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Liquid−Liquid Phase Equilibria of the System Ethanol (1) + Water (2) + Tripotassium Citrate (3)

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Liquid−liquid equilibria (LLE) for the system ethanol (1) + water (2) + tripotassium citrate (3) were measured at temperatures of (288.15, 298.15, and 308.15) K. The dependence of temperature on the LLE data is very small in the temperature range studied, but the slopes of the tie lines were found to decrease slightly with decreasing temperature. The binodal curves at each temperature were well fitted by a nonlinear equation with four parameters, and the tie line data were successfully correlated with the Othmer−Tobias and Bancroft equations. The LLE results were correlated using the nonlinear and Othmer−Tobias equations in addition to the nonlinear and Bancroft equations. Both correlations showed close agreement with experimental values, with (0.27 and 0.26) % as the average deviations of three temperatures, respectively.

Introduction

An aqueous solution of 1-propanol, 2-propanol, or acetone easily separates into two liquid phases upon addition of a salt such as sodium chloride (for example, the phase equilibria of water + 1-propanol + sodium chloride are described by Gomis et al.[1] and Chou et al.,[2] those of water + 2-propanol + sodium chloride by Gomis et al.,[1] Chou et al.,[2] and Wu et al.,[3] and those of water + acetone + sodium chloride by Wu et al.[3]), but an aqueous ethanol solution cannot be phase-separated by an ordinary salt. We have confirmed that the addition of potassium dihydrogen phosphate (KH_2PO_4), disodium hydrogen phosphate (Na_2HPO_4), ammonium chloride, ammonium sulfate, sodium chloride, or sodium sulfate causes no phase separation of the aqueous ethanol solution; furthermore, we also found that aqueous ethanol was separated into two phases upon addition of either dipotassium hydrogen phosphate (K_2HPO_4) or sodium dihydrogen phosphate (NaH_2PO_4).[4] The solubilities of potassium dihydrogen phosphate, disodium hydrogen phosphate, ammonium chloride, ammonium sulfate, sodium chloride, and sodium sulfate in water, which are (20.0, 17.1, 28.2, 43.3, 26.43, and 21.9) wt % at 298.2 K, respectively,[5] are smaller than those of dipotassium hydrogen phosphate and sodium dihydrogen phosphate in water, which are (62.0 and 48.5) wt % at 298.2 K, respectively.[5] Only a few salts, indicating great solubility in water, cause a phase separation of an ethanol solution by addition.

To our knowledge, in addition to dipotassium hydrogen phosphate and sodium dihydrogen phosphate, magnesium sulfate, cesium sulfate, and potassium carbonate have been reported as salts that can cause the phase separation of an ethanol solution.[6–8] We found that tripotassium phosphate (K_3PO_4) also gives a phase separation of both aqueous ethanol and methanol solutions, but no formal experiments have yet been carried out.

In the present work, liquid−liquid equilibria (LLE) for the system ethanol + water + tripotassium citrate ($C_6H_5O_7K_3$) were measured at temperatures of (288.15, 298.15, and 308.15) K, and their phase diagrams were investigated. Tripotassium citrate is the only known organic salt that makes the two-phase separation of an aqueous ethanol solution, and it is also a gentle substance for the environment. It was confirmed that trisodium citrate does not make a two-phase separation of the solution.

When two polymers, such as polyethylene glycol (PEG) and dextran, or a polymer and a salt, such as PEG and potassium sulfate, were added to water, the liquid phase is separated into two phases, consisting of either two different polymer-rich phases or one polymer-rich and one salt-rich phase. In both cases, the water content in each phase constitutes a major component.[9] The systems (polymer + polymer + water) and (polymer + salt + water) as described are known as aqueous two-phase systems (ATPS). The systems (aliphatic alcohol, especially ethanol, + salt + water) which cause phase separations are defined as nonpolymer ATPS because both of their two phases have high water content, with one of the phases forming the alcohol-rich phase and the other the salt-rich phase.

