

Solubility of Anthracene in C₁–C₃ Alcohols from (298.2 to 333.2) K and Their Mixtures with 2-Propanone at 298.2 K

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The anthracene solubility in methanol, ethanol, 1-propanol, and 2-propanol from (298.2 to 333.2 K) and also in binary solvent mixtures of 2-propanone + C₁–C₃ alcohols at 298.2 K was reported. The van't Hoff equation was used for correlation of the solubility data in monosolvents at different temperatures. The solubility of anthracene in mixed solvents was predicted using previously developed quantitative structure–property relationships (QSPR) that required knowledge of the solubility data in monosolvents. The overall mean percentage deviation of the correlated solubilities in monosolvents at different temperatures was 4.8 %. The corresponding value for solubility prediction methods in solvent mixtures varied from (8.5 to 11.9) %. Using ab initio prediction methods, the overall mean percentage deviations varied from (14.7 to 15.8) %.

Introduction

Solubility is an important consideration in many chemical and pharmaceutical applications, including crystallization, separation, decontamination, liquid extraction, and drug formulation design. Solubility data of anthracene are essential in the petroleum industry and in environmental decontamination. In many instances, the solubility in a neat solvent is not sufficient for the desired application. Temperature alteration and solvent mixing are the common methods to enhance the solubility of lowly soluble compounds. However, there is a significant lack of solubility data for many solutes; therefore, efforts have been devoted to developing predictive mathematical models that require a minimum number of experimental input values.¹

Experimental solubility data of anthracene in a number of binary and ternary solvent mixtures have been reported in alcohols, ethers, alkenes, esters, and aromatic compounds by Acree and co-workers. Recently, the anthracene solubility in ternary solvent mixtures of 2,2,4-trimethylpentane + 2-propanone + alcohols² and quaternary solvent mixtures of 2,2,4-trimethylpentane + 2-propanone + methanol + alcohols was reported.³ Experimental anthracene solubilities in binary ketone + alcohol solvent mixtures have not been reported to date.

A numerical method was developed employing the Jouyban–Acree model, the Abraham solute parameters, and the Abraham solvent coefficients. This combination enables one to predict the solubility of a solute in nonaqueous solvent mixtures employing the experimental and/or computed solubility data in monosolvents. The basic Jouyban–Acree model is⁴

$$\ln C_m^{\text{Sat}} = x_1 \ln C_1^{\text{Sat}} + x_2 \ln C_2^{\text{Sat}} + x_1 x_2 \sum_{i=0}^2 J_i (x_1 - x_2)^i \quad (1)$$

where C_m^{Sat} is the molar solubility of the solute in the binary solvent mixtures; x_1 and x_2 are the mole fractions of solvents 1 and 2 in the absence of the solute; C_1^{Sat} and C_2^{Sat} denote the molar solubility of the solute in neat solvents 1 and 2; and J_i is the solvent–solvent and solute–solvent interaction terms. In a previous work, quantitative structure–property relationship (QSPR) models were proposed to calculate the numerical values of the J_i terms using Abraham's water-to-solvent and gas-to-solvent coefficients of the solvents and Abraham's parameters of the solutes.⁵ These parameters (both solute and solvent parameters) are used as the representative descriptors for possible interactions between solvents 1 and 2 and the solute in the solution.

The purposes of the present study are to report the experimental solubility data of anthracene in methanol, ethanol, 1-propanol, and 2-propanol at different temperatures and in the binary mixtures of acetone and C₁–C₃ alcohols at 298.2 K. The experimental results are used to assess the applicability of previously reported QSPR models for predicting the solubility of solutes in binary solvent mixtures.

Experimental Method

Materials. Anthracene (purity 0.96 in mass fraction) was purchased from Fluka, and it was recrystallized several times from 2-propanone and ethyl acetate to yield a purified sample having a melting point of 488 K. Its purity was checked by a thin layer chromatography method,⁶ and also the measured solubilities in a number of monosolvents were compared with the corresponding data from the literature. Methanol (0.995 in mass fraction), ethanol (0.999 in mass fraction), 2-propanol (0.995 in mass fraction), and 2-propanone (0.995 in mass

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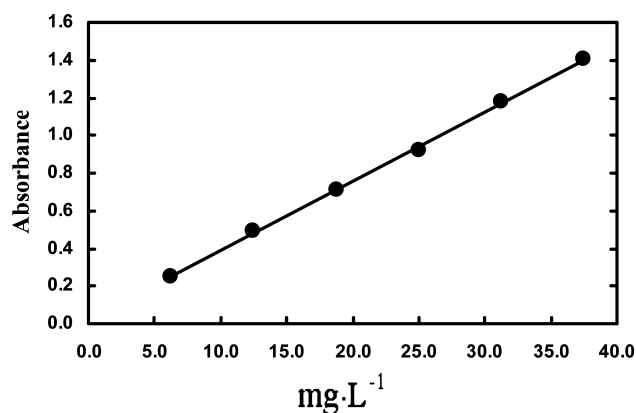


Figure 1. UV calibration curve for anthracene in 2-propanone at 357 nm.

fraction) were purchased from Scharlau Chemie (Spain) and 1-propanol (0.995 in mass fraction) from Merck (Germany).

