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Diffusion Coefficients of Methyl Orange in Dense Carbon Dioxide with the Micelle-Forming Surfactant Dehypon Ls-54

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The diffusion coefficient of methyl orange in dense carbon dioxide (CO_2) has been measured from (87.6 to 226) bar and from (20 to 40) °C. A hydrodynamic model and the hard sphere diffusivity model have been fit to the measured data. The effect of the micelle-forming, nonionic, nonfluorous surfactant Dehypon Ls-54 was also investigated. Effective diffusivity of methyl orange in the $CO_2 + \text{Ls-54} + H_2O$ system was measured at 35 °C and 220 bar at Ls-54 concentrations up to 0.165 M. A simple volumetric-based model based on the binary diffusivity data fits the effective diffusivity well, given realistic property estimates.

Introduction

Solvatochromic dyes have recently been used in fundamental supercritical carbon dioxide (CO_2) research, particularly in systems using micelle-forming surfactants that create micelles with hydrophilic cores.[1,2] Hydrophilic micelle cores act as polar microdomains within the CO_2 phase; the presence of such micelles can greatly increase the ability of CO_2 to solubilize polar compounds such as proteins. Methyl orange is a solvatochromic dye that has seen extensive use in examining the phase equilibria of CO_2 micelle systems.[3−6] Solvatochromic dyes such as methyl orange can be used to indicate the presence of polar micelle cores in organic solvents by measuring the shift in the maximum UV absorbance wavelength. The magnitude of this shift is proportional to the polarity of the dye's local environment. In this capacity, methyl orange has also been used to estimate the chemical nature of these microenvironments.[3−6]

Most research to date has focused on the phase equilibria of CO_2 micelle systems, and studies on the mass transfer characteristics of these systems are few. Predictions of mass transfer behavior are rooted in measurements of the diffusion coefficient; without accurate experimental values and predictive correlations, serious error can result when modeling these systems. Using the Taylor−Aris dispersion technique, basic diffusivity measurements for methyl orange in pure CO_2 have been obtained. This study also investigates the effective diffusivity of methyl orange in the CO_2 + Dehypon Ls-54 micelle system. The methyl orange + CO_2 diffusivity measurements are fit to a simple hydrodynamic model and the hard sphere tracer diffusion model. The effective diffusivities in the CO_2 + Dehypon Ls-54 micelle system can be successfully correlated using a simple volume fraction-based model based on the methyl orange diffusivity in the bulk CO_2, the micelle diffusivity, and other standard micelle parameters.

Experimental Section

Sodium *p*-dimethylaminoazobenzenesulfonate, commonly identified as methyl orange, was used as supplied (Aldrich 234109, certified, 91%, CASRN 547580). Dehypon Ls-54 (Lot# 519120, >99.5%, CASRN 68439-51-0) was supplied by Cognis Corporation of Hoboken, NJ. Supercritical grade carbon dioxide (AGA, 99.9995%) was used for the mobile phase. Deionized water was prepared using an ion-exchange cartridge (Barnstead, D0809).

The surfactant chosen for this study, Dehypon Ls-54 (CASRN 68439-51-0), is an ethoxylated alcohol with the structure[4] $H-(CH_2)_{12}-(OCH_2CH_2)_5-O-(CHCH_3CH_2O)_4-H$. This nonfluorous surfactant closely resembles other ethoxylated alcohol surfactants with the structure $H-(CH_2)_i-(OCH_2CH_2)_j-H$, that are commonly referred to as C_iE_j. Like other ethoxylated alcohol surfactants, Ls-54 is readily available in quantity, unlike some other classes of CO_2-soluble surfactants reported in the literature. Ls-54 has been shown to solubilize methyl orange into micelles in CO_2 at 35 °C and (200 to 220) bar.[4] The micelle radii have been determined through SAXS to be in the range (20.4 to 25.2) Å for corrected water to surfactant molar ratios, W_0^C, of 4 to 8.[4] The corrected water-to-surfactant ratio subtracts out water that is solubilized into the bulk CO_2.

