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ab initio Solubility Prediction of Non-Electrolytes in Ternary Solvents Using a Combination of Jouyban-Acree and Abraham Models

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Applicability of a previously trained model derived from a combination of the Jouyban-Acree and Abraham models for predicting the solubility of non-electrolytes in ternary solvent mixtures was shown using 80 data sets of anthracene and pyrene. A sample program written in SPSS environment was presented in Appendix. The accuracy criterion was mean percentage deviation (MPD) and using experimental solubility data in neat solvents, *i.e.* three data points for each set, the overall MPDs (±SD) were 7.5 (± 5.5) % and 20.2 (± 13.0) %, respectively for water to solvent and gas to solvent parameters of Abraham model. The corresponding MPDs for full predictive versions of the models were 27.5 (± 18.2) % and 20.8 (± 19.5) %. The results showed that one can use the proposed method to predict solubility of non-electrolytes in ternary solvents and the expected prediction errors are quite acceptable specially with employing experimental solubility of solutes in neat solvents as input data.

Key Words: Solubility, Prediction, Cosolvency, Non-electrolytes, Abraham model, Jouyban-Acree model.

INTRODUCTION

The solubility of a solute is one of the most important physico-chemical properties and increased/decreased solubility is required in various chemical processes. Addition of the second solvent is the most common solubility alteration method. When binary solvents are not able to dissolve a desired amount of a solute in a given volume, addition of the third solvent can be used. Experimental solubility of anthracene and pyrene in ternary solvents

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was reported in previous papers[1-18]. In addition to the experimental efforts, a number of computational methods have been developed to calculate the solubility in ternary solvent systems[19-23].

The experimental solubility of a solute in a ternary solvent system could be represented by various mathematical models that are briefly discussed in a previous paper[20]. The most reliable model was the Jouyban-Acree model:

$$\ln X_m = f_1 \ln X_1 + f_2 \ln X_2 + f_3 \ln X_3 + f_1 f_2 \sum_{i=0}^{n} B_i (f_1 - f_2)^i + f_1 f_3 \sum_{i=0}^{n} B_i' (f_1 - f_3)^i + f_2 f_3 \sum_{i=0}^{n} B_i'' (f_2 - f_3)^i \quad (1)$$

where X is the mole fraction solubility of the solute, f denotes the mole fraction of the solvents 1-3 in the solvent mixture, subscripts m and 1-3 are the mixed solvent and solvents 1, 2 and 3, respectively and B_i, B_i' and B_i'' are the sub-binary interaction terms[1-18,20]. Eqn. 1 provided good correlations between solvent composition and the solubility of solutes in ternary solvent mixtures[1-18,20,21]. The main drawback of the equation was its constant terms, which should be calculated using experimental solubility data. In a previous report, we have shown that the model is able to provide reasonably accurate predictions using a minimum number of experimental data in binary and ternary solvents[21]. To overcome this limitation of the Jouyban-Acree model, the sub-binary interaction terms were correlated with the solvent coefficients of Abraham model, therefore, no need for experimental data for calculating the model constants[23]. The equation obtained using water to solvent coefficients (c, r, s, b and v) was as:

$$B_{j,i} = A_{0,i} + A_{1,i}(c_1 - c_2)^2 + A_{2,i}(r_1 - r_2)^2 + A_{3,i}(s_1 - s_2)^2 + A_{4,i}(b_1 - b_2)^2 + A_{5,i}(v_1 - v_2)^2 \quad (2)$$

The corresponding equation for gas to solvent coefficients (c, r, s, b and l) was:

$$B_{j,i} = J_{0,i} + J_{1,i}(c_1 - c_2)^2 + J_{2,i}(r_1 - r_2)^2 + J_{3,i}(s_1 - s_2)^2 + J_{4,i}(b_1 - b_2)^2 + J_{5,i}(l_1 - l_2)^2 \quad (3)$$

The B terms from eqns. 2 and 3 were used along with experimental solubility of anthracene in neat solvents 1-3 to predict the solubility of anthracene in ternary solvent systems where the overall mean percentage deviations (MPD) were 10.7 and 9.3 %, respectively[23]. To provide an *ab initio* solubility prediction method, the solubility of anthracene was predicted using basic models of Abraham[24]:

$$\log\left(\frac{C_S}{C_W}\right) = c + r \cdot R_2 + s \cdot \pi_2^H + a \cdot \sum \alpha_2^H + b \cdot \sum \beta_2^H + v \cdot V_X \tag{4}$$

$$\log\left(\frac{C_S}{C_G}\right) = c + r \cdot R_2 + s \cdot \pi_2^H + a \cdot \sum \alpha_2^H + b \cdot \sum \beta_2^H + l \cdot \log L^{(16)} \tag{5}$$

where C_S and C_W are the solute solubility in the organic solvent and water (in mole per liter), respectively, C_G is the gas phase concentration of the solute, R_2 is the excess molar refraction, π_2^H is dipolarity/polarizability of solute, $\Sigma\alpha_2^H$ denotes the solute's hydrogen-bond acidity, $\Sigma\beta_2^H$ stands for the solute's hydrogen-bond basicity, V_X is the McGowan volume of the solute, and $\log L^{(16)}$ is the logarithm of the solute gas-hexadecane partition coefficient at 298.15 K. In eqns. 4 and 5 the coefficients c, r, s, a, b, v and l are the model constants (*i.e.* solvent's coefficients), which depend upon the solvent system under consideration. Numerical values of the model constants have been reported in the literature[24] for several water-to-organic solvent and gas-to-organic solvent systems.

The overall MPDs of eqns. 2 and 3 were 23.9 and 22.5 %, respectively[23]. As noted above, these equations were obtained using solubility of anthracene in various non-aqueous binary solvents. In a recent paper[25], the computed B terms using experimental solubility data of five polycyclic aromatic hydrocarbons were employed to construct quantitative structure property relationship (QSPR) models using Abraham model coefficients of the solvents and solute's descriptors. The general forms of the proposed QSPR models were:

$$\begin{aligned} B_{j,i} &= M_{0,i} + M_{1,i}(c_1 - c_2)^2 + M_{2,i}R_2(r_1 - r_2)^2 \\ &\quad + M_{3,i}\pi_2^H(s_1 - s_2)^2 + M_{4,i}\sum \beta_2^H(b_1 - b_2)^2 + M_{5,i}V_X(v_1 - v_2)^2 \end{aligned} \tag{6}$$

and

$$\begin{aligned} B_{j,i} &= K_{0,i} + K_{1,i}(c_1 - c_2)^2 + K_{2,i}R_2(r_1 - r_2)^2 \\ &\quad + K_{3,i}\pi_2^H(s_1 - s_2)^2 + K_{4,i}\sum \beta_2^H(b_1 - b_2)^2 + K_{5,i}\log L^{(16)}(l_1 - l_2)^2 \end{aligned} \tag{7}$$

where $M_{0,i}$-$M_{5,i}$ and $K_{0,i}$-$K_{5,i}$ are the model constants, $(c_1–c_2)^2$, $R_2(r_1–r_2)^2$, $\pi_2^H(s_1–s_2)^2$, $\Sigma\beta_2^H(b_1–b_2)^2$, $V_X(\nu_1–\nu_2)^2$ and $\log L^{(16)}(l_1–l_2)^2$ terms could be calculated using the numerical values of the solute's descriptors and solvents' coefficients. The advantages of eqns. 6 and 7 are that they possess a number of solute's descriptors and their applicability could be extended to the solutes other than anthracene. The overall MPD of the combined version of the Jouyban-Acree model with eqns. 6 and 7 for predicting 194 data sets of anthracene, pyrene, benzil, *trans*-stilbene and thianthrene were 4.7 and

11.2 % using experimental solubility of solutes in mono-solvent systems. The applicability of eqns. 6 and 7 has not been checked on solubility data of solutes in ternary solvents. The aim of this work is to check such ability using available solubility data in ternary solvents and this extends the applicability of the Abraham parameters to predict the solubility in ternary solvents.

