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Correlation of surface tension of mixed solvents with solvent composition

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Dedicated to the memory of Professor DJ. Farid, who passed away on the 23rd of November 2003

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A simple computational method for calculating surface tension of solvent mixtures based on the Redlich-Kister extension was proposed. The model was applied to the experimental surface tension of binary solvent mixtures and showed accurate results. Overall average percentage deviation (APD) between calculated and experimental surface tensions was calculated as an accuracy criterion. The overall APD for correlating surface tensions in binary solvents was 4.30%. The accuracy of the proposed model has also been compared with those of previously published models and the results showed that the proposed model was superior and capable of providing more accurate results. An extension of the model was also proposed to correlate surface tension of ternary solvents and the overall APD for ternary solvent data was 2.06%.

1. Introduction

In the simplest applications of the surface tension in the pharmaceutical area, it could be considered as the most important parameter for counting the number of drops per ml from aqueous/mixed solvent based ear/eye/oral drop formulations. Surface tension determines the degree of dispersion and the dynamics of motion of liquids in nanopores. It has also been employed in the theoretical description of retention in high performance liquid chromatography (Cheong and Carr 1987).

Ab initio prediction of the surface tension of solvent systems is not possible at the present time, however, it is possible to calculate surface tension after collecting a number of experimental data and then prediction of surface tension using interpolation techniques. The aim of this work was to provide an accurate model to correlate surface tension of mixed solvents and test its applicability on real experimental data collected from the literature.

2. Investigations, results and discussion

2.1. Computational methods

A solution model (i.e. the combined nearly ideal binary solvent/Redlich-Kister equation, CNIBS/R-K) presented by Acree (1992) was used to correlate different physicochemical properties in mixed solvent systems; including the solubility of polycyclic aromatic hydrocarbons in nonaqueous binary solvent mixtures (Acree 1995), solubility of drugs in water-cosolvent mixtures (Jouyban-Gh. et al. 1999), electrophoretic mobility of analytes in mixed solvent electrolyte systems (Jouyban-Gh. et al. 2000a; 2003a; 2003b), the instability rate constants in binary solvent systems (Jouyban et al. 2002a), the acid dissociation constants in water-organic solvent mixtures (Jouyban et al. 2002b) and the dielectric constants of solvent mixtures (Jouyban et al. 2004). Theoretical basis of the CNIBS/R-K equation for describing the chemical potential of solutes dissolved in mixed solvents (Acree 1992) and acid dissociation constants in aqueous-organic mixtures (Jouyban et al. 2002b) has been provided in earlier papers. As noted above, Jouyban et al. extended the applications of the basic solution model; therefore, it could be called the Jouyban-Acree model (JAM). The model constants of the JAM represent differences in the various solute-solvent and solvent-solvent interactions in the mixture (Acree 1992). Therefore, it should be possible to calculate any other physico-chemical property in mixed solvents, which is a function of solute-solvent and/or solvent-solvent interactions. Based on this assumption, an adopted form of the equation representing the surface tension of a binary solvent mixture is proposed as:

$$\ln\gamma_m = f_1 \ln\gamma_1 + f_2 \ln\gamma_2 + f_1 f_2 \sum_{i=0}^{2} K_i (f_1 - f_2)^i \quad (1)$$

Where γ_m, γ_1 and γ_2 are the surface tensions of the mixture and solvents 1 and 2, respectively, f_1 and f_2 are the volume (weight or mole) fractions of solvents 1 and 2 in the mixture and K_i represent the model constants calculated using a no intercept least square method (Jouyban-Gh and Hanaee 1997).

Extension of the model to surface tensions of binary mixtures departs from previous applications, which were limited for the most part to partial molar quantities. Surface tension of binary mixtures is more of an integral “extra-thermodynamic” property (Acree and Bertrand 1983). The theoretical basis of extra-thermodynamic properties is not as rigorous as that of true thermodynamic properties. Consequently, it might be best to view eq. (1) more as a mathematical representation, rather than an equation derived from a rigorous thermodynamic model. This would be consistent with our application of the JAM to physical properties like dielectric constants.

