Asian Journal of Chemistry Vol. 19, No. 3 (2007), 1853-1862

Solubility Prediction of Pyrene in Non-Aqueous Solvent Mixtures Using Jouyban-Acree Model

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Applicability of a previsouly presented quanitative structure property relationship was extended to predict the solubility of pyrene in binary solvent mixtures at 26°C using solubility parameter, boiling point, vapour pressure and density of solvents as independent variables employing 63 solubility data sets. The mean percentage deviation of experimental and predicted solubilities was computed as a measure of accuracy where the mean percentage deviations were *ca.* 6 and *ca.* 10 %, respectively for pyrene solubillity data in binary and ternary solvents for the best prediction method.

Key Words: Solubility, Modeling, Prediction, Solvent mixtures, Jouyban-Acree model, Pyrene.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are hydrophobic compounds and persist in ecosystem because of their poor aqueous solubilities. They present in contaminated soil, waters and sediments and play a significant role in the environment safety and human health[1]. Solubility data is one of the key information in chemical industries and it is usually determined in water and many organic solvents. When the desired amount of a solute is not soluble, mixing solvents or cosolvency, is the main solubility enhancement method. Solubility of pyrene in non-aqueous mixed solvents has been extensively studied by our research group. In addition to experimental solubility determinations, the data has been correlated using the Jouyban-Acree model. The general form of the model is:

$$\ln X_m = f_1 \ln X_1 + f_2 \ln X_2 + f_1 f_2 \sum_{j=0}^{2} B_i (f_1 - f_2)^j \quad (1)$$

where X is the mole fraction solubility of the solute, f denotes the volume fraction of the solvents 1 and 2 in the solvent mixture, subscripts m, 1 and

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2 are the mixed solvent and solvents 1 and 2, respectively and B_i is the model constants which is calculated using a no intercept least square analysis[2]. The model can be readily extended to ternary solvent systems as:

$$\ln X_m = f_1 \ln X_1 + f_2 \ln X_2 + f_3 \ln X_3 + f_1 f_2 \sum_{i=0}^{n} B_i (f_1 - f_2)^i + f_1 f_3 \sum_{i=0}^{n} B_i' (f_1 - f_3)^i + f_2 f_3 \sum_{i=0}^{n} B_i'' (f_2 - f_3)^i \quad (2)$$

where f_3 stands for the mole/volume fraction of the solvent 3 in the mixed solvent, B_i, B_i' and B_i'' are the sub-binary interaction terms.

Although the Jouyban-Acree model was proposed for modeling of polycyclic aromatic hydrocarbons solubilities in non-aqueous solvent mixtures, it has been shown that the model is capable of calculating the solubility of polar and/or semi-polar compounds in aqueous solvent mixtures[3,4]. It has also been shown that the model is applicable for modeling physico-chemical properties other than solubility in solvent mixtures[5].

The main drawback of the Jouyban-Acree model is its curve-fitting parameters which needs a minimum number of experimental data points for training the model. With known model constants and solubility of a solute in mono-solvent systems, it is possible to predict the solubility of the solute in mixed solvent systems. In a previous work, a quantitative structure property relationship (QSPR) has been proposed to compute the model constants using a minimum number of data points[6]. The model has been evaluated using a limited number of anthracene solubility data sets in mixed solvents and the solubility parameters of the solvents and that of anthracene have been used as independent variables. To continue our previous studies on solubility prediction methods, in an earlier work, a QSPR model has been proposed using a larger number of anthracene solubility data in binary solvents using additional independent variables. The proposed method in previous work showed significant improvement in the results. The aim of this work is to extend applicability of the QSPRs to predict solubility of pyrene in non-aqueous solvent mixtures. In this work, no experimental solubility data of pyrene in mixed solvents has been used in training process of the QSPRs, therefore, the method could be considered as a full predictive method.

