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Liquid−Liquid Equilibria for Methanol/Ethanol + Rubidium Carbonate + Water Systems at Different Temperatures

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Liquid−liquid equilibria for the two ternary systems of $CH_3OH + Rb_2CO_3 + H_2O$ and $CH_3CH_2OH + Rb_2CO_3 + H_2O$ were measured at $T = (298.15, 308.15, \text{and } 318.15)$ K. Binodal curves, tie-lines, and integrated phase diagrams for the two ternary systems are given. The experimental binodal curve data are correlated using a three- parameter equation, and the tie-line data are corrected by the Othmer−Tobias and Bancroft equations. The salting-out effect of ethanol is greater than that for methanol, but the effect of temperature on the equilibrium was not significant.

Introduction

The phase equilibria of mixtures that include one salt dissolved in aqueous−organic mixed solvents have significant importance in scientific research and the chemical industry, such as recycling of an organic solvent in a mixed-solvent system, design of extractors, separation of mixtures, biological processes, etc.[1−3] During the past decade, there have been many studies in this area. For example, Gomis and co-workers have investigated five ternary systems of water + sodium chloride + 1-pentanol/2-pentanol/3-pentanol/2-methyl-1-butanol/2-methyl-2- butanol at 25 °C.[4] They obtained the complete phase diagrams for the examined systems and concluded that the salting-out effect of the five pentanols is similar except for 2-methyl-2-butanol which presents the most significant decrease in the solubility of water. Moreover, they also studied the influence of the cationic radius on the phase equilibria of MCl ($M = Na^+$, K^+, and Li^+) + 1/2-propanol + water ternary systems.[5,6] Similarly, Taboada reported the phase equilibria of the 1-propanol + lithium sulfate + water system at different temperatures.[7] Furthermore, the temperature effect on the liquid−liquid equilibria for some aliphatic alcohols + water + K_2CO_3 systems have been determined by Salabat.[8] In our series of work, we have systematically investigated the phase equilibria for quite a number of ternary or quaternary systems[9−14] consisting of rubidium and cesium salts, water, and organic solvents (e.g., methanol, ethanol, 1-propanol, 2-propanol, as well as poly(ethylene glycol)). Since the solubility of Rb_2CO_3 is much larger than that of other rubidium and cesium salts, the aim of this work is to extend our previous research and enrich the available data on the solubility and other information of rare alkali metal salts. In this research, the liquid−liquid equilibrium (LLE) data for the ternary systems $CH_3OH + Rb_2CO_3 + H_2O$ and $CH_3CH_2OH + Rb_2CO_3 + H_2O$ at (298.15, 308.15, and 318.15) K are reported. A three-parameter equation was successfully applied for the correlation of the experimental LLE data. Furthermore, the tie line data were corrected by the familiar Othmer−Tobias and Bancroft equations.

Experimental Section

Materials. The methanol and ethanol used in this work were analytical reagent grade with purity > 99.5 % (Xi'an Chemical Reagent Factory), and rubidium carbonate was supplied by the Shanghai China Lithium Industrial Co., Ltd. (purity > 99.5 %). Specpure potassium dichromate (Tianjin Chemical Factory) was used without further purification. Double-distilled deionized water was used in our experiments. Its specific conductance was approximately $(1.0 \text{ to } 1.2)\cdot 10^{-4}$ $S\cdot m^{-1}$ and measured every two days.

Apparatus and Experimental Procedure. For LLE experiments, the apparatus and approach have been described in a previous paper,[9] so only a brief description of the experimental procedure is present here. Samples were prepared by adding the weighed methanol or ethanol and water to a known mass of rubidium carbonate. The samples were fixed to a carrier plate and stirred for 48 h at one of the three reaction temperatures [(298.15, 308.15, or 318.15) K], and the solution was allowed to settle for a further 24 h to reach equilibrium. After equilibrium was achieved, the bottom and top phases were sampled, respectively, for determination of the salt concentration. The binodal curves were obtained by the cloud point method means of titration.[2] A 25 mL glass vessel equipped with a Teflon-coated magnetic stirrer was used to carry out the phase equilibrium determinations. It was provided with an external jacket in which water at constant temperature was circulated from a controlled thermostat. The temperature was controlled in this manner to within ± 0.1 K. A known mass of methanol or ethanol was added to the salt solution, or vice versa, until the turbidity appeared. The solution composition at each turbidity point was calculated from the mass of methanol or ethanol and the quantity of the salt solution.

