3896 J. Chem. Eng. Data **2010,** *55,* 3896–3900

Solubilities of Some Disperse Yellow Dyes in Supercritical CO_2

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The solubilities of color index disperse yellow 232 and 184 and their modified forms in supercritical carbon dioxide have been measured at different temperatures of (308, 318, 328, 338, and 348) K and in the pressure range of (12.1 to 35.5) MPa. Solubility data were correlated using four semiempirical models (Chrastil, Kumar−Johnston, Bartle, and Mendez-Santiago−Teja). A comparison among the four models revealed that these models showed good correlations of the solubility data with an overall average absolute relative deviation (AARD) of 0.155, 0.16, 0.184, and 0.157, respectively. Using the correlation results, the enthalpies of dye-CO_2 solvation and dye vaporization were separately approximated. Their values were found to be in the range of (12.5 to 19.4) and (70.6 to 102.7) $kJ\cdot mol^{-1}$.

Introduction

Supercritical fluid dyeing (SFD) technology has received attention in the textile industry due to increased environmental concerns, as reflected by a number of research communications dedicated in this field.[1−10]

This innovation method, as comparing with traditional dyeing process, is capable of substantially reducing waste discharge and energy consumption.[11−13]

Carbon dioxide is a benign, nonflammable, and inexpensive solvent and acts as a useful processing fluid for SFD at supercritical conditions. Supercritical carbon dioxide ($scCO_2$) can be separated easily from the dyeing medium to recycle perfectly by expansion. Moreover, the SFD process can save and reuse the additives and dyes more than the wet dyeing process. Basically, higher solubilities of dyestuffs in supercritical solvents are favorable to promote the efficiency of the dyeing process. The solubility of a solute in a supercritical fluid is the most important thermophysical property which needs to be determined and modeled as a first step to develop any supercritical fluid (SCF) technology. Experimental solid solubility data for different compounds in supercritical CO_2 have been reported by several authors.[14−16]

It is observed that more experimental data are still required for some chemicals and there is a need for generalized correlation models. Density-based approaches show their outstanding simplicity in correlating the solute solubility and only use readily available independent variables (pressure, temperature, and density of pure $scCO_2$). They do not require physical properties such as critical properties, acentric factors, and so forth. These equations possess three or more parameters which are being adjusted to the experimental data. On the other hand, thermodynamic models have the ability to describe and predict the solubility behavior of a solid in compressed gas solvents. For example, the Peng−Robinson equation of state model is a very successful engineering equation of state model for the supercritical solubility systems, but its application requires a number of the dye physical properties (e.g., critical temperature, critical pressure, acentric factor, and sublimation pressure) which are not experimentally available in the literature for a lot of compounds because of the measurement difficulty. Prior to using the model, these parameters thus are obtained through certain estimation methods. Subsequently, these approximated parameters have unsatisfactorily introduced ambiguous uncertainty into the model. Therefore, these thermodynamic models are rarely used for process design applications.[17]

Experimental Section

Materials. High-performance liquid chromatography (HPLC) grade methanol (Merck, Darmstadt, Germany) was used as received. Carbon dioxide with a minimum mass fraction purity of 0.9999 was purchased from Sabalan Co. (Tehran, Iran) and used for all of the extractions. Disperse yellow 232 [D_3, CAS No: 35773-43-4; IUPAC name: 3-(5-chloro-2-benzoxazolyl)-7-(diethylamino)-2*H*-1-benzopyran-2-one], modified disperse yellow 232 [D_1; IUPAC name: 5-diethylamino-2-[(5-choloro-2-hydroxy-phenylimino)-methyl]-phenol], disperse yellow 184 [D_4, CAS No: 164578-37-4; IUPAC name; 3-(5-methyl-2-benzoxazolyl)-7-(diethylamino)-2*H*-1-benzopyran-2-one], and modified disperse yellow 184 [D_2; IUPAC name: 5-diethylamino-2-[(2-hydroxy-5-methyl-phenylimino)-methyl]-phenol] were obtained from the Iran Color Research Center (Tehran, Iran) with an assessed minimum purity of 0.99 (mass fraction). Some basic information for these disperse dyes is given in Table 1.

