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Phase Diagrams of Some Aliphatic Alcohols + Potassium or Sodium Citrate + Water at 25 °C

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Phase diagrams for some alcohols + potassium or sodium citrate + water systems were measured at 25 °C. The alcohols used are ethanol, 1-propanol, 2-propanol, 2-butanol, and 2-methyl-2-propanol. For these systems, binodal and tie line data were determined at 25 °C. Binodal curves were described using a three-parameter equation. Tie lines were satisfactorily described using the Othmer and Tobias equations.

Introduction

Aqueous two-phase systems (ATPS) formed by mixtures of two polymers or of one polymer and one inorganic salt are important for the separation and purification of biological materials such as enzymes and proteins.[1,2] The salts used are usually phosphates, sulfates, and citrates. In recent years, many research groups have focused on the measurement of new phase equilibrium data for aqueous polymer + salt systems.[3−8] Recently, Zafarani-Moattar and co-workers have reported liquid−liquid equilibrium (LLE) data for aqueous poly(ethylene glycol) (PEG) + ammonium dihydrogen phosphate or diammonium hydrogen phosphate[9] and PEG + potassium or sodium citrate systems.[10,11]

For large-scale processes, methods for recycling chemicals have been developed.[12,13] Greve and Kula[13] have described the use of some two-phase systems composed of lower aliphatic alcohols + water + inorganic salts for the extraction of salt from the primary bottom phase of protein extraction processes in aqueous PEG + salt systems. In this respect, Zafarani-Moattar and co-workers have reported LLE data for some aliphatic alcohols + magnesium sulfate + water[14] and aliphatic alcohols + ammonium dihydrogen phosphate or diammonium hydrogen phosphate + water systems.[15]

In this work, we have studied the phase diagrams of alcohols + sodium or potassium citrate + water systems at 25 °C. The alcohols used are ethanol, 1-propanol, 2-propanol, 2-butanol, and 2-methyl-2-propanol. As far as we know, there is no report on the phase diagrams of the aforementioned systems. There are only a few binodal data for the ethanol + sodium citrate + water system at pH 7.5 and 25 °C.[16]

In the following text, we describe the phase diagrams of these systems in some detail. These results can be used to develop thermodynamic models of aqueous two-phase systems.[17−19]

Experimental Section

Materials. The alcohols were obtained from Merck with the listed mass fractions (ethanol GR, 0.995; 1-propanol GR, 0.995; 2-propanol GR, 0.997; 2-butanol GR, 0.99; 2-methyl-2-propanol GR, 0.995). Sodium and potassium citrate with a minimum mass fraction of 0.99 were obtained from Fluka. All of the alcohols and salts were used without further purification, and double-distilled deionized water was used. From experimental refractive index measurement at 25 °C, we obtained values of 1.3594, 1.3836, 1.3753, 1.3954, and 1.3852 for ethanol, 1-propanol, 2-propanol, 2-butanol, and 2-methyl-2-propanol, respectively. These values compare well with the literature values[20] (1.35941, 1.38370, 1.3752, 1.3950, and 1.3851 for ethanol, 1-propanol, 2-propanol, 2-butanol, and 2-methyl-2-propanol, respectively).

Apparatus and Procedure. The experimental apparatus employed is essentially similar to the one used previously.[14] A glass vessel, volume 50 cm^3, was used to carry out the phase equilibrium determinations. The glass vessel was provided with an external jacket in which water at constant temperature was circulated using a thermostat. The temperature was controlled to within ±0.05 °C. The binodal curves were determined by a titration method. A salt solution of known concentration was titrated with the alcohol or vice versa until the solution turned turbid. The composition of the mixture was determined by mass using an analytical balance with an uncertainty of $\pm 1 \times 10^{-7}$ kg. For the determination of the tie lines, feed samples (about 20 cm^3) were prepared by mixing appropriate amounts of alcohol, salt, and water in the vessel. The thermostat was set at a desired temperature, and the sample was stirred for 1 h. Then the mixture was allowed to settle for 24 h. After the separation of the two phases, the concentration of sodium or potassium citrate in the top and bottom phases was determined by flame photometry (Jenway model PFP7, U.K.). The uncertainty in the measurements of the mass fraction of the salt was estimated to be ±0.0002. The concentration of alcohol in both phases was determined by refractive index measurements performed at 25 °C using a refractometer (Quartz RS-232, Ceti, Belgium). The uncertainty in the measurement of the refractive index was found to be ±0.0001. For dilute aqueous solutions containing an alcohol and a salt, the relation between the refractive index, n_D, and the mass fractions of alcohol, w_1, and salt, w_2, is given by