COMPUTATIONAL METHOD

The model constants of the Jouyban-Acree model were computed using eqns. 6 and 7, then the solubility of the solute in ternary solvents (details of the solutes and solvent systems studied were listed in Table-1) were predicted employing eqn. 1. Details of the QSPR models for computing B terms were reported in Table-2. Using B terms computed by the developed models, the unknown parameters to predict the solubility of a solute in ternary solvent system are the numerical values of the solute solubility in pure solvents, *i.e.* X_1-X_3 (numerical methods I and II). In order to provide a full predictive model (without any experimentally determined parameter), C_S values of solute in the solvents under consideration were computed using Abraham's models, *i.e.* eqns. 4 and 5. The calculated molar solubilities were converted to the mole fraction solubilities using the density of the pure organic solvent (instead of saturated solution) and then the computed X_1-X_3 values were used to predict the solubility in ternary solvents using the Jouyban-Acree model (numerical methods III and IV). Summaries of the numerical methods were shown in Figs. 1 and 2. A sample SPSS program file for calculation of the solubility of anthracene in 1, 4-dioxane + 1-butanol + cyclohexane (data set number 1 in Table-1) and the output file using SPSS software are reported in Appendix.

The predicted B terms and experimental/predicted (X_1-X_3) values, were used to compute the solubility of the solute in ternary solvent mixtures and the mean percentage deviation (MPD) of experimental and calculated solubilities were computed using the following:

$$\text{MPD} = \frac{100}{N}\sum \frac{\left|(X_m)_{\text{calulated}} - (X_m)_{\text{experimental}}\right|}{(X_m)_{\text{experimental}}}$$

where N is the number of data points. To check the variability of each solubility data point with the corresponding experimental value, individual percentage deviation (IPD) was also calculated using the following:

$$\text{IPD} = 100\left(\frac{\left|(X_m)_{\text{calulated}} - (X_m)_{\text{experimental}}\right|}{(X_m)_{\text{experimental}}}\right)$$

TABLE-1
DETAILS OF SOLUBILITY DATA SETS IN TERNARY SOLVENTS, THE EXPERIMENTAL SOLUBILITY IN NEAT SOLVENTS 1-3

No.	Solute	Solvent 1	Solvent 2	Solvent 3	X_1	X_2	X_3	Ref.
1	Anthracene	1,4-Dioxane	1-Butanol	Cyclohexane	0.0083	0.0008	0.0016	1
2	Anthracene	1,4-Dioxane	1-Butanol	Heptane	0.0083	0.0008	0.0016	2
3	Anthracene	1,4-Dioxane	1-Butanol	2,2,4-Trimethylpentane	0.0083	0.0008	0.0011	3
4	Anthracene	1,4-Dioxane	1-Propanol	2,2,4-Trimethylpentane	0.0083	0.0006	0.0011	1
5	Anthracene	1,4-Dioxane	1-Propanol	Cyclohexane	0.0083	0.0006	0.0016	2
6	Anthracene	1,4-Dioxane	1-Propanol	Heptane	0.0083	0.0006	0.0016	3
7	Anthracene	1,4-Dioxane	2-Butanol	2,2,4-Trimethylpentane	0.0083	0.0006	0.0011	1
8	Anthracene	1,4-Dioxane	2-Butanol	Cyclohexane	0.0083	0.0006	0.0016	2
9	Anthracene	1,4-Dioxane	2-Butanol	Heptane	0.0083	0.0006	0.0016	3
10	Anthracene	1,4-Dioxane	2-Methyl-1-propanol	2,2,4-Trimethylpentane	0.0083	0.0005	0.0011	1
11	Anthracene	1,4-Dioxane	2-Methyl-1-propanol	Cyclohexane	0.0083	0.0005	0.0016	2
12	Anthracene	1,4-Dioxane	2-Methyl-1-propanol	Heptane	0.0083	0.0005	0.0016	3
13	Anthracene	1,4-Dioxane	2-Propanol	2,2,4-Trimethylpentane	0.0083	0.0004	0.0011	1
14	Anthracene	1,4-Dioxane	2-Propanol	Cyclohexane	0.0083	0.0004	0.0016	2
15	Anthracene	1,4-Dioxane	2-Propanol	Heptane	0.0083	0.0004	0.0016	3
16	Anthracene	1-Butanol	2-Butanol	Cyclohexane	0.0008	0.0006	0.0016	4
17	Anthracene	1-Butanol	2,2,4-Trimethylpentane	Cyclohexane	0.0008	0.0011	0.0016	5
18	Anthracene	1-Butanol	Heptane	Cyclohexane	0.0008	0.0016	0.0016	6
19	Anthracene	1-Propanol	2-Propanol	Cyclohexane	0.0006	0.0004	0.0016	4
20	Anthracene	1-Propanol	1-Butanol	Heptane	0.0006	0.0008	0.0016	7
21	Anthracene	1-Propanol	1-Butanol	2,2,4-Trimethylpentane	0.0006	0.0008	0.0011	8
22	Anthracene	1-Propanol	2,2,4-Trimethylpentane	Cyclohexane	0.0006	0.0011	0.0016	5
23	Anthracene	1-Propanol	2-Butanol	Heptane	0.0006	0.0006	0.0016	7
24	Anthracene	1-Propanol	2-Butanol	2,2,4-Trimethylpentane	0.0006	0.0006	0.0011	8

No.	Solute	Solvent 1	Solvent 2	Solvent 3	X_1	X_2	X_3	Ref.
25	Anthracene	1-Propanol	Heptane	Cyclohexane	0.0006	0.0016	0.0016	6
26	Anthracene	2-Butanol	2,2,4-Trimethylpentane	Cyclohexane	0.0006	0.0011	0.0016	5
27	Anthracene	2-Butanol	Heptane	Cyclohexane	0.0006	0.0016	0.0016	6
28	Anthracene	2-Propanol	1-Butanol	Heptane	0.0004	0.0008	0.0016	7
29	Anthracene	2-Propanol	1-Butanol	2,2,4-Trimethylpentane	0.0004	0.0008	0.0011	8
30	Anthracene	2-Propanol	2,2,4-Trimethylpentane	Cyclohexane	0.0004	0.0011	0.0016	5
31	Anthracene	2-Propanol	2-Butanol	Heptane	0.0004	0.0006	0.0016	7
32	Anthracene	2-Propanol	2-Butanol	2,2,4-Trimethylpentane	0.0004	0.0006	0.0011	8
33	Anthracene	2-Propanol	Heptane	Cyclohexane	0.0004	0.0016	0.0016	6
34	Anthracene	Dibutyl ether	1-Butanol	2,2,4-Trimethylpentane	0.0036	0.0008	0.0011	9
35	Anthracene	Dibutyl ether	1-Butanol	Cyclohexane	0.0036	0.0008	0.0016	10
36	Anthracene	Dibutyl ether	1-Butanol	Heptane	0.0036	0.0008	0.0016	11
37	Anthracene	Dibutyl ether	1-Propanol	2,2,4-Trimethylpentane	0.0036	0.0006	0.0011	9
38	Anthracene	Dibutyl ether	1-Propanol	Cyclohexane	0.0036	0.0006	0.0016	10
39	Anthracene	Dibutyl ether	1-Propanol	Heptane	0.0036	0.0006	0.0016	11
40	Anthracene	Dibutyl ether	2-Butanol	2,2,4-Trimethylpentane	0.0036	0.0006	0.0011	9
41	Anthracene	Dibutyl ether	2-Butanol	Cyclohexane	0.0036	0.0006	0.0016	10
42	Anthracene	Dibutyl ether	2-Butanol	Heptane	0.0036	0.0006	0.0016	11
43	Anthracene	Dibutyl ether	2-Methyl-1-propanol	2,2,4-Trimethylpentane	0.0036	0.0005	0.0011	9
44	Anthracene	Dibutyl ether	2-Methyl-1-propanol	Cyclohexane	0.0036	0.0005	0.0016	10
45	Anthracene	Dibutyl ether	2-Methyl-1-propanol	Heptane	0.0036	0.0005	0.0016	11
46	Anthracene	Dibutyl ether	2-Propanol	2,2,4-Trimethylpentane	0.0036	0.0004	0.0011	9
47	Anthracene	Dibutyl ether	2-Propanol	Cyclohexane	0.0036	0.0004	0.0016	10
48	Anthracene	Dibutyl ether	2-Propanol	Heptane	0.0036	0.0004	0.0016	11
49	Anthracene	Methyl tert-butyl ether	1-Butanol	Heptane	0.0031	0.0008	0.0016	12
50	Anthracene	Methyl tert-butyl ether	1-Butanol	2,2,4-Trimethylpentane	0.0031	0.0008	0.0011	13
51	Anthracene	Methyl tert-butyl ether	1-Butanol	Cyclohexane	0.0031	0.0008	0.0016	14
52	Anthracene	Methyl tert-butyl ether	1-Propanol	Heptane	0.0031	0.0006	0.0016	12