For experiments with Ls-54 + H_2O modified CO_2, known water-to-surfactant ratio solutions were prepared by adding the proper weight of deionized water to a known weight of surfactant; weights were determined using a precision balance (Sartorius, E5500S, ±0.005 g). All water-to-surfactant ratios used in this study provided single-phase solutions. The proper mass of the water + surfactant solution is then withdrawn using a syringe and added to a stainless steel loading tube (see Figure 1). The loading tube is switched inline with the CO_2 supply to the high-pressure pump. The liquid CO_2 feed to the pump displaces the surfactant solution from the vertical loading tube directly into the syringe pump. The pump is brought to the desired pressure at the pump feed temperature (0 °C, Fisher Scientific, Isotemp 9000, ±0.1 °C) by manually opening the feed valve only when more CO_2 feed is needed. The syringe

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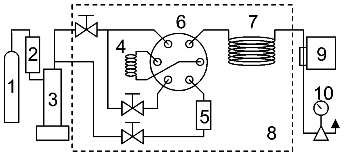

Figure 1. Schematic of the Taylor−Aris dispersion apparatus: 1, liquid CO_2 supply; 2, surfactant loading tube; 3, syringe pump; 4, sample loop; 5, sample tube; 6, sample injection valve; 7, dispersion tube; 8, thermostated bath; 9, UV detector; 10, pressure regulator.

pump is manually agitated and left undisturbed for at least 6 h to allow adequate time for micelle formation. After the period of equilibration, the resulting solution is pumped through the sample column and equilibrated again for at least 2 h at the pressure and temperature of the experiment. The resulting saturated micelle solution is flushed through the sample loop, at which time the experiment is begun.

Apparatus

The Taylor−Aris dispersion apparatus is shown in Figure 1. A high-pressure syringe pump (Isco, μLC-500) delivers liquid CO_2 at a constant flow rate (±0.5%). The entire apparatus between the pump and detector is submerged in a constant temperature glycol bath (Lindberg, WB1130, ±0.05 K). The CO_2 flow is split to either the sampling valve or the sample tube (316 stainless steel, 14 cm × 0.9 cm) containing methyl orange powder. The methyl orange powder is packed loosely in the tube with glass wool on each end and a 2-μm fritted disk on the exit. The mobile phase flows to a six-port high-pressure valve (Valco, AC6W); the flow path is designed so either pure CO_2 directly enters the sample valve or the CO_2 is diverted through the sample tube to become saturated with methyl orange before entering the sample valve. A 20 μL sample loop is attached to the sampling valve. The sample loop is in line with the sample tube when the valve is in “load” mode, allowing CO_2 saturated with methyl orange to be loaded into the loop. The moving injection technique is used for sample injection; the valve is actuated for a short duration (<1 s) to inject a very square pulse of methyl orange saturated CO_2 into the mobile phase.[7] Using helium (supplied at 5.5 bar) as the actuating gas for the sample valve significantly decreases response time and allows for smooth, repeatable sample injections at small time frames.

From the sample valve, the mobile phase flows through a deactivated, 0.2 mm i.d. fused silica capillary column 12.3 m in length (Supelco, custom capillary). Then the end of the column passes though a UV detector (Linear, UVIS 200, ±1.0 nm, <1% linearity) modified to use a capillary column. Wavelengths for the experiments ranged from (390 to 450) nm, as the adsorption maximum of methyl orange changed with increasing water concentrations. Zero dead-volume fittings are used to eliminate peak broadening due to fitting effects. The flow within the dispersion column was maintained such that $DeSc^{1/2} < 10$ to eliminate flow effects due to the coiling of the column.[7]

System pressure is maintained by a dome pressure regulator after the detector. The regulator is controlled using nitrogen gas (National Welders, Industrial Grade) and a hand-operated syringe pump (High-Pressure Equipment, 62-6-10). System pressure is monitored with a digital pressure transducer (Paroscientific Digiquartz, model 740, ±0.01%) positioned after the detector.

