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Solubility of Red 153 and Blue 1 in Supercritical Carbon Dioxide

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ABSTRACT: Solubilities of red 153, (3-[[4-[[5,6(or 6,7)-dichloro-2-benzothiazolyl]azo]phenyl]ethylamino]propanenitrile), an azo compound, and disperse blue 1 (1,4,5,8-tetraaminoantraquinone) in supercritical carbon dioxide (SC CO_2) were measured at $T = (333.2 \text{ to } 393.2)$ K over the pressure range (12.0 to 40.0) MPa by a flow type apparatus. The solubility of red 153 ($0.985 \cdot 10^{-6}$ to $37.2 \cdot 10^{-6}$) in the overall region of measurements is found to be significantly higher than that of disperse blue 1 ($1.12 \cdot 10^{-7}$ to $4.89 \cdot 10^{-7}$). The solubility behavior of disperse red 153 follows the general solubility trend displayed by disperse dyes with a crossover pressure at about 20 MPa. On the other hand, blue 1, which is a disperse anthraquinone dye, exhibits unexpected behavior not recorded previously—there is no crossover pressure at the temperature and pressure ranges studied, and the dye's solubility at $T = 333.2$ K practically does not increase with pressure. To the best of our knowledge, there are no previous measurements of blue 1 solubility in SC CO_2 reported in the literature. The experimental data were correlated by using the Soave−Redlich−Kwong equation of state (EoS) with the one-fluid van der Waals mixing rule, and an acceptable correlation of the solubility data for both dyes was obtained.

INTRODUCTION

Supercritical dyeing of textiles was initially developed for the dyeing of some synthetic polymers using mainly disperse dyes but has also been tested with natural fibers and different classes of dyes.[1] Dry dyeing processes involving supercritical carbon dioxide (SC CO_2) are considered green techniques and have received considerable attention due to increased environmental concerns.[2,3] The development and application of these processes require detailed knowledge of the phase equilibrium between dyes and the supercritical solvent, and a number of studies, discussing the dependence of dye solubility in SC CO_2, both on temperature and pressure, have been presented in the literature.

In particular, several authors have studied the solubility of disperse anthraquinones and azo dyes in SC CO_2 and have demonstrated that the solubility of the former are higher.[4−15] Both static and dynamic methods, with different improvements, have been applied to measure the dyes' solubility. The methods employed by the investigators to recover the solutes and the way the analyses of the composition were performed differ a great deal and can explain, to a certain extent, the errors and deviations of the solubility data reported in literature.

Our work is devoted to the experimental measurement and the thermodynamic modeling of the solubility of red 153 (3-[[4-[[5,6(or 6,7)-dichloro-2-benzothiazolyl]azo]phenyl]ethylamino]-propanenitrile), an azo compound, and blue 1 (1,4,5,8-tetraaminoantraquinone), an amino-anthraquinone-based dyestuff representative of disperse anthraquinone dyes, in SC CO_2. The solubility of red 153 has been studied previously[7] and solubility data at $T = (353.2 \text{ and } 393.2)$ K and pressures from (15 to 30) MPa were presented. Disperse blue 1 is used for coloring textiles and also as a component in hair dyes and other cosmetic products; still, to the best of our knowledge, there are no studies devoted to the measuring and correlating of its solubility in SC CO_2 available in the open literature.

We apply the Soave−Redlich−Kwong cubic equation of state (SRK EoS) with the one-fluid van der Waals mixing rule to correlate the solubility of the two dyes. The thermophysical properties of red 153 and of blue 1, which are required for the modeling, but not available, are estimated and reported.

EXPERIMENTAL SECTION

Chemicals. High purity CO_2 (99.995 % purity) was supplied by Air Liquide (Portugal). Blue 1 (> 0.97 mass fraction), was purchased from Fluka and disperse red 153 (> 0.94 mass fraction), was obtained from DyStar−Anilinas Têxteis (Unip. Lda, Portugal; Portuguese dye industry). The structures and the melting temperatures of the pure components are listed in Table 1.

Equipment and Procedure. The supercritical fluid experiments were performed in a flow-through apparatus. A detailed description of the equipment has been reported previously.[15,16] This equipment permits carrying studies at temperatures up to 393.2 K and pressures up to 60.0 MPa. The uncertainties of the temperature and pressure measurements were ± 1 K and ± 0.1 MPa, respectively. The uncertainty associated with volume measurements was ± 0.5 % in the total volume.

