium data for the system water (1) + formamide (2) + cyclohexane (3) + C_4E_2 (4) (the abbreviation for diethylene glycol monobutyl ether) near the tricritical point. No data for this quaternary system was found in the literature. Following Kahlweit et al.,[6] a binary mixture of water and formamide was considered to be a quasi-“pure” component (abbreviated as A). The mass fraction (denoted as w) of formamide in the quasi-pure component determines the property of this component and thereby determines the temperature range and the composition range of the three coexisting phases of the system and controls the approach to the tricritical point.

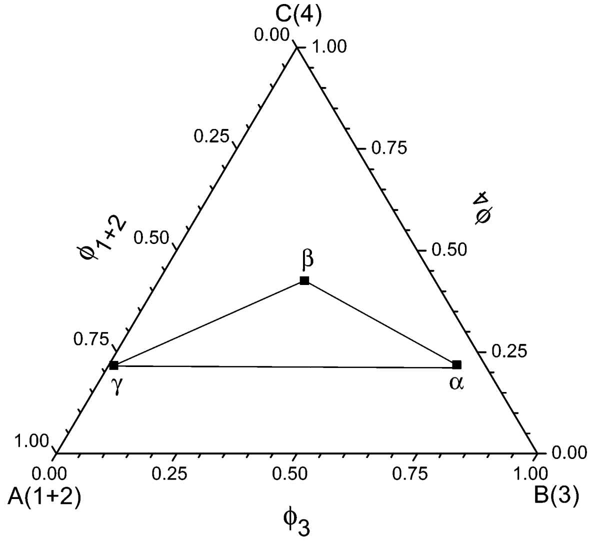

Figure 1. Isothermal quasi-ternary phase diagram for the system water (1) + formamide (2) + cyclohexane (3) + C_4E_2 (4) at t = 43.502 °C and w = 0.066. The mixture of water and formamide is taken as pure component A.

Table 1. Experimental Values of the Density of Pure Components at 20 °C

	ρ/g·cm^{-3}	
liquid	this work	literature
C_4E_2	0.9535	0.9527[a]
cyclohexane	0.7783	0.7785[b]
formamide	1.1328	1.13339[c]

[a] Reference 13. [b] Reference 14. [c] Reference 15.

Experimental Section

Experimental Principle. The compositions in the three coexisting phases of quasi-ternary mixtures were determined by an indirect method that was similar to that proposed by Knobler and Scott[7] to study the tricritical phenomena in quasi-binary mixtures.[8−11] A point in the triangle ABC in Figure 1 represents the composition of a ternary mixture at a certain temperature. Any mixture with a total composition in triangle $\alpha\beta\gamma$ separates into three phases, the concentrations of which are specified by the three vertices of the triangle. If the excess volumes are ignored and the cross-sectional area of the sample cell is

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Table 2. Volume Fractions of the Coexisting Three Phases at Different Temperatures with Different Values of *w* for the System Water (1) + Formamide (2) + Cyclohexane (3) + C_4E_2 (4)

