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Liquid–liquid equilibrium of novel aqueous two-phase systems and evaluation of salting-out abilities of salts

Research Article

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Abstract: Binodal data are beneficial to the design of aqueous two-phase extraction and the establishment of thermodynamic models. For the 2-propanol + Li_2SO_4/Na_2SO_4/ $(NH_4)_2SO_4$/K_3PO_4 + water systems and 1-propanol + K_3PO_4/$K_3C_6H_5O_7$/$(NH_4)_3C_6H_5O_7$ + water systems, binodal data were determined at 298.15 K. The binodal data were correlated by a theoretical equation on the basis of statistical geometry. The salting-out abilities of salts and the phase-separation abilities of alcohols were evaluated by the effective excluded volume of salt and the binodal curves plotted in molality. A simple Hofmeister series of cations and anions were obtained. The organic salt, $K_3C_6H_5O_7$, shows as high a salting-out ability as K_3PO_4.

Keywords: *Aqueous two-phase system • Binodal data • Salting-out ability • 1-Propanol • 2-Propanol*

1. Introduction

Aqueous two-phase systems (ATPS) have been developed in recent years, such as polymer–polymer/salt ATPS, ionic liquid (IL)–salt ATPS and small molecule alcohol–salt ATPS. These novel extraction systems have great potential in biological fields due to their biocompatibility [1,2]. The mutual competition between hydrophilic solvent and salt for water molecules leads to the exclusion of hydrophilic solvents or the crystallization of salts. Reliable binodal data are beneficial to the design of aqueous two-phase extraction and the establishment of thermodynamic models. Meanwhile, these binodal data can also be used to evaluate the salting-out abilities of salts and the phase-separation abilities of hydrophilic solvents.

The study on the salting-out effect of salts or ions is prevalent in various fields. The selection of phase-separation salt with a high salting-out ability is significant for designing aqueous two-phase extraction (ATPE) systems. However, there are no convenient and efficient parameters to evaluate the salting-out abilities of salts for the exclusion of hydrophilic solvents. The published papers always discussed the salting-out abilities of salts or ions by the following two methods:

(1) Comparing the locations of binodal curves plotted in mass fraction is a widely used method [3-5]. So the series of salting-out abilities of salts or ions are obtained under the same mass fraction, not the same molar concentration. It can not exactly reflect the interaction between ions and water molecules, and the obtained series are not exact.

(2) Terms like "Hofmeister series", "kosmotropic ions" or "chaotropic ions" always appear in papers. Some authors discussed the salting-out abilities of salts according to Hofmeister series proposed in other different systems. In fact, many researchers have proved that there's no unique Hofmeister series [6,7]. It will change with the experimental systems. Moreover, the distinction between kosmotropic ions and chaotropic ions is still debated.

In this paper, we extended the application of excluded volume theory [8] to evaluate the salting-out abilities of salts and the phase-separation abilities of hydrophilic solvents. The effective excluded volume (EEV) of salt

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was calculated on the basis of experimental binodal data. Na_2SO_4, Li_2SO_4, $(NH_4)_2SO_4$ and K_2SO_4 were used in order to investigate the salting-out abilities of Na^+, Li^+, NH_4^+ and K^+, which were widely investigated in other fields. Potassium phosphate has shown a high salting-out ability in polymer–salt [9,10] and IL–salt [11-13] ATPS. Citrates have the advantages of biodegradability and are nontoxic compared to traditional phase-forming inorganic salts. The salting-out abilities of K_3PO_4, $K_3C_6H_5O_7$ and $(NH_4)_3C_6H_5O_7$ were also compared in order to discuss the feasibility of organic salts as a substitute for inorganic salts for phase-separation salts.

2. Experimental Procedure

2.1. Chemicals

In this paper, Li_2SO_4, Na_2SO_4, K_2SO_4, $(NH_4)_2SO_4$, $K_3PO_4 \cdot 3H_2O$, $K_3C_6H_5O_7 \cdot H_2O$, $(NH_4)_3C_6H_5O_7$, 1-propanol and 2-propanol were supplied by Sinopharm Chemical Reagent Co., Ltd. with a minimum purity of 99.0%, 99.0%, 99.0%, 99.0%, 99.0%, 99.5%, 99.0%, 99.0% and 99.7%, respectively. All the chemicals were used without further purification. Double distilled and deionized water was used throughout the entire experiment.