Analytical Methods. The concentration of methanol or ethanol was determined by using $K_2Cr_2O_7$ as oxidant.[15,16] The uncertainty of the determination of alcohol mass fraction was less than ± 0.5 %. The concentration of inorganic salt was determined utilizing an atomic absorption spectrometer (AAS). The AAS measurements were performed on a TAS-986 (Beijing Purkinje General Instrument Co., Ltd.) atomic absorption spectrometer, and the uncertainty was within ±

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Table 1. Binodal Curve Data as Mass Fraction for CH_3OH/C_2H_5OH (1) + Rb_2CO_3 (2) + H_2O (3) Systems at (298.15, 308.15, and 318.15) K

w_1	w_2	w_1	w_2	w_1	w_2	w_1	w_2
CH_3OH (T = 298.15 K)							
56.87	19.72	46.9	25.62	35.38	30.24	21.79	40.61
54.77	21.52	46.39	25.06	31.91	32.06	19.48	43.00
53.51	21.89	44.65	25.67	28.86	34.85	18.02	44.58
51.57	22.48	42.34	26.87	26.72	36.01	17.04	44.71
50.85	22.94	40.61	27.66	24.98	37.21	16.61	47.37
48.98	23.87	38.87	28.27	23.53	38.79	14.87	49.96
CH_3OH (T = 308.15 K)							
52.39	19.65	39.29	25.65	26.05	31.44	15.69	37.29
48.73	22.84	37.54	26.57	23.07	33.33	15.46	39.62
47.99	22.32	35.85	26.5	23.05	32.08	15.2	40.59
46.94	22.91	34.79	26.97	21.11	33.79	15.19	40.71
46.08	23.24	33.05	28.02	20.19	34.86	14.52	42.23
44.76	24.43	32.49	27.36	19.78	33.17	12.81	40.89
43.79	24.34	30.99	29.07	19.34	37.07		
43.47	25.15	27.55	29.72	16.25	36.61		
CH_3OH (T = 318.15 K)							
53.09	18.30	28.58	26.97	18.58	33.85	12.60	42.50
48.19	19.96	28.34	26.53	17.82	35.60	11.64	42.96
42.00	21.96	27.27	28.64	15.96	38.05	11.33	45.61
38.50	23.39	25.34	29.99	14.17	38.15	9.95	48.53
34.41	24.47	22.68	30.50	14.10	40.26	8.96	49.09
32.49	27.07	21.97	31.58	14.01	42.01	7.08	51.82
30.64	26.34	19.59	32.19	13.03	39.57	6.38	54.26
53.09	18.30	28.58	26.97	18.58	33.85	12.60	42.50
C_2H_5OH (T = 298.15 K)							
54.99	4.95	37.92	11.19	29.65	15.11	14.52	25.87
50.68	5.85	37.06	11.46	28.8	15.76	12.28	28.02
46.09	7.1	36.75	11.71	26.1	17.26	9.17	32.03
45.22	7.71	35.56	11.97	24.76	17.87	6.93	33.88
41.23	9.56	34.53	12.29	23.84	19.01	5.62	36.33
40.54	10.49	33.96	12.44	22.5	20.62	4.69	39.09
39.61	10.77	32.76	13.56	18.95	21.57	3.82	42.49
39.35	10.83	30.99	13.92	16.27	23.42		
C_2H_5OH (T = 308.15 K)							
62.13	4.22	34.15	12.38	24.76	14.52	16.5	19.9
52.37	5.4	33.39	11.37	23.4	15.53	14.77	21.35
50.1	6.78	31.41	12.44	23.07	15.94	13.23	22.82
46.98	7.91	30.3	11.78	22.68	16.35	11.88	25.33
42.53	8.49	29.92	11.57	22.07	17.76	9.19	27.3
38.57	9.61	28.66	12.18	21.33	17.19	8.27	29.59
35.81	11.21	28.37	14.28	19.93	18.24	6.08	31.83
35.45	11.58	28.29	14.35	18.53	18.61	5.66	34.33
34.73	12.2	27.17	13.44	17.18	21.12	4.73	37.43
C_2H_5OH (T = 318.15 K)							
51.45	4.09	30.3	8.25	20.19	11.33	10.96	18.18
45.22	4.84	29.65	8.89	19.59	12.82	8.77	19.29
41.18	5.9	27.51	9.27	18.96	12.75	7.85	21.01
37.64	6.69	26.11	10.69	17.51	13.22	6.07	23.24
35.45	7.05	25.1	9.91	16.77	13.93	4.73	24.53
33.73	7.82	22.68	10.32	15.02	15.55	3.89	27.3
32.34	7.91	22.07	11.54	12.31	16.43		

