J. Chem. Eng. Data **2009,** *54,* 2881–2886

Solubility of Carbon Dioxide in Water and Hexadecane: Experimental Measurement and Thermodynamic Modeling

C. Eduardo Pereira Siqueira Campos,*,† H. Gomes D'Amato Villardi,‡ F. Luiz Pellegrini Pessoa,‡ and A. Maria Cohen Uller†

PEQ/COPPE/UFRJ, Cidade Universitária, CT, Bl.G, Sala 115, CEP 21949-900, RJ, Brasil, and DEQ/EQ/UFRJ, Cidade Universitária, CT, Bl.E, Sala 207, Gabinete K, CEP 21945-970, RJ, Brasil

Direct contact between oil, gases, and water often occurs in petroleum reservoirs and in many refining operations, where the phase behavior study is fundamental to design and optimization. Accurate experimental data and thermodynamic models are required for a satisfactory prediction of phase equilibrium. The objective of this work is to obtain experimental solubility data for the system of carbon dioxide + hexadecane since this organic compound has properties similar to the average properties of Brazilian heavy oil. The carbon dioxide + water system was used to validate the experimental apparatus and to obtain additional data to complete literature reports. The operational conditions used were a temperature range from (303.2 to 323.2) K and a low-pressure range from (51.7 to 535.4) kPa. Moreover, the results were correlated using a simple thermodynamic model to describe the behavior of both systems, demonstrating satisfactory bubble point calculation measurements.

Introduction

In recent years, the petroleum industry has presented a significant growth. The characterization of all possible present phases becomes essential to design and to optimize the processes. A satisfactory representation of the phase behavior requires complete information about the phase equilibrium, to which experimental data and thermodynamic models are fundamental.

Direct contact between oil, gas, and water is often observed in a petroleum reservoir and in many unit process operations, where the solubility of gases in liquids has an important role in the description of the phase behavior of these systems. The most common gases are carbon dioxide and methane. To represent the oil, hexadecane was used since the Brazilian petroleum can be characterized as heavy oil with average properties similar to those of hexadecane.

In the literature, there are many experimental data on the gas solubility in liquids. For the carbon dioxide + water system, there is a satisfactory amount of available data but mostly at elevated pressures and temperatures, as shown in Table 1. However, for the carbon dioxide + hexadecane system, there are fewer data sets available, as can be observed in Table 2.

Thus, due to the importance of gas solubilities in liquids for correct characterization of the phase behavior in the petroleum industry systems and the limited quantity of experimental data at low pressures of particular systems, the objective of this work is to build an experimental apparatus to determine gas solubilities in liquids, applied to the carbon dioxide + hexadecane system. The carbon dioxide + water system was used to validate the equipment and to obtain some experimental data at low pressures to complete the literature reports. Furthermore, a simple and predictive theoretical model is presented to calculate the phase equilibrium.

Table 1. Literature Data for the Carbon Dioxide + Water System in Different Experimental Ranges

reference	*T*/K	*P*/MPa
Dalmolin et al.[1]	288.0 to 323.0	0.092 to 0.473
Bermejo et al.[2]	296.7 to 369.7	1.55 to 8.34
Someya et al.[3]	282.0 to 293.0	7 to 12
Valtz et al.[4]	278.2 to 318.2	0.465 to 7.963
Chapoy et al.[5]	273.2 to 373.2	0.190 to 9.333
Kiepe et al.[6]	313.2 to 393.2	0.01 to 9.26
Servio and Englezos[7]	274.0 to 283.2	2 to 6
Bamberger et al.[8]	323.2 to 353.1	4.05 to 14.11
Teng et al.[9]	278.0 to 293.0	6.44 to 29.49
Coan and King[10]	298.2 to 373.2	1.7 to 5.2
Stewart and Munjal[11]	273.2 to 298.2	1.0 to 4.5
Dodds et al.[12]	273.2 to 393.2	up to 71
Wiebe and Gaddy[13]	323.2 and 373.2	2 to 71

Table 2. Literature Data for the Carbon Dioxide + Hexadecane System in Different Experimental Ranges

reference	*T*/K	*P*/MPa
Nieuwoudt and Rand[14]	313.2 to 323.2	8.5 to 16.8
Rincón and Trejo[15]	308.2	8.296 to 12.418
Breman et al.[16]	305.7 to 512.3	1.24 to 3.29
Tanaka et al.[17]	313.2	1.726 to 6.473
Spee and Schneider[18]	294.4 to 413.3	up to 25
Steen et al.[19]	283.2 to 306.1	4.44 to 7.55
Schwarz and Prausnitz[20]	295.8 to 297.4	0.43 to 1.67
Charoensombut-Amon et al.[21]	308.2 to 343.2	0.690 to 25.810

Experimental Section

Materials. Carbon dioxide was supplied by Linde Gas Ltd. and hexadecane from Vetec Química Fina Ltd., both with 99.9 % purity, and water was once distilled.