The equilibrium cell is charged with approximately (3.5 to 4) g of dye mixed with glass spheres of (0.71 to 1.18) mm in diameter, resulting in an overall bed height of about (15 to 18) cm. The conditions for the solubility studies were described in detail in a previous work;[16] hence a very concise description will be given here. The solubility of the dyes in supercritical carbon dioxide was measured over a wide range of contact times, from (640 to

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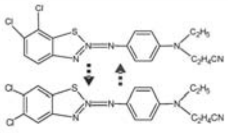
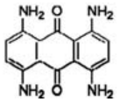
Table 1. Structure, Molar Mass, and Melting Temperature (According to the Supplier) for the Dyes

Name	Structure	Empirical Formula (Molar mass)	Melting Point
Red 153		$C_{18}H_{15}N_5SCl_2$ (404.32 g/mol)	440.2 K
Blue 1		$C_{14}H_{12}N_4O_2$ (268.27g/mol)	605.2 K

2300) s. The reason behind that was to ensure the saturation of the compound in the supercritical phase, as it had already been demonstrated that the equilibrium state was attained when the lengths of the contact time are longer than 900 s.[16]

The dyes were collected using an ice bath in a tube with three filters to make certain that the last filter was not colored. The three-way valve assures that at the end of the run the expansion valve, connecting lines, and the three filters in the tube were washed with an appropriated organic solvent (e.g., methanol) to a fixed volume to guarantee a complete recovery of the dye. The total amount of the dye collected was quantified by UV−visible equipment (Thermo Electron Corporation, model Nicolat evolution 300LC), using a calibration curve to each dye with standard solutions.

Three replicates were performed at each experimental condition, and the solubility obtained is the average of these results. The average uncertainty of the solubility measurements for red 153 isotherms was 9.5 % and for blue 1 was 4.6 %.

■ EXPERIMENTAL RESULTS

The solubility of red 153 and blue 1 in SC CO_2 was measured at T = (333.2 to 393.2) K over the pressure range (12.0 to 40.0) MPa, and the experimental data obtained are summarized in Table 2.

According to the experiments, the solubility of red 153 increases with pressure at a constant temperature owing to an increase in the fluid density, and a crossover of the respective isotherms, at around 20.0 MPa, is exhibited. A comparison of our data with those reported in a previous study[7] shows that the solubility values obtained by us are considerably higher (with a deviation of up to 130 %); the agreement is good only for the low pressure ranges. It should be noted, however, that other authors,[11] who have measured the solubility of several dyes in SC CO_2, ascertained similar considerable deviations for disperse blue 79 (110 % higher values), when they compared their solubility results with those of this particular study.[7] A detailed discussion and analysis of the possible reasons why such large deviations in the solubility values published in the literature by different authors are observed is presented by Huang et al.[12] Still we would like to give two additional possible sources for such a discrepancy, namely: (i) the purity of the solute (which the authors did not not refer to in their paper[7]); (ii) the recovery of the expanded mixture into the two test tubes with the organic solvent (it might have not been completed).

Table 2. Solubility, y_2, for Red 153, and Blue 1[a,b] in SC CO_2 (1) from T = (323.2 to 393.2) K

T	P	red 153	blue 1	ρ_1
K	MPa	$10^6 \cdot y_2$	$10^7 \cdot y_2$	$kg \cdot m^{-3}$
323.2	20.0	0.985 ± 0.12		784.29
	25.0	1.86 ± 0.2		834.19
	30.0	2.68 ± 0.4		870.43
	35.0	3.77 ± 0.1		899.23
	40.0	4.74 ± 0.5		923.32
333.2	20.0		1.12 ± 0.004	723.68
	22.5		1.07 ± 0.05	758.69
	25.0		1.02 ± 0.01	786.55
	30.0		1.19 ± 0.1	829.71
	35.0		1.40 ± 0.1	862.94
	40.0		1.45 ± 0.2	890.14
	15.0	0.084 ± 0.010		427.15
353.2	20.0	0.870 ± 0.08	1.07 ± 0.05	593.89
	22.5		1.46 ± 0.03	646.04
	25.0	2.45 ± 0.4	1.86 ± 0.1	686.22
	30.0	5.82 ± 0.4	2.53 ± 0.1	745.60
	35.0	8.37 ± 0.6	3.77 ± 0.04	788.97
	40.0	11.7 ± 0.6	4.89 ± 0.3	823.16
373.2	20.0		0.663 ± 0.01	480.53
	22.5		1.03 ± 0.05	539.89
	25.0		1.44 ± 0.1	588.45
	30.0		2.31 ± 0.1	661.87
	35.0		3.70 ± 0.2	715.26
	40.0		4.45 ± 0.3	756.67
393.2	20.0	0.602 ± 0.05		401.15
	25.0	3.29 ± 0.3		505.56
	30.0	10.2 ± 0.9		585.22
	35.0	22.5 ± 2.6		645.54
	40.0	37.2 ± 3.7		692.87

[a] Average values of mole fraction taken from triplicate runs. [b] ± Uncertainties refer to standard deviation.