EXPERIMENTAL

The model constants of the Jouyban-Acree model represent the extent of solvent-solvent and solvent-solute interactions in the solution and these interactions could be related to the physico-chemical properties of solvents and solutes in order to establish a quantitative structure property relationship approach. In an earlier paper[6], the differences in solubility parameters of the solvents with that of solute and their square values have

been used to correlate the binary interaction terms (B_i) of the Jouyban-Acree model as:

$$B_0 = 0.081(\delta_1 - \delta_s) + 0.315(\delta_2 - \delta_s) + 0.159(\delta_1 - \delta_s)^2 - 0.129(\delta_2 - \delta_s)^2 \quad (3)$$

$$B_1 = -0.120(\delta_1 - \delta_s) + 0.047(\delta_2 - \delta_s) - 0.073(\delta_1 - \delta_s)^2 + 0.052(\delta_2 - \delta_s)^2 \quad (4)$$

$$B_2 = 0.048(\delta_1 - \delta_s) - 0.012(\delta_2 - \delta_s) + 0.025(\delta_1 - \delta_s)^2 - 0.031(\delta_2 - \delta_s)^2 \quad (5)$$

in which δ_1 and δ_2 are the solubility parameters of solvents 1 and 2, respectively, δ_s is the solute's solubility parameter and the model constants calculated using experimental data of 30 data sets[6]. As a general rule, the more independent variables the more accurate the correlation and the more accurate the predictions. In a recent work[7], the experimentally obtained B_i values for 116 data sets of anthracene solubility in non-aqueous binary solvents, were regressed against $(\delta_1-\delta_2)^2$, $(BP_1\text{-}BP_2)^2$, $(VP_1\text{-}VP_2)^2$ and $(\rho_1-\rho_2)^2$ in which BP, VP and ρ are boiling point, vapour pressure and density of solvents, respectively and subscripts 1 and 2 denote solvents 1 and 2. The resulted equations for calculating B_i terms are:

$$B_0 = 0.062(\delta_1 - \delta_2)^2 + 0.000093(BP_1 - BP_2)^2 - 0.0000299(VP_1 - VP_2)^2 + 16.597(\rho_1 - \rho_2)^2 \quad (6)$$

$$B_1 = 0.009(\delta_1 - \delta_2)^2 + 0.000048(BP_1 - BP_2)^2 - 0.0000147(VP_1 - VP_2)^2 + 3.330(\rho_1 - \rho_2)^2 \quad (7)$$

$$B_2 = 0.019(\delta_1 - \delta_2)^2 + 0.000029(BP_1 - BP_2)^2 - 0.00000139(VP_1 - VP_2)^2 + 3.374(\rho_1 - \rho_2)^2 \quad (8)$$

In developing equations 6-8, no pyrene solubility data were used, so the model could be considered as a predictive model for pyrene data.

The predicted B_i values, have been used to predict the solubility of pyrene and mean percentage deviation (MPD) have been computed using:

$$MPD = \frac{100}{N} \sum \frac{\left|(X_m)_{cal.} - (X_m)_{obs.}\right|}{(X_m)_{obs.}}$$

where N is the number of data points.

RESULTS AND DISCUSSION

The solubility of pyrene in 63 different binary solvent mixtures (Table-1) has been collected from the literature[8-16]. The solubility parameter of pyrene [= 10.6 $(cal/cm^3)^{1/2}$] was taken from a reference[17]. The computed B_i terms using two QSPR methods were used to predict the solubility of pyrene in binary solvent mixtures and mean percentage deviation (MPD) values among numerical values of the employed independent variables were listed in Table-2. For B_i terms computed using equations 3-5, the lowest and highest MPDs were 0.3 (for solubility of pyrene in 2-butanol + 1-pentanol) and 32.5 (for solubility of pyrene in benzene + hexane) % and the overall MPD (± SD) was 10.3 ± 8.7. The required information for predicting solubility of pyrene in binary solvents is the numerical values of X_1 and X_2, *i.e.*, two points for each binary solvent systems.