Apparatus and Procedures. The solubility of anthracene in methanol, ethanol, 1-propanol, and 2-propanol was measured at (298.2, 303.2, 313.2, 323.2, and 333.2) K. An excess amount of the solid was placed into each of the monosolvents and incubated in ovens at (298.2 ± 0.2, 303.2 ± 0.2, 313.2 ± 0.4, 323.2 ± 0.4, and 333.2 ± 0.6) K (Behdad, Tehran, Iran) at least for five days. The binary solvent mixtures were prepared by mixing the appropriate volumes of the solvents, and then the mole fractions of the solvents were computed from the densities of the neat solvents. The solvent composition could be calculated to 0.001 mole fraction. The solubility of anthracene in mixed solvents was determined by equilibrating an excess amount of the solid with the binary solvent mixtures using a shaker (Behdad, Tehran, Iran) placed in an incubator equipped with a temperature controlling system at 298.2 ± 0.2 K (Nabziran, Tabriz, Iran). Attainment of the equilibrium was verified by measurements at different times after 72 h. The solutions were filtered using hydrophobic Durapore filters (0.45 μm, Millipore, Ireland) and then diluted with 2-propanone for spectrophotometric analysis at 357 nm with a UV-vis spectrophotometer (Beckman DU-650, Fullerton, USA). The calibration graph is shown in Figure 1. Concentrations of the dilute solutions were determined from a UV absorbance calibration graph with the molar absorptivities of anthracene ranging from 667 ε/(L·mol⁻¹·cm⁻¹) to 713 ε/(L·mol⁻¹·cm⁻¹) for the anthracene concentrations ranging from (0.0004 to 0.0021) mol·L⁻¹. Each experimental data point is an average of at least three independent measurements with the measured mol·L⁻¹ solubilities being reproducible to within 2.3 % and 2.4 % in monosolvents at different temperatures and mixtures of solvents, respectively. Calculated standard deviations of independent solubility measurements for each data point ranged from σ_{n-1} = 0.0000 (i.e., < 0.00005) to σ_{n-1} = 0.0010 mol·L⁻¹ and σ_{n-1} = 0.0001 to σ_{n-1} = 0.0032 mol·L⁻¹ in monosolvents at different temperatures and in solvent mixtures at 298.2 K, respectively. Densities of the saturated solutions were measured using a 5 mL pycnometer with the uncertainty of 0.0001 g·cm⁻³.

Computational Methods

For correlation of experimental solubility data in monosolvents at different temperatures, the van't Hoff equation is used⁷

$$\ln C_T^{\text{Sat}} = \frac{a}{T} + b \quad (2)$$

where C_T^{Sat} is the saturated molar solubility at temperature T , and a and b are the model constants calculated using a least-squares method.

The J_i terms of the Jouyban–Acree model were computed using eqs 2s to 4s (Supporting Information, SI) and then used to predict the anthracene solubility data in binary solvent mixtures using eq 1, employing experimental solubilities of anthracene in monosolvents. This numerical analysis was called method I. The same computations were carried out employing eqs 5s to 7s (SI). The latter computation method is referred to as numerical method II. To further reduce the experimental data requirement in the prediction process, it is possible to use the Abraham solvation models to predict the solubilities of anthracene in the monosolvents, that is, C_1^{Sat} and C_2^{Sat} . The Abraham model for the water-to-solvent process is

$$\log\left(\frac{C_S}{C_W}\right) = c + e \cdot E + s \cdot S + a \cdot A + b \cdot B + v \cdot V \quad (3)$$

and for the gas-to-solvent process

$$\log\left(\frac{C_S}{C_G}\right) = c + e \cdot E + s \cdot S + a \cdot A + b \cdot B + l \cdot L \quad (4)$$

