Liquid−Liquid Equilibria for Aliphatic Alcohols + Dipotassium Oxalate + Water

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Liquid−liquid equilibria (LLE) in ternary mixtures of water + dipotassium oxalate + alcohol (2-propanol, 1-propanol, 2-methyl-2-propanol, and 2-butanol) have been studied at 298.15 K and atmospheric pressure. The experimental binodal and tie line data were determined at 298.15 K. The binodal curves were correlated using a three-parameter equation, and tie lines were satisfactorily correlated using the Othmer−Tobias, Bancroft, and modified Setschenow equations. The phase separation abilities of the investigated alcohols are in the order of 2-butanol > 2-methyl-2-propanol > 1-propanol > 2-propanol. The effect of temperature on the cloud points as a function of alcohol mole fraction has been studied at temperature range (293.15 to 328.15) K at 5 K intervals. The results show that the lower alcohol concentration is required for phase separation at higher temperatures.

Introduction

When one short-chain alcohol and one salt are dissolved in water above a critical concentration, two immiscible phases are usually formed[1,2] which is referred to as aqueous biphasic systems (ABSs). These types of systems have shown great potential for efficient extraction and purification of biological products because of biocompatibility and low cost. Greve and Kula have described the use of some aqueous biphasic systems containing short-chain alcohols + water + inorganic salts for the extraction of salt from the primary bottom phase of protein extraction processes in polymer + salt systems.[3]

Liquid−liquid equilibria (LLE) in ABSs are due to intermolecular forces, predominantly hydrogen bonding and ion−dipole interactions. Addition of a salt to such a system introduces ion−dipole interactions which alter the structure of the hydrophilic solvents such as alcohols in equilibrium. The molecules of water that surround the ions become unavailable for the hydrophilic solvents, and it becomes “salted out” from the aqueous phase. The opposite process can occur if a polar organic solvent is added to an aqueous salt solution. It captures part of the molecules of water that have been solvating the ions; hence, the salt crystallizes. This effect is used in a process called extractive crystallization in which salts can be recovered from concentrated aqueous solutions by the addition of a polar organic solvent, rather than by the more expensive evaporation process. On the other hand, the salting out effect is utilized for removing organic compounds from water.[4]

Liquid−liquid equilibria in aqueous biphasic systems containing aliphatic alcohols + potassium salts + water have been investigated by several workers. For example, the liquid−liquid equilibria of aqueous 2,3-butanediol−potassium chloride,[5] 1-butanol−potassium iodide,[4] aliphatic alcohols−potassium salts,[6] aliphatic alcohols−potassium citrate,[7] aliphatic alcohols−dipotassium hydrogen phosphate,[8,9] and aliphatic alcohols−potassium carbonate[10,11] systems have been reported. The liquid−liquid equilibria of polymer−salt aqueous two-phase systems have been extensively studied.[12−15] Regarding the polymer−potassium oxalate aqueous two-phase systems, there are only LLE data for the polyvinylpyrrolidone + dipotassium oxalate + water system at different temperatures.[15] As far as we know, there is no report on the LLE data for aliphatic alcohols and aqueous solution of dipotassium oxalate salt. The advantage of using the oxalate anion in aqueous biphasic systems is that it is biodegradable and therefore can be discharged into biological wastewater plants in the separation of biological materials. Due to the salting out effect, adding dipotassium oxalate into the alcohol−water system leads to an organic-rich phase with negligible salt and a salt-rich phase with negligible alcohol, so a large amount of water can be separated from alcohols by simple and efficient phase separation.

In this work, LLE data for the ternary systems 2-propanol/1-propanol/2-methyl-2-propanol/2-butanol + dipotassium oxalate + water at 298.15 K have been reported. The experimental LLE data were correlated with the Othmer−Tobias,[16] Bancroft,[17] and modified Setschenow equations.[18] The data obtained from these and further similar experiments will serve as a base for mathematical modeling of the salt effect on liquid−liquid equilibria.