Proteins are liable to undergo denaturation in organic solvents. According to Scopes,[10] the stability of the native conformation of glyceraldehyde-3-phosphate dehydrogenase decreased in the following alcohol solvents (in the order listed): methanol, ethanol, 2-propanol, 1-propanol, 1-butanol, and 1-pentanol. In other words, with the exception of water, the enzyme is the most stable in methanol, then in ethanol, etc. Some kinds of proteins are expected to maintain a stable state in an aqueous ethanol solution. The knowledge of the phase equilibria measured on nonpolymer ATPS can be applied to the separation and purification of biomolecules such as proteins and amino acids and of pharmaceutical intermediates by liquid−liquid extraction. Biomolecules in nonpolymer ATPS are expected to have different distributions than the ones in ATPS.

Experimental Section

Materials. Ethanol and tripotassium citrate monohydrate ($C_6H_5O_7K_3H_2O$) were purchased from Wako Pure Chemical Industries Ltd. The minimum purities were stated as (99.5 and 99.0) mass %, respectively, and these were used without further purification. 1-Propanol, used as a gas chromatography standard, was supplied from Tokyo Kasei Industry Co. Ltd. (stated to have a minimum purity of 99.0 mass %) and used after dehydration with 0.3 nm molecular sieves.

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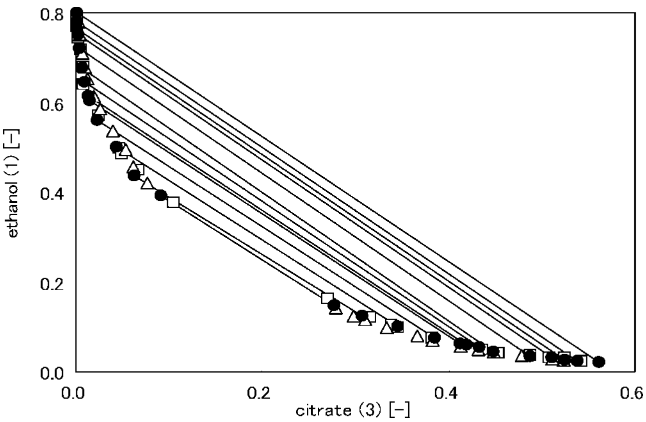

Figure 1. LLE data for the system ethanol (1) + water (2) + potassium citrate (3). □, 288.15 K; ●, 298.15 K; △, 308.15 K; solid tie line, 298.15 K.

Table 1. LLE for the System Ethanol (1) + Water (2) + Potassium Citrate (3)

ethanol-rich phase			citrate-rich phase		
w_1^t	w_2^t	w_3^t	w_1^b	w_2^b	w_3^b
		288.15 K			
0.7845	0.2141	0.0014	0.0241	0.4330	0.5429
0.7680	0.2301	0.0019	0.0315	0.4441	0.5244
0.7435	0.2538	0.0027	0.0302	0.4618	0.5080
0.7166	0.2780	0.0054	0.0370	0.4749	0.4881
0.6778	0.3146	0.0076	0.0402	0.5054	0.4544
0.6405	0.3513	0.0082	0.0494	0.5141	0.4365
0.5709	0.4042	0.0249	0.0757	0.5421	0.3822
0.4986	0.4555	0.0459	0.0992	0.5550	0.3458
0.4847	0.4664	0.0489	0.103	0.558	0.339
0.4485	0.4845	0.0670	0.121	0.563	0.316
0.377	0.518	0.105	0.162	0.567	0.271
		298.15 K			
0.7998	0.1992	0.0010	0.0215	0.4173	0.5612
0.7792	0.2199	0.0009	0.0224	0.4387	0.5389
0.7641	0.2340	0.0019	0.0267	0.4487	0.5246
0.7514	0.2460	0.0026	0.0301	0.4581	0.5118
0.7211	0.2747	0.0042	0.0347	0.4769	0.4884
0.6768	0.3158	0.0074	0.0445	0.5067	0.4488
0.6454	0.3454	0.0092	0.0540	0.5134	0.4326
0.6147	0.3711	0.0142	0.0593	0.5214	0.4193
0.6048	0.3805	0.0147	0.0622	0.5248	0.4130
0.5602	0.4161	0.0237	0.0759	0.5386	0.3855
0.5015	0.4541	0.0444	0.100	0.554	0.346
0.4370	0.4998	0.0632	0.124	0.569	0.307
0.3914	0.5169	0.0917	0.147	0.576	0.277
		308.15 K			
0.8074	0.1909	0.0017	0.0261	0.4510	0.5229
0.7803	0.2173	0.0024	0.0287	0.4600	0.5113
0.7505	0.2454	0.0042	0.0364	0.4859	0.4777
0.7099	0.2830	0.0071	0.0444	0.5077	0.4479
0.6798	0.3113	0.0089	0.0499	0.5182	0.4319
0.6537	0.3344	0.0119	0.0573	0.5295	0.4132
0.6147	0.3663	0.0190	0.0702	0.5475	0.3823
0.5867	0.3879	0.0254	0.0790	0.5550	0.3660
0.5371	0.4239	0.0390	0.0975	0.5687	0.3338
0.4967	0.4493	0.0540	0.117	0.573	0.310
0.4579	0.4809	0.0612	0.125	0.576	0.299
0.4214	0.5018	0.0768	0.143	0.578	0.279