where c , e , s , a , b , v , and l are the model constants (i.e., the Abraham solvent coefficients), E , S , A , B , V , and L are the Abraham work parameters, C_S (the same term as C_1^{Sat} and C_2^{Sat} from this work) and C_W are the solute solubility in the organic solvent and water (in mol·L⁻¹), respectively, and C_G is the gas phase concentration of the solute.⁸ The basic Abraham model involves two different transfer processes, one for solute transfer between two condensed phases (such as water-to-organic solvent) and a separate equation for solute transfer to a condensed phase from the gas phase. The originating phase for the solute transfer process is essentially a reference point, and one can use the correlation to predict the solute properties in the final phase, provided that the solute properties in the originating phase are known. Equation 3 requires the aqueous solubility of the solute (C_W), and eq 4 requires the C_G value. Both equations have been used to correlate and to estimate the solubility of crystalline solutes in organic solvents.⁸⁻¹¹ The numerical values of the solute's Abraham experimental parameters for anthracene are: $\log C_W = -6.430$, $E = 2.290$, $S = 1.340$, $A = 0.000$, $B = 0.280$, $V = 1.454$, and $L = 7.568$.⁸ The $\log C_G = -9.460$ was taken from a published paper by Acree and Abraham.⁸ The predicted C_1^{Sat} and C_2^{Sat} from eq 3 and J_i terms computed using eqs 2s to 4s (SI) were used to predict the C_m^{Sat} , and this numerical method was called method III. A similar numerical analysis employing eqs 5s to 7s (SI) and eq 4 was called method IV.

All calculated C_m^{Sat} solubilities were compared with the corresponding experimental values, and the mean percentage deviation (MPD) was calculated as an accuracy criterion by

$$\text{MPD} = \frac{100}{N} \sum \frac{|(C_m^{\text{Sat}})_{\text{calculated}} - (C_m^{\text{Sat}})_{\text{experimental}}|}{(C_m^{\text{Sat}})_{\text{experimental}}} \quad (5)$$

Table 1. Experimental (expt.) and Correlated (cor) Molar Solubilities^a of Anthracene in Methanol, Ethanol, 1-Propanol, and 2-Propanol Monosolvents, Densities at (298.2 to 333.2) K, *a* and *b* Values (\pm Standard Errors) of Equation 2, and the Mean Percentage Deviation (MPD)

<i>T</i>	$C_T^{\text{Sat}}/\text{mol}\cdot\text{L}^{-1}$		100·deviation ^b
	ρ		
K	$\text{g}\cdot\text{cm}^{-3}$	expt. cor	
Methanol			
$\{a = (-2.31 \pm 0.15) \cdot 10^3, b = 2.7 \pm 0.5 \text{ K}\}$			
298.2	0.7883	0.0063 0.0064	1.6
303.2	0.7842	0.0073 0.0073	0.1
313.2	0.7793	0.0085 0.0093	9.7
323.2	0.7761	0.0114 0.0117	2.7
333.2	0.7704	0.0144 0.0145	0.8
		MPD	3.0
Ethanol			
$\{a = (-3.27 \pm 0.22) \cdot 10^3, b = 6.2 \pm 0.7 \text{ K}\}$			
298.2	0.7844	0.0087 0.0085	2.1
303.2	0.7824	0.0094 0.0102	8.6
313.2	0.7761	0.0128 0.0144	12.5
323.2	0.7732	0.0193 0.0199	3.1
333.2	0.7684	0.0265 0.0269	1.7
		MPD	5.6
1-Propanol			
$\{a = (-3.72 \pm 0.25) \cdot 10^3, b = 7.7 \pm 0.8 \text{ K}\}$			
298.2	0.7995	0.0090 0.0084	8.3
303.2	0.7975	0.0101 0.0104	2.6
313.2	0.7922	0.0143 0.0153	7.3
323.2	0.7875	0.0229 0.0222	3.3
333.2	0.7834	0.0326 0.0313	4.0
		MPD	5.1
2-Propanol			
$\{a = (-4.09 \pm 0.25) \cdot 10^3, b = 8.5 \pm 0.8 \text{ K}\}$			
298.2	0.7813	0.0056 0.0054	3.0
303.2	0.7790	0.0076 0.0068	10.4
313.2	0.7740	0.0102 0.0105	2.7
323.2	0.7670	0.0155 0.0157	1.2
333.2	0.7633	0.0253 0.0229	9.3
		MPD	5.3
		overall	4.8

^a Calculated as:

$$\ln C_{298.2}^{\text{Sat}} = \frac{-2310}{298.2} + 2.7 = -5.0465 \rightarrow C_{298.2}^{\text{Sat}} = 0.0064$$

^b Deviation is calculated as:

$$100 \cdot \text{deviation} = 100 \cdot \frac{|0.0064 - 0.0063|}{0.0063} = 1.6 \%$$

