JOURNAL OF CHEMICAL & ENGINEERING DATA

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Phase Diagram Data for Ethanol/Propan-1-ol/Propan-2-ol−2,3-Dihydroxybutanedioate Aqueous Two-Phase Systems at 298.15 K and Data Correlation

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ABSTRACT: Binodal data for the ethanol/propan-1-ol/propan-2-ol−dipotassium 2,3-dihydroxybutanedioate aqueous two-phase system (ATPS), the propan-2-ol−disodium 2,3-dihydroxybutanedioate/diammonium 2,3-dihydroxybutanedioate ATPS, and the ethanol−potassium sodium 2,3-dihydroxybutanedioate ATPS were determined at 298.15 K. An empirical equation was used to correlate binodal data, and it showed satisfactory accuracy in binodal data fitting for all of the determined systems. The phase-separation abilities of the investigated salts and the water-miscible alcohols were compared by the plotting of binodal curves in mass fraction. The phase-separation abilities of the investigated alcohols are in the order propan-1-ol > propan-2-ol > ethanol, and those of salts are in the order disodium 2,3-dihydroxybutanedioate > potassium sodium 2,3-dihydroxybutanedioate > dipotassium 2,3-dihydroxybutanedioate > diammonium 2,3-dihydroxybutanedioate. However, the two-phase area of the investigated alcohol−dipotassium 2,3-dihydroxybutanedioate ATPS is higher than that of the alcohol−disodium 2,3-dihydroxybutanedioate/potassium sodium 2,3-dihydroxybutanedioate/diammonium 2,3-dihydroxybutanedioate ATPS. The salting-out abilities of the common cations (K^+, Na^+, and NH_4^+) were discussed by comparing their effective excluded volume (EEV) in the propan-2-ol−water component solvent and the binodal curves plotted in molality. They are in the order $Na^+ \approx K^+ > NH_4^+$.

INTRODUCTION

Aqueous two-phase extraction (ATPE) has become an important downstream technology for separation and purification of biological products. Aqueous two-phase systems (ATPS's) are usually composed of two polymers or a polymer with a salt. Compared with traditional organic solvent extraction technology, ATPE technology has the advantages of a mild operation environment and low interfacial tension. Reliable phase diagram data are essential to the design of ATPE and the establishment of thermodynamic models.

In recent years, a large number of studies have been conducted on the phase diagrams of ATPS's, including the polymer−polymer ATPS, the polymer−salt ATPS, the ionic liquid−salt ATPS, and the water-miscible alcohol−salt ATPS.[1−4] So far, the phase diagrams of a series of the water-miscible alcohol−salt ATPS have been determined, and the studied systems are listed in Table 1. As shown in Table 1, the investigated phase-separation salts are mainly inorganic salts. Up to now, there has been relatively little research taking the organic salts as the phase-separation salts in the water-miscible alcohol−salt ATPS. The selection of an appropriate phase-separation salt in the ATPE process always takes into account its salting-out ability, production cost, environmental pollution, and the pH value of salt solution, so a comprehensive research on the phase-separation salts should be made to find the optimum one for the corresponding ATPS.

In this paper, binodal data for the ethanol/propan-1-ol/propan-2-ol−dipotassium 2,3-dihydroxybutanedioate ATPS, the propan-2-ol−disodium 2,3-dihydroxybutanedioate/diammonium 2,3-dihydroxybutanedioate ATPS, and the ethanol−potassium sodium 2,3-dihydroxybutanedioate ATPS were determined at 298.15 K. A nonlinear equation was used to correlate the determined binodal data. On the basis of the binodal curves, the phase-separation abilities of the investigated salts and alcohols were discussed. The salting-out abilities of the common cations (K^+, Na^+, and NH_4^+) were discussed by comparing the binodal curves plotted in molality as well as the effective excluded volume (EEV) of dipotassium 2,3-dihydroxybutanedioate, disodium 2,3-dihydroxybutanedioate, and diammonium 2,3-dihydroxybutanedioate in the propan-2-ol−water component solvent.