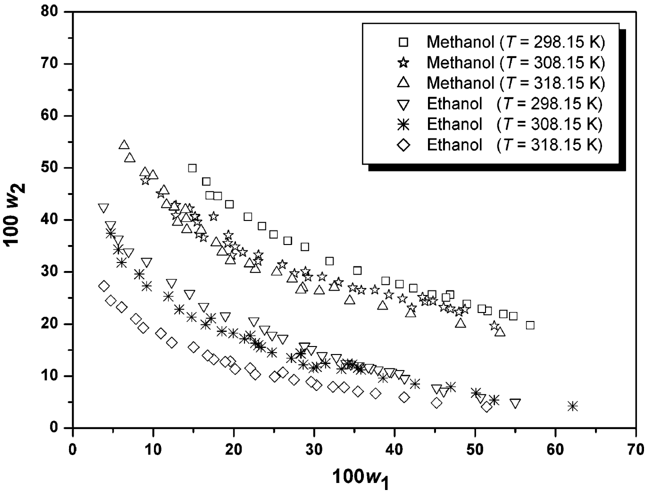

Figure 1. Binodal curves for $CH_3OH/C_2H_5OH + Rb_2CO_3 + H_2O$ at different temperatures.

Table 2. Values of the Parameters for Equation 1[a]

systems	T/K	a	b	c	100δ
	298.15	6.700	−0.507	−0.002	0.100
CH_3OH	308.15	6.620	−0.497	−0.021	0.956
	318.15	6.416	−0.587	−0.007	0.575
	298.15	4.545	0.096	−0.084	0.280
C_2H_5OH	308.15	4.418	−0.063	−0.071	0.110
	318.15	4.268	−0.034	−0.106	0.409

[a] $\delta = \Sigma((w_1^{cal} - w_1^{exp})^2/N)^{0.5}$ where N is the number of binodal data.

0.5 %. Finally, the water concentration was computed using mass-balance equations. The solid phase in equilibrium was analyzed by Thermogravimetric Analysis (TA-SDT Q600). The TGA results show that the solid phases are $Rb_2CO_3 \cdot 3.5H_2O$ (298.15 K) and $Rb_2CO_3 \cdot 3H_2O$ [(308.15 and 318.15) K], respectively.

Results and Discussion

The binodal curve data for the $CH_3OH + Rb_2CO_3 + H_2O$ and $C_2H_5OH + Rb_2CO_3 + H_2O$ ternary systems at three temperatures studied in this work are listed in Table 1 and depicted in Figure 1 in mass fraction. It should be pointed out that it is different from the $CH_3OH + Cs_2CO_3 + H_2O$ system[12] we previously reported, as the CH_3OH/H_2O mixtures are split into two liquid phases by the addition of the Rb_2CO_3 salt. In our opinion, this result is mainly attributed to the greater solvation ability of Rb^+ since the solubility of Cs_2CO_3 is slightly larger than Rb_2CO_3 at the same temperature. Moreover, the effect of the temperature on the phase equilibria is insignificant within the investigated range, as

Table 3. Tie-Line Data As Mass Fraction for CH_3OH/C_2H_5OH (1) + Rb_2CO_3 (2) + H_2O (3) Systems at (298.15, 308.15, and 318.15) K