The calculated surface tensions were compared with experimental (observed) values and mean of the absolute percentage deviation (APD) was used as an accuracy criterion. The APD was calculated using:

$$\mathrm{APD} = \frac{100}{(\mathrm{N}-\mathrm{Q})} \sum \frac{|\mathrm{Calculated}-\mathrm{Observed}|}{\mathrm{Observed}} \qquad (2)$$

where N is the number of data points in each set and Q is the number of constant terms of the corresponding equation. The previously presented equations for correlating surface tension of mixed solvents from the literature are as follows:

Cheong and Carr (1987) used the Redlich-Kister extension to correlate the excess surface tension of the mixture (γ^E) with the solvent composition. γ^E is:

$$\gamma^E = \gamma_m - (f_1\gamma_1 + f_2\gamma_2) \qquad (3)$$

and the Redlich-Kister extension is:

$$\gamma^E = (f_1(1-f_1))\,[S_0 + S_1(1-2f_1) + S_2(1-2f_1)^2] \qquad (4)$$

Where S_0–S_2 are the model constants calculated by regressing $\gamma^E/[f_1(1-f_1)]$ versus $(1-2f_1)$ and $(1-2f_1)^2$ using a least square analysis (Cheong and Carr 1987). Another numerical method has been proposed to compute the model constants of Redlich-Kister (1948) extension which regresses γ^E versus $f_1(1-f_1)$, $f_1(1-f_1)(1-2f_1)$ and $f_1(1-f_1)(1-2f_1)^2$ using a no intercept least square analysis (Jouyban-Gh and Hanaee 1997).

Cheong and Carr (1987) also reported the surface tension of mixed solvent as a polynomial function of solvent composition as:

$$\ln\gamma_m = A + \sum_{i=1}^{m} B_i f_1^i \qquad (5)$$

where A and B_i are the model constants.

Connors and Wright (1989) proposed eq. (6):

$$\gamma_m = \gamma_1 - \left[1 + \frac{bf_1}{1-af_1}\right] f_2(\gamma_1 - \gamma_2) \qquad (6)$$

where a and b are the model constants. It has been used in many published works (e.g. Alvarez et al. 1998; 2003; Vazquez et al. 1996; 1997) as eq. (7).

$$\frac{\gamma_1 - \gamma_m}{\gamma_1 - \gamma_2} = \left(1 + \frac{bf_1}{1-af_2}\right) f_2 \qquad (7)$$

In addition to the above-mentioned equations, there are a number of complicated equations of state (e.g. see Escobedo and Manssori 1998), which require a high degree of knowledge of physico-chemical properties such as critical temperature, critical pressure, acentric factor, and the critical compressibility factor, which are not easily available. In addition the models require complex computational procedures that are not suitable in the pharmaceutical area where the researchers would like to employ an easy to use model. Therefore, we ignored these equations in the comparison of the models in this study.

2.2. Computational results and discussion

Details of experimental surface tensions for various aqueous binary solvents were shown in Table 1. Different equations were fitted to experimental data and the back-calculated surface tensions were used to calculate APDs, which were shown in Table 2. The minimum and maximum APDs for the proposed model were 0.33 (water-acetonitrile, 25 °C) and 11.62% (water-acetone, 38 °C), respectively. The overall APD ± standard deviation was 4.30 ± 2.91%. The minimum APDs for eqs. (4, original version), (4, modified version), (5) and (6) were 1.06, 0.58, 0.50 and 0.02%, respectively, and the corresponding maximum values were 99.06, 19.76, 12.72 and 63.87%. As noted in the computational methods, the model constants of eq. (4) could be calculated using a classical least square method (Cheong and Carr 1987) and a no intercept least square method (Jouyban-Gh and Hanaee 1997). The latter provided more accurate results, therefore, it is suggested to use a no intercept least square method in using Redlich-Kister extension for describing the mixed solvent systems behaviors. The only difference between JAM and eq. (4) is that, in JAM, we have used a logarithmic transformation of the surface tensions.

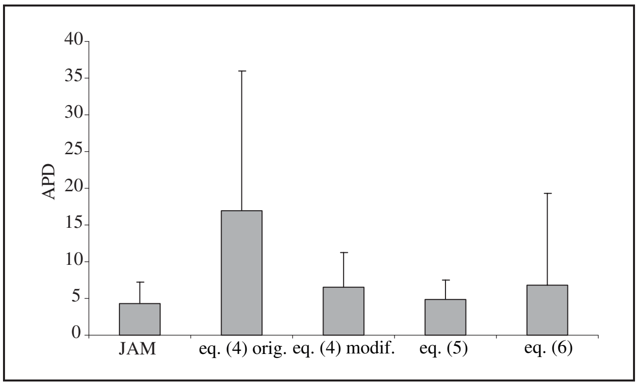

Fig. 1: Mean and standard deviation of average percentage deviation (APD) values for eqs. studied