EXPERIMENTAL SECTION

Materials. Dipotassium 2,3-dihydroxybutanedioate hemihydrate, disodium 2,3-dihydroxybutanedioate dihydrate, diammonium 2,3-dihydroxybutanedioate, potassium sodium 2,3-dihydroxybutanedioate tetrahydrate, ethanol, propan-1-ol, and propan-2-ol were supplied by the Sinopharm Chemical Reagent Co., Ltd. with a minimum mass fraction of 0.990, 0.990, 0.990, 0.990, 0.997, 0.990, and 0.997, respectively. All chemicals were used without further purification. Double-distilled and deionized water was used throughout the entire experiment.

Apparatus and Procedures. The binodal curves were determined by the titration method (cloud point method). A dipotassium 2,3-dihydroxybutanedioate/disodium 2,3-dihydroxybutanedioate/diammonium 2,3-dihydroxybutanedioate/potassium sodium 2,

Received: January 8, 2011
Accepted: February 1, 2011
Published: February 22, 2011

dx.doi.org/10.1021/je2000233 | *J. Chem. Eng. Data* 2011, 56, 622–626

Table 1. Determination of Phase Diagram Data for the Water-Miscible Alcohol−Salt ATPS

ATPS	T/K	ref
ethanol−Cs_2CO_3	273.15, 293.15, 313.15	5
ethanol−Rb_2CO_3	298.15, 308.15, 318.15	6
methanol−Rb_2CO_3	298.15, 308.15	6
propan-1-ol/propan-2-ol−$(NH_4)_2HPO_4$	298.15	7
propan-1-ol−$NH_4H_2PO_4$	298.15	7
ethanol/propan-1-ol/propan-2-ol−$(NH_4)_2SO_4$	298.15	8
ethanol−Mg_2SO_4	298.15, 308.15	9
propan-1-ol/propan-2-ol−Mg_2SO_4	298.15	9
propan-1-ol/propan-2-ol−NaCl/(NaCl+RbCl)	298.15	10
methanol/ethanol−K_2HPO_4	288.15, 298.15, 308.15	2
ethanol−Na_2HPO_4	288.15, 298.15, 308.15	2
propan-1-ol/propan-2-ol−K_2HPO_4	288.15, 298.15, 308.15	11
propan-1-ol−KCl + CsCl	308.15, 318.15	12
propan-2-ol−Na_2CO_3/ K_2CO_3	298.15	13
ethanol−Na_2CO_3	298.15	13
ethanol/propan-1-ol/propan-2-ol−$K_3C_6H_5O_7$	298.15	14, 15
ethanol/propan-1-ol/propan-2-ol−$Na_3C_6H_5O_7$	298.15	14, 15
ethanol/propan-2-ol−$(NH_4)_3C_6H_5O_7$	298.15	15

Table 2. Binodal Data for the Ethanol/Propan-1-ol/Propan-2-ol (1)−Dipotassium 2,3-Dihydroxybutanedioate (2) ATPS at 298.15 K