Experimental Section

Chemicals. The chemicals used in this work were 1-propanol (GR, 99.5 % in mass fraction) and 2-propanol (GR, 99.7 % in mass fraction) from Rankem (India), 2-butanol (GR, >99 % in mass fraction) and 2-methyl-2-propanol (GR, >99 % in mass fraction) from Lobachemie (India), and dipotassium oxalate monohydrate (GR, >99.5 % in mass fraction) from Merck. These chemicals were used without further purification. The doubly distilled deionized water with specific conductance about 0.7 0 $\mu S \cdot cm^{-1}$ was used in all experiments.

Apparatus and Procedure. A double-wall glass vessel was used to carry out the binodal curve measurements on a magnetic stirrer plate. The cell temperature was controlled at constant temperature with circulation of water using a thermostat (JULABO model ED, Germany) with an accuracy of ± 0.03 K. The binodal curves were determined by the cloud point

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Table 1. The Coefficients of Equation 1 for the Investigated Systems at 298.15 K

system	a_0	a_1	a_2
2-propanol + dipotassium oxalate + water	1.3325 ± 0.0001	0.0866 ± 0.0021	0.1353 ± 0.0050
1-propanol + dipotassium oxalate + water	1.3325 ± 0.0001	0.0905 ± 0.0020	0.1347 ± 0.0047
2-methyl-2-propanol + dipotassium oxalate + water	1.3325 ± 0.0001	0.1062 ± 0.0014	0.1314 ± 0.0039
2 -butanol + dipotassium oxalate + water	1.3325 ± 0.0001	0.1059 ± 0.0025	0.1435 ± 0.0060

method. A known concentration of aqueous dipotassium oxalate solution was titrated with alcohol until the solution was turned turbid, which indicated the formation of a two-liquid phase, following which the solution was back-titrated by adding water until the turbidity vanished. The composition before the addition of the last drop before clearing was a point on the binodal curve. The composition of the mixture was measured by mass using an analytical balance (Sartorius model TE214S, Switzerland) with a precision of $\pm 1 \cdot 10^{-7}$ kg.

For determination of the tie-line systems studied, feed samples (less than 10 cm^3) were prepared by mixing appropriate amounts of alcohol, salt, and water in the glass cell. Once the samples were shaken vigorously (2400 $\text{cycles} \cdot \text{min}^{-1}$) for 3 min using a shaker (Labtron model LS-100, Iran), the glass cells were placed in a water bath thermostatted at 298.15 K for (6 to 8) h. The samples were shaken thoroughly once an hour after being immersed in the bath. Longer shaking and settling periods did not result in any sensible change in the phase compositions. After the necessary rest time, the mixture was split into clear and transparent liquid phases with a well-defined interface, and equilibrium state was achieved. The samples were carefully withdrawn using long needle syringes. Then both phases were diluted with doubly distilled water by a mass factor of about 1 to 10 for refractive index measurements. The top phase was sampled first, with care being taken to leave a layer of material at least 0.5 cm thick above the interface. For the withdrawn bottom phase, a tiny bubble of air was retained at the needle tip and expelled in the bottom phase to prevent contamination from upper phase material. After the separation of the two phases, the concentration of potassium oxalate 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 percent of the salt was estimated to be better than ± 0.3 %, and the accuracy of the chemical analysis has been confirmed by analyzing mixtures via gravimetric preparation. The concentration of alcohol in both phases was determined by refractive index measurements performed at 298.15 K using a refractometer (Atago, model DR-A1, Japan). The uncertainty in the measurement of the refractive index was found to be ± 0.0002. The mass fraction of alcohol in two phases in the dilute region at equilibrium was calculated from refractive index measurements by using the following equation

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

where n_D is refractive index and w_1 and w_2 are the mass fractions of alcohol and dipotassium oxalate salt, respectively.