The purities of the ethanol and 1-propanol reagents were checked using a Shimadzu GC-8A gas chromatograph fitted with a thermal conductivity detector and mounted with two stainless steel columns (2 m length × 3 mm I.D. each), with Porapak Q and with PEG 20 M coated on Shin Carbon A. The purities were confirmed as exceeding 99.9 mass % with both the Porapak Q and PEG 20 M columns. Distilled water was used throughout.

Apparatus and Procedure. The experimental method and apparatus were similar to those described by Katayama and Ichikawa[11] and Katayama and Kitagawa.[12] The temperature was measured using an F25 platinum resistance thermometer (supplied by Automatic System Laboratories, Ltd.), with a stated accuracy of ± 0.03 K and a stated resolution of 0.001 K. The temperature fluctuations of the water bath were within ± 0.08 K. An aqueous storage solution saturated with potassium citrate was prepared by dissolving the reagent of tripotassium citrate monohydrate in water. Twenty mixtures of 50 cm^3 total were made by mixing (15 to 30) cm^3 for ethanol, (8 to 22) cm^3 for water, and (10 to 20) cm^3 for the storage solution. Each mixture was poured into six flasks sealed with glass stopcocks, and the flasks were immersed in the bath. The mixtures were agitated for 4 h and then allowed to settle for more than 12 h. Only the LLE data in which both the binodal points and tie line slopes shift regularly on the graphs relating w_1 and w_3 (similar to Figure 1) were selected from the more than 40 data points at each temperature.

First, 1 cm^3 samples were withdrawn from each phase using long-needle syringes and were poured into small bottles which contained 1 cm^3 of 6 wt % 1-propanol aqueous solution. The addition of the solution allowed the samples to be in a homogeneous state. 1-Propanol was used as an internal standard for gas chromatography. The ethanol and water contents were analyzed using the Shimadzu GC-8A gas chromatograph fitted with a thermal conductivity detector and a stainless steel column packed with Porapak Q already described. The gas chromatography unit was cleaned by periodically eliminating contaminants of the citrate from an injection port of the unit and the entrance end of the stainless steel column.

Next, 10 cm^3 and 3 cm^3 amounts of the solutions were withdrawn from the upper and lower layers, respectively, and dried for more than 2 h at a temperature of 463 K in an oven, after which the contents of the citrate were determined gravimetrically on an electronic balance (ER-180A, A & D Company Ltd., Tokyo) with a precision of ± 0.0001 g. The crystals of potassium citrate are known to convert from a monohydrate state to an anhydrous state at a temperature of 453 K, decomposing at 503 K.[13] Phase compositions were obtained, in terms of mass fraction, from combining the gas chromatographic and gravimetric analyses.[4,12] The ethanol and water contents obtained from gas chromatography were measured with a mass fraction precision of 0.0004, and the contents of the citrate from gravimetry were measured with a mass fraction of 0.0001. However, because of the reproducibility of the LLE measurements, the mass fractions of the major components were estimated to be significant to three decimal places, and those of the minor components to four decimal places. The major components for the ethanol-rich phase correspond to ethanol and water, and those for the citrate-rich phase to citrate and water. The minor component of the former phase corresponds to citrate, and that of the latter phase to ethanol.

