Solubility of Anthracene in Multicomponent Solvent Mixtures Containing Propanol, Butanol, and Alkanes

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Experimental solubilities are reported for anthracene dissolved in ternary solvent mixtures 1-propanol + 2-propanol + cyclohexane and 1-butanol + 2-butanol + cyclohexane, for anthracene dissolved in the heptanary solvent mixture 1-propanol + 2-propanol + 1-butanol + 2-butanol + cyclohexane + heptane + 2,2,4-trimethylpentane, and for anthracene dissolved in binary solvent heptane + 2,2,4-trimethylpentane at 298.15 K. For the two ternary solvent systems anthracene mole fraction solubilities are reported at 19 different ternary compositions spanning the entire mole fraction range. In the case of heptanary solvent system, solubility data was measured at eight solvent compositions. Results of these measurements are used to test the predictive ability of the combined nearly ideal multiple solvent (NIMS)/Redlich−Kister and combined NIMS/BAB equations. Computations showed that both solution models provided reasonably accurate predictions for how the mole fraction solubility varied with solvent compositions. Deviations between predicted and experimental values were on the order of ±0.95% for the better of the two predictive equations.

Introduction

Solid−liquid equilibrium data of organic nonelectrolyte systems are becoming increasingly important in the petroleum industry, particularly in light of present trends toward heavier feedstocks and the known carcinogenicity/mutagenicity of many of the larger polycyclic aromatic compounds. Solubility data for a number of polycyclic aromatic hydrocarbons (i.e., anthracene and pyrene) and heteroatom polynuclear aromatics (i.e., carbazole, dibenzothiophene, and xanthene) have been published in recent chemical literature. For a listing of references see Acree (1994, 1995a,b). Despite efforts by experimentalists and scientific organizations, both in terms of new experimental measurements and critically evaluated data compilations, there still exist numerous systems for which solubility data are not readily available.

To address this problem, researchers have turned to group contribution methods and semiempirical equations to predict desired quantities from either pure component properties or measured binary data. In earlier studies we have used the binary solvent reduction of a predictive expression derived from a combined two- and three-body interactional mixing model as a mathematical representation for describing how the measured isothermal solubility of a crystalline solute varies with binary solvent composition. The binary reduction, referred to as the combined NIBS/Redlich−Kister equation (NIBS = nearly ideal binary solvent), was found to accurately describe the observed solubility behavior in a large number of different binary solvent systems (Acree, 1992; Acree and Zvaigzne, 1991; Acree et al., 1991). Our mathematical representation, when extended to ternary solvent mixtures, was recently found to provide reasonably accurate predictions for the mole fraction solubility of anthracene, $x_A^{sat.}$, in 8 different ternary two alkane + alcohol (Deng and Acree, 1998a; Deng et al., 1999b) and 12 different ternary alkane + two alcohol (Deng and Acree, 1998b; Deng et al., 1999a,b) solvent mixtures

$$\ln x_A^{sat.} = x_B^\circ \ln(x_A^{sat.})_B + x_C^\circ \ln(x_A^{sat.})_C + x_D^\circ \ln(x_A^{sat.})_D +$$

$$x_B^\circ x_C^\circ \sum_{i=0}^{r} S_{i,BC}(x_B^\circ - x_C^\circ)^i + x_B^\circ x_D^\circ \sum_{j=0}^{s} S_{j,BD}(x_B^\circ - x_D^\circ)^j +$$

$$x_C^\circ x_D^\circ \sum_{k=0}^{t} S_{k,CD}(x_C^\circ - x_D^\circ)^k \quad (1)$$

using curve-fit parameters ($S_{i,BC}$, $S_{j,BD}$, and $S_{k,CD}$) deduced from the measured anthracene solubility data in the three contributing subbinary solvent systems. In the above expression x_B°, x_C°, and x_D° denote the initial mole fraction compositions of the ternary solvent mixture calculated in the absence of the solute. In keeping with our past terminology, eq 1 will be referred to hereafter as the ternary solvent form of the more general combined nearly ideal multiple solvent (NIMS)/Redlich−Kister model.