$$n_D = a_0 + a_1 w_1 + a_2 w_2 \tag{1}$$

This equation has been used for the phase analysis of the

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Table 1. Coefficients of Equation 1

system	a_0	a_1	a_2
ethanol + potassium citrate + H_2O	1.3325	0.0699	0.1537
1-propanol + potassium citrate + H_2O	1.3325	0.0947	0.1537
2-propanol + potassium citrate + H_2O	1.3325	0.0934	0.1537
2-butanol + potassium citrate + H_2O	1.3325	0.1072	0.1537
2-methyl-2-propanol + potassium citrate + H_2O	1.3325	0.1058	0.1537
ethanol + sodium citrate + H_2O	1.3325	0.0699	0.1807
1-propanol + sodium citrate + H_2O	1.3325	0.0947	0.1807
2-propanol + sodium citrate + H_2O	1.3325	0.0934	0.1807
2-butanol + sodium citrate + H_2O	1.3325	0.1072	0.1807
2-methyl-2-propanol + sodium citrate + H_2O	1.3325	0.1058	0.1807

poly(ethylene glycol) + NaCl + H_2O system by Cheluget et al.[21] We found that the same method of calibration plots and evaluation of parameters given by Cheluget et al.[21] can also be used for the analysis of alcohols + potassium or sodium citrate + water systems. However, it was found that this equation is valid only for mass fractions of $w_1 \leq 0.1$ and $w_2 \leq 0.05$, for which a linear calibration plot of the refractive index of the solution against alcohol and salt are obtained. Therefore, before refractive index measurements, it was necessary to dilute the samples to the above mass fraction range. The values of coefficients a_0, a_1, and a_2 for the studied systems are listed in Table 1. In the determination of alcohol composition by the above procedure, the uncertainty in the mass fraction was ±0.002. The uncertainty in the temperature was ±0.05 K.

Results and Discussion

For ternary aqueous sodium and potassium citrate systems containing different alcohols, the binodal data at 25 °C are given respectively in Tables 2 and 3. The tie line data for these ternary systems containing sodium and potassium salts are given at 25 °C in Tables 4 and 5, respectively. From the comparison of our binodal data for ethanol + sodium citrate + water with those given by Greve and Kula[16] at pH 7.5 and 25 °C, we find that whereas there is fairly good agreement at low salt concentration a noticeable difference exists at higher salt concentrations. This may be due to the different experimental conditions (pH 7.5) that were imposed by Greve and Kula.[16]

The binodal curves were fitted using the following nonlinear expression of Mistry et al.:[22]

$$\ln w_1 = a + b w_2^{0.5} + c w_2^{3} \tag{2}$$

Recently, this empirical equation has been used for the correlation of binodal curves of the systems PEG + Na_2CO_3 + H_2O,[23] 1-propanol + ammonium dihydrogen phosphate or diammonium hydrogen phosphate + water, and 2-propanol + ammonium dihydrogen phosphate or diam-

Table 2. Binodal Data as Mass Fraction for the Alcohols (1) + Sodium Citrate (2) + H_2O (3) System at 25 °C