TABLE-1
LIST OF SOLVENTS AND THE REFERENCES OF SOLUBILITY DATA IN BINARY SOLVENTS

No.	Solvent 1	Solvent 2	Reference
1	1-Butanol	1-Pentanol	8
2	1-Butanol	1-Propanol	9
3	1-Butanol	2-Propanol	9
4	1-Octanol	1-Propanol	9
5	1-Octanol	2-Propanol	9
6	1-Propanol	1-Pentanol	8
7	2,2,4-Trimethyl pentane	1-Octanol	10
8	2,2,4-Trimethyl pentane	1-Propanol	11
9	2,2,4-Trimethyl pentane	2-Butanol	12
10	2,2,4-Trimethyl pentane	2-Propanol	11
11	2,2,4-Trimethyl pentane	Dibutyl ether	13
12	2-Butanol	1-Pentanol	8
13	2-Butanol	1-Propanol	9
14	2-Butanol	2-Propanol	9
15	2-Methyl-1-propanol	1-Pentanol	8
16	2-Methyl-1-propanol	1-Propanol	9
17	2-Methyl-1-propanol	2-Propanol	9
18	2-Propanol	1-Pentanol	8
19	2-Propanol	1-Propanol	9
20	Benzene	2,2,4-Trimethyl pentane	14
21	Benzene	Cyclohexane	14
22	Benzene	Heptane	14
23	Benzene	Hexane	14
24	Benzene	Octane	14
25	Cyclohexane	1-Butanol	15
26	Cyclohexane	1-Octanol	10
27	Cyclohexane	1-Propanol	11
28	Cyclohexane	2- Butanol	12
29	Cyclohexane	2,2,4-Trimethyl pentane	16
30	Cyclohexane	2-Methyl-1-propanol	15
31	Cyclohexane	2-Propanol	11
32	Cyclohexane	Dibutyl ether	13
33	Cyclohexane	Heptane	16
34	Cyclohexane	Hexane	16
35	Cyclohexane	Octane	16
36	Dibutyl ether	Heptane	13
37	Heptane	1-Butanol	15
38	Heptane	1-Octanol	10
39	Heptane	1-Propanol	11
40	Heptane	2-Butanol	12
41	Heptane	2-Methyl-1-propanol	15

No.	Solvent 1	Solvent 2	Reference
42	Heptane	2-Propanol	11
43	Hexane	1-Butanol	15
44	Hexane	1-Octanol	10
45	Hexane	1-Propanol	11
46	Hexane	2-Butanol	12
47	Hexane	2-Methyl-1-propanol	15
48	Hexane	2-Propanol	11
49	Hexane	Dibutyl ether	13
50	Methyl cyclohexane	1-Butanol	15
51	Methyl cyclohexane	1-Octanol	10
52	Methyl cyclohexane	1-Propanol	11
53	Methyl cyclohexane	2-Butanol	12
54	Methyl cyclohexane	2-Methyl-1-propanol	15
55	Methyl cyclohexane	2-Propanol	11
56	Methyl cyclohexane	Dibutyl ether	13
57	Octane	1-Butanol	15
58	Octane	1-Octanol	10
59	Octane	1-Propanol	11
60	Octane	2-Butanol	12
61	Octane	2-Methyl-1-propanol	15
62	Octane	2-Propanol	11
63	Octane	Dibutyl ether	13

The same calculations were done using equations 6-8 and MPD values were also reported in Table-2. The lowest and highest MPDs were 0.2 (for solubility of pyrene in 2-butanol + 1-pentanol) and 17.6 (for solubility of pyrene in 2,2,4-trimethyl pentane + 1-octanol). The overall MPD (± SD) was 6.4 ± 4.7%.

There was significant difference between MPD differences of two studied method (paired t-test, $p < 0.002$) revealing that the method derived from equations 6-7 provides more accurate results in comparison with previous method. Fig. 1 showed the relative frequency of MPD values sorted in three subgroups, *i.e.*, < 10, 10-30 and > 30%. All MPD values of the predictions derived from equations 6-8 lied in < 30 % category while it was 96.8 % for the previous method.

The computed B_i terms using two QSPR methods were used to predict the solubility of pyrene in 17 different ternary solvent mixtures (Table-3) and MPD values were calculated and listed in Table-3. The minimum and maximum MPDs for predicted solubilities derived from equations 3-5 were 23.9 and 54.6 %, respectively and the overall MPD was 41.8 ± 8.8 %. The corresponding values for predictions derived from equations 6-8 were 1.3 and 19.4 % with overall MPD of 10.1 ± 5.8 %.