Methyl Orange + CO_2 Diffusivity

Mutual diffusion coefficients, *D*, are measured using the Taylor−Aris dispersion method. In this method, a mobile phase flows through a column of radius *R* and length *L* at a laminar velocity, *u*. The tracer solute is imposed at the column inlet as a pulse. The averaged concentration at the detector is given by[7]

$$C(t) = \frac{M_2}{2\pi R^2\sqrt{\pi K t}}\exp\left(\frac{-(L-ut)^2}{4Kt}\right) \qquad (1)$$

where M_2 is the amount of solute in the injected pulse in excess of that present in the same volume of the carrier gas. The dispersion coefficient, *K*, can be related to the molecular diffusivity, *D*, by[7]

$$K = \frac{R^2u^2}{48D} + D \qquad (2)$$

To obtain the diffusion coefficient, eq 1 is fit to the response curve, $C(t)$, including any corrections for baseline drift and offset; the molecular diffusion coefficient can be obtained directly from eq 2 using the fitted dispersion coefficient, *K*.

Since the conditions in this study are sufficiently removed from the critical point, the near-critical behavior of the diffusion coefficient is not significant.[7] The hydrodynamic approach is used to correlate the solute diffusion coefficient to the solvent viscosity:[7]

$$\frac{10^9 D}{T} = \alpha\mu_{CO_2}^{\beta} \qquad (3)$$

In eq 3, α and β are constants fitted for the system of interest. For this study, the constants were fit using diffusivity (*D*) in square meters per second, temperature (*T*) in kelvin, and CO_2 viscosity (μ_{CO_2}) in micropascal second.

The hard sphere tracer diffusivity model was also used to fit the data:[8]

$$\frac{D}{\sqrt{T}} = b(V^{2/3} - 1.271{V_0}^{2/3}) \qquad (4)$$

The constants *b* and V_0 were fit using diffusivity (*D*) in square meters per second, temperature (*T*) in kelvin, and CO_2 molar volume (*V*) in cubic meters per mole.

Effective Diffusivity in a Micelle System

In the presence of a micelle-forming surfactant, the Taylor dispersion technique measures an effective diffusivity of the solute. In a micelle system, the solute is partitioned between the bulk solvent phase and the micelle cores. To define an expression for the effective diffusivity, we consider the system as two-phase flow within the dispersion tube. Within the tube, an immiscible micelle core phase is evenly dispersed throughout a continuous solvent phase. Let *C* be the concentration of the solute in the continuous solvent phase and *M* be the solute concentration in the micelle core phase. Likewise, D_{AB} is the diffusivity of the solute in the bulk solvent, and D_M is the diffusivity of the micelles. With this terminology, we can define an average concentration, *U*, on the basis of the volume fraction of the micelle core phase, φ:

$$U = (1-\varphi)C + \varphi M \qquad (5)$$

A differential mass balance on the fluid in the tube gives

$$\frac{\partial U}{\partial t} = (1-\varphi)D_{AB}\frac{\partial^2 C}{\partial x^2} + \varphi D_M\frac{\partial^2 M}{\partial x^2} \qquad (6)$$

where x is the axial distance down the tube and t is the time. In the derivation of eq 6, the diffusion coefficients are assumed constant. Assuming the transfer of solute between the micelle cores and the bulk solvent is very fast, C and M can be taken to be in equilibrium. This equilibrium is described using a partition ratio, Π:

$$M = \Pi C \tag{7}$$

Using eq 7, M and C can be cast as functions of U and eq 6 can be transformed into

$$\frac{\partial U}{\partial t} = \frac{(1-\varphi)D_{\mathrm{AB}} + \varphi\Pi D_{\mathrm{M}}}{(1-\varphi)+\varphi\Pi}\frac{\partial^2 U}{\partial x^2} \tag{8}$$