$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$
Ethanol + Sodium Citrate + H_2O											
26.95	14.10	28.90	12.38	31.17	10.73	36.14	7.49	42.74	4.25	47.17	2.86
27.40	13.48	29.18	12.20	32.93	9.49	36.73	7.18	44.40	3.66	49.11	2.33
27.71	13.35	30.67	11.03	34.04	8.87	39.36	5.83	45.81	3.13	51.11	1.92
1-Propanol + Sodium Citrate + H_2O											
14.49	9.55	19.76	7.27	23.36	6.23	26.34	5.50	30.71	4.45	36.47	3.45
17.47	8.08	21.55	6.76	23.98	6.07	29.34	4.87	33.69	3.97	40.13	2.77
18.26	7.53	22.26	6.52	25.65	5.60						
2-Propanol + Sodium Citrate + H_2O											
13.19	16.79	21.57	11.34	31.30	6.55	35.68	4.8	39.95	3.5	44.06	2.44
14.68	15.65	25.36	9.40	32.95	5.89	37.57	4.24	42.44	2.86	45.38	2.19
17.83	13.47	29.04	7.62								
2-Butanol + Sodium Citrate + H_2O											
4.56	13.04	8.74	6.59	10.16	4.78	12.09	2.97	13.23	2.01	14.27	1.40
7.08	8.58	9.98	5.31	11.39	3.72	12.56	2.69	13.83	1.46	15.00	0.93
8.26	6.91										
2-Methyl-2-Propanol + Sodium Citrate + H_2O											
13.6	9.53	16.83	7.56	21.12	6.15	26.86	4.63	35.27	2.98	42.82	1.8
14.6	8.87	17.65	7.33	23.67	5.44	30.08	3.84	39.57	2.28	45.86	1.41
14.78	8.79	19.98	6.45	23.8	5.17	32.14	3.39				

Table 3. Binodal Data as Mass Fraction for the Alcohols (1) + Potassium Citrate (2) + H_2O (3) System at 25 °C

$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$
Ethanol + Potassium Citrate + H_2O											
33.44	12.62	34.96	11.59	37.77	9.75	43.59	6.51	47.97	4.53	50.72	3.33
33.87	12.27	36.36	10.63	40.68	8.01	46.11	5.31	49.59	3.87	53.00	2.75
1-Propanol + Potassium Citrate + H_2O											
15.48	10.09	24.39	6.88	36.80	4.02	43.94	2.64	47.70	2.10	49.61	1.79
19.06	8.62	28.84	5.73	39.13	3.53	46.06	2.28	48.34	1.95	51.36	1.56
21.15	7.87	32.49	4.96	42.15	2.96						
2-Propanol + Potassium Citrate + H_2O											
20.32	13.69	30.00	8.85	39.59	4.94	45.51	3.29	50.35	2.10	52.38	1.74
24.27	11.62	34.73	6.80	42.49	4.10	47.79	2.67	51.63	1.92	52.74	1.54
2-Butanol + Potassium Citrate + H_2O											
4.40	15.32	8.62	8.04	10.59	5.14	11.75	3.99	13.37	2.35	15.22	1.35
5.84	12.77	9.85	6.31	11.39	4.35	13.09	2.66	14.56	1.69	17.03	0.46
7.39	10.06										
2-Methyl-2-propanol + Potassium Citrate + H_2O											
14.92	9.55	23.70	6.01	34.20	3.75	40.8	2.57	44.64	1.96	47.65	1.53
18.81	7.62	29.28	4.78	37.94	3.03	42.55	2.27	46.20	1.74		

Table 4. Tie Line Data as Mass Fraction for the Alcohols (1) + Sodium Citrate (2) + H_2O (3) System at 25 °C

alcohol-rich phase				water-rich phase			
$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$
1-Propanol + Sodium Citrate + H_2O							
59.43	0.66	53.93	1.07	6.55	16.42	9.31	13.37
56.74	0.92	47.42	1.96	7.98	14.78	13.31	11.96
2-Propanol + Sodium Citrate + H_2O							
52.94	1.22	34.75	5.09	3.29	26.88	12.65	16.54
48.66	1.73	30.09	6.50	4.11	25.36	15.08	14.86
43.66	2.87			6.13	22.20		
2-Butanol + Sodium Citrate + H_2O							
74.15	0.005	69.08	0.03	10.58	1.61	12.87	3.36
73.67	0.004	68.51	0.03	11.90	2.28	13.88	4.39
69.54	0.03			12.04	2.93		
2-Methyl-2-propanol + Sodium Citrate + H_2O							
47.50	1.53	58.08	0.52	19.47	6.90	10.73	11.35
55.66	0.67	60.55	0.62	12.71	10.08	7.37	13.51