On the other hand blue 1 exhibits unexpected solubility behavior which, to the best of our knowledge, has not been recorded previously for any other disperse anthraquinone dye. Namely: (i) there is no crossover pressure at the temperature and pressure ranges studied; (ii) solubility at higher temperatures, for example, T = (353.2 and 373.2) K, increases with pressure, but at a lower temperature, T = 333.2 K, the solubility values are practically constant and almost do not increase with pressure.

A possible reason for this unusual solubility behavior of blue 1 (but just one among many possible others) could be its melting temperature (605 K). If it is compared with the melting temperature of red 153 or with the temperatures of other disperse anthraquinone dyes (e.g., quinizarine, disperse red 9 and disperse blue 14, which vary in the interval (440 to 493) K), the solubility of which was reported earlier,[16] it is obvious that the melting temperature of blue 1 is unusually high for this class of compounds. A high melting temperature is usually an indicator of stability and strong intermolecular forces of a solid and in the case of blue 1 might influence the pattern of its solubility in SC CO_2.

Finally, the blue 1 solubility is about an order of magnitude lower than that of red 153.

THERMODYNAMIC MODELING

Thermodynamic Framework. To compute the solubility (mole fraction) of a solid solute in the fluid phase at a specified temperature, pressure, and overall composition, an appropriate thermodynamic model is required. Within the terms of an EoS framework, the standard formulation of this problem is based on the equi-fugacity condition for the solute; that is, denoting by the superscript "S" the solid solute and by the superscript "F" the fluid phase:

$$f^{\mathrm{S}}(T, P) = f^{\mathrm{F}}(T, P, \mathbf{y}, V) \tag{1}$$

where f^{S} is the fugacity of the solute in the pure solid phase, f^{F} is fugacity of the solute in the fluid-phase solution, $\mathbf{y} = (y_1, y_2, ..., y_{\mathrm{Nc}})^{\mathrm{T}}$ is the vector of fluid-phase mole fractions, and V is the molar volume of the fluid from the EoS model. Additional relationships that must be satisfied are the summation to one of the fluid-phase mole fractions.

The more common approach to the solution of the above problem is to take a popular EoS and use it directly in solid−fluid equilibrium calculations by introducing a solid-phase fugacity function defined in terms of a fluid-phase reference state. Two different approaches are popular. According to the first approach, originally proposed by McHugh et al.,[17] the solid vapor pressure is used as the reference fugacity of the solid, while the second implements the fugacity of a hypothetical subcooled liquid phase, as the reference of the solid phase fugacity, and in this study we use the latter approach.

Thus, (i) assuming an EoS model for the fluid phase and (ii) implementing the fugacity of a hypothetical subcooled liquid phase, as the reference of the solid phase fugacity, the solid-phase fugacity function, f^{S}, for a pure solute solid phase at temperature T and pressure P, defined in terms of a hypothetical liquid−phase fugacity as a reference state, disregarding the change in specific heat, is given as:[18]

$$f^{\mathrm{S}} = f^{\mathrm{SCL}}(P, T)\exp\left(\int_{P^{\mathrm{s}}_{\mathrm{subl}}}^{P} \frac{(V^{\mathrm{S}} - V^{\mathrm{SCL}})}{RT}\mathrm{d}P + \frac{\Delta H_{\mathrm{fus}}}{R}\left(\frac{1}{T_{\mathrm{m}}} - \frac{1}{T}\right)\right) \tag{2}$$

Assuming further that there are no solid−solid phase transitions and provided the solid specific volume at the subcooled liquid state V^{SCL} is only weakly dependent on pressure, eq 2 can be written as follows:

$$f^{\mathrm{S}} = f^{\mathrm{SCL}}(P, T)\exp\left(\frac{(V^{\mathrm{S}} - V^{\mathrm{SCL}})}{RT}(P - P^{\mathrm{S}}_{\mathrm{subl}}(T)) + \frac{\Delta H_{\mathrm{fus}}}{R}\left(\frac{1}{T_{\mathrm{m}}} - \frac{1}{T}\right)\right) \tag{3}$$

Often this approach should be preferred, particularly at higher pressures where the first part of the exponential term in eq 3, $\Delta V_{\mathrm{fus}} = V^{\mathrm{S}} - V^{\mathrm{SCL}}$, is important and should not be eliminated.