It was found that eq 1 is valid only for mass fraction 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 concentration is 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. Data used for calibration of refractive indices in terms of alcohol and salt mass fractions for the determination of phase compositions are given in the Supporting Information tables. In the determination of alcohol composition by the above procedure, the uncertainty in the mass fraction was better than ± 0.002. For the effects of temperature on the cloud point, a known concentration of aqueous dipotassium oxalate solution was titrated with alcohol, until the solution turned turbid, and then temperature was decreased at 5 K intervals until the turbidity vanished and the addition of alcohol repeated until the solution turned turbid again. In this work, the relative mole fraction of two components remained constant, and the mole fraction of the third component (alcohol) was changed.

Results and Discussion

The experimental data for the binodal curves of 2-propanol/1-propanol/2-methyl-2-propanol/2-butanol + dipotassium ox-

Table 2. Binodal Curve Data for Alcohol (1) + Dipotassium Oxalate (2) + Water (3) Systems as a Function of Mass Fractions at 298.15 K

100 w_1	100 w_2	100 w_1	100 w_2	100 w_1	100 w_2	100 w_1	100 w_2	100 w_1	100 w_2	100 w_1	100 w_2
2-Propanol + Dipotassium Oxalate + Water											
2.019	24.306	9.694	16.831	17.702	11.743	24.301	8.580	32.740	5.421	39.205	3.441
3.688	22.457	11.707	15.321	20.374	10.340	27.594	7.392	36.145	4.456	41.274	3.017
5.634	20.498	13.919	13.752	21.958	9.593	30.379	6.259	37.185	4.077	42.129	2.914
7.222	18.822	16.104	12.537								
1-Propanol + Dipotassium Oxalate + Water											
4.918	20.588	10.208	12.412	15.978	8.55	23.587	5.935	32.063	4.229	39.960	2.981
5.826	18.329	11.105	11.696	16.701	8.132	25.490	5.498	33.565	3.980	40.571	2.846
6.735	17.043	11.976	10.910	17.960	7.589	27.108	5.150	35.304	3.663	41.731	2.656
7.852	15.269	13.551	9.722	19.711	6.972	29.256	4.748	36.857	3.373	43.999	2.208
8.787	14.244	14.767	9.034	21.934	6.351	30.663	4.487	39.165	3.094	47.536	1.976
2-Methyl-2-propanol + Dipotassium Oxalate + Water											
3.479	20.779	10.35	11.578	13.839	8.802	19.648	6.071	25.055	4.654	31.594	3.502
4.567	19.434	10.994	11.055	14.762	8.254	20.442	5.829	26.619	4.362	32.858	3.300
6.335	16.443	11.575	10.396	15.972	7.520	21.511	5.501	27.467	4.201	34.517	3.108
8.933	13.443	12.21	9.858	17.603	6.844	22.566	5.248	28.447	4.020	37.673	2.839
9.567	12.681	12.713	9.541	18.735	6.410	23.432	5.020	30.209	3.716		
2-Butanol + Dipotassium Oxalate + Water											
2.004	23.938	6.339	10.010	7.572	7.776	8.887	5.811	10.229	4.165	11.752	2.770
3.601	15.790	6.664	9.333	7.917	7.168	9.366	5.432	10.550	3.947	12.216	2.369
4.969	12.486	7.025	8.542	8.193	6.663	9.621	5.017	10.844	3.674	12.451	2.232
5.560	11.431	7.333	8.107	8.498	6.233	10.108	4.558	11.416	2.959	12.866	2.023
5.938	10.709										

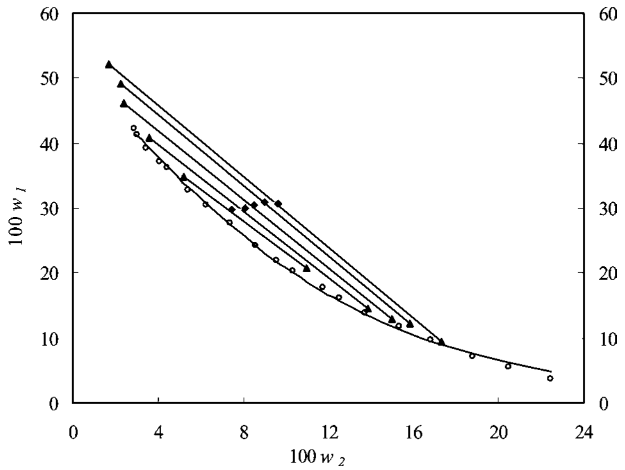

Figure 1. Binodal curve and tie lines for 2-propanol (1) + dipotassium oxalate (2) + water (3) at 298.15 K. ○, experimental data of the binodal curve; −, calculated from eq 2; ▲, tie line data; ♦, initial total composition.