In the present investigation we report the solubility of anthracene in ternary 1-propanol + 2-propanol + cyclohexane and 1-butanol + 2-butanol + cyclohexane solvent mixtures and in the heptanary 1-propanol + 2-propanol + 1-butanol + 2-butanol + cyclohexane + heptane + 2,2,4-trimethylpentane solvent system. Also reported are anthracene solubilities in binary heptane + 2,2,4-trimethylpentane. These measurements were performed so that we could better assess the predictive accuracy of the general solution model from which eq 1 was derived. To our knowledge, this study represents the first time that anyone has applied predictive solubility equations to a heptanary solvent mixture.

Experimental Methods

Anthracene (Acros, 99.9+%) was recrystallized three times from 2-propanone. 1-Propanol (Aldrich, 99+%, an-

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Table 1. Experimental Mole Fraction Solubilities of Anthracene ($x_A^{sat.}$) in Ternary Solvent Mixtures at 298.15 K

$x_B°$	$x_C°$	$x_A^{sat.}$	$x_B°$	$x_C°$	$x_A^{sat.}$
1-Propanol (B) + 2-Propanol (C) + Cyclohexane (D)
0.3804	0.3570	0.000897	0.1946	0.1880	0.001395
0.1650	0.7220	0.000610	0.7350	0.1554	0.000709
0.2990	0.2887	0.001126	0.5465	0.2649	0.000812
0.2773	0.5317	0.000761	0.1323	0.2490	0.001389
0.7297	0.1170	0.000782	0.1167	0.7275	0.000664
0.7111	0.2081	0.000661	0.2603	0.1395	0.001389
0.2164	0.7038	0.000567	0.1342	0.4639	0.001053
0.1313	0.5520	0.000924	0.4666	0.1318	0.001107
0.4179	0.5031	0.000604	0.5560	0.1282	0.000990
0.5153	0.4032	0.000617			
1-Butanol (B) + 2-Butanol (C) + Cyclohexane (D)
0.3531	0.3475	0.001060	0.1728	0.1666	0.001520
0.1557	0.7135	0.000806	0.7165	0.1532	0.000909
0.2727	0.2707	0.001284	0.5219	0.2567	0.000991
0.2623	0.5162	0.000938	0.1154	0.2233	0.001476
0.7231	0.1023	0.000975	0.1074	0.7174	0.000846
0.7134	0.1988	0.000849	0.2269	0.1127	0.001508
0.2038	0.7107	0.000738	0.1140	0.4305	0.001248
0.1126	0.5298	0.001104	0.4313	0.1126	0.001307
0.4087	0.5061	0.000792	0.5324	0.1103	0.001188
0.5102	0.4045	0.000789			

hydrous), 2-propanol (Aldrich, 99+%, anhydrous), 1-butanol (Aldrich, HPLC, 99+%), 2-butanol (Aldrich, 99+%, anhydrous), cyclohexane (Aldrich, HPLC, 99.9+%), heptane (Aldrich, HPLC, 99+%), and 2,2,4-trimethylpentane (Aldrich, HPLC) were stored over molecular sieves and distilled shortly before use. Gas chromatographic analysis showed solvent purities to be 99.7 mol % or better. All solvent mixtures were prepared by mass so that compositions could be calculated to 0.0001 mole fraction. The methods of sample equilibration and spectrophotometric analysis are discussed in an earlier paper (Powell et al., 1997). Experimental anthracene solubilities are listed in Tables 1−3. Numerical values represent the average of between four and eight independent determinations, with the measured values being reproducible to within ±1.5%.