Results and Discussion

The LLE data selected for the system ethanol (1) + water (2) + potassium citrate (3) are shown in Table 1 and Figure 1, in terms of mass fractions of w_1, w_2, and w_3. Subscripts of 1, 2, and 3 denote the components of ethanol, water, and potassium citrate, respectively, and superscripts t and b denote the top phase (ethanol-rich phase) and the bottom phase (citrate-rich phase), respectively.

Figure 1 shows that the effect of temperature on the LLE data is very small in the temperature range (288.15 to 308.15) K. The binodal curves of the three temperatures are almost equal to one another. To simplify the figure, only tie lines for the

Table 2. Values of Parameters in Equation 1

T/K	a	b	c	d	R^2
288.15	−0.1952	−2.0346	−0.8756	−5.3250	0.9983
298.15	−0.1696	−2.5483	0.2769	−6.3669	0.9995
308.15	−0.1463	−2.2230	−0.8975	−5.2704	0.9998

Table 3. Values of Parameters in Equations 2 and 3

T/K	k_1	n	R^2	k_2	r	R^2
288.15	0.362	1.571	0.998	1.711	0.588	0.998
298.15	0.362	1.561	0.998	1.711	0.590	0.996
308.15	0.292	1.612	0.997	1.907	0.567	0.996

Table 4. Deviations of LLE Correlations of the System Ethanol (1) + Water (2) + Potassium Citrate (3) by Using Equations 1 and 2 and Using Equations 1 and 3

	equations 1 and 2			equations 1 and 3		
T/K	dev(1)	dev(2)	dev(3)	dev(1)	dev(2)	dev(3)
288.15	0.19	0.33	0.29	0.24	0.22	0.33
298.15	0.25	0.34	0.25	0.24	0.30	0.26
308.15	0.24	0.33	0.20	0.22	0.29	0.19
	tavdev[a]	0.27		tavdev[a]	0.26	

[a] The total average deviation, tavdev, is defined by the arithmetic mean values of nine dev(i) described above.

temperature of 298.15 K were drawn. The temperature dependence of the slopes of the tie lines is also very small, but the slopes for the temperature of 288.15 K are somewhat smaller than those of the other two. The slopes of the tie lines decrease slightly with decreasing temperature. The reason for this is explained in the following manner. The decrease of temperature causes a decrease of solubility of the citrate in water. That is, the affinity of the citrate to water is weakened as the temperature is lowered. At the same time, the temperature decrease enhances the intermolecular hydrogen bonding between ethanol and water. At this point, some molecules of water in the lower phase move into the upper phase when the temperature is decreased. The same trend has already been observed for the systems ethanol + water + dipotassium phosphate or sodium dihydrogen phosphate[4] and the systems 1-propanol or 2-propanol + water + dipotassium hydrogen phosphate.[12]

The binodal curves were fitted with the following nonlinear equation:[7]

$$\ln w_1 = a + b w_3^{0.5} + c w_3 + d w_3^2 \tag{1}$$

The coefficients of eq 1 for the system ethanol + water + potassium citrate, along with the coefficient of determination, R^2, of w_1, are given in Table 2. The R^2 values are almost unity, which demonstrates that the experimental values are fitted well with eq 1.

The compositions of the tie lines were correlated with the equations of Othmer−Tobias (eq 2)[14] and of Bancroft (eq 3)[15]

$$\left(\frac{1 - w_1^{\mathrm{t}}}{w_2^{\mathrm{t}}}\right) = k_1 \left(\frac{1 - w_3^{\mathrm{b}}}{w_3^{\mathrm{b}}}\right)^n \tag{2}$$

$$\left(\frac{w_2^{\mathrm{b}}}{w_3^{\mathrm{b}}}\right) = k_2 \left(\frac{w_2^{\mathrm{t}}}{w_1^{\mathrm{t}}}\right)^r \tag{3}$$

where the values k_1 and n are the fit parameters of eq 2, and k_2 and r are the parameters of eq 3. These parameters, along with the R^2 values obtained from the least-squares method, are listed in Table 3. From the values of R^2 (= 0.996 − 0.998), the tie lines of the system ethanol + water + citrate are found to be satisfactorily correlated by both eqs 2 and 3.