Table 5. Tie Line Data as Mass Fraction for the Alcohols (1) + Potassium Citrate (2) + H_2O (3) System at 25 °C

alcohol-rich phase				water-rich phase			
$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$
Ethanol + Potassium Citrate + H_2O							
64.67	1.04	48.42	4.39	5.36	44.05	9.48	34.44
55.48	2.68	44.54	5.89	7.49	38.01	12.49	30.88
1-Propanol + Potassium Citrate + H_2O							
59.50	0.85	53.63	1.57	9.27	14.48	15.37	10.08
56.42	1.24	44.79	2.77	12.15	12.88	21.42	7.82
2-Propanol + Potassium Citrate + H_2O							
65.85	0.87	55.06	2.22	9.39	19.94	12.85	15.68
62.69	1.20	46.16	3.62	10.12	18.71	24.00	10.31
60.07	1.43			11.13	17.36		
2-Butanol + Potassium Citrate + H_2O							
49.39	0.04	48.19	0.02	11.63	4.06	14.72	1.58
49.13	0.03	47.36	0.02	12.78	3.06	15.79	0.76
48.64	0.03			12.69	2.65		
2-Methyl-2-Propanol + Potassium Citrate + H_2O							
62.90	0.32	54.88	0.94	9.76	14.17	15.27	9.43
57.68	0.62	50.75	1.38	12.73	11.21	19.96	7.54

Table 6. Coefficients of Equation 2

system	a	b	c	sd[a]
ethanol + potassium citrate + H_2O	−0.28	−2.12	−30.49	0.15
1-propanol + potassium citrate + H_2O	−0.08	−4.59	−342.80	0.44
2-propanol + potassium citrate + H_2O	−0.26	−2.93	−97.80	0.29
2-butanol + potassium citrate + H_2O	−1.56	−2.92	−118.17	0.12
2-methyl-2-propanol + potassium citrate + H_2O	−0.06	−5.27	−264.11	0.66
ethanol + sodium citrate + H_2O	−0.34	−2.46	−20.21	0.16
1-propanol + sodium citrate + H_2O	0.18	−6.40	−186.68	0.46
2-propanol + sodium citrate + H_2O	−0.30	−3.30	−81.80	0.16
2-butanol + sodium citrate + H_2O	−1.57	−3.20	−168.19	0.20
2-methyl-2-propanol + sodium citrate + H_2O	−0.04	−5.96	−160.02	0.58

[a] sd = $[(\sum_i(100w_1^{\text{calcd}} - 100w_1^{\text{exptl}})^2)/N]^{0.5}$, where N is the number of binodal data.

monium hydrogen phosphate + water.[15] The coefficients of eq 2 along with the corresponding standard deviations for the investigated systems are given in Table 6. On the basis of obtained standard deviations, we conclude that eq 2 can be satisfactorily used to correlate the binodal curves of the investigated systems. The experimental and calculated binodal curves for the studied systems containing sodium or potassium citrate are shown respectively in Figures 1 and 2.

The solubility of an aliphatic alcohol in water and the mutual miscibility depend on the chain length and decrease with an increasing number of carbon atoms in the chain.

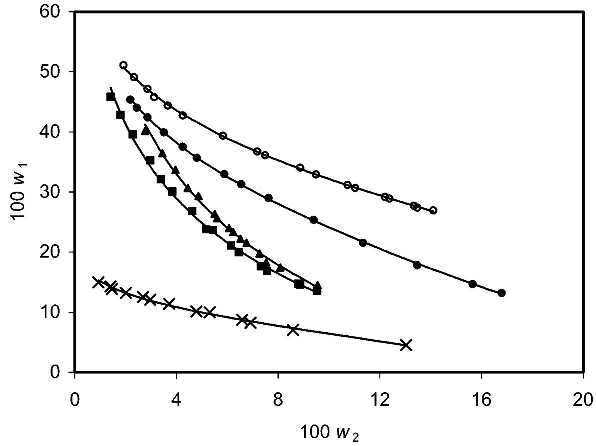

Figure 1. Comparison of binodal curves for the alcohols (1) + sodium citrate (2) + H_2O (3) systems at 25 °C: ○, ethanol; ●, 2-propanol; ▲, 1-propanol; ×, 2-butanol; ■, 2-methyl-2-propanol; −, calculated from eq 2.