2.2. Apparatus and procedures

The binodal curves were plotted by titration method (cloud point method). The Li_2SO_4, Na_2SO_4, $(NH_4)_2SO_4$, K_3PO_4, $K_3C_6H_5O_7$ or $(NH_4)_3C_6H_5O_7$ solution of known concentration was titrated with 2-propanol/1-propanol 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 cm³) was used to carry out the experiment, and the temperature was maintained within 298.15±0.1 K in a water bath. It must be emphasized that the appearance of turbidity is attributed to the exclusion of hydrophilic alcohol, not the crystallization of salts.

3. Excluded volume theory

Excluded volume theory was originally presented in 1993 [8]. A theoretical description of the binodal curves was developed using statistical geometry. 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 is a geometrically saturated solution of one solute in the presence of the other. As for a hydrophilic alcohol (1)–salt (2)–water (3) ternary system, the probability [$P(\underline{V} \ge V_{213})$] of there being no species 2 in an arbitrarily located volume V_{213} is also given by applying Poisson distribution as binary system

$$P(\underline{V} \ge V_{213}) = \frac{(v_1 V_{213})^k}{k!} e^{-v_1 V_{213}} = e^{-v_1 V_{213}} \quad (k=0) \qquad (1)$$

where V_{213} is the "effective excluded volume " of salt and v_1 is the number density of alcohol. The volume of the solution (V) can be divided into two parts

$$V = V(\underline{V} \ge V_{213}) + V(\underline{V} \le V_{213}) \qquad (2)$$

where $V(\underline{V} \ge V_{213})$ is the effective available volume of salt, $V(\underline{V} \le V_{213})$ is the effective unavailable volume of salt in the ternary system. If $P(\underline{V} \ge V_{213})$ is equal to the volume fraction of the effective available volume of salt, then

$$(3) \qquad P(\underline{V} \ge V_{213}) = \frac{V(\underline{V} \ge V_{213})}{V}$$

In combination with Eq. 1 and Eq. 3, the following equation can be deduced

$$(4) \qquad e^{-v_1 V_{213}} = v_2 V_{213}$$

where v_2 is the number density of salt. Considering the existence of space after tight assembly of molecules, Eq. 4 can be expressed by

$$(5) \qquad e^{-v_1 V_{213}} = v_2 V_{213} + f_{213}$$

where f_{213} is the volume fraction of unfilled effective available volume after tight packing of salt into the network of hydrophilic alcohol.

Using the transformation relationship between molecular number density and mass fraction of components

$$v_s = \frac{\rho N_a w_s}{M_s} \quad s=1,2 \qquad (6)$$

where w_s is the mass fraction of component s, ρ is the density of solution, N_a is Avogadro's constant, and M_s is the molar mass of components. The density of solution is treated as a constant, and the scaled effective excluded volume (V_{213}^*) defined by Guan *et al.* is expressed as

$$(7) \qquad V_{213}^* = \rho N_a V_{213}$$

By applying Eq. 6 and 7 in Eq. 5 or 4, two equations can be given respectively

$$(8) \qquad \ln\left(V_{213}^* \frac{w_2}{M_2} + f_{213}\right) + V_{213}^* \frac{w_1}{M_1} = 0$$

$$(9) \qquad \ln\left(V_{213}^* \frac{w_2}{M_2}\right) + V_{213}^* \frac{w_1}{M_1} = 0$$

In the original application, Eq. 9 was used to correlate binodal data of the polymer–polymer systems due to the marked difference in size between the two components. f_{213} will be very small and consequently can be neglected. The f_{213} value depends on the relative geometric shape, size and interaction of unlike molecules. In our application, Eq. 8 was used to calculate the scaled EEV of salts.

Effective excluded volume represents the smallest spacing of hydrophilic alcohol which will accept an individual salt, so it reflects the compatibility of components in the same system. The binodal model developed by Guan *et al*. was used to correlate binodal data of polymer–polymer systems. The scaled EEV of salt has been used to evaluate the salting-out abilities of salts for the exclusion of polymer [10,14], while we extend the application of this model to evaluate the salting-out abilities of salts for the exclusion of hydrophilic solvents and the phase-separation abilities of investigated alcohols. For different salts in the same hydrophilic alcohol–water component solvent, the salting-out ability of salt increases with the increase of EEV; while for the same salt in different component solvents, a larger EEV of salt indicates a higher phase-separation ability of alcohol.