Methods. The constructed equipment was similar to the one proposed by Oliveira and Uller[22] shown in Scheme 1 as a schematic flowchart. The equipment was submerged in a thermostatic bath[1] with digital temperature control. The equi-

---

* Corresponding author. Phone: +55 21 2562-7315 (R.205). Fax: +55 21 2562-8300. E-mail address: carlos@peq.coppe.ufrj.br.

† PEQ/COPPE/UFRJ.

‡ DEQ/EQ/UFRJ.

10.1021/je800966f CCC: $40.75 © 2009 American Chemical Society
Published on Web 06/04/2009

Scheme 1. Schematic: 1, Thermostatic Bath; 2, Equilibrium Cell; 3, Quick Connection; 4, Pressure Transducer; 5, Gas Reservoir; 6, Gas Cylinder; 7, Vacuum Pump Protection; 8, Vacuum Pump; 9 to 12, Two-Way Valve; 13 and 14, Three-Way Valve

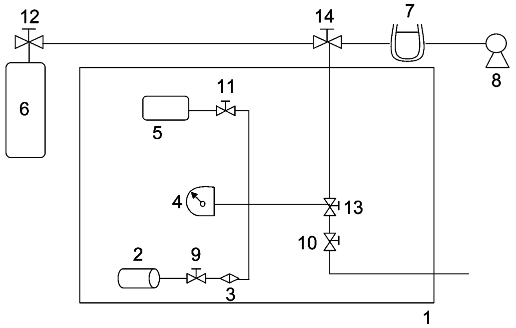


Table 3. Carbon Dioxide (1) + Water (2) System Experimental Data: Apparatus Validation

T/K = 298.2		T/K = 313.2		T/K = 323.2	
P/kPa	10^4x_1	P/kPa	10^4x_1	P/kPa	10^4x_1
59.6	3.4	96.7	3.9	107.0	3.4
118.2	7.3	96.8	3.9	199.1	7.1
159.1	8.8	97.8	3.8	228.1	8.1
175.9	10.8	100.5	4.0	228.3	8.2
180.3	11.4	224.5	10.0	229.0	8.3
252.0	16.3	263.4	11.9	246.4	8.0
271.6	17.6	264.5	11.8	248.4	7.8
279.7	18.1	279.2	11.3	249.2	9.1
280.1	18.1			249.6	9.1
380.5	25.0			252.4	8.8
500.5	33.2			357.0	11.4
				357.8	11.7
				357.9	11.5

librium cell[2] was made of stainless steel, with a total internal volume of approximately 30 cm^3 and an external magnetic stirrer. The valve[9] allowed feeding gas into the cell and prevented any solvent diffusion through the lines, while a Swagelok quick connection[3] was used to remove the mobile phase. An absolute digital pressure transducer[4] (Smar, model LD301) was used to measure system pressure. All lines were 0.635 cm OD stainless steel tubing.

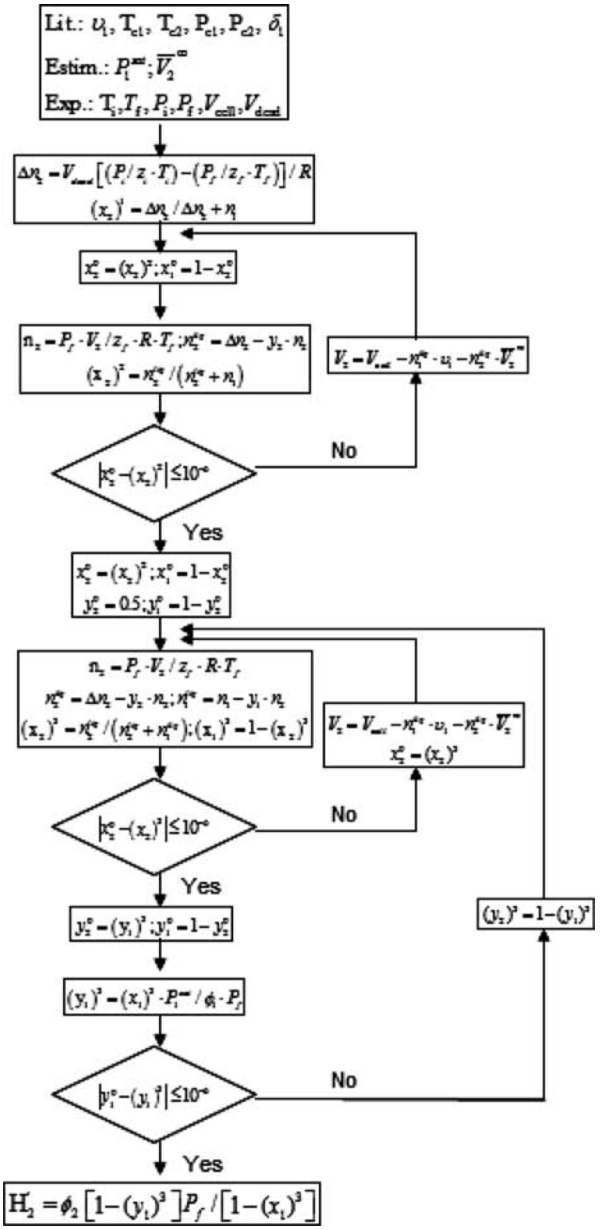
To store the gas in the equipment section inside the bath, a reservoir cell[5] was used with a total capacity of approximately 100 cm^3, connected by a valve.[11] A carbon dioxide cylinder[6] and a vacuum pump[8] with a protection[7] were connected to the system via a three-way valve.[14]