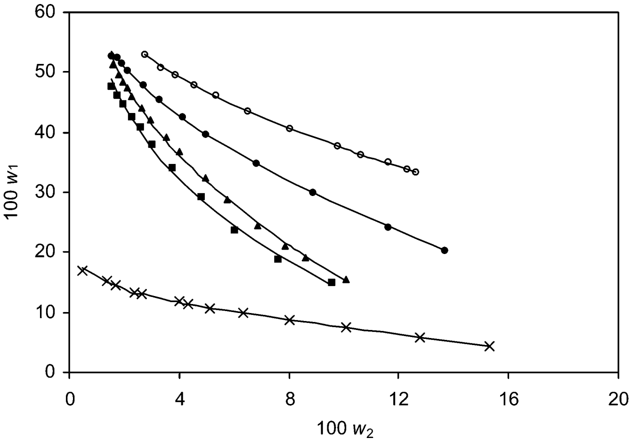

Figure 2. Comparison of binodal curves for the alcohols (1) + potassium citrate (2) + H_2O (3) systems at 25 °C: ○, ethanol; ●, 2-propanol; ▲, 1-propanol; ×, 2-butanol; ■, 2-methyl-2-propanol; −, calculated from eq 2.

At room temperature, 2-butanol is not completely miscible with water. For both the aqueous sodium and potassium citrate + alcohol ternary mixtures, the effect of the higher polarity of 2-propanol as compared with that of 1-propanol is visible from the shift in the binodals as shown in Figures 1 and 2. Greve and Kula[16] have found the same trend for the binodals of the systems 1-propanol + potassium phosphate + water and 2-propanol + potassium phosphate + water.

Similarly, in comparison with the ternary solutions containing 2-butanol, in the phase diagrams for aqueous 2-methyl-2-propanol + sodium or potassium citrate + water systems the binodals are displaced (Figures 1 and 2), indicating the higher polarity of 2-methyl-2-propanol, which leads to improved miscibility and solubility of water and sodium or potassium citrate. As expected, the binodal is displaced even further in the ternary system ethanol + sodium or potassium citrate compared with the corresponding aqueous sodium or potassium citrate solutions containing other alcohols (Figures 1 and 2). However, in contrast to ethanol + potassium citrate + water, which forms two liquid phases, the ternary ethanol + sodium citrate + water system is completely different in that only a solubility limit is observed and the solution is in equilibrium with a solid phase of sodium citrate. This behavior was also reported for the system ethanol + sodium citrate + water at pH 7.5 and 25 °C by Greve and Kula.[16] In the

Table 7. Values of the Parameters of Equation 3

system	k	n	k_1	r	sd_1[a]	sd_2[a]
ethanol + potassium citrate + H_2O	0.403	1.427	1.693	0.616	0.81	0.64
1-propanol + potassium citrate + H_2O	0.164	0.811	7.777	1.026	0.66	0.35
2-propanol + potassium citrate + H_2O	0.276	1.078	3.082	0.779	0.82	0.51
2-butanol + potassium citrate + H_2O	0.885	0.047	11.440	21.422	1.31	0.08
2-methyl-2-propanol + potassium citrate + H_2O	0.173	0.688	10.342	1.241	0.53	0.22
1-propanol + sodium citrate + H_2O	0.080	1.176	6.396	0.711	0.39	0.25
2-propanol + sodium citrate + H_2O	0.294	1.159	2.811	0.740	0.72	0.47
2-methyl-2-propanol + sodium citrate + H_2O	0.160	0.740	9.901	1.119	0.56	0.22

[a] $sd_j = \{{}^1\!/_2\sum_{Li=1}^{L} [(100w_{i,j,\mathrm{calcd}}^{t} - 100w_{i,j,\mathrm{exptl}}^{t})^2 + (100w_{i,j,\mathrm{calcd}}^{b} - 100w_{i,j,\mathrm{exptl}}^{b})^2]\}^{0.5}$, where L is the number of tie lines and $j = 1$ and 2. sd_1 and sd_2 represent the mass % standard deviations for alcohol and salt, respectively.