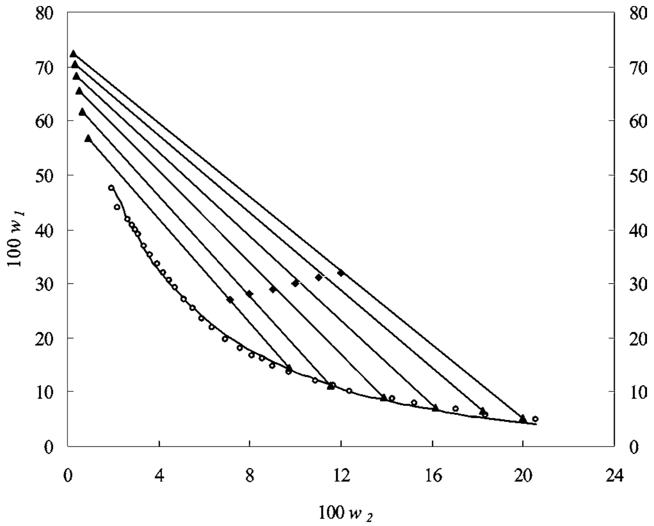

Figure 2. Binodal curve and tie lines for 1-propanol (1) dipotassium oxalate (2) + water (3) at 298.15 K. ○, experimental data of the binodal curve; −, calculated from eq 2; ▲, tie line data; ♦, initial total composition.

alate + water at 298.15 K are listed in Table 2. The binodal curves of the investigated ternary systems are plotted in Figures 1 to 4. These figures represent the effect of alcohol and salt on the phase separation. The addition of salt to aqueous alcohol solution leads to the migration of water molecules away from alcohol molecules to ions of salt and more alcohol molecules will be excluded from the salt-rich phase to the alcohol-rich phase. As shown in these figures, the mass fraction of alcohol in the top phase increases with addition of dipotassium oxalate. This trend was also observed for ammonium sulfate in aqueous ethanol, 1-propanol, and 2-propanol solutions.[20]

The comparison of binodal curves of 2-propanol/1-propanol/2-methyl-2-propanol/2-butanol + dipotassium oxalate + water was depicted in Figure 5 at 298.15 K. This shows the phase separation abilities of different alcohols in alcohol + dipotassium oxalate + water systems. As can be seen from Figure 5, the single-phase areas of the investigated two-phase systems have the order: 2-propanol > 1-propanol > 2-methyl-2-propanol > 2-butanol. Therefore, the phase separation capability of 2-butanol is higher than the other investigated alcohols. This phenomenon can be explained by the different molecular interactions occurring in the solutions (hydrogen bonding between alcohol and water and ion−dipole interactions).[19] The magnitude of the hydrogen bonding and ion−dipole interactions is in accordance with relative permittivity and boiling point of solvent molecules as described by Wang and et al.[20]