Experimental Procedure. As the first experimental step, a sample of liquid was weighed inside the cell using an analytical digital balance. Once the sample was frozen, the cell was connected to the system and was degassed three times. After degassing, the system was completely evacuated; carbon dioxide was filled into the reservoir cell; and the bath temperature was adjusted to the set point.

The initial pressure was first set to 0.133 MPa by feeding the gas into the lines, and the valve[9] was periodically opened to prevent any solvent diffusion until the pressure was stabilized (final pressure). Thus, at the end of each experimental point, a pressure drop was observed, and discounting the dead volume, one can obtain the real pressure drop referring to gas solubility in the liquid, evaluating its mole fraction.

To measure, consecutively, additional five equilibrium pressures using only one liquid sample, other higher initial pressures were set to (0.2000, 0.2700, 0.4000, 0.5330, and 0.667) MPa. Each experiment was conducted three times to obtain repeatability of the proposed data.

Scheme 2. Flow Chart for Data Reduction of Gas Solubility[a]



[a] Subscript 1 denotes the liquid and 2 denotes the gas; superscript 1 denotes solubility first approximation, 2 denotes the second, and 3 denotes the complete calculation; T_c and P_c are critical properties; υ is the molar volume; δ_1 is the solubility parameter; P^{sat} is the saturation pressure; $\bar{V}^{\infty}$ is the partial molar volume at infinite dilution calculated as presented by Handa et al.;[29] T_i, T_f, P_i, and P_f are measured experimental variables; V_{cell} is the equilibrium cell volume; V_{dead} is the dead volume of the entire system; H' is the apparent Henry constant; z is the compressibility factor; and ϕ is the fugacity coefficient calculated through a cubic equation of state.

The estimated accuracy for each measurement was 0.1 K for the temperature, 0.04 % for the pressure, and 0.1 mg for the weighed mass. The uncertainty for the mass determination was 0.05 mg and for the mole fraction of each substance was $5 \cdot 10^{-5}$.

Results and Discussion

Experimental Apparatus Validation. All necessary data input and the calculation sequence for each experimental data reduction are presented in Scheme 2.

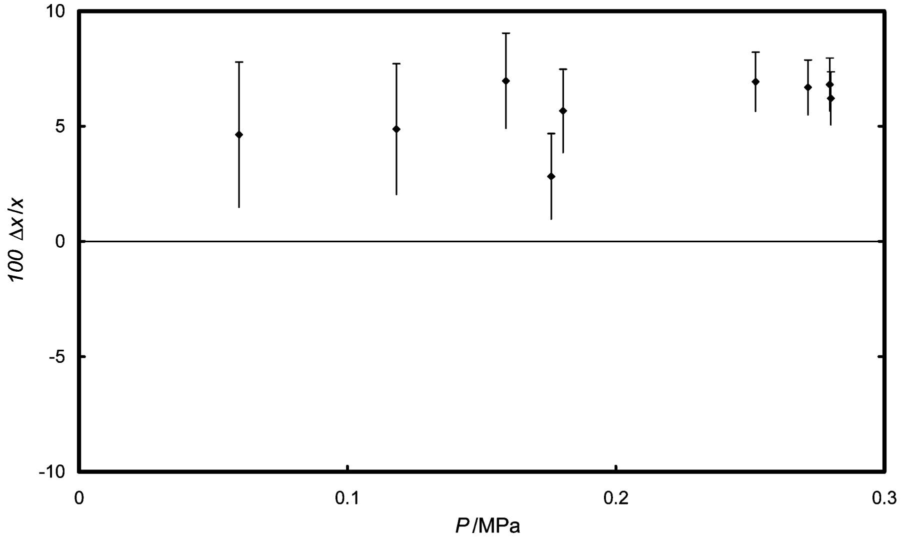


Figure 1. Fractional deviations $\Delta x = x(\text{this work}) - x(\text{literature})$ of CO_2 (1) solubility in H_2O (2) at $T = 298.2$ K obtained from ◆, Dalmolin et al.[1]