The overall APDs and their standard deviations were shown in Fig. 1. The proposed equation (JAM) produced the most accurate results, followed by eqs. (5), (4, modified), (6) and (4, original). The mean APD differences between JAM and eqs. (4, original and modified versions) and (6) were statistically significant (paired t-test, $p < 0.004$). In addition the mean difference between eqs. (4, modified) and (4, original) was statistically significant (paired t-test, $p < 0.0005$). However, there was no significant difference between eq. (5) and JAM (paired t-test, $p > 0.11$). Employing more curve-fitting parameters provides more accurate results for JAM. Fig. 2 shows APD values for JAM and eq. (5) using different numbers of

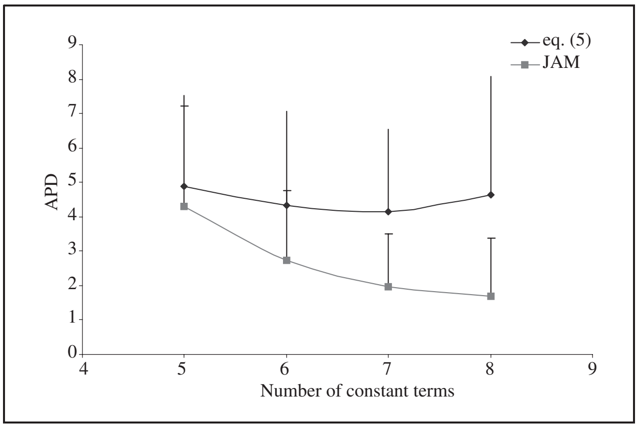

Fig. 2: Mean and standard deviation of average percentage deviation (APD) values for JAM and eq. (5) for various number of constant terms

Table 1: Details of experimental data sets including the name of cosolvent, references, the number of data points in each set (N), temperature (T) and surface tensions of water (solvent 1) and cosolvent (solvent 2)

No.	Cosolvent (solvent 2)	Reference	N[a]	T	γ_1	γ_2
1	Acetone	Howard and McAllister 1957	16	20	72.74	23.24
2	Acetone	Howard and McAllister 1957	9	25	71.94	22.57
3	Acetone	Howard and McAllister 1957	16	38	69.91	20.88
4	Acetone	Howard and McAllister 1957	18	50	68.05	19.57
5	Acetonitrile	Cheong and Carr 1987	13	25	71.66	28.49
6	1-Amino-2-propanol	Alvarez et al. 2003	12	25	72.01	37.38
7	1-Amino-2-propanol	Alvarez et al. 2003	12	30	71.21	36.03
8	1-Amino-2-propanol	Alvarez et al. 2003	12	35	70.42	34.89
9	1-Amino-2-propanol	Alvarez et al. 2003	12	40	69.52	33.68
10	1-Amino-2-propanol	Alvarez et al. 2003	12	45	68.84	32.49
11	1-Amino-2-propanol	Alvarez et al. 2003	12	50	67.92	31.28
12	2-Amino-2-methyl-1-propanol	Vazquez et al. 1997	14	25	72.01	31.37
13	2-Amino-2-methyl-1-propanol	Vazquez et al. 1997	14	30	71.21	30.80
14	2-Amino-2-methyl-1-propanol	Vazquez et al. 1997	14	35	70.42	30.20
15	2-Amino-2-methyl-1-propanol	Vazquez et al. 1997	14	40	69.52	29.64
16	2-Amino-2-methyl-1-propanol	Vazquez et al. 1997	14	45	68.84	29.04
17	2-Amino-2-methyl-1-propanol	Vazquez et al. 1997	14	50	67.92	28.44
18	3-Amino-1-propanol	Alvarez et al. 2003	12	25	72.01	43.90
19	3-Amino-1-propanol	Alvarez et al. 2003	12	30	71.21	42.77
20	3-Amino-1-propanol	Alvarez et al. 2003	12	35	70.42	41.61