	α (upper phase)				β (middle phase)				γ (lower phase)			
t/°C	ϕ_3	ϕ_1	ϕ_2	ϕ_4	ϕ_3	ϕ_1	ϕ_2	ϕ_4	ϕ_3	ϕ_1	ϕ_2	ϕ_4
$w = 0$												
39.427	0.835	0.032	0.000	0.141	0.125	0.546	0.000	0.330	0.057	0.605	0.000	0.340
39.436	0.837	0.028	0.000	0.145	0.128	0.545	0.000	0.324	0.053	0.606	0.000	0.340
39.453	0.828	0.030	0.000	0.151	0.145	0.511	0.000	0.341	0.048	0.706	0.000	0.244
39.473	0.815	0.028	0.000	0.155	0.150	0.506	0.000	0.344	0.041	0.719	0.000	0.241
39.502	0.816	0.032	0.000	0.155	0.159	0.458	0.000	0.384	0.039	0.721	0.000	0.238
39.527	0.812	0.034	0.000	0.154	0.166	0.441	0.000	0.392	0.037	0.734	0.000	0.230
39.568	0.807	0.035	0.000	0.154	0.179	0.429	0.000	0.393	0.032	0.744	0.000	0.223
39.692	0.808	0.040	0.000	0.156	0.199	0.397	0.000	0.404	0.023	0.768	0.000	0.208
39.817	0.803	0.039	0.000	0.161	0.220	0.370	0.000	0.410	0.016	0.786	0.000	0.198
39.970	0.799	0.036	0.000	0.163	0.244	0.342	0.000	0.414	0.011	0.799	0.000	0.190
40.121	0.784	0.042	0.000	0.174	0.269	0.315	0.000	0.416	0.010	0.809	0.000	0.180
40.295	0.773	0.043	0.000	0.184	0.302	0.284	0.000	0.414	0.006	0.821	0.000	0.174
40.461	0.755	0.049	0.000	0.196	0.333	0.258	0.000	0.409	0.004	0.826	0.000	0.164
40.617	0.738	0.053	0.000	0.209	0.369	0.229	0.000	0.402	0.003	0.831	0.000	0.167
40.778	0.697	0.060	0.000	0.239	0.426	0.197	0.000	0.379	0.002	0.847	0.000	0.160
40.815	0.686	0.062	0.000	0.244	0.443	0.182	0.000	0.375	0.001	0.851	0.000	0.157
40.832	0.686	0.068	0.000	0.253	0.438	0.184	0.000	0.372	0.001	0.852	0.000	0.157
40.840	0.666	0.071	0.000	0.263	0.457	0.171	0.000	0.372	0.001	0.852	0.000	0.156
$w = 0.066$												
42.983	0.772	0.033	0.002	0.193	0.180	0.404	0.025	0.390	0.041	0.649	0.041	0.269
43.244	0.742	0.045	0.003	0.210	0.248	0.312	0.020	0.421	0.014	0.717	0.045	0.224
43.366	0.731	0.051	0.003	0.215	0.269	0.275	0.017	0.439	0.011	0.726	0.046	0.217
43.502	0.721	0.055	0.003	0.218	0.303	0.255	0.016	0.426	0.011	0.726	0.046	0.217
43.749	0.651	0.081	0.005	0.263	0.394	0.193	0.012	0.413	0.011	0.739	0.046	0.196
43.847	0.622	0.091	0.006	0.281	0.430	0.173	0.011	0.386	0.011	0.739	0.046	0.197
$w = 0.086$												
43.902	0.733	0.039	0.003	0.215	0.159	0.434	0.036	0.379	0.079	0.558	0.047	0.317
43.912	0.734	0.036	0.003	0.215	0.159	0.427	0.036	0.371	0.069	0.554	0.046	0.317
43.927	0.727	0.051	0.004	0.218	0.166	0.412	0.035	0.387	0.059	0.603	0.051	0.288
44.113	0.715	0.061	0.005	0.218	0.229	0.334	0.028	0.410	0.040	0.649	0.054	0.257
44.279	0.706	0.062	0.005	0.227	0.266	0.293	0.025	0.417	0.033	0.670	0.056	0.242