Results and Discussion

Conceptually, eq 1 traces back to when Acree and Bertrand (1981) employed the NIBS model to estimate the solubility of benzoic acid in ternary solvent mixtures. The authors derived expressions based upon the assumption that the carboxylic acid solute existed in solution entirely as monomers or completely in dimeric form. The assumptions represented the two limiting conditions. For molecules having identical molar volumes, the predictive expression derived by the authors can be written as

$$\ln x_A^{sat.} = x_B° \ln(x_A^{sat.})_B + x_C° \ln(x_A^{sat.})_C + x_D° \ln(x_A^{sat.})_D + G_{BCD}^E (RT)^{-1} \quad (2)$$

a weighted mole fraction average of the logarithmic solute solubilities in the three pure solvents plus a term containing the excess Gibbs free energy of the ternary solvent mixtures. Equation 2 is derived for solutes having very limited saturation solubilities.

Thermodynamic properties of ternary mixtures were relatively scarce in 1981, and the required G_{BCD}^E values had to be estimated from available binary vapor−liquid equilibria (VLE) data. Several predictive models had been suggested for estimating ternary thermodynamic and physical properties from measured binary data. Equation

Table 2. Experimental Mole Fraction Solubilities of Anthracene ($x_A^{sat.}$) in Heptanary 1-Propanol (B) + 2-Propanol (C) + 1-Butanol (D) + 2-Butanol (E) + Cyclohexane (F) + Heptane (G) + 2,2,4-Trimethylpentane (H) Solvent Mixtures at 298.15 K

solvent composn/solubility	solvent composn/solubility
mixture 1	mixture 2
$x_B° = 0.1474$	$x_B° = 0.0794$
$x_C° = 0.1419$	$x_C° = 0.0719$
$x_D° = 0.1452$	$x_D° = 0.0729$
$x_E° = 0.1400$	$x_E° = 0.0728$
$x_F° = 0.1421$	$x_F° = 0.0747$
$x_G° = 0.1429$	$x_G° = 0.0855$
$x_H° = 0.1405$	$x_H° = 0.5428$
$x_A^{sat.} = 0.001\ 116$	$x_A^{sat.} = 0.001\ 164$
mixture 3	mixture 4
$x_B° = 0.5344$	$x_B° = 0.0745$
$x_C° = 0.0727$	$x_C° = 0.5387$
$x_D° = 0.0985$	$x_D° = 0.0733$
$x_E° = 0.0742$	$x_E° = 0.0944$
$x_F° = 0.0731$	$x_F° = 0.0743$
$x_G° = 0.0736$	$x_G° = 0.0730$
$x_H° = 0.0735$	$x_H° = 0.0718$
$x_A^{sat.} = 0.000\ 875$	$x_A^{sat.} = 0.000\ 784$
mixture 5	mixture 6
$x_B° = 0.0753$	$x_B° = 0.0722$
$x_C° = 0.0765$	$x_C° = 0.0724$
$x_D° = 0.0752$	$x_D° = 0.5364$
$x_E° = 0.5466$	$x_E° = 0.0742$
$x_F° = 0.0751$	$x_F° = 0.0985$
$x_G° = 0.0762$	$x_G° = 0.0744$
$x_H° = 0.0751$	$x_H° = 0.0719$
$x_A^{sat.} = 0.000\ 888$	$x_A^{sat.} = 0.000\ 996$
mixture 7	mixture 8
$x_B° = 0.0731$	$x_B° = 0.0884$
$x_C° = 0.0689$	$x_C° = 0.0681$
$x_D° = 0.0760$	$x_D° = 0.0741$
$x_E° = 0.0795$	$x_E° = 0.0752$
$x_F° = 0.5513$	$x_F° = 0.0777$
$x_G° = 0.0765$	$x_G° = 0.5432$
$x_H° = 0.0747$	$x_H° = 0.0733$
$x_A^{sat.} = 0.001\ 467$	$x_A^{sat.} = 0.001\ 412$

Table 3. Experimental Mole Fraction Solubilities of Anthracene ($x_A^{sat.}$) in Binary Heptane (B) + 2,2,4-Trimethylpentane (C) Solvent Mixtures at 298.15 K