ethanol−dipotassium 2,3-dihydroxybutanedioate				propan-1-ol−dipotassium 2,3-dihydroxybutanedioate				propan-2-ol−dipotassium 2,3-dihydroxybutanedioate			
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
2.20	55.07	18.01	24.12	0.56	53.17	11.20	15.07	0.61	53.82	24.46	12.21
3.29	49.44	20.63	21.79	0.80	48.56	12.17	14.40	0.99	49.35	28.32	10.27
3.24	48.82	23.31	19.31	1.13	44.72	15.56	11.50	1.44	45.55	31.02	8.80
3.57	47.62	27.38	15.59	1.52	41.22	18.87	9.73	1.91	42.24	33.81	7.51
4.70	44.66	30.64	12.97	1.99	38.05	22.13	8.35	2.66	39.26	36.21	6.49
5.08	43.20	33.49	10.96	2.49	35.38	26.17	7.11	3.27	36.59	38.25	5.71
6.27	40.65	39.81	7.27	3.01	33.08	29.79	6.14	4.07	34.14	40.07	5.07
7.10	38.70	43.32	5.67	3.54	30.99	32.85	5.37	5.39	31.13	41.61	4.56
8.01	37.20	46.11	4.61	4.77	28.22	36.86	4.51	7.39	27.36	43.30	4.07
8.79	35.81	48.28	3.90	5.73	24.88	41.01	3.64	9.37	24.28	45.96	3.34
10.06	33.84	50.41	3.29	6.73	22.22	44.25	3.02	11.58	21.63	48.18	2.82
10.78	32.60	52.31	2.80	6.69	22.21	46.98	2.56	13.57	19.54	50.08	2.42
11.54	31.30	54.27	2.36	7.83	20.00	49.23	2.21	15.95	17.57	51.68	2.12
13.05	29.81	56.02	2.03	8.95	18.08	51.15	1.94	18.32	15.86	53.04	1.88
14.44	28.28	57.46	1.77	10.13	16.38	52.86	1.72	20.38	14.51	54.29	1.68
16.13	26.06							22.63	13.24		

3-dihydroxybutanedioate solution of known concentration was titrated with ethanol/propan-1-ol/propan-2-ol until the clear solution turned turbid. The compositions of the mixture were determined by a Sartorious analytical balance (model BS 124S) with a precision of ± 0.0001 g. A conical flask (50 cm^3) was used to carry out the experiment, and the temperature was maintained within (298.15 ± 0.1) K in a water bath.

RESULTS AND DISCUSSION

Binodal Data and Correlation. For the ethanol/propan-1-ol/propan-2-ol−dipotassium 2,3-dihydroxybutanedioate ATPS, the propan-2-ol−disodium 2,3-dihydroxybutanedioate/diammonium 2,3-dihydroxybutanedioate ATPS, and the ethanol−potassium sodium 2,3-dihydroxybutanedioate ATPS, the binodal data determined at 298.15 K are listed in Tables 2 and 3. For all of the investigated systems, binodal data were correlated by the following equation

$$w_1 = \exp(a + bw_2^{0.5} + cw_2 + dw_2^{2}) \qquad (1)$$

where w_2 is the mass fraction of salts, w_1 is the mass fraction of alcohols, and a, b, c, and d are fitting parameters. The fitting parameters, coefficients of determination (R^2) and standard deviations (sd) are listed in Table 4. As shown in Table 4, the coefficients of determination (R^2) of eq 1 for the investigated systems are all larger than 0.9992, and the standard deviations are all smaller than 0.47, which indicate that eq 1 can be satisfactorily used to correlate

Table 3. Binodal Data for the Propan-2-ol (1)−Disodium 2,3-Dihydroxybutanedioate/Diammonium 2,3-Dihydroxybutanedioate (2) ATPS and the Ethanol (1)−Potassium Sodium 2,3-Dihydroxybutanedioate (2) ATPS at 298.15 K