No.	Solute	Solvent 1	Solvent 2	Solvent 3	X_1	X_2	X_3	Ref.
53	Anthracene	Methyl tert-butyl ether	1-Propanol	2,2,4-Trimethylpentane	0.0031	0.0006	0.0011	13
54	Anthracene	Methyl tert-butyl ether	1-Propanol	Cyclohexane	0.0031	0.0006	0.0016	14
55	Anthracene	Methyl tert-butyl ether	2-Butanol	Heptane	0.0031	0.0006	0.0016	12
56	Anthracene	Methyl tert-butyl ether	2-Butanol	2,2,4-Trimethylpentane	0.0031	0.0006	0.0011	13
57	Anthracene	Methyl tert-butyl ether	2-Butanol	Cyclohexane	0.0031	0.0006	0.0016	14
58	Anthracene	Methyl tert-butyl ether	2-Methyl-1-propanol	Heptane	0.0031	0.0005	0.0016	12
59	Anthracene	Methyl tert-butyl ether	2-Methyl-1-propanol	2,2,4-Trimethylpentane	0.0031	0.0005	0.0011	13
60	Anthracene	Methyl tert-butyl ether	2-Methyl-1-propanol	Cyclohexane	0.0031	0.0005	0.0016	14
61	Anthracene	Methyl tert-butyl ether	2-Propanol	Heptane	0.0031	0.0004	0.0016	12
62	Anthracene	Methyl tert-butyl ether	2-Propanol	2,2,4-Trimethylpentane	0.0031	0.0004	0.0011	13
63	Anthracene	Methyl tert-butyl ether	2-Propanol	Cyclohexane	0.0031	0.0004	0.0016	14
64	Pyrene	1-Butanol	Cyclohexane	Heptane	0.0062	0.0110	0.0110	15
65	Pyrene	1-Propanol	1-Butanol	2,2,4-Trimethylpentane	0.0043	0.0062	0.0072	16
66	Pyrene	1-Propanol	1-Butanol	Cyclohexane	0.0043	0.0062	0.0110	17
67	Pyrene	1-Propanol	1-Butanol	Heptane	0.0043	0.0062	0.0110	18
68	Pyrene	1-Propanol	2-Butanol	2,2,4-Trimethylpentane	0.0043	0.0044	0.0072	16
69	Pyrene	1-Propanol	2-Butanol	Cyclohexane	0.0043	0.0044	0.0110	17
70	Pyrene	1-Propanol	2-Butanol	Heptane	0.0043	0.0044	0.0110	18
71	Pyrene	1-Propanol	Cyclohexane	Heptane	0.0043	0.0110	0.0110	15
72	Pyrene	2-Butanol	Cyclohexane	Heptane	0.0044	0.0110	0.0110	15
73	Pyrene	2-Methyl-1-propanol	Cyclohexane	Heptane	0.0033	0.0110	0.0110	15
74	Pyrene	2-Propanol	1-Butanol	2,2,4-Trimethylpentane	0.0029	0.0062	0.0072	16
75	Pyrene	2-Propanol	1-Butanol	Cyclohexane	0.0029	0.0062	0.0110	17
76	Pyrene	2-Propanol	1-Butanol	Heptane	0.0029	0.0062	0.0110	18
77	Pyrene	2-Propanol	2-Butanol	2,2,4-Trimethylpentane	0.0029	0.0044	0.0072	16
78	Pyrene	2-Propanol	2-Butanol	Cyclohexane	0.0029	0.0044	0.0110	17
79	Pyrene	2-Propanol	2-Butanol	Heptane	0.0029	0.0044	0.0110	18
80	Pyrene	2-Propanol	Cyclohexane	Heptane	0.0029	0.0110	0.0110	15

TABLE-2
PROPOSED QSPR MODELS FOR CALCULATING THE JOUYBAN-ACREE'S MODEL CONSTANTS (B TERMS) USING WATER TO SOLVENT AND GAS TO SOLVENT PROCESSES

Water to solvent (eqn. 6)
$B_0 = 0.028 + 2.123(c_1 - c_2)^2 - 0.160R_2(r_1 - r_2)^2 + 0.282\pi_2^H(s_1 - s_2)^2 + 1.713\sum\beta_2^H(b_1 - b_2)^2 + 2.006V_X(v_1 - v_2)^2$
$B_1 = 0.033 + 0.670(c_1 - c_2)^2 - 0.477R_2(r_1 - r_2)^2 + 0.051\pi_2^H(s_1 - s_2)^2 + 0.476\sum\beta_2^H(b_1 - b_2)^2 - 0.234V_X(v_1 - v_2)^2$
$B_2 = 0.022 + 2.024(c_1 - c_2)^2 - 0.204R_2(r_1 - r_2)^2 + 0.034\pi_2^H(s_1 - s_2)^2 + 0.243\sum\beta_2^H(b_1 - b_2)^2 + 0.848V_X(v_1 - v_2)^2$
$B_0^{'} = 0.028 + 2.123(c_1 - c_3)^2 - 0.160R_2(r_1 - r_3)^2 + 0.282\pi_2^H(s_1 - s_3)^2 + 1.713\sum\beta_2^H(b_1 - b_3)^2 + 2.006V_X(v_1 - v_3)^2$
$B_1^{'} = 0.033 + 0.670(c_1 - c_3)^2 - 0.477R_2(r_1 - r_3)^2 + 0.051\pi_2^H(s_1 - s_3)^2 + 0.476\sum\beta_2^H(b_1 - b_3)^2 - 0.234V_X(v_1 - v_3)^2$
$B_2^{'} = 0.022 + 2.024(c_1 - c_3)^2 - 0.204R_2(r_1 - r_3)^2 + 0.034\pi_2^H(s_1 - s_3)^2 + 0.243\sum\beta_2^H(b_1 - b_3)^2 + 0.848V_X(v_1 - v_3)^2$
$B_0^{''} = 0.028 + 2.123(c_2 - c_3)^2 - 0.160R_2(r_2 - r_3)^2 + 0.282\pi_2^H(s_2 - s_3)^2 + 1.713\sum\beta_2^H(b_2 - b_3)^2 + 2.006V_X(v_2 - v_3)^2$
$B_1^{''} = 0.033 + 0.670(c_2 - c_3)^2 - 0.477R_2(r_2 - r_3)^2 + 0.051\pi_2^H(s_2 - s_3)^2 + 0.476\sum\beta_2^H(b_2 - b_3)^2 - 0.234V_X(v_2 - v_3)^2$
$B_2^{''} = 0.022 + 2.024(c_2 - c_3)^2 - 0.204R_2(r_2 - r_3)^2 + 0.034\pi_2^H(s_2 - s_3)^2 + 0.243\sum\beta_2^H(b_2 - b_3)^2 + 0.848V_X(v_2 - v_3)^2$