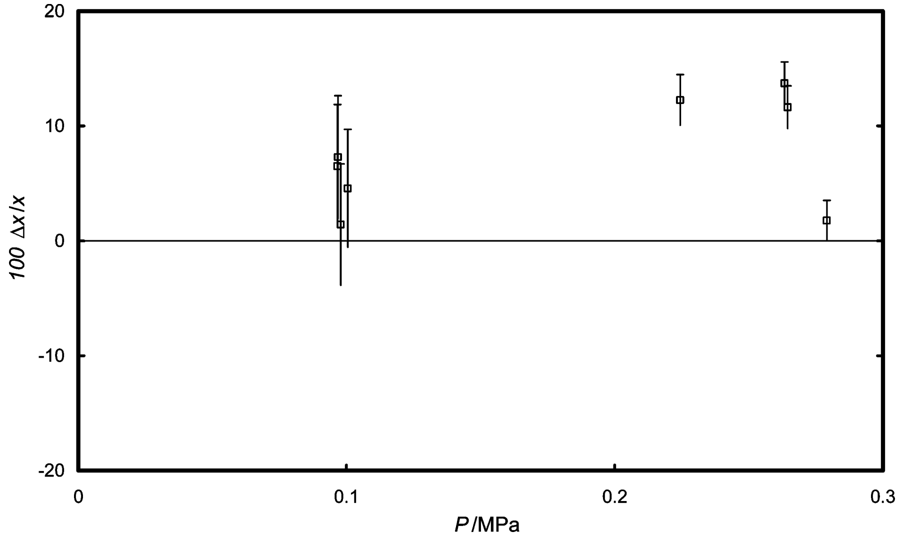


Figure 2. Fractional deviations $\Delta x = x(\text{this work}) - x(\text{literature})$ of CO_2 (1) solubility in H_2O (2) at $T = 313.2$ K obtained from □, Kiepe et al.[6]

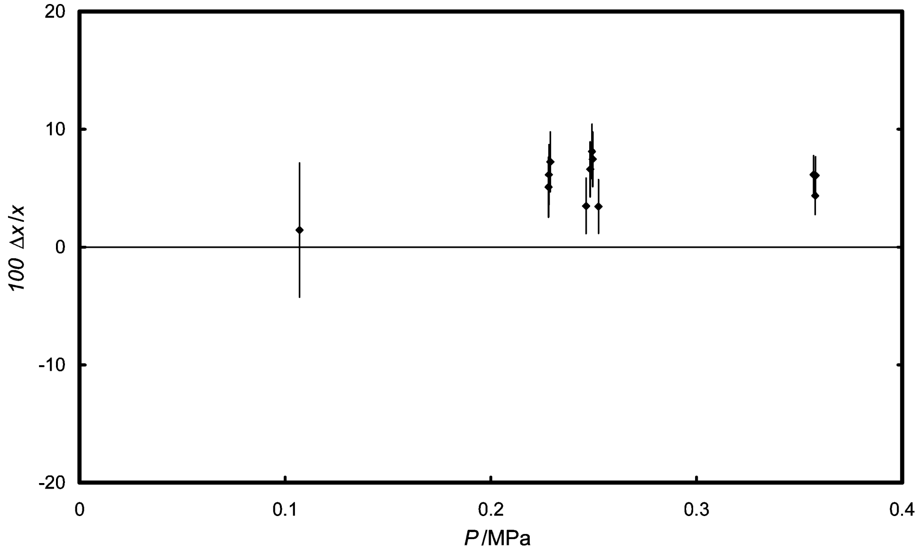


Figure 3. Fractional deviations $\Delta x = x(\text{this work}) - x(\text{literature})$ of CO_2 (1) solubility in H_2O (2) at $T = 323.2$ K obtained from ◆, Dalmolin et al.[1]

To validate the equipment, some experimental data at low pressures reported in the literature were reproduced for the carbon dioxide + water system. The experimental data presented by Dalmolin et al.[1] and Kiepe et al.[6] were used,

Table 4. Carbon Dioxide (1) + Water (2) System Solubility Data

T/K = 303.2		T/K = 313.2		T/K = 323.2	
P/kPa	$10^4 x_1$	P/kPa	$10^4 x_1$	P/kPa	$10^4 x_1$
63.8	3.1 ± 0.2	68.3	2.5 ± 0.1	74.7	2.0 ± 0.1
124.0	6.7 ± 0.3	129.0	5.4 ± 0.1	135.8	4.5 ± 0.1
187.5	10.4 ± 0.4	192.6	8.4 ± 0.1	199.4	7.1 ± 0.2
280.7	16.1 ± 0.5	288.0	13.1 ± 0.2	296.8	11.1 ± 0.3
391.8	22.9 ± 0.7	401.2	18.6 ± 0.3	411.7	15.8 ± 0.4
511.0	30.7 ± 1.6	524.3	24.6 ± 0.4	535.4	21.0 ± 0.6

Table 5. Carbon Dioxide (1) + Hexadecane (2) System Solubility Data

T/K = 303.2		T/K = 313.2		T/K = 323.2	
P/kPa	$10^3 x_1$	P/kPa	$10^3 x_1$	P/kPa	$10^3 x_1$
51.7	6.6 ± 0.8	55.0	4.9 ± 1.0	58.6	3.9 ± 0.8
106.5	15.7 ± 1.0	110.2	12.6 ± 2.0	115.0	10.9 ± 1.4
165.4	25.9 ± 1.2	169.6	21.3 ± 3.6	175.0	18.9 ± 2.6
252.1	41.7 ± 1.2	257.0	35.3 ± 5.8	263.7	31.9 ± 4.0
355.7	62.2 ± 1.2	361.2	53.5 ± 7.6	369.9	48.7 ± 5.6
470.8	86.7 ± 1.0	476.3	75.6 ± 9.0	486.4	69.2 ± 7.2