As general conclusion, the extended method was able to compute

TABLE-2
SOLUBILITY PARAMETER (δ), BOILING POINTS (BP), VAPOUR PRESSURE (VP) AND DENSITY (ρ) OF SOLVENTS 1 AND 2, NUMBER OF EXPERIMENTAL DATA POINTS IN EACH SET (N) AND MEAN PERCENTAGE DEVIATIONS (MPD) FOR PREDICTED SOLUBILITY USING THE JOUYBAN-ACREE MODEL WITH MODEL CONSTANTS FROM EQUATIONS 3-5 AND 6-8

No.[a]	δ_1 $(Cal/cm^3)^{1/2}$	δ_2 $(Cal/cm^3)^{1/2}$	BP_1 (°C)	BP_2 (°C)	VP_1 (torr)	VP_2 (torr)	ρ_1 (g/cm^3)	ρ_2 (g/cm^3)	N	Equations 3-5	Equations 6-8
1	11.39	11.10	117.7	137.8	6.180	2.350	0.806	0.812	9	1.6	1.8
2	11.39	11.98	117.7	97.2	6.180	20.850	0.806	0.800	9	3.2	0.8
3	11.39	11.49	117.7	82.3	6.180	45.160	0.806	0.781	9	1.9	1.7
4	10.32	11.98	195.2	97.2	0.075	20.850	0.822	0.800	9	14.8	6.4
5	10.32	11.49	195.2	82.3	0.075	45.160	0.822	0.781	9	19.0	9.0
6	11.98	11.10	97.2	137.8	20.850	2.350	0.800	0.812	9	13.7	8.9
7	6.84	10.32	99.2	195.2	49.000	0.075	0.688	0.822	9	17.5	17.6
8	6.84	11.98	99.2	97.2	49.000	20.850	0.688	0.800	9	18.7	13.6
9	6.84	10.80	99.2	99.6	49.000	18.290	0.688	0.803	9	11.7	2.4
10	6.84	11.49	99.2	82.3	49.000	45.160	0.688	0.781	9	11.0	2.6
11	6.84	7.77	99.2	142.2	49.000	12.500	0.688	0.764	7	5.1	1.0
12	10.80	11.10	99.6	137.8	18.290	2.350	0.803	0.812	9	0.3	0.2
13	10.80	11.98	99.6	97.2	18.290	20.850	0.803	0.800	9	3.1	1.7
14	10.80	11.49	99.6	82.3	18.290	45.160	0.803	0.781	9	0.9	2.8
15	11.20	11.10	107.7	137.8	10.220	2.350	0.798	0.812	9	1.7	3.6
16	11.20	11.98	107.7	97.2	10.220	20.850	0.798	0.800	9	4.3	1.0
17	11.20	11.49	107.7	82.3	10.220	45.160	0.798	0.781	9	1.6	1.9
18	11.49	11.10	82.3	137.8	45.160	2.350	0.781	0.812	9	2.8	3.2
19	11.49	11.98	82.3	97.2	45.160	20.850	0.781	0.800	9	5.0	0.4