propan-2-ol−diammonium 2,3-dihydroxybutanedioate				ethanol−potassium sodium 2,3-dihydroxybutanedioate		propan-2-ol−disodium 2,3-dihydroxybutanedioate			
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
7.11	31.73	36.29	8.59	7.39	36.60	5.87	25.93	29.17	7.72
8.92	28.10	40.29	7.03	11.75	29.76	8.15	22.23	31.31	6.78
10.13	25.87	42.26	6.28	15.90	24.81	9.32	20.82	33.07	6.08
10.85	25.07	43.51	5.88	19.86	20.59	10.78	19.19	34.65	5.48
12.85	22.52	46.05	5.05	22.20	18.30	13.31	16.88	36.47	4.91
13.50	21.76	48.27	4.41	24.39	16.18	15.22	15.44	38.82	4.18
15.14	20.24	49.81	4.00	26.33	14.48	16.18	14.70	40.80	3.62
17.33	18.42	52.44	3.35	28.02	13.03	18.67	13.20	42.50	3.20
20.10	16.61	54.64	2.87	29.87	11.51	19.58	12.61	43.96	2.85
22.50	15.15	56.63	2.48	32.09	10.01	20.79	11.90	48.71	1.94
21.38	15.74	58.30	2.18	35.20	8.15	22.41	11.05	50.73	1.64
25.32	13.54	59.51	1.98	37.62	6.86	23.72	10.40	52.33	1.41
28.94	11.81	61.90	1.59	39.79	5.87	24.77	9.81	53.81	1.24
30.80	10.96	64.03	1.33	41.65	5.14	26.91	8.76	55.10	1.10
32.09	10.39	65.78	1.13	43.31	4.53				
34.92	9.26								

Table 4. Values of Parameters (a, b, c, d) of eq 1, Coefficients of Determination (R^2), and Standard Deviations (sd) for the Ethanol/Propan-1-ol/Propan-2-ol−Dipotassium 2,3-Dihydroxybutanedioate/Disodium 2,3-Dihydroxybutanedioate/Diammonium 2,3-Dihydroxybutanedioate/Potassium Sodium 2,3-Dihydroxybutanedioate ATPS at 298.15 K[a]

alcohol−salt systems	a	B	c	d	R^2	100 sd
ethanol−dipotassium 2,3-dihydroxybutanedioate	−0.1423	−3.4443	2.7844	−9.4697	0.9999	0.16
propan-1-ol−dipotassium 2,3-dihydroxybutanedioate	−0.2583	−1.6063	−9.5944	7.3248	0.9992	0.47
propan-2-ol−dipotassium 2,3-dihydroxybutanedioate	−0.2979	−2.1928	−1.9031	−8.5115	0.9998	0.25
propan-2-ol−diammonium 2,3-dihydroxybutanedioate	−0.1817	−2.0689	−2.1057	−7.7044	0.9995	0.38
ethanol−potassium sodium 2,3-dihydroxybutanedioate	−0.0032	−5.0438	5.9179	−12.8643	1.0000	0.06
propan-2-ol−disodium 2,3-dihydroxybutanedioate	−0.2318	−3.6329	1.4394	−18.0670	0.9999	0.16

[a] sd = $(\sum_{i=1}^{n}(w_1^{\text{cal}} - w_1^{\text{exp}})^2/N)^{0.5}$, where w_1 and N represent the mass fraction of ethanol/propan-1-ol/propan-2-ol and the number of binodal data, respectively. w_1^{exp} is the experimental mass fraction of alcohols listed in Tables 2 and 3; w_1^{cal} is corresponding data calculated using eq 1, and the fitting parameters are listed in Table 4.

the binodal data for the investigated ethanol/propan-1-ol/propan-2-ol−2,3-dihydroxybutanedioate ATPS, which can also be proved by the binodal curves reproduced by eq 1 in Figure 1. We have also used eq 1 to correlate binodal data of the ethanol/propan-1-ol/propan-2-ol−$K_3C_6H_5O_7$/$Na_3C_6H_5O_7$/$(NH_4)_3C_6H_5O_7$/Na_2CO_3/K_2CO_3 ATPS.[13,15] On the basis of the experimental results (R^2 and sd), it can be seen that eq 1 also shows satisfactory accuracy in binodal data fitting for these systems. So it can be concluded that eq 1 can be taken as a universally applicable equation to correlate binodal data for the water-miscible alcohol−salt ATPS.