top phase			bottom phase		
100 w_1	100 w_2	100 w_3	100 w_1	100 w_2	100 w_3
CH_3OH (T = 298.15 K)					
65.13	18.75	16.12	10.87	62.51	26.62
58.78	20.01	21.21	11.46	57.83	30.71
54.33	22.16	23.51	13.21	55.55	31.24
50.24	23.73	26.03	14.11	50.65	35.24
CH_3OH (T = 308.15 K)					
70.73	18.14	11.13	4.91	69.21	25.88
62.60	19.56	17.84	5.67	63.20	31.12
56.26	20.91	22.83	7.79	58.91	33.29
52.53	21.63	25.85	8.06	55.46	36.48
CH_3OH (T = 318.15 K)					
73.74	13.82	12.44	3.40	69.51	27.09
71.04	14.33	14.63	4.27	65.89	29.84
65.07	15.80	19.13	4.91	62.78	32.31
61.87	16.55	21.58	6.89	58.06	35.06
C_2H_5OH (T = 298.15 K)					
82.24	0.55	17.21	0.11	57.95	41.94
76.05	1.54	22.41	0.10	53.64	46.27
65.93	2.54	31.53	2.54	46.59	50.87
56.72	4.49	38.79	4.33	41.20	54.47
C_2H_5OH (T = 308.15 K)					
81.64	0.27	18.09	0.15	66.83	33.02
76.61	0.81	22.58	0.82	63.22	35.96
71.74	1.73	26.53	1.42	56.79	41.79
67.43	2.22	30.34	2.22	53.27	44.51
57.58	4.91	37.50	3.59	46.05	50.35
C_2H_5OH (T = 318.15 K)					
78.98	0.15	20.88	0.16	65.66	33.08
68.32	0.85	30.82	0.28	57.27	42.46
59.42	2.85	37.74	0.83	51.41	47.75
49.26	4.66	46.08	2.96	43.10	53.94