Gas to solvent (eqn. 7)
$B_0 = 0.062 + 0.118(c_1 - c_2)^2 - 0.332R_2(r_1 - r_2)^2 + 0.410\pi_2^H(s_1 - s_2)^2 + 2.399\sum\beta_2^H(b_1 - b_2)^2 + 15.715\log L^{(16)}(l_1 - l_2)^2$
$B_1 = 0.103 - 1.864(c_1 - c_2)^2 - 1.590R_2(r_1 - r_2)^2 + 0.119\pi_2^H(s_1 - s_2)^2 + 1.010\sum\beta_2^H(b_1 - b_2)^2 - 9.493\log L^{(16)}(l_1 - l_2)^2$
$B_2 = -0.008 + 1.075(c_1 - c_2)^2 + 0.053R_2(r_1 - r_2)^2 + 0.084\pi_2^H(s_1 - s_2)^2 + 0.414\sum\beta_2^H(b_1 - b_2)^2 + 7.727\log L^{(16)}(l_1 - l_2)^2$
$B_0^{'} = 0.062 + 0.118(c_1 - c_3)^2 - 0.332R_2(r_1 - r_3)^2 + 0.410\pi_2^H(s_1 - s_3)^2 + 2.399\sum\beta_2^H(b_1 - b_3)^2 + 15.715\log L^{(16)}(l_1 - l_3)^2$
$B_1^{'} = 0.103 - 1.864(c_1 - c_3)^2 - 1.590R_2(r_1 - r_3)^2 + 0.119\pi_2^H(s_1 - s_3)^2 + 1.010\sum\beta_2^H(b_1 - b_3)^2 - 9.493\log L^{(16)}(l_1 - l_3)^2$
$B_2^{'} = -0.008 + 1.075(c_1 - c_3)^2 + 0.053R_2(r_1 - r_3)^2 + 0.084\pi_2^H(s_1 - s_3)^2 + 0.414\sum\beta_2^H(b_1 - b_3)^2 + 7.727\log L^{(16)}(l_1 - l_3)^2$
$B_0^{''} = 0.062 + 0.118(c_2 - c_3)^2 - 0.332R_2(r_2 - r_3)^2 + 0.410\pi_2^H(s_2 - s_3)^2 + 2.399\sum\beta_2^H(b_2 - b_3)^2 + 15.715\log L^{(16)}(l_2 - l_3)^2$
$B_1^{''} = 0.103 - 1.864(c_2 - c_3)^2 - 1.590R_2(r_2 - r_3)^2 + 0.119\pi_2^H(s_2 - s_3)^2 + 1.010\sum\beta_2^H(b_2 - b_3)^2 - 9.493\log L^{(16)}(l_2 - l_3)^2$
$B_2^{''} = -0.008 + 1.075(c_2 - c_3)^2 + 0.053R_2(r_2 - r_3)^2 + 0.084\pi_2^H(s_2 - s_3)^2 + 0.414\sum\beta_2^H(b_2 - b_3)^2 + 7.727\log L^{(16)}(l_2 - l_3)^2$

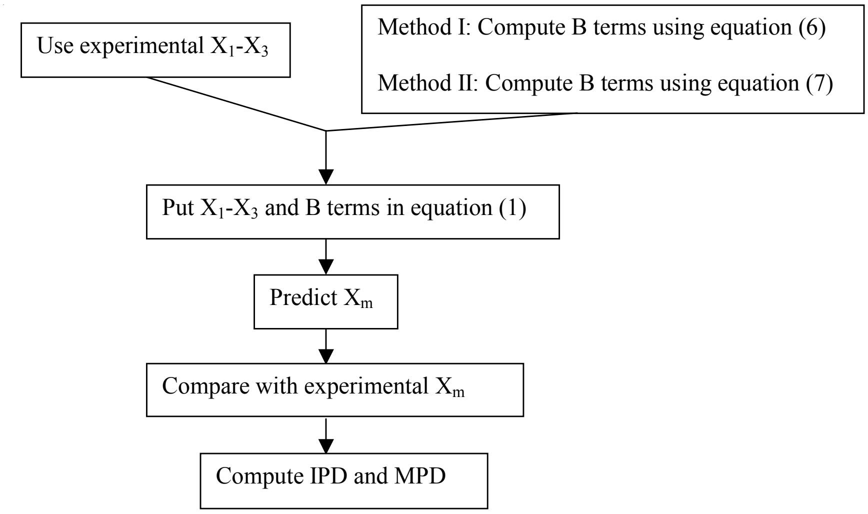

Fig. 1. Flow chart of the numerical methods I and II used in this study

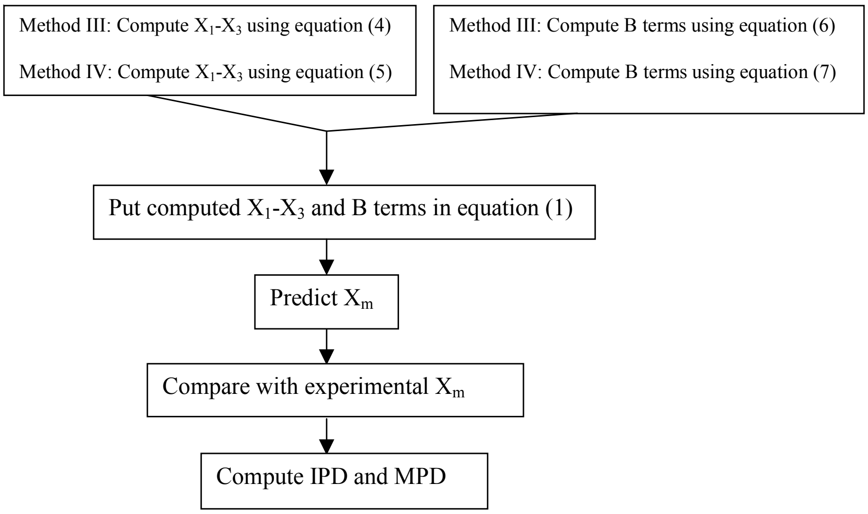

Fig. 2. Flow chart of the numerical methods III and IV used in this study

TABLE-3
MOLECULAR DESCRIPTORS AND INPUT PROPERTIES FOR SOLUTES STUDIED IN THIS WORK AND THEIR REFERENCES

Solute	R_2	π_2^H	$\Sigma\alpha_2^H$	$\Sigma\beta_2^H$	V_X	log C_W	log $L^{(16)}$	log C_G	Ref.
Anthracene	2.290	1.340	0.000	0.280	1.4540	-6.430	7.5680	-9.460	24
Pyrene	2.808	1.710	0.000	0.280	1.5850	-6.150	8.8330	-9.650	26

TABLE-4
COEFFICIENTS IN EQNS. 1 AND 2 FOR WATER TO SOLVENT AND GAS TO SOLVENT PROCESSES OF THE SOLVENTS USED IN THIS STUDY[a]

No.	Water to solvent	c	r	s	a	b	v
1	1,4-Dioxane (dry)	0.098	0.350	-0.083	-0.556	-4.826	4.172
2	1-Butanol (dry)	0.152	0.437	-1.175	0.098	-3.914	4.119
3	1-Propanol (dry)	0.148	0.436	-1.098	0.389	-3.893	4.036
4	2-Butanol (dry)	0.106	0.272	-0.988	0.196	-3.805	4.110
5	2-Methyl-1-propanol (dry)	0.177	0.335	-1.099	0.069	-3.570	3.990
6	2-Propanol (dry)	0.063	0.320	-1.024	0.445	-3.824	4.067
7	Cyclohexane	0.159	0.784	-1.678	-3.740	-4.929	4.577
8	Dibutyl ether (dry)	0.203	0.369	-0.954	-1.488	-5.426	4.508
9	Heptane	0.325	0.670	-2.061	-3.317	-4.733	4.543
10	2,2,4-Trimethylpentane	0.288	0.382	-1.668	-3.639	-5.000	4.461
11	Methyl *tert*-butyl ether	0.376	0.264	-0.788	-1.078	-5.030	4.410
No.	Gas to solvent	c	r	s	a	b	l
1	1,4-Dioxane (dry)	-0.034	-0.354	1.674	3.021	0.000	0.919
2	1-Butanol (dry)	-0.039	-0.276	0.539	3.781	0.995	0.934
3	1-Propanol (dry)	-0.028	-0.185	0.648	4.022	1.043	0.869
4	2-Butanol (dry)	-0.013	-0.456	0.780	3.753	1.064	0.906
5	2-Methyl-1-propanol (dry)	0.012	-0.407	0.670	3.645	1.283	0.895
6	2-Propanol (dry)	-0.060	-0.335	0.702	4.017	1.040	0.893
7	Cyclohexane	0.163	-0.110	0.000	0.000	0.000	1.013
8	Dibutyl ether (dry)	0.165	?0.421	0.760	2.102	?0.664	1.002
9	Heptane	0.275	?0.162	0.000	0.000	0.000	0.983
10	2,2,4-Trimethylpentane	0.275	?0.244	0.000	0.000	0.000	0.972
11	Methyl *tert*-butyl ether	0.278	?0.489	0.801	2.495	0.000	0.993

[a] Data taken from reference[24].