To arrive at this equation, we have assumed the micelle core volume fraction, φ, and the diffusion coefficients, D_{AB} and D_{M}, are constant. From eq 8, the effective diffusivity, D_{E}, at the average concentration, U, is given by

$$D_{\mathrm{E}} = \frac{(1-\varphi)D_{\mathrm{AB}} + \varphi\Pi D_{\mathrm{M}}}{(1-\varphi)+\varphi\Pi} \tag{9}$$

Because of the large size of the micelles relative to the solvent molecules, the Stokes–Einstein equation can be used to predict the micelle diffusivity with good accuracy:[9]

$$D_{\mathrm{M}} = \frac{\kappa T}{6\pi R_{\mathrm{M}}\mu_{\mathrm{CO_2}}} \tag{10}$$

In eq 10, κ is Boltzmann's constant, T is the system temperature (on an absolute scale), $\mu_{\mathrm{CO_2}}$ is the viscosity of CO_2, and R_{M} is the radius of the micelle.

The micelle core volume fraction, φ, can be computed from the surfactant concentration:

$$\varphi = (C_{\mathrm{S}} - C_{\mathrm{cmc}})\frac{1}{n_{\mathrm{S}}}\left(\frac{4}{3}\pi R_{\mathrm{M}}^2\right)N_{\mathrm{A}} \tag{11}$$

Here, C_{S} is the total surfactant concentration, C_{cmc} is the critical micelle concentration, n_{S} is the number of surfactant molecules per micelle, R_{M} is the micelle radius, and N_{A} is Avogadro's number.

Equations 9 and 11 are used to calculate the effective diffusivity, so there are three parameters that are unknown for this system: the partition ratio, Π, the critical micelle concentration, C_{cmc}, and the number of surfactant molecules per micelle, n_{S}. To model the effective diffusivity, Π and C_{cmc} are estimated from literature data and n_{S} is fit to the experimental data.

The estimate of the partition ratio is based on the solubility of methyl orange in each phase. The solubility of methyl orange in the micelle cores is difficult to determine experimentally. It is common to approximate the environment inside the micelle cores as a bulk aqueous phase. The micelle cores in this system are water with solubilized CO_2 and methyl orange. The solubility of methyl orange in water is a function of pH.[10] When water is in contact with CO_2 at the conditions of interest, it becomes acidic with a pH of about 2.8.[11] Since the Ls-54 surfactant is not ionic, we assume that the surfactant headgroups will not significantly alter the pH of the cores and the methyl orange solubility is taken to be 3.1×10^{-5} M

The solubility of methyl orange in CO_2 is unknown. For this study, we use the solubility of *p*-dimethylaminoazobenzene in CO_2 as an estimate for that of methyl orange. The solubility of *p*-dimethylaminoazobenzene in CO_2 at 203 bar is 3.45×10^{-7} M.[12] Using these approximate values, Π is calculated to be 90. Because methyl orange is the sodium salt, the actual solubility is expected to be lower than that of *p*-dimethylaminoazobenzene The value of Π is not sensitive to as much as a 25% lower solubility.

Table 1. Experimental Conditions and Measured Diffusion Coefficients, *D*, for Methyl Orange in SCF Grade CO_2

T (°C)	P (bar)	10^9D ($m^2 \cdot s$)	T (°C)	P (bar)	10^9D ($m^2 \cdot s$)
40	214	7.8	20	146	6.8
40	214	7.8	20	146	6.9
40	214	7.7	20	146	6.7
40	214	7.8	20	146	6.7
40	163	9.0	20	146	6.5
40	163	8.4	20	117	7.1
40	163	9.3	20	117	7.2
40	163	8.8	20	117	6.9
40	163	8.2	20	117	6.8
40	115	11.2	20	117	7.0
40	115	11.6	20	96	7.2
40	115	10.5	20	96	7.6
40	115	11.1	20	96	7.3
40	103	12.5	20	96	7.4
40	103	13.0	20	96	7.3
40	103	12.2	20	88	8.6
40	97	13.1	20	88	9.1
40	97	12.6	20	88	8.8
40	97	12.4	20	205	6.2
40	97	12.3	20	205	6.6
40	97	12.2	20	205	6.3
20	227	6.1	20	205	6.9
20	227	5.8	20	205	6.4
20	227	6.3	20	88	6.4
20	227	6.2	20	88	6.5
20	182	6.3	35	222	7.0
20	182	6.5	35	222	7.3
20	182	6.7	35	222	7.3
20	182	6.6	35	222	7.1
20	182	6.4	35	222	7.4