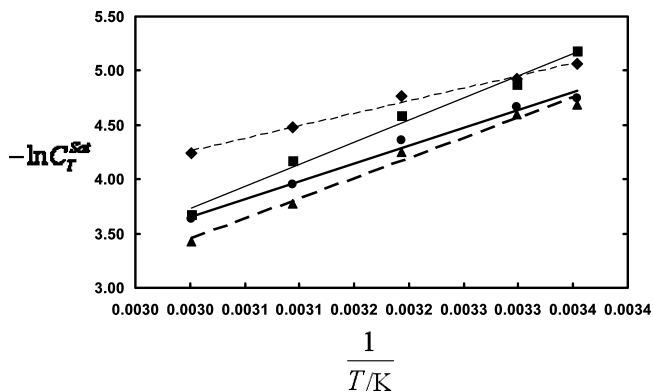


Figure 2. Logarithm of the solubility of anthracene ($-\ln C_T^{\text{Sat}}$) in \blacklozenge , methanol; \bullet , ethanol; \blacktriangle , 1-propanol; \blacksquare , 2-propanol; and their related calculated solubility data: - - -, methanol; thick —, ethanol; - - -, 1-propanol; thin —, 2-propanol, using eq 2.

Results and Discussion

Table 1 gives the experimental molar solubilities of anthracene in methanol, ethanol, 1-propanol, and 2-propanol at the five temperatures studied, measured densities of the saturated

Table 2. Experimental (expt.) and Predicted Molar Solubilities of Anthracene in 2-Propanone (1) + Alcohols (2) Solvent Mixtures and Their Related Saturated Solution Densities at 298.2 K

x_2	ρ		$C_m^{\text{Sat}}/\text{mol}\cdot\text{L}^{-1}$			
	$\text{g}\cdot\text{cm}^{-3}$	expt.	method I ^a	method II ^a	method III ^a	method IV ^a
2-Propanone (1) + Methanol (2)						
1.000	0.8195	0.0063	0.0063	0.0063	0.0070	0.0069
0.942	0.8190	0.0088	0.0078	0.0084	0.0087	0.0090
0.879	0.8227	0.0112	0.0097	0.0110	0.0109	0.0117
0.809	0.8242	0.0153	0.0121	0.0144	0.0138	0.0150
0.731	0.8234	0.0196	0.0153	0.0185	0.0175	0.0189
0.644	0.8240	0.0233	0.0193	0.0236	0.0223	0.0237
0.547	0.8230	0.0308	0.0246	0.0246	0.0286	0.0293
0.437	0.8237	0.0374	0.0314	0.0376	0.0370	0.0358
0.312	0.8275	0.0427	0.0398	0.0464	0.0476	0.0427
0.168	0.8153	0.0470	0.0486	0.0538	0.0591	0.0478
0.000	0.7956	0.0527	0.0527	0.0527	0.0652	0.0449
2-Propanone (1) + Ethanol (2)						
1.000	0.8198	0.0087	0.0087	0.0087	0.0132	0.0091
0.919	0.8201	0.0136	0.0107	0.0110	0.0159	0.0113
0.835	0.8208	0.0166	0.0131	0.0137	0.0192	0.0139
0.746	0.8218	0.0231	0.0161	0.0172	0.0231	0.0171
0.654	0.8221	0.0246	0.0197	0.0215	0.0277	0.0210
0.558	0.8223	0.0291	0.0240	0.0268	0.0332	0.0256
0.457	0.8215	0.0372	0.0292	0.0331	0.0395	0.0310
0.351	0.8209	0.0431	0.0352	0.0402	0.0466	0.0368
0.240	0.8203	0.0432	0.0417	0.0471	0.0541	0.0421
0.123	0.8161	0.0470	0.0480	0.0522	0.0609	0.0456
0.000	0.7956	0.0527	0.0527	0.0527	0.0652	0.0449
2-Propanone (1) + 1-Propanol (2)						
1.000	0.8012	0.0090	0.0090	0.0090	0.0106	0.0067
0.898	0.8012	0.0144	0.0116	0.0118	0.0136	0.0088
0.797	0.8000	0.0198	0.0146	0.0151	0.0172	0.0115
0.696	0.7991	0.0239	0.0181	0.0190	0.0214	0.0147
0.596	0.7973	0.0334	0.0221	0.0237	0.0264	0.0186
0.496	0.7957	0.0371	0.0268	0.0291	0.0322	0.0231
0.396	0.7949	0.0413	0.0321	0.0350	0.0388	0.0282
0.296	0.7939	0.0444	0.0378	0.0411	0.0459	0.0335
0.197	0.7927	0.0458	0.0435	0.0467	0.0532	0.0387
0.098	0.7917	0.0469	0.0488	0.0509	0.0599	0.0428
0.000	0.7956	0.0527	0.0527	0.0527	0.0652	0.0449
2-Propanone (1) + 2-Propanol (2)						
1.000	0.7834	0.0056	0.0056	0.0056	0.0069	0.0050
0.826	0.7834	0.0105	0.0094	0.0097	0.0115	0.0086
0.678	0.7834	0.0151	0.0139	0.0146	0.0171	0.0129
0.551	0.7833	0.0222	0.0191	0.0203	0.0235	0.0178
0.441	0.7842	0.0269	0.0247	0.0264	0.0304	0.0230
0.345	0.7844	0.0307	0.0306	0.0325	0.0377	0.0282
0.260	0.7856	0.0341	0.0363	0.0384	0.0448	0.0332
0.184	0.7871	0.0414	0.0416	0.0435	0.0514	0.0374
0.116	0.7890	0.0444	0.0462	0.0477	0.0571	0.0409
0.055	0.7943	0.0474	0.0500	0.0508	0.0617	0.0434
0.000	0.7956	0.0527	0.0527	0.0527	0.0652	0.0449