TABLE-5
MEAN PERCENTAGE DEVIATIONS (MPD) FOR SOLUBILITY PREDICTION IN TERNARY SOLVENTS USING VARIOUS NUMERICAL METHODS AND SOLUBILITY PREDICTION IN NEAT SOLVENTS 1-3 USING WATER TO SOLVENT AND GAS TO SOLVENT PARAMETERS

No.[a]	Water to solvent		Gas to solvent		Water to solvent			Gas to solvent		
	I	III	II	IV	X_1	X_2	X_3	X_1	X_2	X_3
1	17.1	26.9	4.8	8.5	20.5	26.2	147.9	20.0	0.5	8.4
2	7.3	6.4	7.0	14.6	20.5	26.2	15.9	20.0	0.5	27.5
3	10.9	9.7	6.4	17.5	20.5	26.2	40.7	20.0	0.5	36.7
4	16.5	10.9	10.2	19.6	20.5	34.1	40.7	20.0	15.0	36.7
5	21.9	22.9	16.2	14.6	20.5	34.1	147.9	20.0	15.0	8.4
6	13.9	5.1	10.4	17.5	20.5	34.1	15.9	20.0	15.0	27.5
7	17.9	15.8	10.4	30.7	20.5	21.2	40.7	20.0	24.0	36.7
8	22.7	20.2	10.2	18.5	20.5	21.2	147.9	20.0	24.0	8.4
9	12.9	8.6	10.4	27.3	20.5	21.2	15.9	20.0	24.0	27.5
10	8.5	7.0	10.6	17.5	20.5	37.7	40.7	20.0	11.7	36.7
11	14.8	27.7	13.1	10.6	20.5	37.7	147.9	20.0	11.7	22.5
12	9.2	8.3	9.8	16.9	20.5	37.7	15.9	20.0	11.7	27.5
13	22.9	18.5	13.5	28.4	20.5	28.3	40.7	20.0	5.6	36.7
14	27.5	17.8	11.8	16.8	20.5	28.3	147.9	20.0	5.6	8.4
15	19.6	11.3	14.1	26.2	20.5	28.3	15.9	20.0	5.6	27.5
16	6.0	63.7	23.2	11.9	26.2	21.2	147.9	0.5	24.0	8.4
17	9.1	48.8	16.5	8.7	25.5	40.9	149.1	1.0	36.9	7.9
18	8.8	56.6	13.8	7.7	25.5	16.9	149.1	1.0	28.4	7.9
19	2.7	59.7	36.6	23.1	34.1	28.3	147.9	15.0	5.6	8.4
20	6.5	24.6	23.0	6.6	34.1	25.5	16.9	15.0	1.0	28.4
21	6.8	18.6	22.6	4.4	34.1	25.5	40.9	15.0	1.0	36.9
22	7.7	49.9	50.3	23.0	34.1	40.9	149.1	15.0	36.9	7.9
23	6.5	23.2	21.0	8.7	34.1	21.6	16.9	15.0	23.8	28.4
24	6.9	17.9	19.7	8.7	34.1	21.6	40.9	15.0	23.8	36.9
25	9.9	61.8	51.7	27.2	34.1	16.9	149.1	15.0	28.4	7.9
26	6.2	44.3	25.7	9.1	21.6	40.9	149.1	23.8	36.9	7.9
27	4.6	49.0	21.1	8.5	21.6	16.9	149.1	23.8	28.4	7.9
28	3.7	15.7	6.7	4.5	28.5	25.5	16.9	5.4	1.0	28.4
29	3.3	13.5	8.2	5.0	28.5	25.5	40.9	5.4	1.0	36.9
30	2.2	39.9	24.1	9.8	28.5	40.9	149.1	5.4	36.9	7.9
31	2.8	13.1	6.7	13.2	28.5	21.6	16.9	5.4	23.8	28.4
32	2.0	12.3	7.3	14.2	28.5	21.6	40.9	5.4	23.8	36.9
33	2.1	44.4	19.4	7.6	28.5	16.9	149.1	5.4	28.4	7.9
34	4.8	11.4	17.2	6.0	11.4	26.2	40.7	32.6	0.5	36.7
35	2.1	57.3	16.3	5.0	11.4	26.2	147.9	32.6	0.5	8.4
36	6.4	14.0	14.3	7.1	11.4	26.2	16.9	32.6	0.5	28.3
37	3.9	10.2	43.9	8.9	11.4	34.1	40.7	32.6	15.0	36.7
38	3.9	55.4	57.2	30.2	11.4	34.1	147.9	32.6	15.0	8.4
39	7.0	15.8	47.6	12.7	11.4	34.1	16.9	32.6	15.0	28.3

No.[a]	Water to solvent		Gas to solvent		Water to solvent			Gas to solvent		
	I	III	II	IV	X_1	X_2	X_3	X_1	X_2	X_3
40	3.2	7.4	22.1	14.8	11.4	21.2	40.7	32.6	24.0	36.7
41	4.6	48.1	25.4	6.0	11.4	21.2	147.9	32.6	24.0	8.4
42	6.5	8.4	22.0	11.9	11.4	21.2	16.9	32.6	24.0	28.3
43	9.4	16.9	36.3	6.5	11.4	37.7	40.7	32.6	11.7	36.7
44	7.7	75.3	42.4	19.4	11.4	37.7	147.9	32.6	11.7	8.4
45	11.4	24.3	33.9	5.2	11.4	37.7	16.9	32.6	11.7	28.3
46	7.8	7.7	17.2	8.7	11.4	28.3	40.7	32.6	5.6	36.7
47	11.0	39.7	22.2	6.9	11.4	28.3	147.9	32.6	5.6	8.4
48	8.5	4.3	15.9	7.0	11.4	28.3	16.9	32.6	5.6	28.3
49	5.0	15.6	3.3	14.6	22.6	25.5	16.9	24.6	1.0	28.4
50	5.1	11.4	6.5	14.1	22.6	25.5	40.9	24.6	1.0	36.9
51	3.3	54.6	6.3	9.9	22.6	25.5	149.1	24.6	1.0	7.9
52	5.1	16.5	29.3	7.3	22.6	34.1	16.9	24.6	15.0	28.4
53	4.2	11.8	31.5	6.6	22.6	34.1	40.9	24.6	15.0	36.9
54	5.5	52.9	37.4	19.0	22.6	34.1	149.1	24.6	15.0	7.9
55	4.8	11.7	9.0	18.6	22.6	21.6	16.9	24.6	23.8	28.4
56	4.8	8.1	11.8	19.0	22.6	21.6	40.9	24.6	23.8	36.9
57	6.1	46.6	11.0	12.0	22.6	21.6	149.1	24.6	23.8	7.9
58	10.5	27.6	20.2	5.0	22.6	37.3	16.9	24.6	11.9	28.4
59	11.2	20.7	24.0	5.3	22.6	37.3	40.9	24.6	11.9	36.9
60	5.3	71.4	26.2	11.7	22.6	37.3	149.1	24.6	11.9	7.9
61	6.9	9.6	7.3	11.9	22.6	28.5	16.9	24.6	5.4	28.4
62	5.3	7.4	9.1	12.6	22.6	28.5	40.9	24.6	5.4	36.9
63	11.3	39.7	9.7	9.5	22.6	28.5	149.1	24.6	5.4	7.9
64	4.4	23.4	10.1	39.9	33.8	59.5	65.9	42.0	38.1	5.5
65	3.9	40.1	22.2	47.1	21.2	33.8	75.4	32.6	42.0	23.1
66	2.9	14.4	43.5	96.8	21.2	33.8	59.5	32.6	42.0	38.1
67	2.6	37.7	20.3	50.5	21.2	33.8	66.0	32.6	42.0	5.7
68	4.8	40.2	20.4	29.0	21.2	36.8	75.4	32.6	2.8	23.1
69	2.8	15.3	46.9	82.6	21.2	36.8	59.5	32.6	2.8	38.1
70	2.2	39.2	17.2	30.6	21.2	36.8	66.0	32.6	2.8	5.7
71	3.6	20.9	51.3	87.6	21.2	59.5	65.9	32.6	38.1	5.5
72	1.2	28.3	19.8	33.8	36.8	59.5	65.9	2.8	38.1	5.5
73	11.6	22.1	33.6	59.3	26.4	59.5	65.9	21.0	38.1	5.5
74	2.7	44.3	7.5	33.0	27.7	33.8	75.4	41.5	42.0	23.1
75	3.2	17.9	23.1	73.1	27.7	33.8	59.5	41.5	42.0	38.1
76	3.1	42.3	4.6	34.4	27.7	33.8	66.0	41.5	42.0	5.7
77	2.1	44.5	8.6	19.7	27.7	36.8	75.4	41.5	2.8	23.1
78	3.6	20.1	25.3	59.1	27.7	36.8	59.5	41.5	2.8	38.1
79	4.7	44.8	6.0	17.6	27.7	36.8	66.0	41.5	2.8	5.7
80	4.0	26.9	19.2	51.8	27.7	59.5	65.9	41.5	38.1	5.5
OM	7.5	27.5	20.2	20.8	–	–	–	–	–	–

[a] Details of data sets are the same as in Table-1; OM = Overall MPD.