44.444	0.690	0.066	0.006	0.239	0.303	0.257	0.021	0.419	0.022	0.685	0.057	0.236
44.613	0.650	0.077	0.006	0.267	0.357	0.219	0.018	0.406	0.018	0.693	0.058	0.220
44.694	0.627	0.081	0.007	0.285	0.417	0.188	0.016	0.379	0.013	0.701	0.059	0.215
44.719	0.597	0.093	0.008	0.302	0.446	0.175	0.015	0.364	0.012	0.706	0.059	0.214
$w = 0.126$												
46.206	0.716	0.045	0.006	0.215	0.210	0.346	0.045	0.400	0.058	0.566	0.073	0.302
46.286	0.704	0.053	0.007	0.230	0.227	0.314	0.040	0.419	0.042	0.602	0.077	0.280
46.362	0.699	0.054	0.007	0.240	0.248	0.294	0.038	0.422	0.039	0.624	0.080	0.257
46.429	0.684	0.062	0.008	0.246	0.266	0.276	0.036	0.422	0.030	0.629	0.081	0.260
46.488	0.674	0.066	0.008	0.253	0.280	0.264	0.034	0.422	0.028	0.637	0.082	0.253
46.544	0.668	0.065	0.008	0.258	0.295	0.252	0.032	0.421	0.029	0.643	0.083	0.245
46.585	0.660	0.069	0.009	0.262	0.310	0.239	0.031	0.420	0.022	0.648	0.083	0.243
46.628	0.640	0.077	0.010	0.273	0.323	0.230	0.030	0.417	0.022	0.651	0.084	0.237
46.708	0.618	0.082	0.010	0.291	0.357	0.206	0.026	0.413	0.018	0.654	0.084	0.241
46.736	0.590	0.083	0.011	0.306	0.373	0.195	0.025	0.406	0.020	0.654	0.084	0.236
46.739	0.602	0.083	0.011	0.303	0.373	0.196	0.025	0.407	0.020	0.656	0.084	0.236
46.766	0.585	0.091	0.012	0.321	0.394	0.184	0.024	0.398	0.016	0.657	0.084	0.233
$w = 0.186$												
49.617	0.601	0.067	0.014	0.318	0.216	0.291	0.059	0.435	0.091	0.454	0.092	0.363
49.624	0.597	0.068	0.014	0.322	0.229	0.279	0.057	0.434	0.084	0.469	0.095	0.342
49.687	0.589	0.071	0.015	0.325	0.262	0.248	0.050	0.44	0.066	0.501	0.102	0.331
49.696	0.588	0.074	0.015	0.323	0.266	0.246	0.050	0.439	0.065	0.503	0.103	0.329
49.745	0.581	0.073	0.015	0.331	0.293	0.225	0.046	0.436	0.061	0.513	0.105	0.320
49.759	0.581	0.073	0.015	0.332	0.298	0.223	0.046	0.433	0.057	0.515	0.105	0.322
49.826	0.565	0.073	0.015	0.347	0.338	0.190	0.039	0.433	0.044	0.532	0.109	0.315
49.834	0.562	0.075	0.015	0.347	0.343	0.187	0.038	0.432	0.042	0.532	0.109	0.318
49.859	0.549	0.076	0.015	0.355	0.367	0.177	0.036	0.427	0.040	0.537	0.109	0.319
49.874	0.537	0.085	0.017	0.356	0.396	0.164	0.034	0.402	0.036	0.536	0.109	0.312
$w = 0.206$												
50.703	0.568	0.074	0.017	0.324	0.238	0.267	0.062	0.433	0.126	0.419	0.097	0.371
50.749	0.561	0.079	0.018	0.342	0.272	0.242	0.056	0.431	0.106	0.440	0.102	0.362
50.793	0.537	0.084	0.020	0.358	0.309	0.218	0.050	0.423	0.085	0.448	0.104	0.350
50.826	0.513	0.089	0.021	0.377	0.338	0.197	0.045	0.421	0.072	0.473	0.109	0.348