Apparatus and Procedure. A Suprex (Pittsburgh, PA) MPS/225 integrated SFE/SFC system equipped with a modified static system for the solubility determination in the SFE mode was used. The schematic diagram of the experimental apparatus and a detailed description of the apparatus and operating procedures are available in the literature.[18] Solubility measurements were accomplished with a 1 mL extraction vessel in the pressure range from (12.1 to 35.5) MPa at temperatures of (308 to 348) K for a duration of 30 min. The solubilities were calculated by absorbance measurements at λ_{max} of each compound (Table 2) using a model Cecil Aquarius CE 7200 double-beam spectrophotometer (London, UK). The values are obtained from an arithmetic average of three replicate measurements with a

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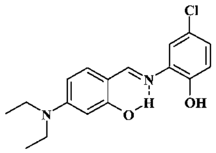
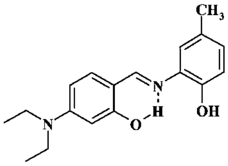
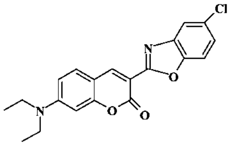
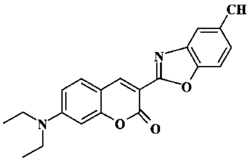
10.1021/je901049r

Table 1. Structure of the Dyes and Their Physicochemical Properties

Compound	Formula	Structure	M/g.mol^{-1}	$^{a}T_m$/K	λ_{max}/nm
D$_1$	$C_{17}H_{19}ClN_2O_2$		318.5	498-500	420
D$_2$	$C_{18}H_{22}N_2O_2$		298	492-494	427
D$_3$	$C_{20}H_{17}ClN_2O_3$		368.5	467-469	446
D$_4$	$C_{21}H_{20}N_2O_3$		348	481-483	450

[a] Melting temperature that was measured by a melting point measurement instrument from GallenKamp Company (U.K.).

relative deviation less than 7 %. The uncertainty of the dissolution temperature and pressure measurements was ± 0.1 K and ± 0.1 MPa, respectively. The densities of CO_2, calculated by the computer system of the Suprex MPS/225 at various pressures and temperatures, were used to determine the solubility of the mentioned dyes. It is noteworthy that the Surpex MPS/225 uses the modified equation of state extended to include high-pressure fluids as proposed by Pitzer et al.[19]

Data Correlation

The correlation of the experimental solubility data was investigated using different density-based models. Chrastil[20] derived an equation which relates the solubility of a solid solute in a SCF with the density of pure SCF and the absolute temperature. This model is expressed by a linear relationship between the logarithm of the solid solubility and the logarithm of the density of the pure CO_2.

$$\ln(S/\text{kg}\cdot\text{m}^{-3}) = a_0 + a_1 \ln(\rho/\text{kg}\cdot\text{m}^{-3}) + \frac{a_2}{T/\text{K}} \quad (1)$$

In this expression, S is the solubility of the solid in the supercritical phase; ρ is the density of the pure supercritical fluid; the parameter a_1 is the association number; a_2 is a constant, equivalent to $\Delta H/R$, where ΔH (kJ·mol^{-1}) is the sum of the enthalpies of vaporization and solvation of the solute and R (J·mol^{-1}·K^{-1}) is the gas constant; and a_0 is another constant related to the molecular weight of the solute and the solvent. The parameters a_0, a_1, and a_2 are determined by fitting to experimental data.