4. Results and Discussion

4.1 Binodal data and binodal curves

Besides K_2SO_4, the aqueous solutions of Li_2SO_4, Na_2SO_4, $(NH_4)_2SO_4$ and K_3PO_4 can all separate into two phases with the addition of 2-propanol at 298.15 K. The addition of K_2SO_4 to aqueous 2-propanol solution does not lead to the exclusion of 2-propanol because the solubility of K_2SO_4 is very low and there are an insufficient number of ions to compete with 2-propanol for water molecules. It cannot be proven that the salting-out abilities of Li^+, Na^+ and NH_4^+ are higher than K^+. For the 2-propanol + Li_2SO_4/Na_2SO_4/$(NH_4)_2SO_4$/K_3PO_4 + water systems and 1-propanol + K_3PO_4/$K_3C_6H_5O_7$/$(NH_4)_3C_6H_5O_7$ systems, the binodal data determined at 298.15 K are listed in Table 1. The binodal curves for the investigated systems are plotted in mass fraction and molality respectively, as shown in Figs. 1 and 2.

4.2 Effective excluded volume and salting-out abilities of salts

The scaled EEV of salts were calculated based on the experimental binodal data, and the results are listed in Table 2. The scaled EEV of K_3PO_4 in the 1-propanol–

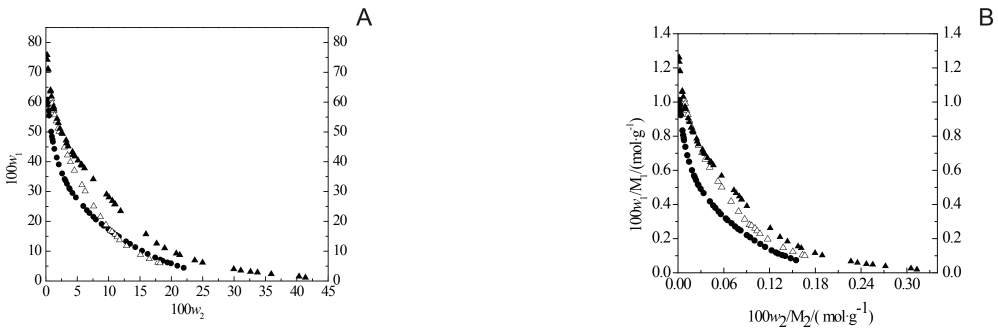

Figure 1. Effect of type of salts on the binodal curves plotted in molality and mass fraction for the 2-propanol (1) + Li_2SO_4/Na_2SO_4/$(NH_4)_2SO_4$ (2) + water (3) systems at 298.15 K. Δ, Li_2SO_4; ●, Na_2SO_4; ▲ $(NH_4)_2SO_4$.

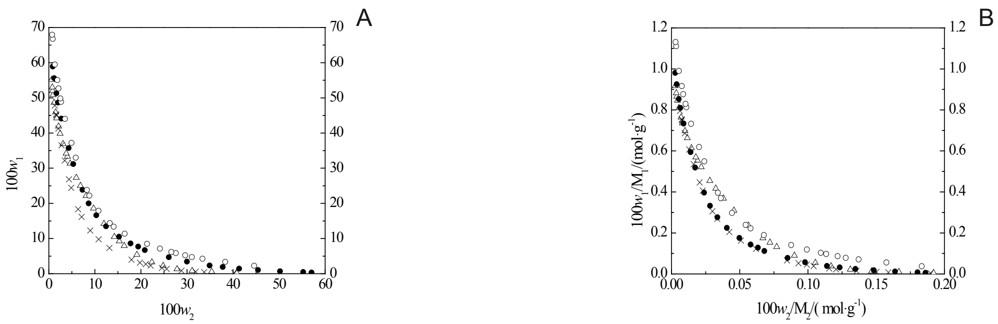

Figure 2. Effect of salt type and alcohols on the binodal curves plotted in mass fraction (a) and molality (b) for the hydrophilic alcohol (1) + salt (2) + water (3) systems at 298.15 K. Δ, 2-propanol–K_3PO_4 system; ×, 1-propanol–K_3PO_4 system; ●, 1-propanol–$K_3C_6H_5O_7$ system; ○, 1-propanol–$(NH_4)_3C_6H_5O_7$ system.

Table 1. Binodal data for the hydrophilic alcohol (1) + salt (2) + water (3) systems at 298.15 K.