uniform, then the total height H_i of a fluid component i in the cell may be expressed as eq 1

$$H_i = h^{\alpha}\varphi_i^{\alpha} + h^{\beta}\varphi_i^{\beta} + h^{\gamma}\varphi_i^{\gamma} \tag{1}$$

where h^{α}, h^{β}, and h^{γ} denote the heights of α, β, and γ phases, $\varphi_i{}^{\alpha}$, φ_i^{β}, and φ_i^{γ} are the volume fractions of component i in the α, β, and γ phases. $H_i = m_i/\rho_i S$ with ρ_i, S, and m_i being the density, cross-sectional area of the sample cell, and mass weighed into the cell. Subscript i denotes A, or cyclohexane, or C_4E_2.

In fact, it was observed that $\Delta H = \sum H_i - (h^{\alpha} + h^{\beta} + h^{\gamma}) \neq 0$ for each sample in the sample cell, which indicated that the excess volume probably could not be neglected. We assumed that the contribution of the excess volume to ΔH may be expressed as eq 2

$$\Delta H = ah^{\alpha} + bh^{\beta} + ch^{\gamma} \tag{2}$$

where a, b, and c are the excess heights of unit h for the α, β, and γ phases at a constant temperature and were obtained by fitting the experimental values of ΔH to eq 2. The combination of eqs 1 and 2 yields eq 3:

$$H_i = h^{\alpha}(1 + a)\varphi_i^{\alpha} + h^{\beta}(1 + b)\varphi_i^{\beta} + h^{\gamma}(1 + c)\varphi_i^{\gamma} \tag{3}$$

The values of h^{α}, h^{β}, and h^{γ} may be measured with a cathetometer, and H_i may be calculated from the mass of each component and its corresponding density. If a series of samples with different overall compositions were made over the three-phase regions, then we may obtain a series of simultaneous equations. The values of the volume fractions in eq 3 may be obtained by fitting the experimental values of h^{α}, h^{β}, and h^{γ} to the equation. The sum of the volume fractions in each phase should equal 1, which is a criterion of good experimental data.

Experimental Materials. The water was distilled and deionized, and its electrolytic conductivity was about 1.8 × 10^{-7} S·cm^{-1}. The formamide and the cyclohexane were of analytical purity grade provided by Xi'an Chemical Reagent Company. The mass fraction purity of nonionic surfactant diethylene glycol monobutyl ether (C_4E_2) obtained from Fluka was ≥99.0%. The densities of these materials were measured over a temperature range of 37 °C to 57 °C with an Anton Paar DMA60 DMA602 vibrating tube densimeter, and the dependences of density ρ on temperature t are expressed below with an uncertainty of better than 0.0001 g·cm^{-3}:

$$\rho_1/\text{g·cm}^{-3} = 1.0020 - 1.25 \times 10^{-4}(t/°\text{C}) - 3.24 \times 10^{-6}(t/°\text{C})^2$$

$$\rho_2/\text{g·cm}^{-3} = 1.1264 + 1.01 \times 10^{-4}(t/°\text{C}) - 9.52 \times 10^{-6}(t/°\text{C})^2$$

$$\rho_3/\text{g·cm}^{-3} = 0.7956 - 8.43 \times 10^{-4}(t/°\text{C}) - 1.31 \times 10^{-6}(t/°\text{C})^2$$

$$\rho_4/\text{g·cm}^{-3} = 0.9711 - 8.83 \times 10^{-4}(t/°\text{C}) + 3.79 \times 10^{-7}(t/°\text{C})^2$$

For the quasi-pure component (A), the dependence of density $\rho_A/(\text{g·cm}^{-3})$ on ρ_1 and ρ_2 with fixed w can be expressed as $\rho_A = \rho_1\rho_2/w\rho_1 + (1 - w)\rho_2$.

Table 1 shows the densities of the four materials at 20 °C together with those reported in the literature for comparison. The density data were used to calculate H_i in eq 3.

Experimental Procedure. The sample cells with Teflon screw plugs were purchased from Ace Glass Inc. The sample cells filled with acetone were weighed repeatedly over 4 days at room temperature to check for leakage from the cells. No significant changes in the mass were observed. The sample cells were calibrated by measuring the heights of the cyclohexane weighed into each cell. A uniform cross-sectional area was then obtained for each sample cell with an uncertainty of 0.001 cm^2, which was used to calculate H_i in eq 3. The quasi-pure component was prepared by mixing the proper amounts of water and formamide in a flask to form a binary solution with the desired mass fraction w. Five samples with various overall compositions were prepared by weighing A, cyclohexane, and C_4E_2 into the sample cells, and the cells were set in a holder immersed in a water bath with a temperature stability of 0.002 K. The holder was adjusted to be vertical to a cathetometer, which was used to measure the heights of the three coexisting phases with a precision of ±0.001cm after phase equilibrium was reached. This procedure was repeated for various temperatures in the three-phase region. After each change of temperature, we waited for 1 h for thermal equilibrium, and then the sample cells were shaken for a few minutes in the bath. The cells were set in the water bath for an additional (1 to 3) h after shaking for phase equilibrium. The temperature was measured with a Pt resistance thermometer connected to a multimeter/data acquisition system (Keithley model 2700). The precision of the temperature measurement was about ±0.002 K. The phase heights measured for 10 to 20 samples with various overall compositions at constant w were fitted to eq 3 to obtain φ_i^{α}, φ_i^{β}, and φ_i^{γ} for each temperature. The uncertainty in the determination of the volume fraction for each phase was estimated to be about ±0.005.