Table 6. Apparent Henry's Constant[23] for the Carbon Dioxide + Water System

T/K = 303.2		T/K = 313.2		T/K = 323.2	
P/kPa	k'_H/MPa	P/kPa	k'_H/MPa	P/kPa	k'_H/MPa
63.8	192.0	68.3	246.3	74.7	304.9
124.0	178.2	129.0	224.4	135.8	271.1
187.5	173.8	192.6	217.1	199.4	260.6
280.7	169.0	288.0	211.7	296.8	253.5
391.8	165.5	401.2	208.1	411.7	248.5
511.0	160.5	524.3	205.2	535.4	243.3

Table 7. Apparent Henry's Constant[23] for the Carbon Dioxide + Hexadecane System

T/K = 303.2		T/K = 313.2		T/K = 323.2	
P/kPa	k'_H/MPa	P/kPa	k'_H/MPa	P/kPa	k'_H/MPa
51.7	7.8	55.0	11.4	58.6	15.1
106.5	6.8	110.2	8.8	115.0	10.6
165.4	6.3	169.6	7.9	175.0	9.2
252.1	6.0	257.0	7.2	263.7	8.2
355.7	5.6	361.2	6.7	369.9	7.5
470.8	5.3	476.3	6.1	486.4	6.9

Table 8. Henry's Law Constants

systems	$CO_2 + H_2O$	$CO_2 + C_{16}H_{34}$
T/K	k_H/MPa	k_H/MPa
303.2	193.0	7.9
313.2	248.9	11.6
323.2	309.7	15.0

Table 9. Pure Component Properties[24]

	substances		
properties	CO_2	H_2O	$C_{16}H_{34}$
molar mass/g·gmol^{-1}	44.01	18.01	226.45
critical pressure/MPa	7.4	22.1	1.4
critical temperature/K	304.1	647.1	723.0
acentric factor	0.225	0.344	0.718

but it is important to emphasize that other mentioned works at the same experimental conditions were not ignored and are present in this analysis, especially in the figures.

Table 3 shows the experimental data obtained in the present work for the carbon dioxide + water system at 298.2 K, 313.2 K, and 323.2 K. Figures 1, 2, and 3 illustrate the fractional deviations (Δx) of the solubility (x) of carbon dioxide in water to demonstrate its satisfactory qualitative behavior. The mean deviations achieved were 5.7 %, 7.4 %, and 5.5 % for temperatures of 298.2 K, 313.2 K, and 323.2 K, respectively.

Table 10. Antoine Parameters[24]

substance	A	B	C
water	5.1156	1687.537	230.170
hexadecane	4.1536	1830.510	154.450

Table 11. UNIFAC Group Interactions Parameters[25]

n	m	A_{nm}/K	A_{mn}/K	B_{nm}	B_{mn}
CO_2	H_2O	601.10	271.80	−2.9100	2.7500
CO_2	CH_2	110.60	116.70	0.5003	−0.9106

Table 12. Area and Volume Parameters[24]

substance	R	Q
CO_2	1.2960	1.2610
$C_{16}H_{34}$	11.2438	9.2560
H_2O	0.9200	1.4000

Table 13. Thermodynamic Modeling Results[a]

$CO_2 + H_2O$				$CO_2 + C_{16}$			
T/K	P/kPa	P^{CALC}/kPa	100 MD	T/K	P/kPa	P^{CALC}/kPa	100 MD
303.2	64.7	61.2	6.5	303.2	52.4	45.8	4.5
303.2	125.7	127.2		303.2	107.9	101.8	
303.2	189.9	196.5		303.2	167.6	161.5	
303.2	284.4	301.1		303.2	255.4	249.3	
303.2	396.9	426.6		303.2	360.3	354.2	
303.2	517.8	570.2		303.2	477.0	470.9	
313.2	69.2	60.53	3.6	313.2	55.7	48.2	4.7
313.2	130.7	123.4		313.2	111.7	105.3	
313.2	195.1	189.6		313.2	171.8	165.6	
313.2	291.8	289.2		313.2	260.3	254.2	
313.2	406.5	407.6		313.2	366.0	359.9	
313.2	531.2	537.2		313.2	482.6	476.5	
323.2	75.7	63.2	5.3	323.2	59.4	49.6	5.2
323.2	137.5	125.3		323.2	116.5	109.6	
323.2	202.1	190.1		323.2	177.3	170.9	
323.2	300.7	288.9		323.2	267.2	261.0	
323.2	417.1	406.6		323.2	374.7	368.6	
323.2	542.4	537.0		323.2	492.8	486.6	
	100 GMD		5.2		100 GMD		4.8

[a] T, temperature; P, experimental pressure; P^{CALC}, calculated pressure; MD, mean deviation in each isotherm; GMD, global mean deviation.