To confirm the reliability of measured solubility data, a semiempirical model was proposed first by Bartle et al.[21] given by

$$\ln \frac{y_2 P}{P_{\text{ref}}} = A + b_1(\rho - \rho_{\text{ref}}) \quad (2)$$

where

$$A = b_0 + \frac{b_2}{T} \quad (3)$$

and

$$\ln \frac{y_2 P}{P_{\text{ref}}} = b_0 + b_1(\rho - \rho_{\text{ref}}) + \frac{b_2}{T} \quad (4)$$

where y_2 is the equilibrium mole fraction of the solute in $scCO_2$, with a P_{ref} of 0.1 MPa and a reference density assumed as 700 kg·m^{-3}. In this model, the coefficients b_0, b_1, and b_2 can be obtained by fitting to experimental data. In the first step, from the experimental solubility data, each isotherm is fitted using eq 2, to obtain the values of A and b_1. The b_1 values are averaged, and these values are then used to recalculate the A values for the various isotherms. The A constants are then plotted against $1/T$ and correlated with eq 3, to determine constants b_0 and b_2. Finally, the values b_0, b_1, and b_2 are used to predict the solubility, by applying eq 4.

The parameter b_2 is related to the enthalpy of vaporization of the solid solute, $\Delta_{\text{vap}}H$, by the expression $\Delta_{\text{vap}}H = -Rb_2$. On the basis of the values of $\Delta_{\text{tot}}H$ and $\Delta_{\text{vap}}H$, the heat of solvation can be approximated for each solute$-CO_2$ system.

Table 2. Solubilities of Four Dyes in $scCO_2$ at Various Temperatures and Pressures

			D_1		D_2		D_3		D_4	
T / K	P / MPa	ρ^a / $kg \cdot m^{-3}$	$100\ S$ / $g \cdot L^{-1}$	$10^6\ y_2$	$100\ S$ / $g \cdot L^{-1}$	$10^6\ y_2$	$100\ S$ / $g \cdot L^{-1}$	$10^6\ y_2$	$100\ S$ / $g \cdot L^{-1}$	$10^6\ y_2$
308	12.1	771	0.13	0.23	0.57	1.09	0.30	0.47		
	15.2	818	0.52	0.87	0.89	1.61	0.52	0.75	0.01	0.02
	18.2	850	0.58	0.93	1.20	2.08	0.55	0.77	0.07	0.11
	21.3	876	0.71	1.12	1.42	2.40	0.71	0.96	0.69	0.99
	24.3	897	0.83	1.28	1.52	2.50	0.88	1.17	0.83	1.17
	27.3	916	0.91	1.37	1.85	2.99	0.97	1.27	1.05	1.44
	30.4	931	1.11	1.65	2.07	3.28	1.20	1.54	1.20	1.63
	33.4	946	1.27	1.85	2.17	3.39	1.48	1.86	1.41	1.88
	35.5	955	1.51	2.18	2.54	3.92	1.60	2.00	1.89	2.50
318										
	12.1	661	0.08	0.18	0.50	1.12	0.23	0.42		
	15.2	745	0.48	0.89	0.80	1.58	0.39	0.63		
	18.2	792	0.74	1.29	1.11	2.07	0.71	1.07	0.06	0.10
	21.3	826	0.86	1.44	1.48	2.64	0.82	1.18	0.93	1.42
	24.3	852	1.04	1.68	1.91	3.32	0.96	1.35	0.95	1.41
	27.3	875	1.11	1.75	2.32	3.91	1.21	1.66	1.27	1.84
	30.4	893	1.27	1.96	3.21	5.31	1.50	2.00	1.91	2.71
	33.4	910	1.42	2.15	3.94	6.40	1.80	2.36	2.70	3.76
	35.5	919	1.83	2.75	4.25	6.84	2.22	2.89	3.11	4.28
328										
	12.1	516	0.04	0.10	0.34	0.98				
	15.2	657	0.44	0.93	0.73	1.64	0.32	0.59		
	18.2	726	0.84	1.61	1.05	2.14	0.66	1.08		
	21.3	771	1.03	1.85	1.58	3.03	0.84	1.30	0.37	0.61
	24.3	804	1.28	2.19	2.10	3.85	1.03	1.53	1.07	1.68
	27.3	831	1.67	2.77	3.09	5.49	1.44	2.06	1.60	2.43
	30.4	853	1.94	3.15	4.13	7.14	1.80	2.52	2.07	3.06
	33.4	872	2.11	3.34	4.91	8.32	2.31	3.16	3.05	4.43
	35.5	884	2.48	3.88	6.21	10.37	2.92	3.95	3.76	5.38
338										
	12.1	396			0.30	1.12				
	15.2	561	0.39	0.96	0.80	2.10	0.16	0.34		
	18.2	654	0.74	1.56	1.16	2.61	0.57	1.03		
	21.3	712	1.15	2.23	2.49	5.15	1.02	1.71	0.26	0.47
	24.3	754	1.52	2.79	3.85	7.54	1.30	2.07	1.20	2.02
	27.3	786	1.86	3.27	5.90	11.08	1.89	2.87	1.79	2.88
	30.4	812	2.03	3.46	5.95	10.82	2.83	4.16	2.64	4.12
	33.4	834	2.61	4.33	7.77	13.75	3.46	4.95	4.52	6.85
	35.5	848	3.44	5.60	10.41	18.13	4.21	5.92	4.81	7.17
348										
	12.1	327			0.23	1.03				
	15.2	477	0.38	1.10	0.76	2.36	0.08	0.20		
	18.2	585	1.08	2.54	1.30	3.27	0.47	0.95		
	21.3	652	1.42	3.01	2.59	5.88	1.02	1.87	0.36	0.70
	24.3	702	1.81	3.56	3.90	8.20	1.44	2.44	1.33	2.40
	27.3	740	2.17	4.06	5.90	11.77	2.41	3.88	2.62	4.48
	30.4	772	2.51	4.49	6.57	12.57	3.17	4.91	3.20	5.24
	33.4	796	3.15	5.47	9.43	17.49	4.21	6.31	4.94	7.85
	35.5	811	4.03	6.86	11.30	20.57	5.23	7.70	5.83	9.09