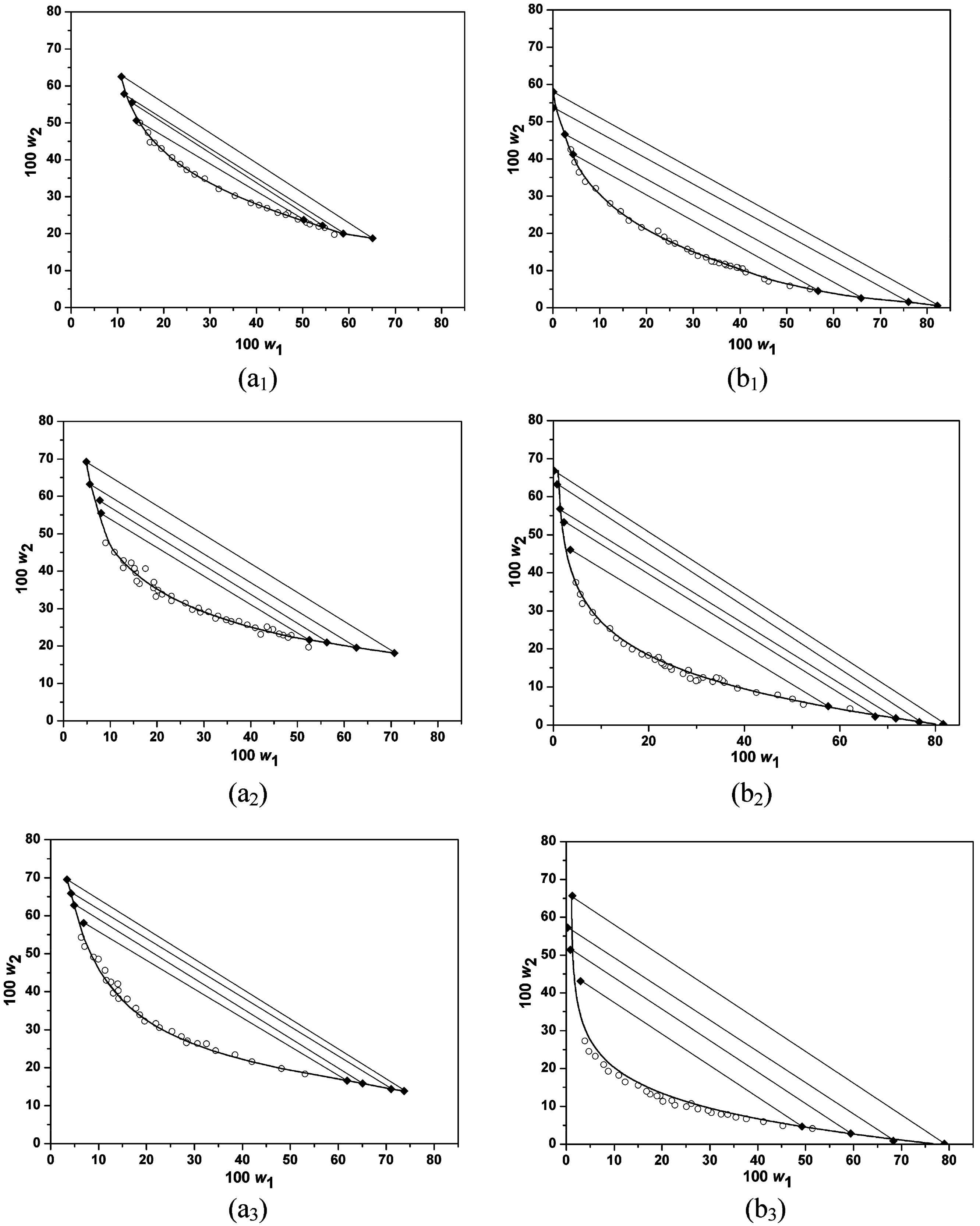

Figure 2. Binodal curves and tie-lines for the CH_3OH (1) + Rb_2CO_3 (2) + H_2O (3) (a_1, a_2, a_3) and C_2H_5OH(1) + Rb_2CO_3(2) + H_2O (3) (b_1, b_2, b_3) systems at T = (298.15, 308.15, and 318.15) K (○, binodal curve points; ■, tie-line points).

can be seen in Figure 1. These trends are in agreement with our experimental results of the aliphatic alcohol + cesium carbonate systems[10,12] and that obtained by Salabat for aliphatic alcohol + K_2CO_3 systems.[8] Additionally, as expected, the two-phase area of the C_2H_5OH + Rb_2CO_3 + H_2O system is larger than that of CH_3OH + Rb_2CO_3 + H_2O because reducing the number of carbon atoms in the chain improves the miscibility.

The binodal curves were given using the following three-parameter nonlinear expression

$$\ln w_1 = a + b w_2^{0.5} + c w_2 \qquad (1)$$

where w_1 and w_2 represent the mass fractions of organic solvents and rubidium carbonate, respectively. The coefficients of eq 1 along with the corresponding standard deviations for the investigated systems are given in Table 2. On the basis of the obtained standard deviations, eq 1 can be satisfactorily used to correlate the binodal curves of the investigated systems.

The compositions of tie lines are given in Table 3. Figure 2 shows the binodal curves, together with the experimental tie lines of the systems at three investigated temperatures. As can be seen from Figure 2, the tie-line plots and binodal curves match well. The reliability of the measured tie-line compositions was ascertained by the correlation equations given by Othmer−Tobias (eq 2)[17] and Bancroft (eq 3).[18]

$$((1 - w_1^{\mathrm{t}})/w_1^{\mathrm{t}}) = k_1((1 - w_2^{\mathrm{b}})/w_2^{\mathrm{b}})^n \qquad (2)$$

$$(w_3^{\mathrm{b}}/w_2^{\mathrm{b}}) = k_2(w_3^{\mathrm{t}}/w_3^{\mathrm{b}})^r \qquad (3)$$

In the above relations, w_1^{t} is the mass fraction of aliphatic alcohol in the top phase; w_2^{b} is the mass fraction of Rb_2CO_3 in the bottom phase; and w_3^{b} and w_3^{t} are the mass fraction of water in the bottom and top phase, respectively. k_1, k_2, n, and r represent fit parameters. The values of these parameters are listed in Table 4. A linear dependency of the plots $\lg((1 - w_1^{\mathrm{t}})/w_1^{\mathrm{t}})$ against $\lg((1 - w_2^{\mathrm{b}})/w_2^{\mathrm{b}})$ and $\lg(w_3^{\mathrm{b}}/w_2^{\mathrm{b}})$ against $\lg(w_3^{\mathrm{t}}/w_3^{\mathrm{b}})$ indicates an acceptable consistency of the results. The corresponding correlation coefficient values, R_1 and R_2, are also given. Furthermore, on the basis of the standard deviations, δ_1 and δ_2, given in Table 4, we conclude that eqs 2 and 3 can be satisfactorily used to correlate the tie-line data of the investigated systems.