No.[a]	δ_1 $(Cal/cm^3)^{1/2}$	δ_2 $(Cal/cm^3)^{1/2}$	BP_1 (°C)	BP_2 (°C)	VP_1 (torr)	VP_2 (torr)	ρ_1 (g/cm^3)	ρ_2 (g/cm^3)	N	Equations 3-5	Equations 6-8
20	9.19	6.84	80.1	99.2	95.200	49.000	0.874	0.688	7	31.2	11.3
21	9.19	8.21	80.1	80.7	95.200	98.000	0.874	0.774	7	27.4	10.8
22	9.19	7.38	80.1	98.4	95.200	45.700	0.874	0.680	7	29.1	5.2
23	9.19	7.28	80.1	68.7	95.200	151.300	0.874	0.655	7	32.5	4.3
24	9.19	7.58	80.1	125.7	95.200	14.000	0.874	0.699	7	25.2	5.9
25	8.21	11.39	80.7	117.7	98.000	6.180	0.774	0.806	9	2.7	6.8
26	8.21	10.32	80.7	195.2	98.000	0.075	0.774	0.822	9	5.3	7.7
27	8.21	11.98	80.7	97.2	98.000	20.850	0.774	0.800	9	7.2	8.5
28	8.21	10.80	80.7	99.6	98.000	18.290	0.774	0.803	9	9.5	15.2
29	8.21	6.84	80.7	99.2	98.000	49.000	0.774	0.688	7	26.5	3.3
30	8.21	11.20	80.7	107.7	98.000	10.220	0.774	0.798	9	6.3	11.7
31	8.21	11.49	80.7	82.3	98.000	45.160	0.774	0.781	9	11.5	14.6
32	8.21	7.77	80.7	142.2	98.000	12.500	0.774	0.764	8	23.9	7.4
33	8.21	7.38	80.7	98.4	98.000	45.700	0.774	0.680	7	23.0	1.3
34	8.21	7.28	80.7	68.7	98.000	151.300	0.774	0.655	7	24.7	1.0
35	8.21	7.58	80.7	125.7	98.000	14.000	0.774	0.699	7	20.9	2.6
36	7.77	7.38	142.2	98.4	12.500	45.700	0.764	0.680	7	21.1	0.7
37	7.38	11.39	98.4	117.7	45.700	6.180	0.680	0.806	9	6.6	2.3
38	7.38	10.32	98.4	195.2	45.700	0.075	0.680	0.822	9	9.2	15.4
39	7.38	11.98	98.4	97.2	45.700	20.850	0.680	0.800	9	1.8	1.9
40	7.38	10.80	98.4	99.6	45.700	18.290	0.680	0.803	9	2.5	8.7
41	7.38	11.20	98.4	107.7	45.700	10.220	0.680	0.798	9	2.0	7.5
42	7.38	11.49	98.4	82.3	45.700	45.160	0.680	0.781	9	4.7	8.5
43	7.28	11.39	68.7	117.7	151.300	6.180	0.655	0.806	9	10.8	1.8

No.[a]	δ_1 $(Cal/cm^3)^{1/2}$	δ_2 $(Cal/cm^3)^{1/2}$	BP_1 (°C)	BP_2 (°C)	VP_1 (torr)	VP_2 (torr)	ρ_1 (g/cm^3)	ρ_2 (g/cm^3)	N	Equations 3-5	Equations 6-8
44	7.28	10.32	68.7	195.2	151.300	0.075	0.655	0.822	9	9.0	14.0
45	7.28	11.98	68.7	97.2	151.300	20.850	0.655	0.800	9	5.8	1.0
46	7.28	10.80	68.7	99.6	151.300	18.290	0.655	0.803	9	3.0	8.6
47	7.28	11.20	68.7	107.7	151.300	10.220	0.655	0.798	9	3.6	7.5
48	7.28	11.49	68.7	82.3	151.300	45.160	0.655	0.781	9	2.5	5.7
49	7.28	7.77	68.7	142.2	151.300	12.500	0.655	0.764	7	13.4	5.8
50	7.82	11.39	100.9	117.7	46.000	6.180	0.765	0.806	9	2.1	5.3
51	7.82	10.32	100.9	195.2	46.000	0.075	0.765	0.822	9	7.3	9.5
52	7.82	11.98	100.9	97.2	46.000	20.850	0.765	0.800	9	4.5	5.3
53	7.82	10.80	100.9	99.6	46.000	18.290	0.765	0.803	9	7.4	13.4
54	7.82	11.20	100.9	107.7	46.000	10.220	0.765	0.798	9	7.5	12.6
55	7.82	11.49	100.9	82.3	46.000	45.160	0.765	0.781	9	10.6	13.8
56	7.82	7.77	100.9	142.2	46.000	12.500	0.765	0.764	7	17.5	4.0
57	7.58	11.39	125.7	117.7	14.000	6.180	0.699	0.806	9	1.8	2.2
58	7.58	10.32	125.7	195.2	14.000	0.075	0.699	0.822	9	7.7	8.1
59	7.58	11.98	125.7	97.2	14.000	20.850	0.699	0.800	9	4.2	4.3
60	7.58	10.80	125.7	99.6	14.000	18.290	0.699	0.803	9	7.0	11.5
61	7.58	11.20	125.7	107.7	14.000	10.220	0.699	0.798	9	6.8	10.7
62	7.58	11.49	125.7	82.3	14.000	45.160	0.699	0.781	9	10.6	12.5
63	7.58	7.77	125.7	142.2	14.000	12.500	0.699	0.764	7	10.6	1.0
									Over all MPD	10.3	6.4