$x_C°$	$x_A^{sat.}$	$x_C°$	$x_A^{sat.}$	$x_C°$	$x_A^{sat.}$
0.0000	0.001 571	0.3713	0.001 328	0.7804	0.001 140
0.0912	0.001 464	0.4721	0.001 280	0.8839	0.001 098
0.1857	0.001 423	0.5705	0.001 230	1.0000	0.001 074

1 can be derived by substituting the Redlich−Kister prediction for

$$G_{BCD}^E = x_B° x_C° \sum_{i=0}^{r} G_{i,BC}(x_B° - x_C°)^i +$$

$$x_B° x_D° \sum_{j=0}^{s} G_{j,BD}(x_B° - x_D°)^j + x_C° x_D° \sum_{k=0}^{t} G_{k,CD}(x_C° - x_D°)^k$$

(3)

into eq 2. The various $G_{i,BC}$, $G_{j,BD}$, and $G_{k,CD}$ values represent coefficients deduced from a regressional analysis of experimental binary VLE data. Our approach differs slightly in application in that we now prefer to determine the Redlich−Kister coefficients from measured solute solubilities in the three contributing subbinary solvent mixtures, rather than from available VLE data. To indicate this practice, we have denoted the curve-fit coefficients in eq 1 as $S_{i,BC}$, $S_{j,BD}$, and $S_{k,CD}$. The binary reduction of eq 1 has been used as a mathematical representation of experi-

Table 4. Summarized Comparison between Experimental Anthracene Solubilities and Predicted Values Based Upon the Combined NIMS/Redlich−Kister and Combined NIMS/BAB Equations

	% dev[a]	
solvent system	NIMS/Redlich−Kister	NIMS/BAB
ternary solvent systems		
1-propanol (B) + 2-propanol (C) + cyclohexane (D)	0.94	0.79
1-butanol (B) + 2-butanol (C) + cyclohexane (D)	1.25	1.50
heptanary solvent system		
1-propanol (B) + 2-propanol (C) + 1-butanol (D) + 2-butanol (E) + cyclohexane (F) + heptane (G) + 2,2,4-trimethylpentane (H)	0.66	1.17
av dev	0.95	1.15

[a] Deviation (%) = $(100/N)\sum |[(x_A^{sat.})^{calc} - (x_A^{sat.})^{exp}]/(x_A^{sat.})^{exp}|$; where N is the number of data points in each data set. For the ternary solvent mixtures $N = 19$, and for the heptanary solvent mixture $N = 8$.

mental isothermal mole fraction solubility data for solutes dissolved in binary solvent mixtures. In the case of solutes dissolved in binary aqueous−organic solvent mixtures, the experimental solubility data were represented by a modified form of eq 1 which contained volume fraction compositions of the various solvent components rather than mole fractions (Jouyban-Gharamaleki and Acree, 1998; Jouyban-Gharamaleki et al., 1998). The modified form is derivable from the general NIBS model by approximating the weighting factors with molar volumes. Equation 1 assumes that the weighting factors of all components are equal.

Careful inspection of eq 1 requires that one must parametrize the measured binary solvent solubility data in the specific form of

$$\ln x_A^{sat.} = x_B^\circ \ln(x_A^{sat.})_B + x_C^\circ \ln(x_A^{sat.})_C +$$
$$x_B^\circ x_C^\circ \sum_{i=0}^{n} S_{i,BC}(x_B^\circ - x_C^\circ)^i \quad (4)$$

as the actual numerical values of the $S_{i,BC}$ coefficients are needed in the calculation. This requirement presents no problems in the present study as the coefficients of eq 4 were given when we communicated the binary solvent solubility data. Not all researchers parametrize measured solubility data in accordance with eq 4. For example, in the pharmaceutical industry isothermal solubility data may be expressed in terms of the extended Hildebrand solubility parameter approach (Barra et al., 1997; Martin et al., 1980, 1982), the double log−log model (Barzegar-Jalali and Hanaee, 1994), modified Wilson model (Jouyban-Gharamaleki, 1997), or as a power series expansion in mole fraction composition of one of the binary solvent components. Solubility data for select crystalline polycyclic aromatic hydrocarbons dissolved in binary aqueous−organic solvent mixtures were described using the Margules equation in a recently published paper (Fan and Jafvert, 1997) appearing in an environmental science journal.