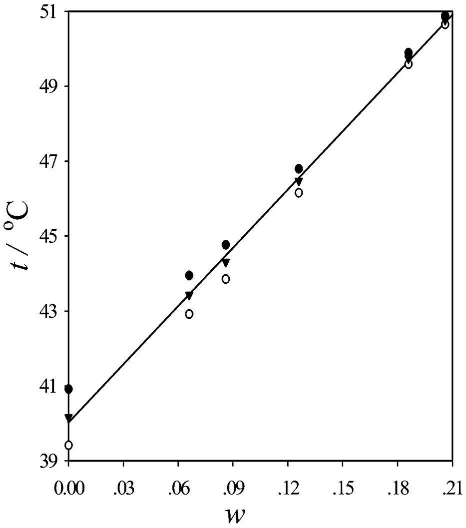

Figure 2. Plots of the upper critical end temperature t_u, lower critical end temperature t_l, and middle temperature t_m vs w for the system water (1) + formamide (2) + cyclohexane (3) + C_4E_2 (4): ●, t_u vs w; ○, t_l vs w; ▼, t_m vs w.

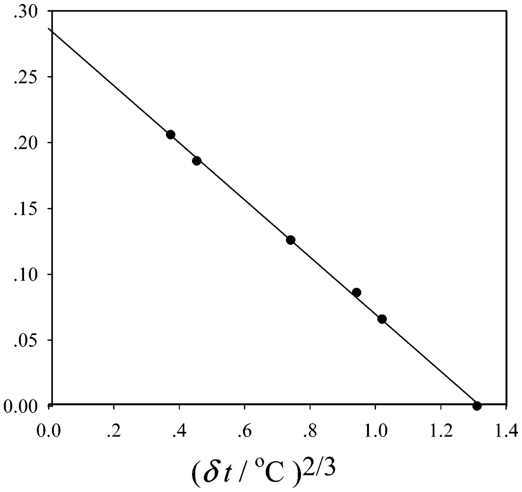

Figure 3. Plot of $(\delta t/°\text{C})^{2/3}$ vs w for the system water (1) + formamide (2) + cyclohexane (3) + C_4E_2 (4).

Result and Discussion

With the value of w being fixed at 0.000, 0.066, 0.086, 0.126, 0.186, and 0.206, six quasi-pure A components were