21	3-Amino-1-propanol	Alvarez et al. 2003	12	40	69.52	40.48
22	3-Amino-1-propanol	Alvarez et al. 2003	12	45	68.84	39.33
23	3-Amino-1-propanol	Alvarez et al. 2003	12	50	67.92	38.16
24	tert-Butanol	Cheong and Carr 1987	13	25	71.66	19.93
25	Diethanolamine	Vazquez et al. 1996	11	25	72.01	47.21
26	Diethanolamine	Vazquez et al. 1996	11	30	71.21	46.46
27	Diethanolamine	Vazquez et al. 1996	11	35	70.42	45.66
28	Diethanolamine	Vazquez et al. 1996	11	40	69.52	44.75
29	Diethanolamine	Vazquez et al. 1996	11	45	68.84	44.05
30	Diethanolamine	Vazquez et al. 1996	11	50	67.92	43.12
31	Dimethyl sulphoxide	Cheong and Carr 1987	13	25	71.66	42.70
32	Dioxane	Hovorka et al. 1936	11	20	72.75	33.39
33	Dioxane	Hovorka et al. 1936	11	25	71.97	32.85
34	Dioxane	Hovorka et al. 1936	11	30	71.18	32.19
35	Dioxane	Hovorka et al. 1936	11	40	69.56	30.73
36	Dioxane	Hovorka et al. 1936	11	50	67.91	29.35
37	Dioxane	Hovorka et al. 1936	11	60	66.18	27.81
38	Dioxane	Hovorka et al. 1936	11	70	64.40	26.25
39	Dioxane	Hovorka et al. 1936	8	80	62.60	24.76
40	Isopropanol	Cheong and Carr 1987	13	25	71.66	20.82
41	Methanol	Weast 1972	9	20	72.75	22.65
42	Methanol	Weast 1972	9	30	71.18	21.58
43	Methanol	Weast 1972	9	50	67.91	19.52
44	Methanol	Cheong and Carr 1987	13	25	71.66	22.35
45	N-Methyldiethanolamine	Alvarez et al. 1998	14	25	72.01	38.90
46	N-Methyldiethanolamine	Alvarez et al. 1998	14	30	71.21	38.10
47	N-Methyldiethanolamine	Alvarez et al. 1998	14	35	70.42	37.62
48	N-Methyldiethanolamine	Alvarez et al. 1998	14	40	69.52	37.31
49	N-Methyldiethanolamine	Alvarez et al. 1998	14	45	68.84	37.16
50	N-Methyldiethanolamine	Alvarez et al. 1998	14	50	67.92	36.88
51	Monoethanolamine	Vazquez et al. 1997	13	25	72.01	48.95
52	Monoethanolamine	Vazquez et al. 1997	13	30	71.21	48.14
53	Monoethanolamine	Vazquez et al. 1997	13	35	70.42	47.34
54	Monoethanolamine	Vazquez et al. 1997	13	40	69.52	46.43
55	Monoethanolamine	Vazquez et al. 1997	13	45	68.84	45.73
56	Monoethanolamine	Vazquez et al. 1997	13	50	67.92	44.81
57	Tetrahydrofuran	Cheong and Carr 1987	13	25	71.66	26.88
58	Triethanolamine	Vazquez et al. 1996	11	25	72.01	45.95
59	Triethanolamine	Vazquez et al. 1996	11	30	71.21	45.16
60	Triethanolamine	Vazquez et al. 1996	11	35	70.42	44.38
61	Triethanolamine	Vazquez et al. 1996	11	40	69.52	43.48
62	Triethanolamine	Vazquez et al. 1996	11	45	63.38	42.80
63	Triethanolamine	Vazquez et al. 1996	11	50	67.92	41.88