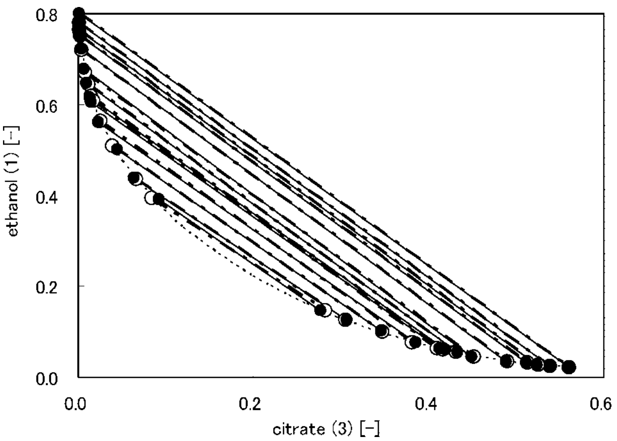

Figure 2. Comparison of experimental and calculated LLE of the system ethanol (1) + water (2) + potassium citrate (3) at 298.15 K. ● and solid tie line, experimental values; ○ and broken tie line, the correlated values from eqs 1 and 3; the dotted binodal curve, the calculated value from eq 1.

Correlations of LLE can be conducted using any two of eqs 1, 2, and 3. The correlated results for the system, using two pairs of three equations, namely, eqs 1 and 2 and eqs 1 and 3, are summarized in Table 4. The compositions used to conduct the correlations are the mean values of the top and bottom experimental compositions. The deviation of calculated and experimental values for the ith component was expressed as a root-mean-square deviation, dev(i), as follows

$$\mathrm{dev}(i) = \frac{\sqrt{\sum_{n=1}^{N} \sum_{k=1}^{2} (w_{i,k,n,\mathrm{calcd}} - w_{i,k,n,\mathrm{exptl}})^2}}{2N} \cdot 100.0 \tag{4}$$

where k is the number of phases (1 to 2); n is the number of LLE data (1 to N = 11 − 13); and calcd and exptl subscripts are calculated and experimental values, respectively.

The dev(i) values from eqs 1 and 2 were almost equal to those from eqs 1 and 3, but the former were slightly inferior to the latter. The total average deviations, tavdev, which are defined as the arithmetic mean values of nine dev(i) (for three components and three temperatures), are (0.27 and 0.26) %, respectively. The correlated results from the third pair of equations (eqs 2 and 3), indicating tavdev = 0.30 %, are inferior to the ones from both of the above pairs. Figure 2 describes a comparison of LLE at 298.15 K between the experimental data and the calculated values from eqs 1 and 3 using the respective parameters listed in Tables 2 and 3.

To our knowledge, seven salts (cesium sulfate,[7] magnesium sulfate,[6] sodium dihydrogen phosphate,[4] dipotassium hydrogen phosphate,[4] potassium carbonate,[8] tripotassium citrate, and tripotassium phosphate) cause an ethanol + water solution to split into two liquid phases. We found that tripotassium phosphate (K_3PO_4) also gives phase separations of both aqueous ethanol and methanol solutions, but no formal experiments have yet been carried out.

Figure 3shows the comparison of binodal curves for one ternary system (ethanol + water + cesium sulfate) at 308.15 K and four ternary systems (ethanol + water + potassium citrate; ethanol + water + sodium dihydrogen phosphate; ethanol + water + dipotassium hydrogen phosphate; ethanol + water + potassium carbonate) at 298.15 K. Because of the complexity of Figure 3, the curve of the system ethanol + water + magnesium sulfate is not described. It partially overlaps the system ethanol + water +