[a] The densities of CO_2 were calculated by the computer system of the Suprex MPS/225 based on ref 19.

Kumar and Johnston[22] proposed the linear relationships between ln y_2 and ρ (denoted as the K-J model) as

$$\ln y_2 = c_0 + c_1\rho + \frac{c_2}{T} \tag{5}$$

Again the coefficients c_0, c_1, and c_2 are obtained by fitting to experimental data. Likewise, the parameter c_2 is related to $\Delta_{\text{tot}}H$ by assumption that $\Delta_{\text{tot}}H = -c_2R$.

Mendez-Santiago and Teja[23] model is based on the theory of dilute solutions and relates the solubility of solid, y_2 (in mole fraction) with the density of SCF ($kg \cdot m^{-3}$). The resulted expression (denoted as the M-T model) contains three adjustable parameters

$$T \ln(y_2P) = d_0 + d_1\rho + d_2T \tag{6}$$

where d_0 to d_2 are the model constants.

Results and Discussion

The experimental solubility data for compounds (in terms of equilibrium mole fraction, y_2, and equilibrium solubility, S ($g \cdot L^{-1}$), in $scCO_2$) are shown in Table 2. The possible intramolecular H-bonding between D_2 molecules weakens the solute−solute interaction, so it possesses the highest solubility. Regarding the same intramolecular H-bonding in D_1, the existence of chlorine substitution instead of a methyl group decreases its solubility in a nonpolar solvent of CO_2. The results presented in Table 2 (also in Figure 1) indicate that the solubility of the analytes in $scCO_2$ always increases as the pressure does; this is because the CO_2 density increases, so the solvent power does also. The parameters of a correlation equation were adjusted by the minimization of the average absolute relative deviation (AARD).