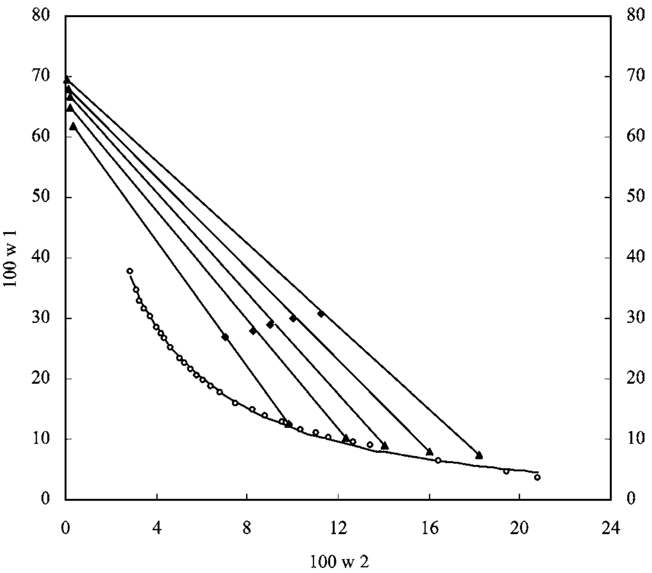

Figure 3. Binodal curve and tie lines for 2-methyl-2-propanol (1) + dipotassium oxalate (2) + water (3) at 298.15 K. ○, experimental data of the binodal curve; −, calculated from eq 2; ▲, tie line data; ♦, initial total composition.

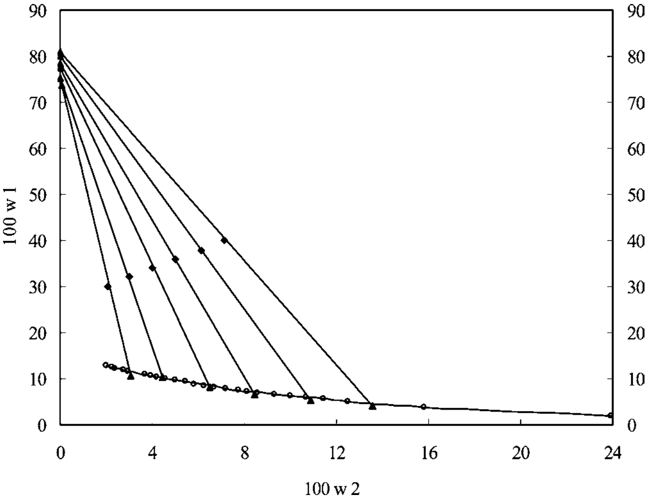

Figure 4. Binodal curve and tie lines for 2-butanol (1) + dipotassium oxalate (2) + water (3) at 298.15 K. ○, experimental data of the binodal curve; −, calculated from eq 2; ▲, tie line data; ♦, initial total composition.

The binodal curves were fitted to the following nonlinear equation

$$\ln w_1 = a + bw_2^{0.5} + cw_2 \qquad (2)$$

where w_1 and w_2 are the mass fraction of alcohol and salt, respectively; and *a*, *b*, and *c* are the fit coefficient of eq 2. The coefficients of eq 2 along with the corresponding standard deviations for the investigated systems are given in Table 4. The standard deviations were calculated using the following equation

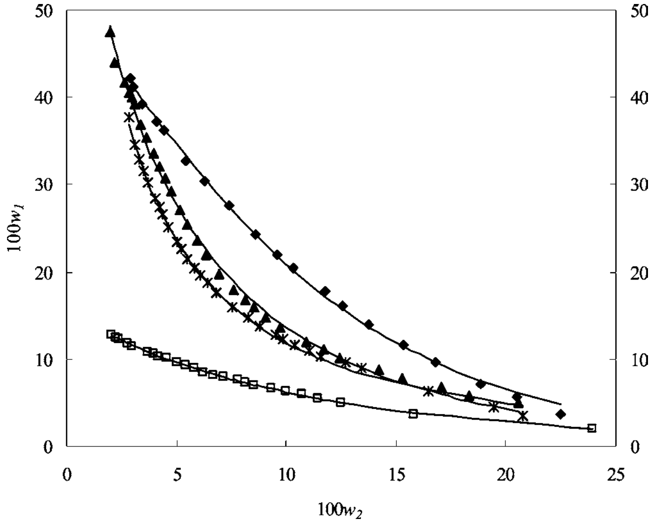

Figure 5. Comparison of binodal curves for the alcohols (1) + dipotassium oxalate (2) + water (3) systems at 298.15 K. ■, 2-propanol; ▲, 1-propanol; ∗, 2-methyl-2-propanol; □, 2-butanol; −, calculated from eq 2.