The C_{cmc} for large nonionic surfactants is usually very low. Surfactants the size of Ls-54 typically have a C_{cmc} below 2×10^{-4} M.[13,14] This represents less than 0.2% of the surfactant concentration range used in these experiments (0 to 0.163 M). Due to the small size of the C_{cmc} relative to the concentrations used, it is assumed zero in our calculations. Using $C_{\mathrm{cmc}} = 0$ and $\Pi = 90$, the volume fraction of the micelle cores, φ, can be computed from eq 11.

Results

Diffusion coefficients for methyl orange in CO_2 are listed in Table 1. The reproducibility for any experimental condition varies less than 7% from the average. For Taylor dispersion measurements, 5% accuracy is considered feasible when the solvent is an incompressible liquid.[15] The somewhat higher variability of measurements in CO_2 is due to the more compressible nature of the fluid under the conditions investigated in this study. For comparison of the experimental data, methyl orange has a reported diffusivity[16] of 1.2×10^{-9} $m^2 \cdot s^{-1}$ in water at 40 °C while the measured diffusivity in CO_2 at 40 °C and 214 bar (854 $kg \cdot m^{-3}$) is 7.8×10^{-9} $m^2 \cdot s^{-1}$.

The hydrodynamic and hard sphere model fits to methylene orange diffusivity are shown in Figures 2 and 3, respectively. The average absolute relative deviation (AARD) for the hydrodynamic model fit is 2.6% using the fitted constants $\alpha = 0.385$ and $\beta = -0.620$. The hard sphere tracer diffusivity model fit gives an AARD of 6.8% with $V_0 = 1.50 \times 10^{-5}$ $m^3 \cdot mol^{-1}$ and $b = 7.30 \times 10^{-7}$ $mol^{2/3} \cdot s^{-1} \cdot K^{-1/2}$.

The effective diffusivities of methyl orange in the CO_2 + Ls-54 system are listed in Table 2 and shown in Figure 4. The data were taken at 35 °C, 220 bar, and $W_0{}^{\mathrm{C}} = 4.0$.

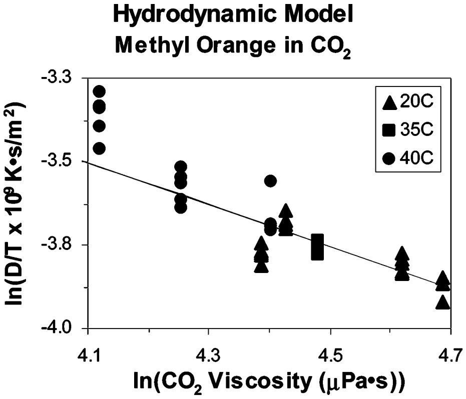

Figure 2. Hydrodynamic model fit of ln(D/T) to ln(η) for the methyl orange + CO_2 diffusion coefficients, where η is the viscosity of CO_2: ▲, 20 °C; ■, 35 °C; ●, 40 °C.

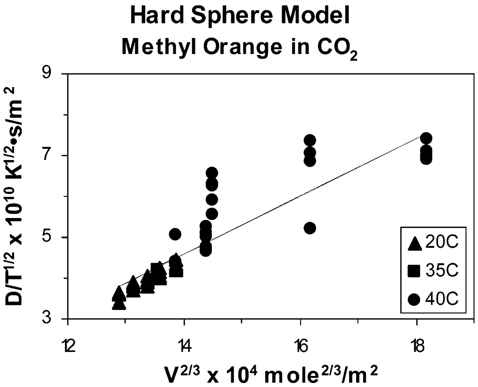

Figure 3. Hard sphere model fit of D/T to $V^{2/3}$ for the methyl orange + CO_2 diffusion coefficients, where V is the molar volume of CO_2: ▲, 20 °C; ■, 35 °C; ●, 40 °C.