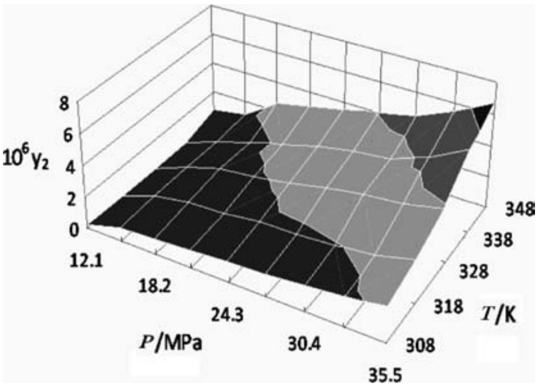


Figure 1. Surface plot of experimental solubility of D_1 in $scCO_2$ versus pressure and temperature.

Table 3. Different Parameters of the Dye + CO_2 Binary Systems Obtained by Using the Chrastil, Bartle, K-J, and M-T Models

Chrastil model	a_0	a_1	a_2/K	AARD
compound				
D_1	−37.1	4.36	−6135	0.113
D_2	−50.1	6.23	−7656	0.172
D_3	−56.5	6.42	−6441	0.098
D_4	−103	12.5	−10817	0.240
average				0.155
Bartle model	b_0	b_1/L·g^{-1}	b_2/K	AARD
compound				
D_1	17.4	0.0101	−8491	0.173
D_2	21.7	0.0119	−9770	0.194
D_3	17.2	0.0122	−8571	0.105
D_4	27.8	0.0182	−12353	0.264
average				0.184
K-J model	c_0	c_1/L·g^{-1}	c_2/K	AARD
compound				
D_1	1.073	0.00026	−6192	0.101
D_2	4.267	0.00036	−7603	0.167
D_3	−0.988	0.00034	−6035	0.129
D_4	7.250	0.00068	−10876	0.240
average				0.160
M-T model	d_0/K	d_1/K·L·g^{-1}	d_2	AARD
compound				
D_1	−10941	0.14	15.7	0.114
D_2	−12545	0.16	19.8	0.171
D_3	−12129	0.18	16.8	0.104
D_4	−18506	0.29	30	0.235
average				0.157

$$\mathrm{AARD} = \frac{1}{N}\sum \frac{|y_{i,\mathrm{Calc}} - y_{i,\mathrm{Obs}}|}{y_{i,\mathrm{Obs}}} \tag{7}$$

where N is the number of data points, $y_{i,\mathrm{Obs}}$ is the experimental solubility of the solid for experimental point i, and $y_{i,\mathrm{Calc}}$ is the calculated solubility corresponding to point i (in the Chrastil model the y parameter is replaced with S). The correlated results of solubilities of the compounds are tabulated in Table 3 using the four density-based models. As shown in these tables, the overall average AARD values are 0.155, 0.184, 0.16, and 0.157 for the Chrastil, the Bartle, the K-J, and the M-T models, respectively.

Table 4 lists the calculated enthalpies for the compounds by the different models in $scCO_2$ system. Since the vaporization is endothermic, whereas the solvation is exothermic, the vaporization heat is higher than the total reaction heat for each dye. The solute−solvent solvation heat is obtained from the difference between vaporization and total reaction heats. It is worthy to note that these thermodynamic functions are of interest for the dyeing process.

Table 4. Approximated Total Reaction, Solvation, and Vaporization Enthalpies for the Dyes

compound	$\Delta H_{\mathrm{total}}{}^{a}$ / kJ·mol^{-1}	$\Delta H_{\mathrm{total}}{}^{b}$ / kJ·mol^{-1}	$\Delta H_{\mathrm{vap}}{}^{c}$ / kJ·mol^{-1}	$\Delta H_{\mathrm{sol}}{}^{d}$ / kJ·mol^{-1}
D_1	51	51.5	70.6	19.3
D_2	63.6	63.2	81.2	17.8
D_3	53.5	50.1	71.2	19.4
D_4	89.9	90.4	102.7	12.5

[a] Obtained from the Chrastil model. [b] Obtained from the K-J model. [c] Obtained from the Bartle model. [d] Obtained from the difference between $\Delta H_{\mathrm{vap}}{}^{c}$ and $(\Delta H_{\mathrm{total}}{}^{a} + \Delta H_{\mathrm{total}}{}^{b})/2$.