In eqs 2 and 3, f^{SCL} is the fugacity of the pure subcooled liquid, ΔH_{fus} is the enthalpy of melting, and $\Delta V_{\mathrm{fus}} = V^{\mathrm{S}} - V^{\mathrm{SCL}}$ is the change in volume, all taken for the solute at its triple point. The fugacity of the pure subcooled liquid is calculated from an EoS[18] (in our case the SRK EoS). The values of V^{SCL} are also evaluated by the EoS assuming that the volume is independent of pressure and temperature and equal to the value at the normal melting point.[18]

In our case of a binary system (SC CO_2 + dye) we denote the solid solute by subscript 2 and then its fugacity in the supercritical phase is:

$$f_2^{\mathrm{F}} = y_2 P \varphi_2^{\mathrm{F}} \tag{4}$$

where y_2 is the solubility (mole fraction) of the solute in the supercritical fluid and φ_2^{F} is its fugacity coefficient, which is calculated from the EoS.

Table 3. Red 153 and Blue 1 Pure Component Properties Estimated

	red 153	blue 1
p_{c}/MPa	1.97	2.158
T_{c}/K	977.15	1098.22
p/MPa (triple point)	$2.88 \cdot 10^{-4}$	$7.75 \cdot 10^{-4}$
$V^{\mathrm{S}}/\mathrm{m}^3 \cdot \mathrm{mol}^{-1}$	$2.87 \cdot 10^{-4}$	$1.96 \cdot 10^{-4}$
ω	0.950	0.925
$P_{\mathrm{s}}^{\mathrm{subl}}$/MPa at T = 323.2 K	$3.19 \cdot 10^{-10}$	
$P_{\mathrm{s}}^{\mathrm{subl}}$/MPa at T = 333.2 K		$6.36 \cdot 10^{-11}$
$P_{\mathrm{s}}^{\mathrm{subl}}$/MPa at T = 353.2 K	$2.57 \cdot 10^{-8}$	$5.39 \cdot 10^{-10}$
$P_{\mathrm{s}}^{\mathrm{subl}}$/MPa at T = 373.2 K		$3.343 \cdot 10^{-9}$
$P_{\mathrm{s}}^{\mathrm{subl}}$/MPa at T = 393.2 K	$2.58 \cdot 10^{-6}$	

Pure Component Property Estimation. To calculate the fugacity of a pure solid solute according to eq 3, the following data are required: its melting properties (ΔH_{fus}, melting temperature (T_{m}), ΔV_{fus}, sublimation pressure ($P_{\mathrm{s}}^{\mathrm{subl}}$), and its critical parameters (T_{c} and P_{c}). Generally, most of these properties are either not available experimentally or are even hypothetical and hence have to be predicted applying different estimation and group-contribution methods.

For the two dyes discussed in our study the experimental data presented in the literature are extremely scarce. The only property of red 153 known is its melting temperature, which was measured by a differential scanning calorimeter (DSC).[7] For blue 1 the melting temperature[19] and its sublimation pressure at 298.15 K[20] are reported. The rest of the properties required for correlating the dyes' solubility in the supercritical solvent are not available. To estimate them the algorithm discussed in detail by Fornari et al.[21] was followed.

Herewith we mark only the relevant main steps: The enthalpies of melting were estimated applying the method of Jain et al.[22] The sublimation pressures at the temperatures of interest were calculated from the Clapeyron equation, applying the data from the melting properties of the two dyes. The solid molar volumes were estimated applying the method of Bondi.[23] The critical parameters were estimated applying the methods suggested by Wakeham et al.[24] and Brauner et al.[25] Finally, the fugacity of the pure solutes in the subcooled liquid phase, the triple-point pressure, and V^{SCL} were all calculated from the EoS.[18]

The pure component properties of the two dyes used in our work are summarized in Table 3.