Table 2. Measured Effective Diffusivities, D_E, of Methyl Orange as a Function of Dehypon Ls-54 Surfactant Concentration at 35 °C, 220 bar, and Corrected Water-to-Surfactant Ratio W_0^C = 4.0

[Ls-54] (M)	10^9D ($m^2 \cdot s$)	[Ls-54] (M)	10^9D ($m^2 \cdot s$)
0	7.0	0.05	5.1
0	7.3	0.05	5.0
0	7.3	0.05	5.0
0	7.1	0.05	5.1
0	7.4	0.05	5.1
0.05	5.4	0.05	5.2
0.05	5.2	0.03	5.3
0.05	4.7	0.03	5.8
0.05	5.0	0.03	5.8
0.05	5.2	0.03	5.6
0.05	5.0	0.16	3.4
0.05	5.0	0.16	3.5
0.05	5.2	0.16	3.1
0.05	5.2		

At these conditions, it is reported that Ls-54 forms spherical micelles with a radius of 20.4 Å.[4] From the Stokes–Einstein equation (eq 10) the micelle diffusivity is calculated to be 1.02×10^{-9} $m^2 \cdot s^{-1}$. The diffusivity of methyl orange in CO_2 in the absence of surfactant was determined experimentally to be 7.23×10^{-9} $m^2 \cdot s^{-1}$ at 35 °C and 220 bar. These values along with the micelle core volume fraction are used to fit eq 9 to the effective diffusivity data by adjusting n_S. The best fit was achieved using $n_S = 189$ and is shown in Figure 4.

An n_S of 189 is a reasonable quantity, as ethoxylated surfactants of similar size ($C_{12}E_4$) can have aggregation numbers of 500 or more.[17] Knowing the micelle radius and using an n_S of 189, the area per surfactant headgroup is calculated to be about 40 Å^2; this is well within the normal range for nonionic surfactants, which is about (30 to 50) Å^2.[17]

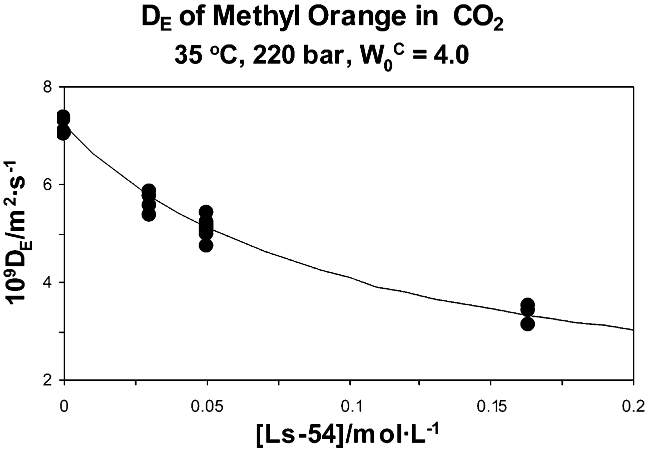

Figure 4. Effective diffusion coefficients for methyl orange in CO_2 + Ls-54 + H_2O at 35 °C, 220 bar, and corrected water-to-surfactant ratio $W_0^C = 4$.

Conclusions

The diffusion coefficients for methyl orange in dense CO_2 have been measured over the ranges (87.6 to 226) bar and (20 to 40) °C. The measured diffusivities are used to fit a simple hydrodynamic model and the hard sphere tracer correlation; the models fit the data with AARDs of 2.6% and 6.8%, respectively. The effective diffusivity of methyl orange in the CO_2 + Ls-54 + H_2O system was measured at 220 bar and 35 °C for surfactant concentrations of (0 to 0.164) M. A simple volume-based model can accurately predict the effective diffusivity in the micelle system from the binary diffusivity data and realistic property estimates.

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