[a] N is the number of data points in each set

Table 2: Average percentage deviation (APD) for different equations

No.[a]	JAM	eq. (4) original	eq. (4) modified	eq. (5)	eq. (6)
1	7.83	36.55	14.95	7.15	1.34
2	7.97	31.80	16.86	12.72	63.87
3	11.62	99.00	19.76	8.52	1.86
4	10.16	99.06	17.58	8.61	1.56
5	0.33	1.88	1.24	0.50	3.19
6	3.72	14.03	5.08	4.44	0.07
7	4.18	16.40	5.72	4.78	0.03
8	4.70	18.95	6.43	5.21	0.04
9	5.00	20.53	6.88	5.48	0.02
10	5.50	23.22	7.60	5.86	0.02
11	6.01	26.28	8.32	6.17	0.10
12	10.22	40.10	14.65	9.13	0.13
13	10.15	40.00	14.64	9.14	0.15
14	10.09	39.74	14.63	9.15	0.16
15	10.00	39.39	14.57	9.15	0.15
16	10.05	39.86	14.74	9.22	0.16
17	9.97	39.61	14.70	9.23	0.18
18	0.89	2.71	1.29	1.57	0.02
19	1.20	3.74	1.68	1.90	0.04
20	1.31	4.16	1.84	2.07	0.02
21	1.52	4.81	2.11	2.33	0.05
22	1.86	6.24	2.55	2.62	0.04
23	2.07	6.92	2.82	2.80	0.04
24	4.95	27.58	14.25	7.74	0.40
25	2.32	6.29	2.85	3.15	10.33
26	2.35	6.40	2.90	3.18	10.51
27	2.39	6.51	2.95	3.22	10.72
28	4.77	7.65	3.53	3.79	10.20
29	2.46	6.77	3.05	3.32	0.03
30	2.49	6.92	3.11	3.37	10.45
31	0.50	1.06	.58	0.99	0.29
32	3.08	7.38	5.39	5.70	0.35
33	3.07	7.48	5.47	5.95	0.47
34	3.17	7.73	5.64	6.11	0.30
35	3.25	7.74	5.76	6.17	0.51
36	3.21	8.11	5.92	6.42	0.55
37	3.43	8.61	6.29	6.60	0.40
38	3.43	8.84	6.45	6.83	0.58
39	3.94	8.98	7.49	8.88	40.35
40	2.67	15.86	8.30	3.84	2.53
41	1.54	2.19	1.76	1.63	20.39
42	1.47	2.42	1.76	1.55	20.46
43	1.68	2.83	1.97	1.81	0.08
44	0.64	2.51	1.24	0.60	0.84
45	6.33	25.89	8.01	4.94	20.85
46	6.46	25.78	8.19	4.95	20.28
47	6.32	23.92	8.04	4.96	20.17
48	5.91	23.22	7.60	4.56	20.96
49	5.85	22.12	7.60	4.65	20.60
50	6.10	22.25	8.01	5.33	20.67
51	1.99	5.04	2.42	2.14	0.04
52	2.01	5.10	2.45	2.17	0.04
53	5.19	8.13	5.81	5.41	0.53
54	2.04	5.21	2.51	2.21	0.04
55	2.06	5.25	2.53	2.24	0.04
56	2.08	5.34	2.56	2.26	0.04
57	2.87	11.05	6.08	3.24	0.71
58	3.37	10.42	4.47	4.74	20.60
59	3.41	10.59	4.54	4.79	0.03
60	3.45	10.76	4.61	4.85	0.04
61	3.49	10.99	4.69	4.91	0.03
62	3.52	11.15	4.75	4.98	0.03
63	3.48	11.43	4.84	5.04	0.12

[a] Details of the set numbers are the same as in Table 1

constant terms. For JAM, two of the constant terms are γ_1 and γ_2, and as shown in the figure for JAM, the more curve-fitting parameters the more accurate the results. This is because of an appropriate independent variable arrangement of JAM. However, this is not the case for eq. (5), as overall APD decreases slightly from five constant terms and then increases again from seven to eight constant terms version of the model.

The JAM could be extended for correlating surface tension of ternary solvents as:

$$\begin{aligned}\mathrm{In}\gamma_m = {} & f_1\ln\gamma_1 + f_2\ln\gamma_2 + f_3\ln\gamma_3 + f_1f_2\sum_{i=0}^{2} K_i\,(f_1 - f_2)^i \\ & + f_1f_3\sum_{i=0}^{2} K'_i\,(f_1 - f_3)^i f_2f_3\sum_{i=0}^{2} K''_i\,(f_2 - f_3) \\ & + f_1f_2f_3\sum_{i=0}^{2} K'''\sum_{i=0}^{2} K'''_i\,(f_1 - f_2 - f_3)^i \qquad (8)\end{aligned}$$

where γ_3 and f_3 are surface tension of solvent 3 and volume (weight or mole) fraction of solvent three in the mixture and K'_i, K''_i and K'''_i are the model constants. The model was applied to the available ternary solvent surface tension data of water – glycerin – ethanol (Ernst et al. 1936), water – *n*-butyl acetate – methanol and water – *n*-pentyl acetate – methanol (Santos et al. 2003) and APDs obtained were 2.98% (N = 66 and Q = 15), 2.34% (N = 83, Q = 15) and 0.87% (N = 42, Q = 15), respectively, and the overall APD was 2.06%. In our past studies involving the mathematical description of solute solubilities in binary solvents, we noted that it was possible to use the calculated regression parameters determined from binary solvent solubility data to predict the solubility of the solute in ternary solvent mixtures (Jouyban-Gh. et al. 2000b). While there is insufficient ternary mixture surface tension data available in the chemical and pharmaceutical literature to test this possible application, we hypothesize that the ternary mixture form of the JAM should provide reasonably accurate predictions of the surface tensions of ternary mixtures.

In conclusion, the JAM is capable of correlating other physico-chemical properties in solvent mixtures, and the results obtained in this study recommend it for correlating surface tension calculations in practice, when an accurate model is needed to screen the experimentally determined surface tensions for detecting possible outliers and/or to predict the surface tension of solvent mixtures at any solvent compositions after training the model using a minimum number of data points, i.e. five points. The best combination of the training set could be considered as: $f_1 = 0.00$, 0.25, 0.50, 0.75 and 1.00 for the proposed model.

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