Phase-Separation Abilities of Alcohols. In a practical ATPE process, the extraction efficiency of a target extract is always compared under the same mass of salt or alcohol to find the optimum quantity for the phase-forming substances, so the phase-separation abilities of alcohols were compared by the binodal curves plotted in mass fraction and not plotted in molality in this paper. Then the obtained series of the salting-out abilities of salts and alcohols would be useful for applying to practical production.

The binodal curves for the ethanol/propan-1-ol/propan-2-ol−dipotassium 2,3-dihydroxybutanedioate ATPS, the propan-2-ol−disodium 2,3-dihydroxybutanedioate/diammonium 2,3-dihydroxybutanedioate ATPS, and the ethanol−potassium sodium 2,3-dihydroxybutanedioate ATPS were plotted in Figure 1. The closer the binodal curves to the origin of coordinates, the less the amount of alcohol required for the formation of ATPS under the same concentration of salt. So the phase-separation abilities of alcohols are in accordance with the locations of the binodal curves at the same concentration of a same salt. The closer the binodal curves to the origin, the higher the phase-separation abilities of alcohols. In comparison of the binodal curves of the ethanol−dipotassium 2,3-dihydroxybutanedioate ATPS, the propan-1-ol−dipotassium 2,3-dihydroxybutanedioate ATPS, and the propan-2-ol− dipotassium 2,3-dihydroxybutanedioate ATPS, it can be concluded that the phase-separation abilities of the investigated alcohols are in the order propan-1-ol > propan-2-ol > ethanol, which is in accordance with the series in the ethanol/propan-1-ol/propan-2-ol−$(NH_4)_2SO_4$ ATPS.[8] Although the phase-separation ability of ethanol is higher than that of propan-1-ol and propan-2-ol, the stability of some biomolecules is ethanol is higher than that in propan-1-ol and propan-2-ol.[2] So the selection of the optimum phase-separation alcohols should take into account their phase-separation abilities and biocompatibility.

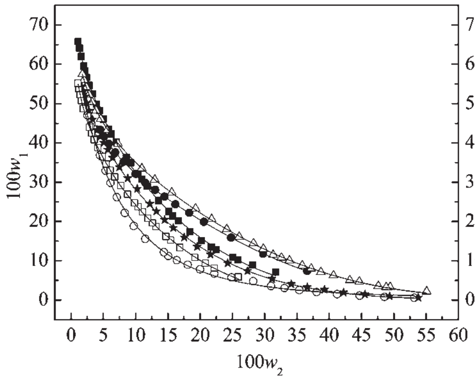

Figure 1. Binodal curves plotted in mass fraction (w) for the alcohols (1) + 2,3-dihydroxybutanedioates (2) ATPS at 298.15 K. ■, propan-2-ol−diammonium 2,3-dihydroxybutanedioate ATPS; ●, ethanol−potassium sodium 2,3-dihydroxybutanedioate ATPS; △, ethanol−dipotassium 2,3-dihydroxybutanedioate ATPS; ○, propan-1-ol−dipotassium 2,3-dihydroxybutanedioate ATPS; ★, propan-2-ol−dipotassium 2,3-dihydroxybutanedioate ATPS; □, propan-2-ol−disodium 2,3-dihydroxybutanedioate ATPS; —, reproduced by eq 1.

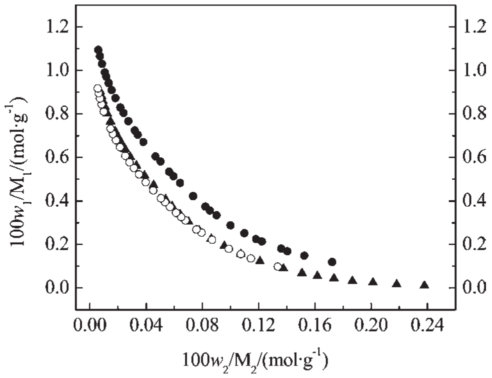

Figure 2. Binodal curves plotted in molality (mass fraction w/molar mass M) for the alcohols (1)−2,3-dihydroxybutanedioates (2) ATPS at 298.15 K. ●, propan-2-ol−diammonium 2,3-dihydroxybutanedioate ATPS; ▲, propan-2-ol−dipotassium 2,3-dihydroxybutanedioate ATPS; ○, propan-2-ol−disodium 2,3-dihydroxybutanedioate ATPS.