Table 14. Parameters Obtained for Each System

system	$F1$	$F2$	$F3$
$CO_2 + H_2O$	9.6132	−6.4628	−1.3075
$CO_2 + C_{16}$	−24.3541	−34.2778	−31.0756

Analyzing the presented results, one can observe an adequate performance of the constructed apparatus in this work.

New Experimental Data. New gas solubility data in liquids were obtained for the carbon dioxide + water system to complete other reports, presented in Table 4, and for the carbon dioxide + hexadecane system, presented in Table 5.

From the results, it is possible to observe that the gas solubilities in both liquids decrease with increasing temperature. On the other hand, it is also detected that carbon dioxide solubilities in both liquids presented small values, demonstrating that any kind of loss or experimental uncertainty leads to deviations in the measured pressure drop. Thus, periodic tests to verify the equipment conditions are recommended during the experiments.

Moreover, apparent Henry constants, as shown by Ohgaki et al.,[23] were evaluated through eq 1, where k'_H represents this constant; y_{gas} is the vapor phase mole fraction of the gas; x_{gas} is the liquid phase mole fraction of the gas; ϕ_{gas} is the fugacity coefficient of the gas; and P is the system equilibrium pressure. Table 6 shows the calculated values of this thermodynamic property for the carbon dioxide + water system, and Table 7 for carbon dioxide + hexadecane. However, when the pressure

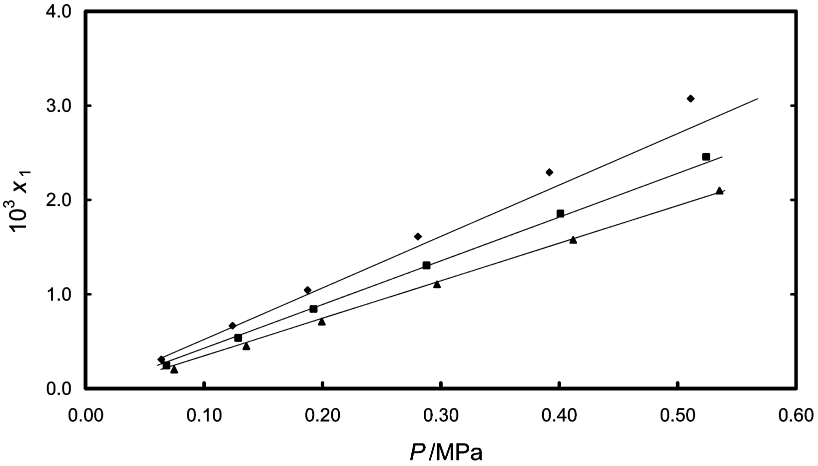


Figure 4. CO_2 (1) solubility in H_2O (2) as function of pressure: ♦, $T = 303.2$ K; ■, $T = 313.2$ K; ▲, $T = 323.2$ K. The solid lines (—) represent the calculated values.

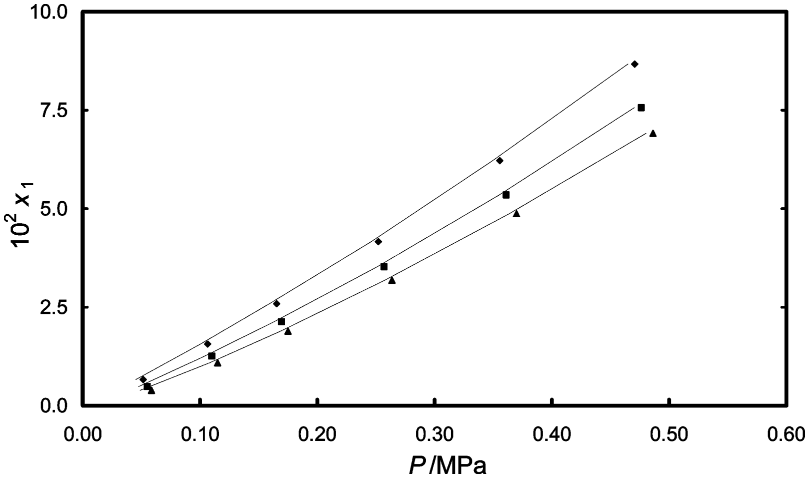


Figure 5. CO_2 (1) solubility in $C_{16}H_{34}$ (2) as function of pressure: ♦, $T = 303.2$ K; ■, $T = 313.2$ K; ▲, $T = 323.2$ K. The solid lines (—) represent the calculated values.

tends to zero, one can determine Henry's law constants, and these values are presented in Table 8.

$$k'_{\mathrm{H}} = \frac{y_{\mathrm{gas}}\phi_{\mathrm{gas}}P}{x_{\mathrm{gas}}} \qquad (1)$$