$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$	$100w_1$	$100w_2$
	2-propanol–Li_2SO_4			71.13	0.34	37.83	6.16	0.23	40.27	12.30	8.99
61.05	0.91	25.09	7.62	70.88	0.36	34.15	7.56	0.45	33.73	16.16	7.12
59.63	1.02	21.54	8.70	64.02	0.75	29.08	9.68	0.57	31.90	18.35	6.36
57.12	1.23	18.84	9.57	63.63	0.76	28.07	10.06	0.87	28.78	24.43	4.85
55.67	1.41	16.91	10.26	61.92	0.90	26.89	10.60	1.36	25.59	26.85	4.38
53.60	1.64	16.59	10.55	58.48	1.21	25.77	11.06	1.63	24.54	32.17	3.45
51.16	1.96	15.64	11.04	58.27	1.24	25.73	10.96	2.30	22.12	36.50	2.75
50.12	2.11	14.73	11.36	57.59	1.30	23.48	11.92	2.74	21.06	41.27	2.06
44.85	2.95	13.73	11.75	54.43	1.78	15.75	15.96	3.13	19.76	45.06	1.62
42.21	3.45	11.80	12.87	53.03	1.99	12.60	17.66	3.99	17.95	45.31	1.57
39.95	3.92	8.83	15.09	51.08	2.26	11.05	18.95	7.34	13.14	47.81	1.31
37.10	4.55	7.40	16.48	49.81	2.52	9.29	20.80	9.78	10.81		
32.20	5.75	6.24	17.86	49.38	2.65	8.75	21.34		1-propanol–$K_3C_6H_5O_7$		
30.16	6.27	6.05	18.20	47.22	3.23	6.99	23.74	0.35	57.04	10.56	15.22
	2-propanol–Na_2SO_4			46.23	3.32	6.18	24.98	0.49	55.23	13.50	12.41
60.83	0.29	22.85	6.94	45.20	3.55	4.02	29.95	0.72	50.19	16.62	10.29
59.09	0.35	21.57	7.57	43.31	4.25	3.55	31.08	1.08	45.41	20.00	8.65
57.12	0.44	20.66	7.96	42.54	4.40	3.11	32.63	1.48	41.31	23.86	7.31
55.49	0.51	19.16	8.89	42.34	4.48	2.92	33.73	1.92	37.76	31.2	5.29
50.12	0.83	18.55	9.21	41.81	4.65	2.35	35.93	2.35	34.90	35.74	4.28
48.49	0.95	17.43	9.96	40.44	5.12	1.55	40.36	3.45	29.99	44.11	2.71
47.48	1.05	16.35	10.62	39.02	5.60	1.15	41.38	4.69	26.06	48.67	1.98
46.68	1.11	15.47	11.19	38.98	5.70			6.73	20.84	51.30	1.63
44.32	1.35	14.92	11.64		2-propanol–K_3PO_4			7.72	19.40	55.60	1.15
41.41	1.72	13.29	12.79	0.32	40.78	27.32	5.96	8.62	17.74	58.94	0.84
39.11	2.05	12.48	13.41	0.60	35.37	31.36	4.63		1-propanol–$(NH_4)_3C_6H_5O_7$		
36.09	2.58	11.38	14.24	0.81	31.14	33.28	4.05	2.29	44.58	22.22	8.79
34.22	2.99	10.19	15.32	1.33	27.83	34.20	3.78	3.39	38.29	23.82	8.27
33.69	3.12	9.12	16.22	2.26	24.83	36.92	3.09	4.28	33.35	33.01	5.84
32.64	3.35	7.90	17.41	3.28	22.37	39.85	2.46	4.75	31.05	37.20	4.91
31.16	3.71	7.19	18.23	5.40	19.12	41.90	2.11	5.24	29.61	44.04	3.49
30.86	3.81	6.78	18.61	7.95	16.37	44.21	1.71	5.88	27.57	48.88	2.66
29.56	4.22	6.25	19.38	9.24	15.31	45.92	1.50	6.20	26.6	49.85	2.49
28.05	4.77	5.94	19.94	10.52	14.22	48.63	1.15	7.16	24.13	52.70	2.09
25.17	5.95	5.12	20.96	14.19	11.97	50.85	0.94	8.50	21.33	55.08	1.84
23.77	6.52	4.44	21.98	18.57	9.72	51.99	0.82	11.40	16.45	59.50	1.30
	2-propanol–$(NH_4)_2SO_4$			22.16	8.09	53.03	0.75	13.37	14.08	66.79	0.84
75.79	0.21	38.92	5.76	25.04	6.88	55.12	0.59	14.38	13.22	67.98	0.72
74.23	0.23	38.74	5.78		1-propanol–K_3PO_4			17.90	10.77		