To the best of our knowledge, for both dyes, there are no estimations of the thermophysical properties required by the model for correlating their solubility available in the literature. Hence, we are not in the position to compare our properties' predictions with those of other authors. Still, it should be noted that for blue 1 the sublimation pressure values estimated by us are in the correct magnitude range, taking into consideration the change of $P_{\mathrm{s}}^{\mathrm{subl}}$ with temperature, as anticipated

Table 4. Binary Interaction Parameters (k_{ij}) and AARD for the Systems Red 153 + CO_2 and Blue 1 + CO_2 at the Temperatures of the Experiment

T	red 153 + CO_2		blue 1 + CO_2	
K	k_{ij}	AARD %	k_{ij}	AARD %
323.2	0.222	1.5		
333.2			0.176	5.8
353.2	0.238	10.3	0.188	6.0
373.2			0.196	5.3
393.2	0.253	4.2		

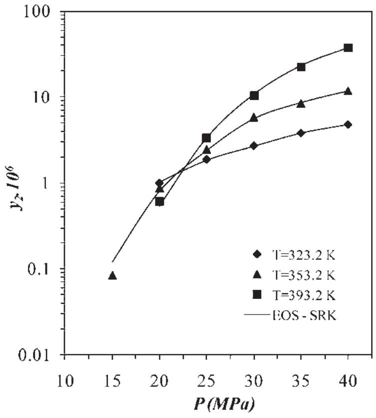


Figure 1. Solubility of red 153 in SC CO_2. Symbols represent the experimental solubility; lines represent the solubility, correlated by the SRK EoS. ♦, *T* = 323.2 K; ▲, *T* = 313.2 K; ■, *T* = 323.2 K.

from the experimental value reported at *T* = 298.2 K, namely, $P_s^{\text{subl}} = 2.4 \cdot 10^{-12}$ MPa.[20]

The thermodynamic model applied was the SRK EoS with the one-fluid van der Waals mixing rule. The binary interaction parameters k_{ij} for the two solids with CO_2 were estimated for each temperature of the experiment from the best fit to the data measured in this work by a standard optimization procedure and are shown in Table 4. The interaction parameters thus obtained represent just one of the possible sets, as a local rather than a global method was employed in the optimization.

The average deviations (AARD) between the solubility values measured experimentally, y_i^{exp}, and those correlated by the model, y_i^{calc}, are calculated according to the following:

$$\text{AARD} = \left(\frac{100}{N}\right)\left(\sum_{j=1}^{N} \frac{|y_i^{\text{exp}} - y_i^{\text{calc}}|}{y_i^{\text{exp}}}\right) \qquad (5)$$

where *N* is the number of data points. The values obtained are presented in Table 4.

The agreement between the experimentally measured and correlated solubilities in SC CO_2 for red 153 (Figure 1) and blue 1 (Figure 2) is acceptable, particularly after the crossover pressure for red 153, where the experimental conditions are far from the CO_2 critical point, and where the cubic EoS performs better.

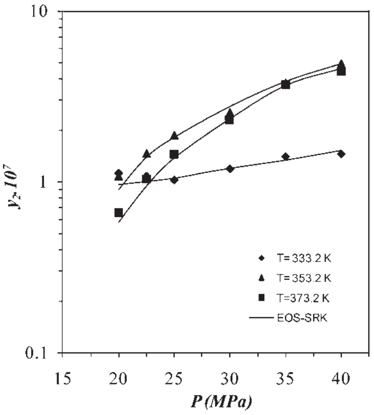


Figure 2. Solubility of blue 1 in SC CO_2. Symbols represent the experimental solubility; lines represent the solubility, correlated by the SRK EoS. ♦, *T* = 333.2 K; ▲, *T* = 353.2 K; ■, *T* = 373.2 K.

■ CONCLUSIONS

In this work, the solid solubilities in SC CO_2 of red 153 and blue 1 are measured in a consistent way in the temperature range from (333.2 to 393.2) K and at pressures from (12.0 to 40.0) MPa. The solubility behavior of disperse red 153 follows the general solubility trend displayed by disperse dyes with a crossover pressure at about 20 MPa. Blue 1, on the other hand, exhibits an unexpected behavior for a disperse anthraquinone dye not recorded previously—namely, there is no crossover pressure at the temperature and pressure ranges studied, and the dye's solubility at *T* = 333.2 K practically does not increase with pressure.

The SRK EoS with the one-fluid van der Waals mixing rule was shown to be able to provide a good correlation of the solubility of the two dyes in SC CO_2. The thermophysical parameters of the pure solid solutes, required by the modeling, for which there were no data available, were estimated and reported.

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