Phase-Separation Abilities of 2,3-Dihydroxybutanedioates. The phase-separation abilities of salts were also determined by the comparison of the binodal curves of the investigated alcohol−2,3-dihydroxybutanedioate ATPS. In comparison of the binodal curves of the ethanol−dipotassium 2,3-dihydroxybutanedioate ATPS and the ethanol−potassium sodium 2,3-dihydroxybutanedioate ATPS, it can be concluded that the phase-separation abilities of potassium sodium 2,3-dihydroxybutanedioate is slightly higher than that of dipotassium 2,3-dihydroxybutanedioate, but the two-phase area of the ethanol−dipotassium 2,3-dihydroxybutanedioate ATPS is larger than that of the ethanol−potassium sodium 2,3-dihydroxybutanedioate ATPS. Meanwhile, in comparison of the bindal curves of the propan-2-ol−dipotassium 2,3-dihydroxybutanedioate ATPS, the propan-2-ol−disodium 2,3-dihydroxybutanedioate ATPS, and the propan-2-ol−diammonium 2,3-dihydroxybutanedioate ATPS, it can also be concluded that the phase-separation abilities of the salts are in the order disodium 2,3-dihydroxybutanedioate > dipotassium 2,3-dihydroxybutanedioate > diammonium 2,3-dihydroxybutanedioate. However, the two-phase area of the propan-2-ol−dipotassium 2,3-dihydroxybutanedioate ATPS is larger than the propan-2-ol−disodium 2,3-dihydroxybutanedioate ATPS. In many ATPE experiments, potassium salts are always chosen as the phase-separation salts. This is because the phase-separation abilities of potassium salts are similar to that of sodium salts and the two-phase area of the alcohol−potassium salt ATPS is always larger than that of the corresponding alcohol−sodium salt ATPS. The selection of the optimum phase-separation salts should take into account their phase-separation abilities, two-phase areas, and the pH value of the solutions.

Table 5. Values of Parameters (Scaled EEV V_{213}^{*}) of eq 2, Coefficients of Determination (R), and Standard Deviations (sd) for the Propan-2-ol (1) + 2,3-Dihydroxybutanedioate (2) + Water (3) Systems at 298.15 K

	V_{213}^{*}		
salt	g·mol^{-1}	R	100 sd[a]
diammonium 2,3-dihydroxybutanedioate	333.41	0.9913	2.53
disodium 2,3-dihydroxybutanedioate	399.96	0.9935	1.87
dipotassium 2,3-dihydroxybutanedioate	389.18	0.9959	2.50

[a] sd = $(\sum_{i=1}^{N}(w_1^{\text{cal}} - w_1^{\text{exp}})^2/N)^{0.5}$, where N represent the number of binodal data. w_1^{exp} is the experimental mass fraction of alcohol listed in Tables 2 and 3; w_1^{cal} is the corresponding data calculated using eq 2.

Salting-Out Abilities of K^+, Na^+, and NH_4^+. In this paper, the salting-out abilities of the common cations, K^+, Na^+, and NH_4^+, were compared by the binodal curves plotted in molality to guarantee that the molality of salt ions is the same in the investigated systems. In fact, it can exactly reflect the nature of interactions between salt ions and solvent molecules. The phase-separation salts, including disodium 2,3-dihydroxybutanedioate, dipotassium 2,3-dihydroxybutanedioate, and diammonium 2,3-dihydroxybutanedioate, share a common anion, so the salting-out abilities of cations are in accordance with the locations of binodal curves plotted in molality. The closer the binodal curves to the origin, the higher the salting-out abilities of cations. So in comparison of the locations of binodal curves plotted in molality in Figure 2, it can also be concluded that the salting-out abilities of the investigated cations are in the order $Na^+ \approx K^+ > NH_4^+$.