RESULTS AND DISCUSSION

Numerical method I

The calculated B terms using eqn. 6 and experimental values of X_1, X_2 and X_3 were used to predict X_m by eqn. 1. The minimum MPD was 1.2 % for solubility of pyrene in 2-butanol + cyclohexane + heptane and the maximum MPD was 27.5 % for solubility of anthracene in 1,4-dioxane + 2-propanol + cyclohexane. The overall MPD (± SD) was 7.5 (± 5.5) %. The corresponding overall MPD (±SD) for 194 solubility data sets in binary solvents from a previous work[25] was 4.7 (± 4.0) % which was significantly ($p < 0.0005$) less than 7.5 % of ternary solvents from this work. A possible reason for this observation could be solvents with high MPD values in ternary solvent compositions. As an example, most of the data sets containing 1,4-dioxane produced MPD > 10 %.

The overall MPD was 10.7 % for 32 solubility data sets using eqn. 2 reported in an earlier paper[23]. There was no significant difference between overall MPD of the same data sets from this work (10.9 %) and 10.7 % of the previous work. However, eqn. 6 is preferred to eqn. 2, since its application could be extended to various solutes dissolved in ternary solvents.

In addition to MPD, which represent average of prediction error for data points of a set and the overall MPD, the IPD was also studied in three subgroups, *i.e.* IPD ≤ 4 % which is comparable with experimental relative standard deviation for repeated experiments, 4 < IPD ≤ 30 % which could be considered as an acceptable error range for many pharmaceutical and industrial applications and IPD > 30 %. Pie chart I of Fig. 3 gives the relative frequency of three subgroups of IPD which is quite reasonable and the probability of solubility prediction with IPD < 30 % is 0.986. The accuracy of numerical method I is further documented in Fig. 4, which compares the predicted $-\ln X_m$ *vs.* the corresponding experimental values. The resulting correlation coefficient between predicted *versus* observed $-\ln X_m$ values was 0.9864. The results of present computations show that when the solubility of a solute in neat solvents 1-3 are available, the numerical method I provided the most accurate predictions in ternary solvents and is recommended for practical applications.

Numerical method II

The calculated B terms using eqn. 7 and experimental values of X_1, X_2 and X_3 were employed to predict X_m using eqn. 1. The minimum and maximum MPDs were 3.3 and 57.2 %, respectively for solubilities of anthracene dissolved in ternary methyl *tert*-butyl ether + 1-butanol + heptane and ternary dibutyl ether + 1-propanol + cyclohexane mixtures. The overall MPD (± SD) was 20.2 (± 13.0) %. Pie chart II of Fig. 3 illustrates the IPD distribution for the numerical method II where the probability of solubility

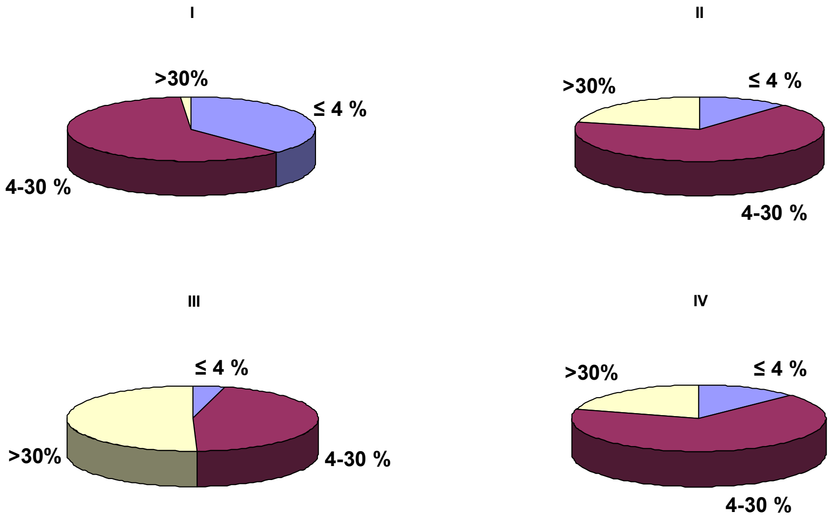

Fig. 3. Distributions of the individual percentage deviations (IPD) for various numerical methods

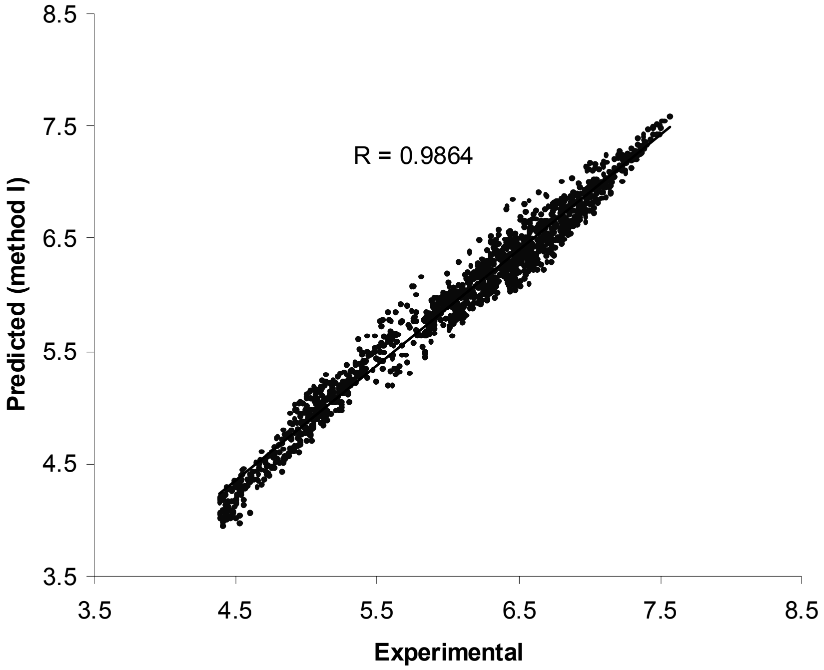

Fig. 4. Predicted -ln X_m using method I *vs.* experimental solubilities in ternary solvents studied

prediction with IPD < 30 % was 0.785. The overall MPD of method II was significantly ($p < 0.0005$) more than that of method I for ternary data. This finding was also confirmed by considering solubility prediction of non-electrolytes in binary solvents using methods I and II from a previous work[25] where the overall MPDs were 4.7 and 11.2 %, respectively. The correlation coefficient of the predicted -ln X_m *vs.* experimental -ln X_m was 0.9823 as shown in Fig. 5.

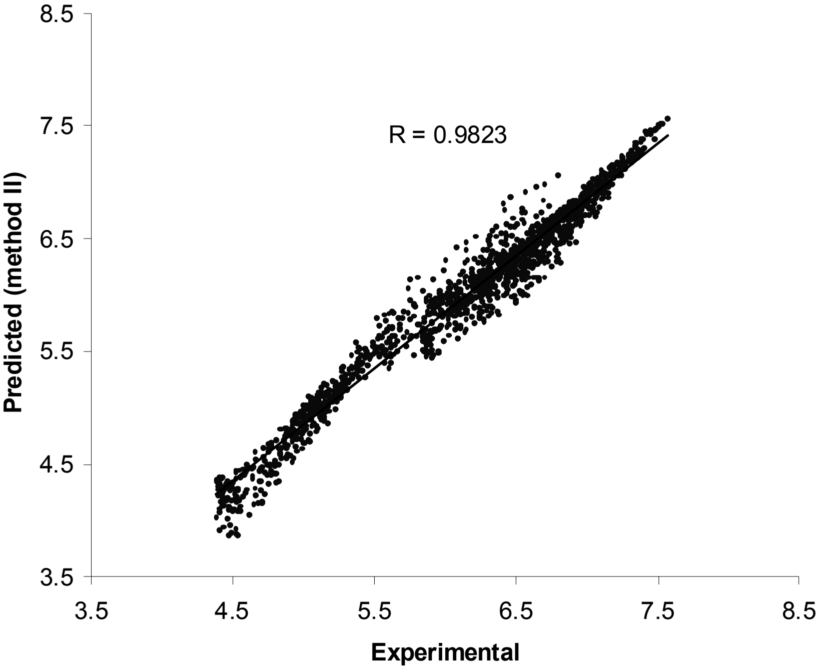

Fig. 5. Predicted -ln X_m using method II *vs.* experimental solubilities in ternary solvents studied