Table 3. Tie-Line Data for the Alcohol (1) + Dipotassium Oxalate (2) + Water (3) Systems as a Function of Mass Fraction at 298.15 K

total composition		top phase		bottom phase	
100 w_1	100 w_2	100 w_1	100 w_2	100 w_1	100 w_2
2-Propanol + Dipotassium Oxalate + Water					
29.85	7.45	34.94	5.22	20.72	10.98
29.96	8.10	40.91	3.58	14.61	13.87
30.52	8.51	46.21	2.40	13.03	14.96
31.02	9.03	49.26	2.25	12.28	15.87
30.70	9.61	52.11	1.68	9.49	17.30
1-Propanol + Dipotassium Oxalate + Water					
26.94	7.13	56.83	0.89	14.37	9.75
28.02	7.99	61.73	0.66	11.19	11.52
29.00	9.00	65.49	0.52	8.93	13.90
30.02	9.99	68.21	0.40	7.13	16.13
31.02	11.01	70.39	0.29	6.57	18.20
26.94	7.13	56.83	0.89	14.37	9.75
2-Methyl-2-propanol + Dipotassium Oxalate + Water					
26.97	7.01	61.86	0.37	12.59	9.83
27.89	8.27	64.80	0.22	10.27	12.33
28.99	9.00	66.65	0.18	8.94	14.04
29.94	10.02	67.94	0.13	8.06	16.05
30.76	11.22	69.55	0.09	7.55	18.22
2-Butanol + Dipotassium Oxalate + Water					
29.96	2.05	73.87	0.03	10.75	3.06
32.07	3.02	75.38	0.02	10.20	4.46
34.16	4.03	77.52	0.03	8.05	6.53
35.95	4.99	78.54	0.02	6.47	8.45
37.93	6.13	80.13	0.02	5.47	10.87
39.96	7.15	80.95	0.019	4.10	13.54

$$\sigma = \sum_{i=1}^{N}\left(\frac{(w_{i,1,\mathrm{cal}} - w_{i,1,\mathrm{exp}})^2}{N}\right)^{0.5} \tag{3}$$

where N is the number of binodal data.

On the basis of the obtained standard deviations, we conclude that eq 2 can be satisfactorily used to correlate the binodal curves of the investigated systems.

The tie-line data for the investigated ternary systems are given at 298.15 K in Table 3. The tie-line data of 2-propanol/1-propanol/2-methyl-2-propanol/2-butanol + dipotassium oxalate + water ternary systems at 298.15 K are given in Table 3 and represented in Figures 1 to 4. The correlation equations[16,17] given by Othmer−Tobias (eq 4a) and Bancroft (eq 4b) have been used to correlate the tie-line compositions.

Table 4. Coefficients of Equation 2 for the Correlation Binodal Curves Data of Investigated Systems at 298.15 K

system	a	b	c	100σ
2-propanol + dipotassium oxalate + water	−0.8891	2.5821	−14.9625	0.468
1-propanol + dipotassium oxalate + water	0.0355	−4.7066	−5.3523	0.675
2-methyl-2-propanol + dipotassium oxalate + water	0.4802	−9.3509	3.4355	0.372
2-butanol + dipotassium oxalate + water	−1.8111	−0.7373	−7.3420	0.096

$$\left(\frac{1 - w_1^{\mathrm{top}}}{w_1^{\mathrm{top}}}\right) = k_1\left(\frac{1 - w_2^{\mathrm{bot}}}{w_2^{\mathrm{bot}}}\right)^n \tag{4a}$$

$$\left(\frac{w_3^{\mathrm{bot}}}{w_2^{\mathrm{bot}}}\right) = k_2\left(\frac{w_3^{\mathrm{top}}}{w_1^{\mathrm{top}}}\right)^r \tag{4b}$$

where k_1, n, k_2, and r represent fit parameters. Superscripts “top” and “bot” represent the top and bottom phases, respectively. The values of parameters are given in Table 5. On the basis of obtained standard deviations, in this table, it can be seen that eq 4 can be satisfactorily used to correlate the tie-line data of the investigated systems. The corresponding standard deviations have been calculated using the following equations.