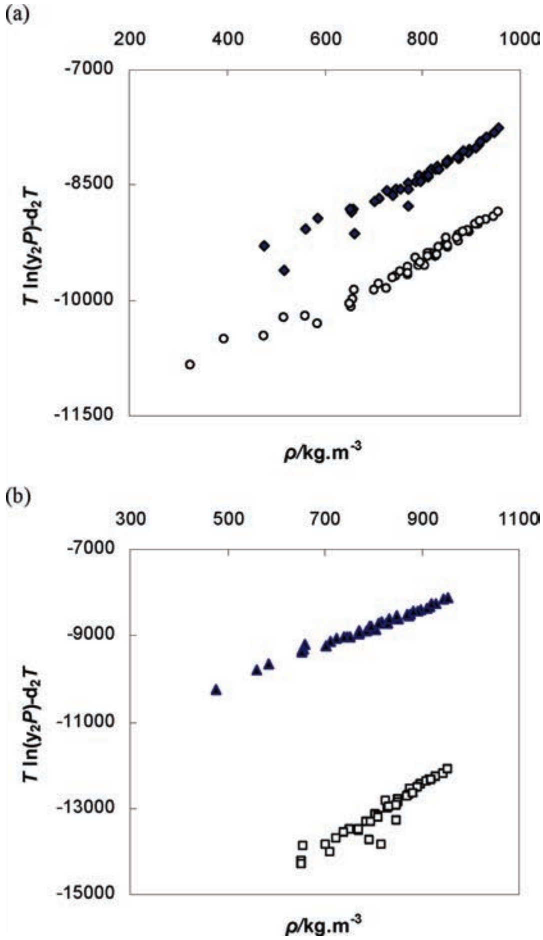


Figure 2. Plot of $T \ln(y_2P) - d_2T$ as a function of ρ (kg·m^{-3}) for (a) D_1 (♦, $R^2 = 0.9455$) and D_2 (○, $R^2 = 0.9642$) and (b) D_3 (▲, $R^2 = 0.9893$) and D_4 (□, $R^2 = 9044$) based on the M-T model.

For the M-T model, eq 6 may be arranged as follows

$$T \ln(y_2P) - d_2T = d_0 + d_1\rho \tag{8}$$

Mendez-Santiago and Teja claimed that eq 8 could be used to check the consistency of experimental data. The experimental solubility data are considered consistently good, if all isotherms collapse to a single straight line on a graph of the left-hand side of eq 8 versus the density of solvent. Also, as an illustrative example, Figure 2 shows that all of the experimental data of CO_2 + disperse dyes follow the predominant trend of linearity.

The correlation coefficients, R^2, and AARDs that were obtained based on M-T model for the four dyes were compared, and the results showed that the best correlation between experimental and predicted data was obtained for D_3 dye (Table 3).

Conclusion

The solubilities of four disperse yellow dyes in pure supercritical CO_2 were investigated at pressures ranging from (12.1 to 35.5) MPa and temperatures of (308 to 348) K. By using four density-based models (Chrastil, Bartle, K-J, and M-T), satisfactory correlation results for solid solubilities were obtained. On the basis of the Bartle model, the vaporization heats of solutes were located in the range of (70.59 to 102.7) $kJ \cdot mol^{-1}$. The solvation heats for various dye + CO_2 systems were also approximated, ranging from (12.52 to 19.4) $kJ \cdot mol^{-1}$.

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Received for review December 14, 2009. Accepted May 10, 2010.

JE901049R