Numerical method III

The main advantage of method III (and also IV) is that there is no need for experimental data. The method is a completely predictive, therefore, the expected prediction error is relatively high. The calculated B terms using eqn. 6 and predicted solubilities in neat solvents 1-3 using eqn. 4 were used to predict X_m by eqn. 1. The minimum MPD was 4.3 % for solubility of anthracene in dibutyl ether + 2-propanol + heptane and the maximum MPD was 75.3 % for solubility of anthracene in dibutyl ether + 2-methyl-1-propanol + cyclohexane mixtures. Solubility of anthracene in cyclohexane was predicted using eqn. 4 with IPD, *i.e.* 145.4 %[24]. The corresponding IPD for solubility of pyrene in cyclohexane was 65 %[26]. These high prediction errors reflected in all solubility data in ternary solvents containing cyclohexane as one of the cosolvents. The overall MPD (± SD) was 27.5 (± 18.2).

One should be able to improve the present approach by developing better methods to predict the solubility in mono-solvent systems. The main solubility prediction methods for mono-solvent systems are: (a) group contribution methods that predict the activity coefficients (for example the UNIFAC and modified UNIFAC (Dortmund models)[27,28]; (b) linear free energy correlations and quantitative structure-property relationships, like the Abraham solvation parameter model and CODESSA approach of Katritzky *et al.*[29-31] and (c) quantum mechanical approaches (for example the COSMO-RS treatment of Klamt and coworkers[32]). Such methods predict the chemical potential of the dissolved solute in the given solvent, or the Gibbs energy of transfer for the solute between two immiscible or partly miscible phases. The solute's enthalpy of fusion and/or vapour pressure is needed to convert the predicted quantity to the desired solubility. Experimental errors in the required solute's enthalpy of fusion and/or vapour pressure do affect the predicted solubility. It is difficult to estimate the error that one could reasonable expect from employing predictive methods to estimate the solubility in the neat organic cosolvents as the published methods have been tested on relatively few of the many possible solute-solvent combinations. On the basis of our review of the published comparisons, we do not think that it would be unreasonable to assign an expected error in the range of 0.1 to 0.3 log units to solubilities predicted by group contribution and linear free energy correlations for many of the simpler systems, though, for aqueous systems a much larger value might be more reasonable. Klamt *et al.*[32] reported a root-mean-squared deviation of about 0.65 log units in using the COSMO-RS model to predict the aqueous solubility of 250 drugs and pesticides.

Numerical method IV

The calculated B terms using eqn. 7 and predicted solubilities in neat solvents 1-3 from eqn. 5 were used to predict X_m using eqn. 1. The minimum MPD was 4.4 % for solubility of anthracene in 1-propanol + 1-butanol + 2,2,4-trimethylpentane and the maximum MPD was 96.8 % for solubility of pyrene in 1-propanol + 1-butanol + cyclohexane mixtures. The overall MPD (± SD) was 20.8 (± 19.5) %. These results showed that as a pure predictive model, the numerical method IV is the most accurate one and is recommended for practical applications when no experimental solubility of a solute in neat solvents 1-3 is available.

Conclusion

The proposed QSPRs provided accurate predictions for solubility of solutes in ternary solvents and could be used to predict the solubility of chemicals in non-aqueous ternary solvent mixtures.

APPENDIX:

Program file in SPSS:

```
data list free/f1 f2 xm.
begin data.
.3714. 3413. 004202
.1720. 6948. 001972
.2908. 2681. 004046
.2791. 5032. 003094
.7231. 1074. 007562
.7159. 1952. 006997
.2179. 6892. 002197
.1276. 5197. 002149
.4196. 4873. 003898
.5203. 3880. 004816
.1852. 1713. 003235
.7253. 1460. 007327
.5313. 2525. 005721
.1316. 2234. 002654
.1199. 7040. 001725
.2390. 1234. 003779
.1300. 4238. 002362
.4455. 1127. 005672
.5417. 1124. 006553
end data.
title 'Anthracene in 1,4-dioxane + 1-butanol + cyclohexane'.
subtitle 'Data taken from J. Chem. Eng. Data, 45, 971 (2000)'.
* Mole fraction solubility of anhtracene in neat solvents 1-3.
compute x1=. 008329.
compute x2=. 000801.
compute x3=. 001553.
* Fraction of the third solvent in the mixture.
compute f3=1-f1-f2.
* Molecular weights of solvents 1-3.
compute mw1=88.107.
compute mw2=74.124.
compute mw3=84.162.
```

```
* Densities of solvents 1-3.
compute ds1=1.02797.
compute ds2=.8060.
compute ds3=.77389.
* Water to solvent parameters.
*Solvent 1.
compute cws1=. 098.
compute rws1=. 35.
compute sws1=-.083.
compute aws1=-.556.
compute bws1=-4.826.
compute vws1= 4.172.
*Solvent 2.
compute cws2=. 152.
compute rws2=. 437.
compute sws2=-1.175.
compute aws2=. 098.
compute bws2=-3.914.
compute vws2= 4.119.
*Solvent 3.
compute cws3=. 159.
compute rws3=. 784.
compute sws3=-1.678.
compute aws3=-3.74.
compute bws3=-4.929.
compute vws3= 4.577.
* Gas to solvent parameters.
*Solvent 1.
compute cgs1=-.034.
compute rgs1=-.354.
compute sgs1= 1.674.
compute ags1= 3.021.
compute bgs1= 0.
compute lgs1=. 919.
*Solvent 2.
compute cgs2=-.039.
compute rgs2=-.276.
compute sgs2=. 539.
compute ags2= 3.781.
compute bgs2=. 995.
compute lgs2=. 934.
*Solvent 3.
```

```
compute cgs3=. 163.
compute rgs3=-.11.
compute sgs3= 0.
compute ags3= 0.
compute bgs3= 0.
compute lgs3= 1.013.
*Abraham solute parameters of Anthracene.
compute r2 = 2.290.
compute pi2h = 1.340.
compute alfa = 0.000.
compute beta = 0.280.
compute vx = 1.454.
compute lcw = -6.430.
compute ll16 = 7.568.
compute lcg = -9.46.
* End of row data.

compute lx1=ln(x1).
compute lx2=ln(x2).
compute lx3=ln(x3).
compute q012=f1*f2.
compute q112=f1*f2*(f1-f2).
compute q212=f1*f2*(f1-f2)*(f1-f2).
compute q013=f1*f3.
compute q113=f1*f3*(f1-f3).
compute q213=f1*f3*(f1-f3)*(f1-f3).
compute q023=f2*f3.
compute q123=f2*f3*(f2-f3).
compute q223=f2*f3*(f2-f3)*(f2-f3).

* Prediction using experimental solubility in neat solvents 1-3.
* Abraham model, Water to solvent.
compute h112=((cws1-cws2)**2).
compute h212=(r2*(rws1-rws2)**2).
compute h312=(pi2h*(sws1-sws2)**2).
compute h412=(beta*(bws1-bws2)**2).
compute h512=(vx*(vws1-vws2)**2).
compute a12q0=. 028+2.123 *h112-0.160
*h212+.282*h312+1.713*h412+2.006*h512.
compute a12q1=0.033 +0.670*h112 -0.477 *h212+.051 *h312+.476 *h412
-0.234 *h512.
compute a12q2=. 022 +2.024*h112 -.204 *h212+.034 *h312+.243
```

```
*h412+.848 *h512.
compute h113=((cws1-cws3)**2).
compute h213=(r2*(rws1-rws3)**2).
compute h313=(pi2h*(sws1-sws3)**2).
compute h413=(beta*(bws1-bws3)**2).
compute h513=(vx*(vws1-vws3)**2).
compute a13q0=. 028+2.123 *h113-0.160
*h213+.282*h313+1.713*h413+2.006*h513.
compute a13q1=0.033 +0.670*h113 -0.477 *h213+.051 *h313+.476 *h413
-0.234 *h513.
compute a13q2=. 022 +2.024*h113 -.204 *h213+.034 *h313+.243
*h413+.848 *h513.
compute h123=((cws2-cws3)**2).
compute h223=(r2*(rws2-rws3)**2).
compute h323=(pi2h*(sws2-sws3)**2).
compute h423=(beta*(bws2-bws3)**2).
compute h523=(vx*(vws2-vws3)**2).
compute a23q0=. 028+2.123 *h123-0.160
*h223+.282*h323+1.713*h423+2.006*h523.
compute a23q1=0.033 +0.670*h123 -0.477 *h223+.051 *h323+.476 *h423
-0.234 *h523.
compute a23q2=. 022 +2.024*h123 -.204 *h223+.034 *h323+.243
*h423+.848 *h523.
compute ypa=a12q0*q012 + a12q1*q112 + a12q2*q212+ a13q0*q013 +
a13q1*q113 + a13q2*q213+ a23q0*q023 + a23q1*q123 + a23q2*q223.
compute lxmpq=f1*lx1+f2*lx2+f3*lx3+ypa.
compute xmpq=exp(lxmpq).
compute aperab1=abs (100*(xmpq-xm)/xm ).
compute lg1p= cws1 +r2*rws1 + pi2h*sws1+beta*bws1 + vx*vws1.
compute lg2p= cws2 +r2*rws2 + pi2h*sws2+beta*bws2 + vx*vws2.
compute lg3p= cws3 +r2*rws3 + pi2h*sws3+beta*bws3 + vx*vws3.
compute lcs1=lg1p+lcw.
compute lcs2=lg2p+lcw.
compute lcs3=lg3p+lcw.
compute cs1=10**(lcs1).
compute cs2=10**(lcs2).
compute cs3=10**(lcs3).
* Conversion of molarity to mole fraction solubility.
compute n1=(1000*ds1)/mw1.
compute x1ab=cs1/(cs1+n1).
compute n3=(1000*ds2)/mw2.
compute x2ab=cs2/(cs2+n3).
```

```
compute n5=(1000*ds3)/mw3.
compute x3ab=cs3/(cs3+n5).
compute lx1ab=ln(x1ab).
compute lx2ab=ln(x2ab).
compute lx3ab=ln(x3ab).
compute lxmpab=f1*lx1ab+f2*lx2ab+f3*lx3ab+ypa.
compute xmpab=exp(lxmpab).
compute aperab2=abs (100*(xmpab-xm)/xm ).

* Abraham model, Gas to solvent.
compute g112=((cgs1-cgs2)**2).
compute g212=(r2*(rgs1-rgs2)**2).
compute g312=(pi2h*(sgs1-sgs2)**2).
compute g412=(beta*(bgs1-bgs2)**2).
compute g512=(ll16*(lgs1-lgs2)**2).
compute i12q0=.062+0.118 *g112-0.332
*g212+.410*g312+2.399*g412+15.715*g512.
compute i12q1=0.103 -1.864*g112 -1.590 *g212+.119 *g312+1.010 *g412
-9.493 *g512.
compute i12q2= -.008 +1.075*g112 +.053 *g212+.084 *g312+.414
*g412+7.727 *g512.
compute g113=((cgs1-cgs3)**2).
compute g213=(r2*(rgs1-rgs3)**2).
compute g313=(pi2h*(sgs1-sgs3)**2).
compute g413=(beta*(bgs1-bgs3)**2).
compute g513=(ll16*(lgs1-lgs3)**2).
compute i13q0=.062+0.118 *g113-0.332
*g213+.410*g313+2.399*g413+15.715*g513.
compute i13q1=0.103 -1.864*g113 -1.590 *g213+.119 *g313+1.010 *g413
-9.493 *g513.
compute i13q2= -.008 +1.075*g113 +.053 *g213+.084 *g313+.414
*g413+7.727 *g513.
compute g123=((cgs2-cgs3)**2).
compute g223=(r2*(rgs2-rgs3)**2).
compute g323=(pi2h*(sgs2-sgs3)**2).
compute g423=(beta*(bgs2-bgs3)**2).
compute g523=(ll16*(lgs2-lgs3)**2).
compute i23q0=.062+0.118 *g123-0.332
*g223+.410*g323+2.399*g423+15.715*g523.
compute i23q1=0.103 -1.864*g123 -1.590 *g223+.119 *g323+1.010 *g423
-9.493 *g523.
compute i23q2= -.008 +1.075*g123 +.053 *g223+.084 *g323+.414
```

```
*g423+7.727 *g523.
compute ypi=i12q0*q012 + i12q1*q112 + i12q2*q212 + i13q0*q013 +
i13q1*q113 + i13q2*q213 + i23q0*q023 + i23q1*q123 + i23q2*q223.
compute lxmpi=f1*lx1+f2*lx2+f3*lx3+ypi.
compute xmpi=exp(lxmpi).
compute aperab3=abs (100*(xmpi-xm)/xm ).
compute lg1i= cgs1 +r2*rgs1 + pi2h*sgs1+beta*bgs1 + ll16*lgs1.
compute lg2i= cgs2 +r2*rgs2 + pi2h*sgs2+beta*bgs2 + ll16*lgs2.
compute lg3i= cgs3 +r2*rgs3 + pi2h*sgs3+beta*bgs3 + ll16*lgs3.
compute lcs1i=lg1i+lcg.
compute lcs2i=lg2i+lcg.
compute lcs3i=lg3i+lcg.
compute cs1i=10**(lcs1i).
compute cs2i=10**(lcs2i).
compute cs3i=10**(lcs3i).
* Conversion of molarity to mole fraction solubility.
compute n1i=(1000*ds1)/mw1.
compute x1abi=cs1i/(cs1i+n1i).
compute n3i=(1000*ds2)/mw2.
compute x2abi=cs2i/(cs2i+n3i).
compute n5i=(1000*ds3)/mw3.
compute x3abi=cs3i/(cs3i+n5i).
compute lx1abi=ln(x1abi).
compute lx2abi=ln(x2abi).
compute lx3abi=ln(x3abi).
compute lxmpabi=f1*lx1abi+f2*lx2abi+f3*lx3abi+ypi.
compute xmpabi=exp(lxmpabi).
compute aperab4=abs (100*(xmpabi-xm)/xm ).
* Generating report.
list f1 f2 aperab1 aperab2 aperab3 aperab4.
descriptive /variable aperab1 aperab2 aperab3 aperab4/sta mean.
```

The output file of SPSS:

```
Anthracene in 1,4-Dioxane+1-Butanol+ Cyclohexane
Data taken from J. Chem. Eng. Data. 45 (2000) 971-973

List
     F1     F2      APERAB1     APERAB2     APERAB3   APERAB4
     . 37   . 34    19.54       21.18       2.28      8.34
     . 17   . 69    15.01       16.39       4.58      9.52
     . 29.  27      22.31       30.29       .94       9.09
```

```
. 28   . 50   19.50   16.16   3.10   10.88
. 72   . 11   14.36   17.22   6.70   10.59
. 72   . 20   15.79   9.20    2.27   13.59
. 22   . 69   18.43   8.49    8.42   13.75
. 13   . 52   11.03   41.61   4.73   1.53
. 42   . 49   21.96   2.85    7.06   16.24
. 52   . 39   19.54   5.47    2.31   13.87
. 19   . 17   17.42   59.56   6.75   3.24
. 73   . 15   14.57   13.74   5.11   11.67
. 53   . 25   19.90   14.15   2.44   10.82
. 13   . 22   12.16   70.29   9.92   . 79
. 12   . 70   10.87   25.96   . 44   4.86
. 24   . 12   19.57   54.40   6.56   4.50
. 13   . 42   11.91   49.33   6.22   . 95
. 45   . 11   20.86   31.86   6.43   7.33
. 54   . 11   20.67   23.34   5.05   9.73
```

Number of cases read: 19 Number of cases listed: 19

Descriptives

Descriptive Statistics

	N	Mean
APERAB1	19	17.1258
APERAB2	19	26.9205
APERAB3	19	4.8051
APERAB4	19	8.4898
Valid N (listwise)	19	

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