$$\sigma_j = \left[\frac{1}{2N}\sum_{i=1}^{N}[(w_{i,j,\mathrm{cal}}^{\mathrm{top}} - w_{i,j,\mathrm{exp}}^{\mathrm{top}})^2 + (w_{i,j,\mathrm{cal}}^{\mathrm{bot}} - w_{i,j,\mathrm{exp}}^{\mathrm{bot}})^2]\right]^{0.5} \tag{5}$$

where σ_j is standard deviation of component j and N is the number of tie lines. The following modified Setschenow equation[18] was also used for the correlation of experimental tie-line data

$$\ln\left(\frac{x_1^{\mathrm{top}}}{x_1^{\mathrm{bot}}}\right) = \beta_2 + k_3(x_2^{\mathrm{bot}} - x_2^{\mathrm{top}}) \tag{6}$$

where k_3 is the salting-out coefficient; β_2 is a constant related to the activity coefficient; and x_1, x_2, respectively, represent the mole fraction of the species 1 and 2 in the top and bottom phases. The values of the parameters for the investigated systems in this work are given in Table 6. On the basis of obtained standard deviations, in this table, it can be seen that eq 7 can be satisfactorily used to correlate the tie-line data of the investigated systems. The corresponding standard deviations have been calculated using the following equations.

$$\sigma_j = \left[\frac{1}{2N}\sum_{i=1}^{N}[(x_{i,j,\mathrm{cal}}^{\mathrm{top}} - x_{i,j,\mathrm{exp}}^{\mathrm{top}}) + (x_{i,j,\mathrm{cal}}^{\mathrm{bot}} - x_{i,j,\mathrm{exp}}^{\mathrm{bot}})]\right]^{0.5} \tag{7}$$

where σ_j is standard deviation of component j and N is the number of tie lines.

The effect of temperature on the investigated systems can be further illustrated if we consider the diagram of cloud-point temperature as a function of alcohol mole fraction. Figure 6 shows the alcohol concentration dependence of cloud points (CP). In Figure 6, the phase diagram of the alcohol mole

Table 5. Values of Parameters of Equation 4 for the Correlation of Tie-Line Data of Investigated Systems at 298.15 K

system	k_1	n	k_2	r	$100\sigma_1$	$100\sigma_2$
2-propanol + dipotassium oxalate + water	0.1186	1.3237	4.5080	0.5677	1.149	0.863
1-propanol + dipotassium oxalate + water	0.1209	0.8167	10.9484	1.0731	0.398	0.234
2-methyl-2-propanol + dipotassium oxalate + water	0.2120	0.4803	20.5862	1.9326	0.252	0.125
2-butanol + dipotassium oxalate + water	0.1458	0.2590	1441.1687	3.8057	0.860	0.200

Table 6. Coefficients of Equation 6 for the Correlation of Tie-Line Data of Investigated Systems at 298.15 K

system	β_2	k_3	$100\sigma_1$	$100\sigma_2$
2-propanol + dipotassium oxalate + water	−0.1870	97.8982	0.535	0.033
1-propanol + dipotassium oxalate + water	0.8368	85.1871	2.060	0.095
2-methyl-2-propanol + dipotassium oxalate + water	1.4221	54.3964	1.276	0.101
2-butanol + dipotassium oxalate + water	2.3157	85.5192	1.216	0.044

Table 7. Cloud-Point (CP) Data for the Alcohol (1) + Dipotassium Oxalate (2) + H_2O (3) Systems as a Function of Alcohol Mole Fraction