The integrated phase diagrams for these two ternary systems at the investigated temperatures are similar. For a sample, the diagram is shown in Figure 3 for the $CH_3OH + Rb_2CO_3 + H_2O$ system at 298.15 K. The letters L and S denote the liquid phase and the solid phase, respectively. Six zones are observed in the diagram. Region L represents the homogeneous zone of unsaturated liquid. In the region 2L, two liquid phases are in equilibrium: a top phase rich in CH_3OH and a bottom phase rich in Rb_2CO_3. Two L + S regions are observed, where the solid phase is $Rb_2CO_3 \cdot 3.5H_2O$, but the difference is that the right represents a saturated liquid with low ethanol concentration. The saturation curve of this region begins at the solubility point of the salt in water (A), to the eutectic point of the aqueous phase. The saturation curve of the zone to the left has a high CH_3OH concentration, and this curve initiates in the solubility point of the salt in CH_3OH (B) and terminates at the vertex of pure CH_3OH. Two liquid and one solid phase exist in the 2L + S region. This curve is initiated in the eutectic point of the organic phase and terminates at the vertex of pure CH_3OH. In the region L + 2S, the presence of two solid phases is observed: one is $Rb_2CO_3 \cdot 3.5H_2O$, and the other is Rb_2CO_3. The phase diagrams for these two systems at (308.15 and 318.15) K are similar to that of 298.15 K, except for the slight shift and enlargement of the 2L region and the alteration of the solid phase in equilibrium ($Rb_2CO_3 \cdot 3.5H_2O$ at 298.15 K and $Rb_2CO_3 \cdot 3H_2O$ at (308.15 and 318.15) K). In comparison, the phase diagram for the $CH_3OH + Cs_2CO_3 + H_2O$ ternary system just presents one L and one L + S region since only one liquid phase is observed.[12]

Table 4. Values of Parameters for Equations 2 and 3[a]

systems	T/K	k_1	n	k_2	r	R_1	R_2	$100\delta_1$	$100\delta_2$
CH_3OH	298.15	1.043	1.226	1.036	0.657	0.993	0.997	0.290	0.214
	308.15	1.221	1.316	0.908	0.487	0.997	0.993	1.100	1.275
	318.15	0.907	1.132	1.067	0.561	0.995	0.997	0.477	1.553
C_2H_5OH	298.15	0.401	1.812	1.599	0.507	0.996	0.992	0.643	0.707
	308.15	0.735	0.421	0.731	0.038	0.991	0.996	0.798	0.200
	318.15	0.706	1.397	1.307	0.722	0.995	0.994	1.907	0.792

[a] $\sigma_j = \{1/2L\sum_{i=1}^{L}[(w_{i,j,\mathrm{cal}}^{\mathrm{t}} - w_{i,j,\mathrm{exp}}^{\mathrm{t}})^2 + (w_{i,j,\mathrm{cal}}^{\mathrm{b}} - w_{i,j,\mathrm{exp}}^{\mathrm{b}})^2]\}^{0.5}$, where L is the number of ties lines and $j = 1$ and 2. δ_1 and δ_2 represent the mass percent standard deviation for alcohol and Rb_2CO_3, respectively.

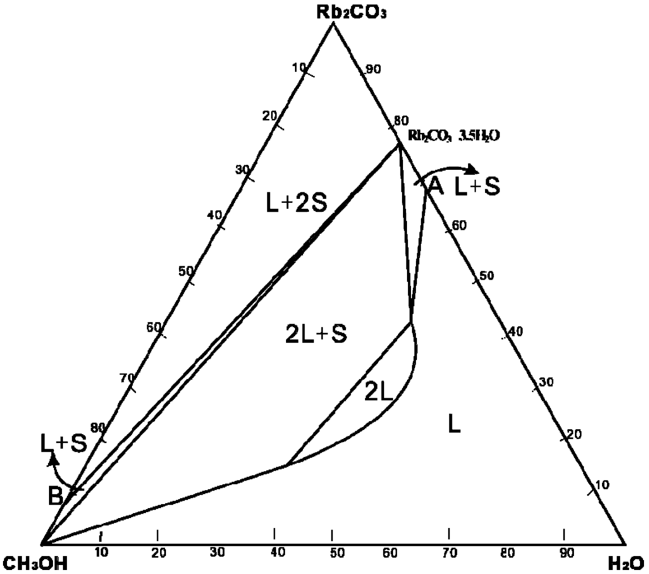

Figure 3. Schematic presentation of the integrated phase diagram for the CH_3OH (1) + Rb_2CO_3 (2) + H_2O (3) system at 298.15 K (S, solid phase; L, liquid phase; A, the solubility of Rb_2CO_3 in water; B, the solubility of Rb_2CO_3 in methanol).

Conclusion

For the methanol + rubidium carbonate + water and ethanol + rubidium carbonate + water ternary systems, we obtained binodal curves and tie-line data at (298.15, 308.15, and 318.15) K. The tie-line data for these systems could be satisfactorily described using the Othmer−Tobias and Bancroft equations. Moreover, the salting-out effect in the two-liquid region was greater for ethanol than for methanol, and the effect of temperature on the liquid−liquid equilibrium was not significant.

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