water solvent differs from that in the 2-propanol–water solvent. The EEV of salt varies with the size and shape of components as well as the interaction between them, so the EEV of the same salt changes in different hydrophilic alcohol–water component solvents. The salting-out abilities of salts should be compared by the EEV of different salts in the same component solvent. The larger the EEV value, the higher the salting-out ability of salt. It can be concluded from Table 2 that the salting-out abilities of investigated salts are in the order Na_2SO_4 > Li_2SO_4 > $(NH_4)_2SO_4$. Many researchers believe that the salting-out ability of salts is related to Gibbs free energy of hydration for the constituent ions (ΔG_{hyd}), and the ions with a higher salting-out ability has a more negative ΔG_{hyd} value [15,16]. The studied salts, Li_2SO_4 and Na_2SO_4, share a common anion but contain different cations with increasing radii. The ΔG_{hyd} values of Na^+ and Li^+ are -364 and -474 kJ mol^{-1}, respectively [17]. The ΔG_{hyd} values of cation are in the decreasing order, while the salting-out abilities of salts are in the order Na_2SO_4 > Li_2SO_4. So the salting-out abilities of salts cannot be directly related to the ΔG_{hyd} of the constituent ions.

Table 2. Scaled effective excluded volume of salts in the investigated system at 298.15 K.

No.	alcohol–salt	V_{213}* /(g mol^{-1})	100sd[a]	simple Hofmeister series
1	2-propanol–Li_2SO_4	350.53	2.73	cation: $Na^+ > Li^+ > NH_4^+$
	2-propanol–Na_2SO_4	421.34	0.75	
	2-propanol–$(NH_4)_2SO_4$	309.31	1.93	
2	1-propanol–$K_3C_6H_5O_7$	532.16	3.95	anion: $PO_4^{3-} > C_6H_5O_7^{3-}$
	1-propanol–$(NH_4)_3C_6H_5O_7$	442.61	3.99	cation: $K^+ > NH_4^+$
	1-propanol–K_3PO_4	587.58	2.99	
	2-propanol–K_3PO_4	479.22	3.01	alcohol: 1-propanol>2-propanol

[a] $sd = (\sum_{i=1}^{N} (w_1^{cal} - w_1^{\exp})^2 / N)^{0.5}$, where N represent the number of binodal data. w_1^{exp} is the experimental mass fraction of alcohol listed in Table 1, w_1^{cal} is corresponding data calculated using Eq. 8.

The salting-out ability of K_3PO_4 is similar to $K_3C_6H_5O_7$, which indicates that organic citrates will become a good substitute for traditional inorganic salts for use in phase-separation salts of ATPS. A simple Hofmeister series of cation or anion can be obtained as listed in Table 2, because these salts share a common anion or cation. One point needs to be emphasized: the salting-out abilities of investigated salts concluded from EEV reflect their overall salting-out abilities in the range of concentration on the binodal curves plotted in molality, because the EEV are calculated on the basis of binodal data.

In many previous papers, the salting-out abilities of salts or ions are always compared by the binodal curves plotted in mass fraction. In fact, this can not exactly reflect the nature of interaction between molecules in the system. The increase in the scaled EEV is reflected by a decrease in the molality of salt required for the formation of aqueous two-phase systems, which indicates a higher salting-out ability of salt. This relationship can also be deduced from Eq. 8 or 9. In comparison of binodal curves in Figs. 1 and 2, it can be seen that the locations of binodal curves plotted in molality are different from that plotted in mass fraction. When comparing the salting-out abilities of K_3PO_4 and $K_3C_6H_5O_7$ by the binodal curves plotted in mass fraction, K_3PO_4 shows a much higher salting-out ability than $K_3C_6H_5O_7$; nevertheless, when comparing their salting-out abilities by the binodal curves plotted in molality, K_3PO_4 only show a slightly higher salting-out ability than $K_3C_6H_5O_7$. The salting-out ability of $K_3C_6H_5O_7$ is similar to K_3PO_4. It can be concluded from Figs. 1 and 2 that the series of salting-out abilities of investigated salts are also in the same order as EEV. The two-phase area of $(NH_4)_2SO_4$ is larger than Na_2SO_4 or Li_2SO_4 due to a higher solubility of $(NH_4)_2SO_4$ compared with the other two salts.