T/K	x_2/x_3	x_1
2-Propanol + Dipotassium Oxalate + Water		
298.15	0.0089	0.167
303.15	0.0089	0.164
308.15	0.0089	0.161
313.15	0.0089	0.163
318.15	0.0089	0.157
323.15	0.0089	0.155
328.15	0.0089	0.153
1-Propanol + Dipotassium Oxalate + Water		
293.15	0.0089	0.101
298.15	0.0089	0.096
303.15	0.0089	0.092
308.15	0.0089	0.089
313.15	0.0089	0.087
318.15	0.0089	0.087
323.15	0.0089	0.087
328.15	0.0089	0.087
2-Methyl-2-propanol + Dipotasium Oxalate + Water		
293.15	0.0089	0.069
298.15	0.0089	0.062
303.15	0.0089	0.055
308.15	0.0089	0.049
313.15	0.0089	0.044
318.15	0.0089	0.041
323.15	0.0089	0.038
328.15	0.0089	0.035
2-Butanol + Dipotassium Oxalate + Water		
293.15	0.0089	0.027
298.15	0.0089	0.025
303.15	0.0089	0.024
308.15	0.0089	0.023
313.15	0.0089	0.022
318.15	0.0089	0.021
323.15	0.0089	0.019
328.15	0.0089	0.018

fractions as a function of the cloud-point temperatures has been shown for the same concentration of aqueous dipotassium oxalate solution. Cloud-point data for the investigated systems are given in Table 7. As shown in Figure 6, the concentration of alcohol required to achieve a phase separation slightly decreases by increasing temperature. Also, the results show that the alcohol mole fraction required for phase separation in the temperature range (293.15 to 328.15) K is in the following order, 2-propanol > 1-propanol > 2-methyl-2-propanol > 2-butanol, which is in agreement with hydrophilic series.

Conclusions

Phase diagrams of some two-phase systems, alcohols + dipotassium oxalate + water, with the alcohols being 2-propanol, 1-propanol, 2-methyl-2-propanol, and 2-butanol, have been determined at 298.15 K, for which the binodal curves and tie lines were obtained. It was found that the solubility of aliphatic alcohol in water and the mutual miscibility depend on the chain length and polarity of alcohol, and solubility decreases with increasing number of carbon atoms of alcohol and in the case of alcohol isomers increases with increasing alcohol polarity. The results show that the lower alcohol concentration is required for phase separation at higher temperatures. The binodal curves of the investigated system were fitted using a nonlinear equation with three parameters, and the tie lines were satisfactorily described with the Othmer−Tobias, Bancroft, and also modified Setschenow equations.

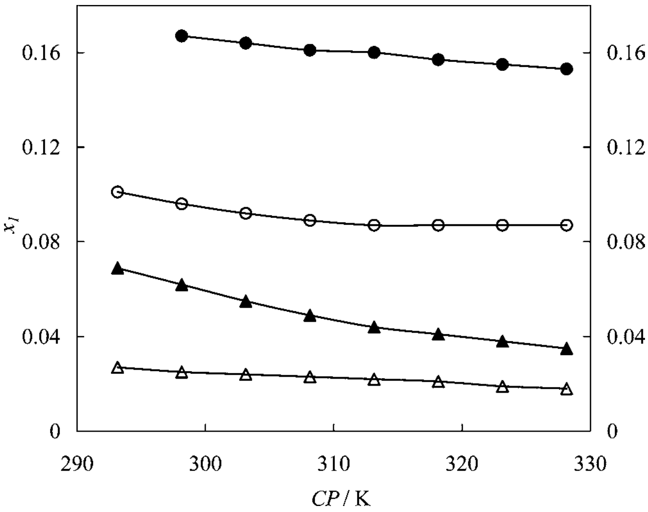

Figure 6. Effect of temperature on cloud point, CP, as a function of alcohol mole fraction, in the presence of aqueous solution of dipotassium oxalate salt: ●, 2-propanol; ○, 1-propanol; ▲, 2-methyl-2-propanol; △, 2-butanol.

Supporting Information Available:

Supplemental Tables 1 and 2. This material is available free of charge via the Internet at http://pubs.acs.org.

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