In the paper, the scaled EEV of disodium 2,3-dihydroxybutanedioate, dipotassium 2,3-dihydroxybutanedioate, and diammonium 2,3-dihydroxybutanedioate in the propan-2-ol−water component solvents was calculated using the model developed by Guan et al.[16] It is based on the concept that macroscopically any molecular species in a solution is distributed at random and every system composition on the binodal curve is a geometrically saturated solution of one solute in the presence of another. The scaled EEV of different salts in the same component solvent have been used to evaluate the salting-out abilities of salts,[3,17] and it can be calculated by the following equation

$$\ln\left(V_{213}^{*}\frac{w_2}{M_2}\right) + V_{213}^{*}\frac{w_1}{M_1} = 0 \qquad (2)$$

where V_{213}^{*}, M_1, and M_2 are the scaled EEV of salts and molar mass of alcohol and salt, respectively. For the investigated salts in the same propan-2-ol−water component solvent, the salting-out abilities of cations increase with the increase of EEV. The calculated results are listed in Table 5. As given in Table 5, the scaled EEV of investigated 2,3-dihydroxybutanedioate in the propan-2-ol−water component solvent is in the order disodium

2,3-dihydroxybutanedioate (399.96 $g \cdot mol^{-1}$) > dipotassium 2,3-dihydroxybutanedioate (389.18 $g \cdot mol^{-1}$) > diammonium 2,3-dihydroxybutanedioate (333.41 $g \cdot mol^{-1}$). Because these salts sharing a common anion, the results also indicate that the salting-out abilities of the investigated cations are in the order $Na^+ \approx K^+ > NH_4^+$, which is in the same order as the locations of the binodal curves plotted in molality.

CONCLUSION

Binodal data for the ethanol/propan-1-ol/propan-2-ol—dipotassium 2,3-dihydroxybutanedioate ATPS, the propan-2-ol—disodium 2,3-dihydroxybutanedioate/diammonium 2,3-dihydroxybutanedioate ATPS, and the ethanol—potassium sodium 2,3-dihydroxybutanedioate ATPS were obtained at 298.15 K. On the basis of the experimental results (R^2 and sd), it can be concluded that the equation $w_1 = \exp(a + bw_2^{0.5} + cw_2 + dw_2^2)$ can be taken as a universally applicable equation to correlate binodal data of the water-miscible alcohol—salt ATPS. In this paper, the salting-out abilities of cations (K^+, Na^+, and NH_4^+) were compared by the scaled EEV of salts (disodium 2,3-dihydroxybutanedioate, dipotassium 2,3-dihydroxybutanedioate, and diammonium 2,3-dihydroxybutanedioate) and the locations of binodal curves plotted in molality. The experimental and calculated results both indicate that the salting-out abilities of the investigated cations are in the order $Na^+ \approx K^+ > NH_4^+$. Considering the application in the practical ATPE process, the phase-separation abilities of alcohols and salts were compared by the binodal curves plotted in mass fraction. It can be concluded from the experimental results that the phase-separation abilities of the investigated salts are in the order disodium 2,3-dihydroxybutanedioate > dipotassium 2,3-dihydroxybutanedioate > diammonium 2,3-dihydroxybutanedioate, and that of the investigated alcohols are in the order propan-1-ol > propan-2-ol > ethanol. The fundamental phase diagrams of ATPS and the series of the phase-separation abilities of the substances in ATPS are both significant to the development of ATPE technology.

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Funding Sources

Financial support from the Natural Science Foundation of China (21076098; 20976074) and the National Science Foundation of Jiangsu Province (BK2010349) is gratefully acknowledged.

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