4.3 Effective excluded volume and phase-separation abilities of alcohols

The EEV of the same salt varies in different hydrophilic alcohol–water component solvents. For the same salt in different component solvents, the phase-separation ability of alcohol increases with the increase of EEV. As proposed by Guan *et al.*, irregular molecular shapes are expected to reduce the EEV value. The EEV of K_3PO_4 in 1-propanol–water component solvent is higher than that in 2-propanol–water component solvent. So the phase-separation abilities of the investigated alcohols are in the order 1-propanol > 2-propanol, which is also proven by the binodal curves plotted in molality in Fig. 2. The increase in EEV is reflected by a decrease in the concentration of alcohol required for the formation of ATPS.

1-propanol and 2-propanol can dissolve in water in any proportion due to “hydrogen bonding” interactions. The magnitude of the “hydrogen bonding” interaction is in accordance with the polarity of alcohol. Permittivity (ε) is an easily available parameter for evaluating the “polarizing capability” of solvents. The ε_r value of 1-propanol and 2-propanol can be calculated with an equation of the form [18]

$$\varepsilon_r(T) = a + bT + cT^2 + dT^3 \qquad (10)$$

The ε_r values are in the order 1-propanol (20.52) > 2-propanol (19.27), which indicates that the polarity of 1-propanol is higher than 2-propanol. Generally speaking, the solvent with higher polarity is easier to dissolve in water. However, when comparing EEV and binodal curves, it can be concluded that 1-propanol is easier to be excluded to the alcohol-rich phase compared with 2-propanol. In fact, the forces of alcohol molecules action on themselves should not be neglected, as it is also very important for phase-separation. “Boiling point” is an easily available and efficient criterion for evaluating the intensity of these self-interaction forces (including Van der Waals forces and hydrogen-bonding forces) between alcohol molecules. The boiling point of 1-propanol is much higher than 2-propanol. The boiling points of 1-propanol and 2-propanol are 97.2 and 82.3°C, respectively. The difference in boiling point temperature between 1-propanol and 2-propanol is approximately 15°C, which reflects that the intensity of acting force between 1-propanol molecules themselves is much higher than 2-propanol molecules. This may lead to an easier exclusion of 1-propanol compared with 2-propanol.

5. Conclusion

Binodal data of the 2-propanol + Li_2SO_4/Na_2SO_4/$(NH_4)_2SO_4$/K_3PO_4 + water systems and the 1-propanol + K_3PO_4/$K_3C_6H_5O_7$/$(NH_4)_3C_6H_5O_7$ systems were experimentally determined at 298.15 K. The obtained binodal data are beneficial to the design of aqueous two-phase extraction and the establishment of thermodynamic models. Organic salts, such as citrates, show great potential for substituting traditional phase-forming inorganic salts due to their biodegradability and nontoxic character, as well as high salting-out abilities.

The effective excluded volume of salts and the locations of binodal curves plotted in molaity are efficient and convenient parameters to evaluate the salting-out abilities of salts and the phase -separation abilities of hydrophilic solvents. A large amount of verified binodal data can be downloaded from the Thermodynamics Research Center (TRC) of the National Institute of Standards and Technology (NIST), which was especially beneficial for this evaluation

Abbreviations

w_s	- Mass fraction
M_s	- Molar mass
ν	- Number density
V_{213}	- Effective excluded volume of salt
$V_{213}{}^*$	- Scaled effective excluded volume of salt
V	- Volume of solution
$V(\underline{V} \geq V_{213})$	- Effective available volume of salt
$V(\underline{V} \leq V_{213})$	- Effective unavailable volume of salt
$P(\underline{V} \geq V_{213})$	- Probability of there being no species 2 in an arbitrarily located volume V_{213}
N_a	- Avogadro's constant
f_{213}	- Volume fraction of unfilled effective available volume
Greek letters	
ρ	- Density of solution
Subscripts	
1	- Hydrophilic alcohol
2	- Salt
3	- Water
Superscripts	
b	- Bottom phase
t	- Top phase
cal	- Calculated value
exp	- Experimental value

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