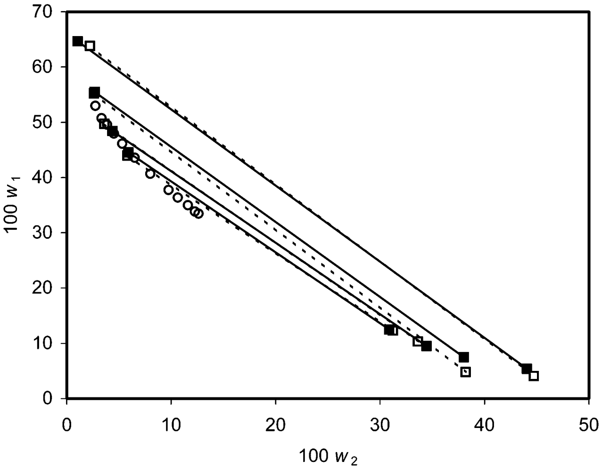

Figure 3. Tie lines for ethanol (1) + potassium citrate (2) + H_2O (3) at 25 °C: -■-, experimental; - -□- -, calculated from eq 3; ○, binodal.

case of the ethanol + potassium citrate + water system, however, we observe an aqueous two-liquid-phase system containing an appreciable amount of potassium citrate in the ethanol-rich phase (Table 5).

The correlation equations[24] given by Othmer and Tobias (eqs 3a and 3b) have been used to correlate the tie line compositions

$$\left(\frac{1 - w_1^{\mathrm{t}}}{w_1^{\mathrm{t}}}\right) = k\left(\frac{1 - w_2^{\mathrm{b}}}{w_2^{\mathrm{b}}}\right)^n \tag{3a}$$

$$\left(\frac{w_3^{\mathrm{b}}}{w_2^{\mathrm{b}}}\right) = k_1\left(\frac{w_3^{\mathrm{t}}}{w_1^{\mathrm{t}}}\right)^r \tag{3b}$$

where k, n, k_1, and r represent fit parameters. Superscripts t and b stand for the alcohol rich-phase and water-rich phase, respectively. The values of the parameters are given in Table 7. Recently, eqs 3a and 3b have been used for the assessment and correlation of tie line compositions of the PEG + $NaNO_3$ + H_2O[3], PEG + Na_2CO_3 + H_2O,[23] PEG + potassium citrate + H_2O,[10] and aliphatic alcohols + ammonium dihydrogen phosphate or diammonium hydrogen phosphate + water[15] systems.

On the basis of standard deviations sd_1 and sd_2 given in Table 7, we conclude that eq 3 can be satisfactorily used to correlate the tie line data of the investigated systems.

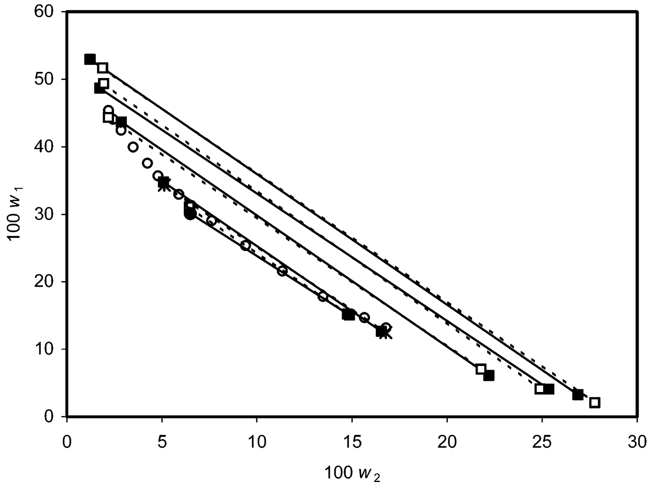

Figure 4. Tie lines for 2-propanol (1) + sodium citrate (2) + H_2O (3) at 25 °C: -■-,experimental; - -□- -, calculated from eq 3; ○, binodal.