Thermodynamic Modeling. To calculate the vapor−liquid equilibrium (VLE), one starts from the isofugacity criteria. Considering the nonideality in the liquid phase, the fugacity for a substance i presented in the liquid phase can be described by eq 2, and the vapor phase, assuming ideal behavior, by eq 3.

$$f_{\mathrm{i}}^{\mathrm{L}} = x_{\mathrm{i}}\gamma_{\mathrm{i}}f_{\mathrm{i}}^{\mathrm{ref}} \qquad (2)$$

$$f_{\mathrm{i}}^{V} = y_{\mathrm{i}}P \qquad (3)$$

Using eqs 2 and 3, one obtains eq 4 to calculate the equilibrium total pressure of a binary system formed by carbon dioxide and a liquid.

$$P = x_{\mathrm{CO_2}}\gamma_{\mathrm{CO_2}}f_{\mathrm{CO_2}}^{\mathrm{ref}} + x_{\mathrm{liquid}}\gamma_{\mathrm{liquid}}f_{\mathrm{liquid}}^{\mathrm{ref}} \qquad (4)$$

Table 9 shows the properties of each substance presented in the studied systems, obtained from Reid et al.[24]

In general, several works in the literature use as the reference state for the substances in the liquid phase a pure liquid at the same pressure and temperature of the system, and its fugacity is calculated through the vapor pressure, as shown in eq 5. Table 10 presents the Antoine parameters[24] for each substance required in eq 5.

$$\log_{10}[P^{\mathrm{vap}}/\mathrm{bar}] = A - \frac{B}{T/\mathrm{K} + C - 273.15} \qquad (5)$$

The UNIFAC group contribution model proposed by Hansen et al.[25] was chosen to represent the nonideality of the liquid phase, and the group interaction parameters presented by Voutsas et al.[26] were applied in this work. Table 11 illustrates these parameters, and Table 12 shows the surface and area parameters obtained from Reid et al.[24]

For the gas, the strategy proposed by Prado et al.[27] and tested in many types of systems was used. The gas fugacity reference was obtained by estimating the parameters F1, F2, and F3 presented in a correlation similar to the one proposed by Prausnitz and Shair[28] as shown in eq 6, using the gas critical properties.

$$\ln\left(\frac{f_{\mathrm{CO_2}}^{\mathrm{ref}}}{P_{\mathrm{c}}}\right) = F1 - \frac{F2}{T_{\mathrm{r}}} - F3\ln T_{\mathrm{r}} \qquad (6)$$

The bubble point was calculated using the experimental data obtained in this work, and the deviations were evaluated using eq 7, where *P* represents the experimental pressure and P^{CALC}, the calculated pressure. Table 13 presents the results obtained for both systems, and Table 14 shows the estimated parameters. These parameters were estimated using all experimental points in each system, achieving a simple group that simultaneously represents all isotherms. The Simplex fitting method was applied in this optimization step, and the objective function is presented in eq 8, where NP represents the number of experimental points.

$$\mathrm{MD} = \left(\frac{|P - P^{\mathrm{CALC}}|}{P}\right) \tag{7}$$

$$F_{\mathrm{obj}} = \sum_{i=1}^{\mathrm{NP}} \left(\frac{P_{\mathrm{i}} - P_{\mathrm{i}}^{\mathrm{CALC}}}{P_{\mathrm{i}}}\right)^2 \tag{8}$$

Analyzing Table 13, one can observe not only that the experimental data studied were modeled with satisfactory results but also the applicability of the developed thermodynamic approach. Figures 4 and 5 illustrate gas solubility dependence on the temperature and equilibrium pressure for the carbon dioxide + water and carbon dioxide + hexadecane systems, respectively.

Conclusions

An equipment setup was built for determination of the gas solubility in liquids. Carbon dioxide solubilities in water and in hexadecane were measured in a temperature range from (303.2 to 323.2) K and pressures from (51.7 to 535.4) kPa. Apparent Henry's constants were calculated for each experimental determination. Furthermore, simple thermodynamic theoretical modeling was done based on the gamma (γ) − phi (ϕ) approach to evaluate bubble point pressure of each experimental datum. For the gas reference fugacity, a hypothetical state was considered, where its fugacity was calculated from an equation similar to the Prausnitz and Shair[28] equation, estimating its parameters. The pressure deviations achieved were around 5 % for both systems compared to experimental data.

Literature Cited

(1) Dalmolin, I.; Skovroinski, E.; Biasi, A.; Corazza, M. L.; Dariva, C.; Oliveira, J. V. Solubility of Carbon Dioxide in Binary and Ternary Mixtures with Ethanol and Water. *Fluid Phase Equilib.* **2006**, *245*, 193–200.

(2) Bermejo, M. D.; Martín, A.; Florusse, L. J.; Peters, C. J.; Cocero, M. J. The Influence of Na_2SO_4 on the CO_2 Solubility in Water at High Pressure. *Fluid Phase Equilib.* **2005**, *238*, 220–228.

(3) Someya, S.; Bando, S.; Chen, B.; Song, Y.; Nishio, M. Measurement of CO_2 Solubility in Pure Water and the Pressure Effect on it in the Presence of Clathrate Hydrate. *Int. J. Heat Mass Transfer* **2005**, *48*, 2053–2507.