We note that the aforementioned solubility study of Acree and Bertrand (1981) contained provisions for mathematical representations other than the Redlich−Kister equation. There is no requirement that the ternary G_{BCD}^E value in eq 2 be estimated with the Redlich−Kister equation. In fact, Acree and Bertrand used the BAB equation to generate the excess Gibbs energy of the ternary solvent in their benzoic acid solubility study. The BAB equation combines binary G_{IJ}^E values at specified solvent concentrations

$$G_{BCD}^E = (x_B^\circ + x_C^\circ)^2(G_{BC}^E)^* + (x_B^\circ + x_D^\circ)^2(G_{BD}^E)^* +$$
$$(x_C^\circ + x_D^\circ)^2(G_{CD}^E)^* \quad (5)$$

rather than using coefficients of a particular mathematical representation. The excess Gibbs of the *ij* binary solvent mixture, $(G_{IJ}^E)^*$, is calculated at a mole fraction composition of $x_i^* = x_I^\circ/(x_I^\circ + x_J^\circ)$. For a ternary solvent mixture, the predictive solubility equation takes the form of

$$\ln x_A^{sat.} = x_B^\circ \ln(x_A^{sat.})_B + x_C^\circ \ln(x_A^{sat.})_C + x_D^\circ \ln(x_A^{sat.})_D +$$
$$(x_B^\circ + x_C^\circ)^2(G_{BC}^E)^*(RT)^{-1} + (x_B^\circ + x_D^\circ)^2(G_{BD}^E)^*(RT)^{-1} +$$
$$(x_C^\circ + x_D^\circ)^2(G_{CD}^E)^*(RT)^{-1} \quad (6)$$

which generalizes to

$$\ln x_A^{sat.} = \sum_I^{solv} x_i^\circ \ln(x_A^{sat.})_i + \sum_I^{solvsolv}\sum_J (x_I^\circ + x_J^\circ)^2(G_{IJ}^E)^*(RT)^{-1} \quad (7)$$

The BAB equation is described in greater detail elsewhere (Bertrand et al., 1983; Acree and Bertrand, 1983). As in the case of eq 1, we will determine the needed $(G_{IJ}^E)^*$ values from measured solubility data in the three binary contributing subbinary solvent systems

$$(G_{IJ}^E)^*(RT)^{-1} = \ln[x_A^{sat.}(\text{at } x_I^*)] - x_I^* \ln(x_A^{sat.})_I -$$
$$x_J^* \ln(x_A^{sat.})_J \quad (8)$$

when using the combined NIMS/BAB equation to predict anthracene solubilities in ternary (and higher-order multicomponent) solvent mixtures. Any mathematical representation can be used for describing how $\ln x_A^{sat.}$ varies with binary solvent composition.

Similarly, the generalized form of the combined NIMS/Redlich−Kister equation is given by

$$\ln x_A^{sat.} = \sum_I^{solv} x_I^\circ \ln(x_A^{sat.})_I +$$
$$\sum_I^{solvsolv}\sum_J [x_I^\circ x_J^\circ \sum_k S_{k,IJ}(x_I^\circ - x_J^\circ)^k] \quad (9)$$

The first summation in eq 9 extends over all solvents in the multicomponent mixture, whereas the double summation extends over all binary combinations of solvents. In a heptanary solvent mixture, as is the case here, there are 21 binary solvent combinations (BC, BD, BE, BF, BG, BH, CD, CE, CF, CG, CH, DE, DF, DG, DH, EF, EG, EH, FG, FH, GH).