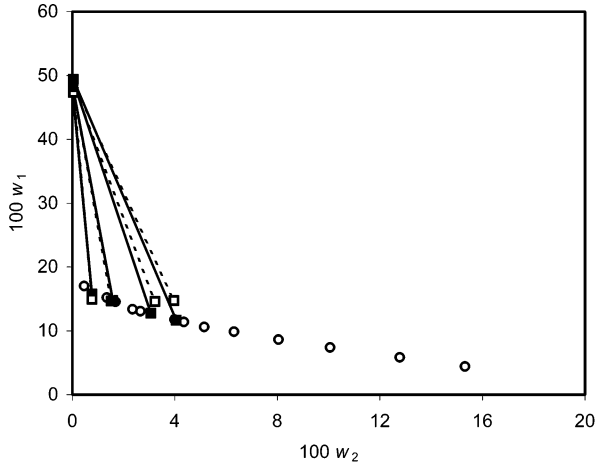

Figure 5. Tie lines for 2-butanol (1) + potassium citrate (2) + H_2O (3) at 25 °C: -■-, experimental; - -□- -, calculated from eq 3; ○, binodal.

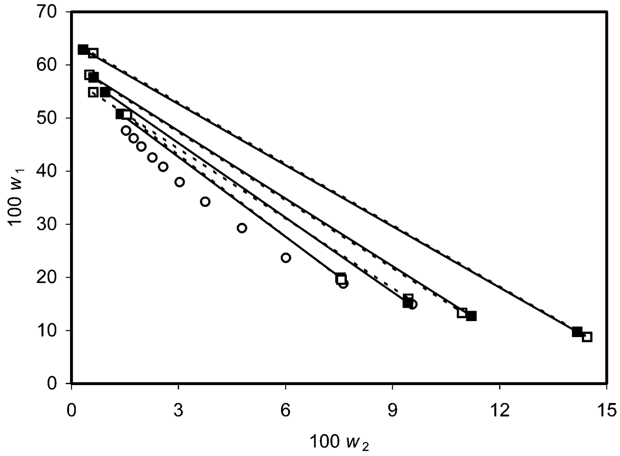

Figure 6. Tie lines for 2-methyl-2-propanol (1) + potassium citrate (2) + H_2O (3) at 25 °C: -■-, experimental; - -□- -, calculated from eq 3; ○, binodal.

In the case of the aqueous 2-butanol + sodium citrate system, however, a relatively higher mass % standard deviation for 2-butanol, $sd_1 = 1.31$, was obtained. To show the reliability of the model, comparison between the experimental and correlated tie lines are shown in Figures 3 to 6 for selected systems, as examples.

Table 8. Plait Points at 25 °C

system	$100w_1$	$100w_2$
ethanol + potassium citrate + H_2O	27.76	18.00
1-propanol + potassium citrate + H_2O	33.71	4.73
2-propanol + potassium citrate + H_2O	27.02	10.46
2-methyl-2-propanol + potassium citrate + H_2O	35.12	3.59
1-propanol + sodium citrate + H_2O	28.79	5.04
2-propanol + sodium citrate + H_2O	23.21	10.61
2-methyl-2-propanol + sodium citrate + H_2O	33.81	3.27

The location of the plait points for these systems was estimated by extrapolation from the midpoints of the different tie lines, and the values are listed in Table 8. In the case of the 2-butanol + potassium citrate system, the plait point cannot be given because there is no complete binodal curve because of the fact that at room temperature 2-butanol is not completely miscible with water. Also, for the 2-butanol + sodium citrate + water system, because only a solubility limit is observed and the solution is in equilibrium with a solid phase of sodium citrate, there is no plait point.

Conclusions

Phase diagrams of ternary alcohols + sodium or potassium citrate + water systems, with the alcohols being ethanol, 1-propanol, 2-propanol, 2-butanol, and 2-methyl-2-propanol, have been studied at 25 °C. These ternary systems form two-phase systems at 25 °C, for which the binodals and tie lines were obtained. The tie lines were satisfactorily correlated with Othmer and Tobias equations. For the system ethanol + sodium citrate + water, however, only a solubility limit was observed, and the solution is in equilibrium with a solid phase of sodium citrate.

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