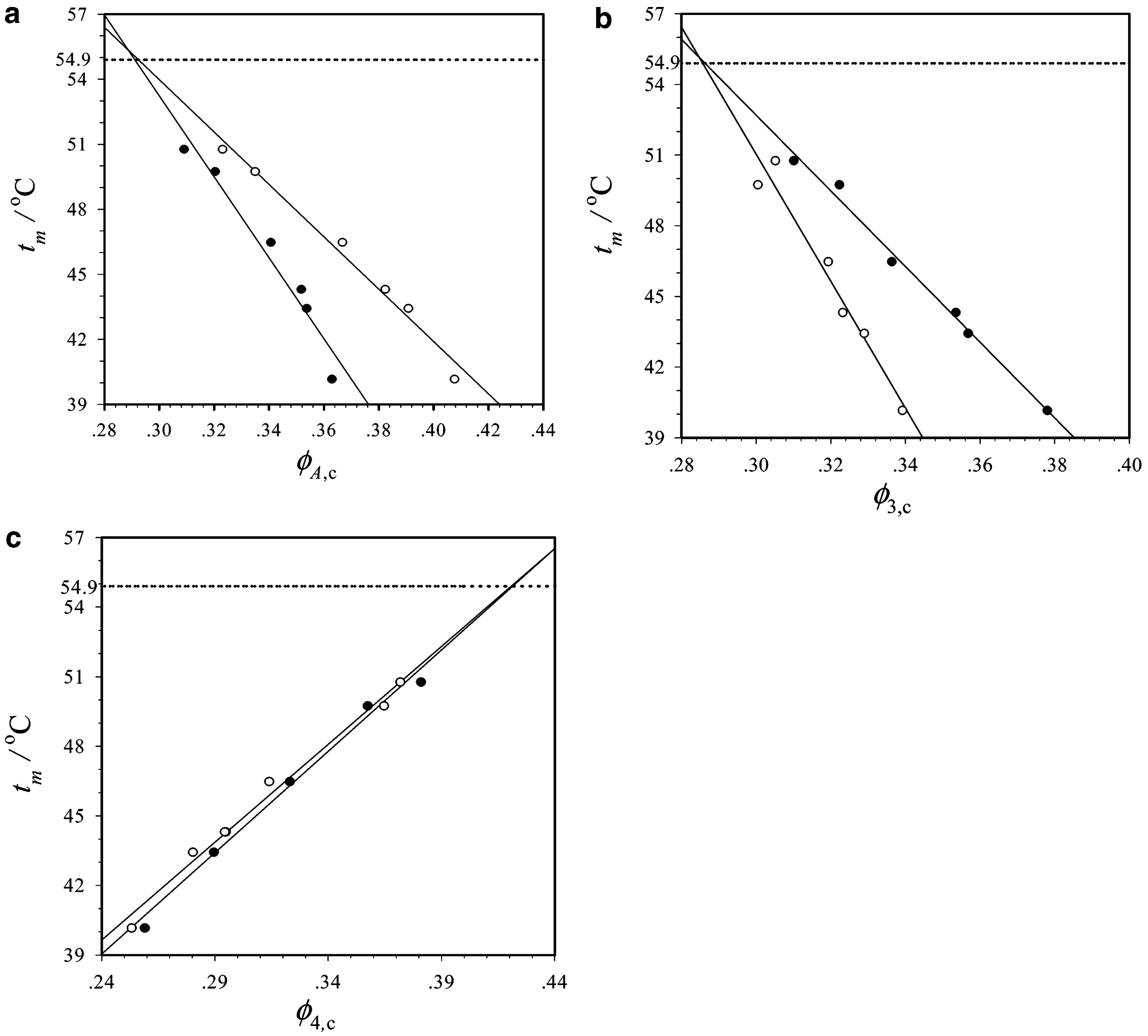

Figure 4. (a) Temperature dependence of the volume fraction $\phi_{A,c}$ of the centroids of the critical end-point tie lines for the system water (1) + formamide (2) + cyclohexane (3) + C_4E_2 (4): ○, centroids of the lower critical end-point tie lines; ●, centroids of the upper critical end-point tielines. The mixture of water and formamide is taken as pure component A. (b) Temperature dependence of the volume fraction $\phi_{3,c}$ of the centroids of the critical end-point tie lines for the system water (1) + formamide (2) + cyclohexane (3) + C_4E_2 (4): ○, centroids of the lower critical end-point tie lines; ●, centroids of the upper critical end-point tie lines. (c) Temperature dependence of the volume fraction $\phi_{4,c}$ of the centroids of the critical end-point tie lines for the system water (1) + formamide (2) + cyclohexane (3) + C_4E_2 (4): ○, centroids of the lower critical end-point tie lines; ●, centroids of the upper critical end-point tie lines.

made separately. Various quasi-ternary solutions of A + cyclohexane + C_4E_2 with different compositions for each of quasi-pure component were made in the three-phase region. The heights of three coexisting phases then were measured to obtain the volume fractions for various temperatures. Most of the sums of three volume fractions obtained from fitting eq 3 were 1.000 ± 0.009, which supports the estimated precision of ±0.005 for the volume fraction determined in this work. The volume fractions of water and formamide then were calculated from the values of volume fraction φ_A of A and w under the assumption of the quasi-ternary system. The results are listed in Table 2.

To test the hypothesis of the quasiternary solutions, eq 3 was used to fit part of the experimental data for components water and formamide to obtain the volume fractions of water and formamide in each phase at certain temperatures. It has been found that the ratios of these two volume fractions and therefore the ratios of the mass fractions of water to formamide in the three phases at each of the temperatures are identical within the experimental uncertainties. This is evidence that the hypothesis of the quasi-ternary solutions is valid.