The predictive abilities of the combined NIMS/Redlich−Kister and combined NIMS/BAB equations are summarized in Table 4 for anthracene dissolved in the two ternary solvent systems and in the heptanary solvent system. For convenience we have listed in Table 5 the

Table 5. Combined NIBS/Redlich−Kister Parameters Calculated from Anthracene Solubilities in the Subbinary Solvent Systems

solvent (I) + solvent (J)	$S_i{}^a$
cyclohexane (I) + 1-propanol (J)	1.121
	0.040
	0.256
heptane (I) + 1-propanol (J)	1.098
	−0.106
	0.324
2,2,4-trimethylpentane (I) + 1-propanol (J)	0.825
	−0.103
	0.291
cyclohexane (I) + 2-propanol (J)	1.589
	−0.143
	0.248
heptane (I) + 2-propanol (J)	1.491
	−0.587
	0.566
2,2,4-trimethylpentane (I) + 2-propanol (J)	1.193
	−0.369
	0.333
cyclohexane (I) + 1-butanol (J)	0.741
	0.345
	0.223
heptane (I) + 1-butanol (J)	0.723
	0.004
	0.201
2,2,4-trimethylpentane (I) + 1-butanol (J)	0.536
	0.151
	0.142
cyclohexane (I) + 2-butanol (J)	1.260
	0.206
heptane (I) + 2-butanol (J)	1.225
	−0.292
2,2,4-trimethylpentane (I) + 2-butanol (J)	1.070
	−0.213
1-propanol (I) + 2-propanol (J)	0.068
	−0.058
1-propanol (I) + 1-butanol (J)	0.117
	−0.015
1-propanol (I) + 2-butanol (J)	0.000
	0.000
2-propanol (I) + 1-butanol (J)	0.243
	0.011
2-propanol (I) + 2-butanol (J)	0.097
	0.013
1-butanol (I) + 2-butanol (J)	−0.072
	0.000
cyclohexane (I) + heptane (J)	0.153
	0.089
	−0.084
cyclohexane (I) + 2,2,4-trimethylpentane (J)	−0.011
	−0.021
heptane (I) + 2,2,4-trimethylpentane (J)	−0.083[b]
	−0.079[b]
	−0.341[b]

^{a} Combined NIBS/Redlich−Kister curve-fit parameters are ordered as S_0, S_1, and S_2. Curve-fit coefficients were taken from Acree et al. (1994), Acree and Zvaigzne (1994), Zvaigzne and Acree (1994), Zvaigzne et al. (1993, 1995), Deng and Acree (1998a), and Deng et al. (1999b). ^{b} Determined as part of this work.

various S_i curve-fit coefficients needed in eq 1 to predict the solubility of anthracene in the solvent systems studied. The measured solubilities in 1-propanol ($x_A^{\text{sat.}}$ = 0.000 591), 2-propanol ($x_A^{\text{sat.}}$ = 0.000 411), 1-butanol ($x_A^{\text{sat.}}$ = 0.000 801), 2-butanol ($x_A^{\text{sat.}}$ = 0.000 585), cyclohexane ($x_A^{\text{sat.}}$ = 0.001 553), heptane ($x_A^{\text{sat.}}$ = 0.001 571), and 2,2,4-trimethylpentane ($x_A^{\text{sat.}}$ = 0.001 074) were taken from our earlier publications (Acree et al., 1994; Acree and Zvaigzne, 1994; Zvaigzne and Acree, 1994; Zvaigzne et al., 1993). Examination of Table 5 reveals that both models provide very reasonable predictions for the solubility behavior of anthracene. The overall average absolute deviation be-

tween experimental and predicted mole fraction solubilities is approximately 0.95 and 1.15% for the combined NIMS/Redlich−Kister and combined NIMS/BAB equations, respectively. Deviations are comparable in magnitude to the ±1.5% experimental uncertainty associated with each measured mole fraction solubility.

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