^a Numerical methods I and III, the J_i terms of eq 1, were computed employing eqs 2s to 4s (SI); methods II and IV, eqs 5s to 7s (SI) were used to calculate the J_i terms. In methods I and II, the experimental values of C_1^{Sat} and C_2^{Sat} were used in eq 1, whereas in methods III and IV, the predicted C_1^{Sat} and C_2^{Sat} respectively by eqs 3 and 4 were used in the predictions.

solutions, the back-calculated solubilities based on eq 2, and the equation coefficients. Equation 2 was found to describe the observed solubilities to within an overall MPD of 4.8%. Figure 2 compares the experimental and back-calculated solubilities at the five temperatures studied.

The experimental and predicted solubilities of anthracene using methods I to IV are tabulated in Table 2, along with the densities of the saturated solutions. Our measured anthracene solubilities in 2-propanone ($0.053 \text{ mol}\cdot\text{L}^{-1}$), methanol ($0.007 \text{ mol}\cdot\text{L}^{-1}$), ethanol ($0.009 \text{ mol}\cdot\text{L}^{-1}$), 1-propanol ($0.009 \text{ mol}\cdot\text{L}^{-1}$), and 2-propanol ($0.006 \text{ mol}\cdot\text{L}^{-1}$) at 298.2 K were in good agreement with published literature values⁸ for anthracene dissolved in 2-propanone ($0.052 \text{ mol}\cdot\text{L}^{-1}$), methanol ($0.006 \text{ mol}\cdot\text{L}^{-1}$), ethanol ($0.008 \text{ mol}\cdot\text{L}^{-1}$), 1-propanol (0.008

Table 3. Numerical Values of the Mean Percentage Deviation (MPD) for the Predicted Solubilities of Anthracene in 2-Propanone (1) + Alcohols (2) Using Various Numerical Methods and Their Overall Values

solvent 2	MPD			
	method I	method II	method III	method IV
1-propanol	16.2	13.4	14.3	29.4
2-propanol	5.3	5.4	20.3	11.7
ethanol	14.2	10.9	18.5	13.0
methanol	12.0	4.3	9.9	4.6
overall MPD	11.9	8.5	15.8	14.7

mol·L⁻¹), and 2-propanol (0.006 mol·L⁻¹). The solubility of anthracene in binary solvent mixtures was predicted using numerical methods I to IV. The predicted solubilities were compared with the corresponding experimental data, and the computed MPD values are listed in Table 3. Method II has the best prediction capability by employing the solubility data in monosolvents, and its overall MPD (8.5 %) is significantly less than that of method I (11.9 %). There is no significant difference between overall MPDs of methods III and IV. Both are ab initio prediction methods, and to improve the prediction capability of these methods, more accurate solubility prediction tools for monosolvents are required.

In conclusion, the experimental solubility is reported for anthracene in four monosolvents at several different temperatures, along with the solubility of anthracene in binary mixtures containing methanol, ethanol, 1-propanol, and 2-propanol with 2-propanone at 298.2 K. The measured solubility data can be used in the chemical or petrochemical industries. The applicability of previously developed models to predict the solubility of anthracene is shown. The careful review of the overall MPDs showed that eq 2, the developed QSPR, and Abraham models can be used for anthracene solubility prediction in studied alcohols at different temperatures and binary solvent mixtures of 2-propanone + alcohols. These computations can be used for solubility predictions of anthracene in the chemical industry.

Supporting Information Available:

Details of the QSPR equations used in this work along with the required input data are included in a Word file. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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