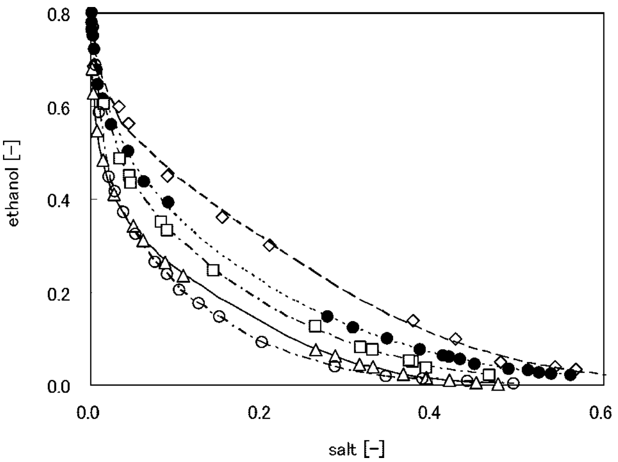

Figure 3. Comparison of LLE of the ternary systems ethanol + water + salt (cesium sulfate, potassium citrate, sodium dihydrogen phosphate, dipotassium hydrogen phosphate, or potassium carbonate). ◇, cesium sulfate (308.15 K) from ref 7; ●, potassium citrate (298.15 K) from this work; □, sodium dihydrogen phosphate (298.15 K) from ref 4; △, dipotassium hydrogen phosphate (298.15 K) from ref 4; ○, potassium carbonate (298.15 K) from ref 8.

Table 5. Solubility Saturated and Crystal State of Seven Salts in Aqueous Solution at 298.2 K

salts	solubility mass fraction of salt in aqueous solution	hydrate number[a]
cesium sulfate[b]	0.645, 0.634 (273.0 K)	0 (anhydrous)
potassium citrate[c]	0.625 (288.2 K)	1
sodium dihydrogen phosphate[b]	0.485	2
dipotassium hydrogen phosphate[b]	0.620	3
potassium carbonate[b]	0.529	3/2
magnesium sulfate[c]	0.640 (313 K)	7
tripotassium phosphate[b]	0.514	7

[a] The hydrate number stands for moles of water per 1 mole of salt (as solid) in equilibrium with saturated water. [b] The values of the substances were cited from ref 5. [c] The values of the substances were cited from ref 13.

dipotassium hydrogen phosphate, but the LLE area of the former is somewhat smaller than that of the latter.

Table 5 lists the solubility and hydrate number of seven salts in an aqueous solution saturated at 298.15 K, with the exception of potassium citrate at 288.2 K and magnesium sulfate at 313 K. All the salts have high solubilities in water. The magnitudes of the LLE areas become greater in the following order: cesium sulfate, tripotassium citrate, sodium dihydrogen phosphate, magnesium sulfate, dipotassium hydrogen phosphate, and potassium carbonate. That is, the cesium sulfate system has the smallest two-phase area of the six systems, and the potassium carbonate system has the largest area of the six. Cesium sulfate has the highest solubility in water among the seven salts, but the crystal of cesium sulfate contacting the saturated water solution has no hydrate. This implies that either cesium sulfate dissolved in water forms no complex with water or the bonding power of the complex is weaker than that of other salts, even if the sulfate forms a complex with water. The order of the hydrate number of salts in the crystal state agrees with the magnitude of the two-phase area, except for potassium carbonate. Combining the solubility of the salt in water and the hydrate number of the salt (solid) serves as an indicator to deduce the magnitude of the two-phase area. Because of the high solubility of the salts in water, it was assumed that the molecules of the salts splitting an aqueous ethanol solution into two phases have not dissociated into ions in the salt-rich phase, but have formed complexes with water.

Conclusions

Liquid−liquid equilibria of the system ethanol + water + potassium citrate were studied at temperatures of (288.15, 298.15, and 308.15) K. It was found that the dependence of temperature on the LLE data is very small in the temperature range studied, but slopes of the tie lines slightly decrease with decreasing temperature. The binodal curves of the system were fitted using a nonlinear equation with four parameters, and the tie lines were satisfactorily described with the Othmer−Tobias and Bancroft equations. LLE calculations could be successfully carried out using any two of three equations (the nonlinear equation, the Othmer−Tobias, and Bancroft equations).

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Supporting Information Available:

The experimental and correlated results for the manuscript are provided in the terms of the Excel spreadsheet format. This material is available free of charge via the Internet at http://pubs.acs.org.

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