[a]Details of data sets are the same as in Table-1.

TABLE-3
LIST OF SOLVENTS AND THE REFERENCES OF SOLUBILITY DATA IN BINARY SOLVENTS AND MEAN PERCENTAGE DEVIATIONS (MPD) FOR PREDICTED SOLUBILITY USING THE JOUYBAN-ACREE MODEL WITH MODEL CONSTANTS FROM EQUATIONS 3-5 AND 6-8 (NUMBER OF DATA POINTS FOR ALL SETS WAS 19)

No.	Solvent 1	Solvent 2	Solvent 3	Reference	Equations 3-5	Equations 6-8
1	1-Butanol	Cyclohexane	Heptane	17	49.0	5.6
2	1-Propanol	1-Butanol	2,2,4-Trimethylpentane	18	36.3	10.3
3	1-Propanol	1-Butanol	Cyclohexane	19	23.9	14.3
4	1-Propanol	1-Butanol	Heptane	20	35.5	1.3
5	1-Propanol	2-Butanol	2,2,4-Trimethylpentane	18	37.6	7.6
6	1-Propanol	2-Butanol	Cyclohexane	19	32.1	12.8
7	1-Propanol	2-Butanol	Heptane	20	38.2	4.0
8	1-Propanol	Cyclohexane	Heptane	17	50.7	7.2
9	2-Butanol	Cyclohexane	Heptane	17	54.6	17.1
10	2-Methyl-1-propanol	Cyclohexane	Heptane	17	53.8	15.1
11	2-Propanol	1-Butanol	2,2,4-Trimethylpentane	18	41.6	4.5
12	2-Propanol	1-Butanol	Cyclohexane	19	34.4	13.9
13	2-Propanol	1-Butanol	Heptane	20	41.1	6.1
14	2-Propanol	2-Butanol	2,2,4-Trimethylpentane	18	42.4	1.6
15	2-Propanol	2-Butanol	Cyclohexane	19	38.5	19.4
16	2-Propanol	2-Butanol	Heptane	20	44.8	12.6
17	2-Propanol	Cyclohexane	Heptane	17	56.1	17.9
				Overall MPD	41.8	10.1

solubility of pyrene in mixed solvent systems using experimental data of the solute in mono-solvent systems and the expected prediction error is *ca.* 6 and *ca.* 10 %, respectively for binary and ternary solvents which is acceptable error range for prediction purposes.

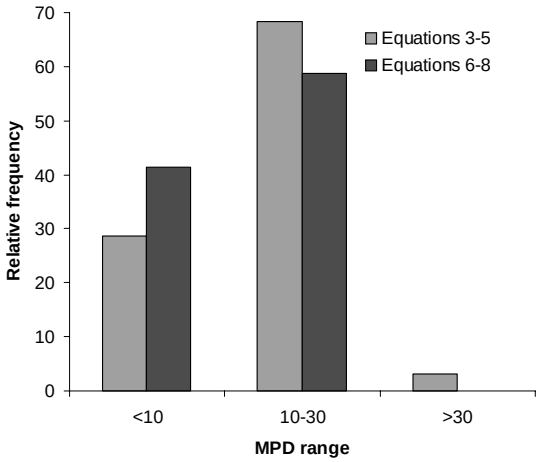

Fig. 1. Relative frequency of mean percentage deviation values for two studied methods

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(*Received*: 14 March 2006; *Accepted*: 13 October 2006) AJC-5191