The volume fractions listed may be converted to the mole fractions of components for the coexisting phases by the following equations:

$$x_A = \frac{M_3 M_4 \rho_A \varphi_A}{M_3 M_4 \rho_A \varphi_A + M_A M_4 \rho_3 \varphi_3 + M_A M_3 \rho_4 \varphi_4} \quad (4a)$$

$$x_3 = \frac{M_A M_4 \rho_3 \varphi_3}{M_3 M_4 \rho_A \varphi_A + M_A M_4 \rho_3 \varphi_3 + M_A M_3 \rho_4 \varphi_4} \quad (4b)$$

$$x_4 = \frac{M_3 M_A \rho_4 \varphi_4}{M_3 M_4 \rho_A \varphi_A + M_A M_4 \rho_3 \varphi_3 + M_A M_3 \rho_4 \varphi_4} \quad (4c)$$

where M_A, M_3, and M_4 are the molar masses of quasi-pure component A, cyclohexane, and C_4E_2, respectively. The mole fractions of water and formamide (x_1 and x_2) were then calculated from the values of x_A and w.

The three-phase equilibria may be illustrated by an "S"-type curve in a plot of temperature against volume fraction. However, only for a sample with a proper overall composition does an entire S-type curve appear, thereby allowing the upper critical end temperature t_u and lower critical end temperature t_l to be obtained by changing the temperature.

We carefully searched for the proper composition using the method described by Shen et al.[12] and determined the t_u, t_l, mean temperature t_m $\{t_m = (t_u + t_l)/2\}$, and critical compositions at the critical end points for each value of w. Figure 2 shows a plot of t_u, t_l, and t_m against w. The three temperatures approached one point called the tricritical point, at which the difference δt of the upper critical temperature and the lower critical temperature approaches zero, the interfaces between the upper and middle phases and the middle and lower phases disappear simultaneously, and the three phases become identical.

A plot of t_m against w yielded a straight line shown in Figure 2, which demonstrates that w is a good variable for controlling the approach to the tricritical point. According to the classical tricritical theory, $(\delta t)^{2/3}$ is proportional to t_m; therefore, plotting $(\delta t)^{2/3}$ against w should also yield a straight line. Figure 3 shows such a plot, which indicates that the classical tricritical theory is valid for our system. From Figure 3, we obtained the tricritical value of w at $\delta t = 0$, which is 0.287 ± 0.003. This value was then used to find the tricritcal temperature in Figure 2, which is (54.9 ± 0.2) °C.

Following Bocko's definition of the centroid compositions of the critical end-point tie lines,[4] we defined the centroid volume fraction as

$$\phi_{i,c} = \left(\frac{2}{3}\right)\phi_{i,ce} + \left(\frac{1}{3}\right)\phi_{i,sp} \qquad (5)$$

with $\phi_{i,ce}$ and $\phi_{i,sp}$ being the volume fractions of the ith component in the critical phase and in the spectator phase of the critical end-point tie lines, respectively. The values of $\phi_{i,ce}$ were obtained by extrapolating the average volume fractions of two coexisting phases close to the critical end points, whereas the values of $\phi_{i,sp}$ were obtained by extrapolating the volume fractions of spectator phases to the lower or upper critical temperatures. The plots of mean temperature t_m against $\phi_{i,c}$ for i = the quasi-pure component (A), cyclohexane, and surfactant diethylene glycol monobutyl ether (C_4E_2) are shown in Figure 4a−c, from which it may be clearly seen that the values of $\phi_{i,c}$ and t_m have linear dependences. Two lines in each Figure representing the centroids of the upper critical end-point and lower critical end-point tie lines intersect almost exactly at the tricritical temperature of 54.9 °C. At this temperature, the volume fractions from the two lines for all three components are consistent within 0.001. These volume fractions are the tricritical values and were found to be 0.292, 0.286, and 0.421 for A, cyclohexane, and C_4E_2, respectively. The sum of these three values of tricritical volume fractions was 0.999, which was consistent with the value of 1 within the experimental uncertainties. With the tricritical values of $w = 0.287$ at $t_t = 54.9$ °C, the tricritical volume fractions for water and formamide were calculated to be 0.215 and 0.077, respectively.

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