(4) Valtz, A.; Chapoy, A.; Coquelet, C.; Paricaud, P.; Richon, D. Vapor-Liquid Equilibria in the Carbon Dioxide-Water System, Measurement and Modeling from 278.2 to 318.2 K. *Fluid Phase Equilib.* **2004**, *226*, 333–344.

(5) Chapoy, A.; Mohammadi, A. H.; Chareton, A.; Todihi, B.; Richon, D. Measurement and Modeling of Gas Solubility and Literature Review of the Properties for the Carbon Dioxide-Water System. *Ind. Eng. Chem. Res.* **2004**, *43*, 1794–1802.

(6) Kiepe, J.; Horstmann, S.; Fischer, K.; Gmehling, J. Experimental Determination and Prediction of Gas Solubility Data for $CO_2 + H_2O$ Mixtures Containing NaCl or KCl at Temperatures Between 313 and 393 K and Pressures up to 10 MPa. *Ind. Eng. Chem. Res.* **2002**, *41*, 4393–4398.

(7) Servio, P.; Englezos, P. Effect of Temperature and Pressure on the Solubility of Carbon Dioxide in Water in the Presence of Gas Hydrate. *Fluid Phase Equilib.* **2001**, *190*, 127–134.

(8) Bamberger, A.; Sieder, G.; Maurer, G. High-pressure (vapor + liquid) equilibrium in binary mixtures of (carbon dioxide + water or acetic acid) at temperatures from 313 to 353 K. *J. Supercrit. Fluids* **2000**, *17*, 97–110.

(9) Teng, H.; Yamasaki, A.; Chun, M. K.; Lee, H. Solubility of liquid CO2 in water at temperatures from 278K to 293K and pressures from 6.44 to 29.49 MPa and densities of the corresponding aqueous solutions. *J. Chem. Thermodyn.* **1997**, *29*, 1301–1310.

(10) Coan, C. R.; King, A. D., Jr. Solubility of Water in Compressed Carbon Dioxide, Nitrous Oxide, and Ethane. Evidence for Hydration of Carbon Dioxide and Nitrous Oxide in the Gas Phase. *J. Am. Chem. Soc.* **1971**, *93*, 1857–1862.

(11) Stewart, P. B.; Munjal, P. Solubility of Carbon Dioxide in Pure Water, Synthetic Sea Water, and Synthetic Sea Water Concentrates at−50 to 250 C. and 10- to 45-Atm. Pressure. *J. Chem. Eng. Data* **1970**, *15*, 67–71.

(12) Dodds, W. S.; Stutzman, L. F.; Sollami, B. J. Carbon Dioxide Solubility in Water. *Ind. Eng. Chem.* **1956**, *1*, 92–95.

(13) Wiebe, R.; Gaddy, V. L. The Solubility in Water of Carbon Dioxide at 50, 75 and 100°, at Pressures to 700 atm. *J. Am. Chem. Soc.* **1939**, *61*, 315–318.

(14) Nieuwoudt, I.; Rand, M. Measurement of Phase Equilibria of Supercritical Carbon Dioxide and Paraffins. *J. Supercrit. Fluids* **2002**, *22*, 185–199.

(15) Rincón, R. E.; Trejo, A. Solubility of n-octadecane in supercritical carbon dioxide at 310, 313, 333, and 353 K, in the range 10−20 MPa. *Fluid Phase Equilib.* **2001**, *185*, 231–239.

(16) Breman, B. B.; Beenackers, A. A. C. M.; Rietjens, E. W. J.; Stege, R. J. H. Gas-Liquid Solubilities of Carbon Monoxide, Carbon Dioxide, Hydrogen, Water, 1-Alcohols ($1 < n < 6$), and n-Paraffins ($2 < n < 6$) in Hexadecane, Octacosane, 1-Hexadecanol, Phenanthrene, and Tetraethylene Glycol at Pressures up to 5.5 MPa and Temperatures from 293 to 553 K. *J. Chem. Eng. Data* **1994**, *39*, 647–666.

(17) Tanaka, H.; Yamaki, Y.; Kato, M. Solubility of Carbon Dioxide in Pentadecane, Hexadecane, and Pentadecane + Hexadecane. *J. Chem. Eng. Data* **1993**, *38*, 386–388.

(18) Spee, M.; Schneider, G. M. Fluid phase equilibrium studies on binary and ternary mixtures of carbon dioxide with hexadecane, 1-dodecanol, 1,8-octanediol and dotriacontane at 393.2 K and at pressures up to 100 MPa. *Fluid Phase Equilib.* **1991**, *65*, 263–274.

(19) Steen, J.; Loos, Th. W.; Arons, J. S. The volumetric analysis and prediction of liquid-liquid-vapor equilibria in certain carbon dioxide + n-alkane systems. *Fluid Phase Equilib.* **1989**, *51*, 353–367.

(20) Schwarz, B. J.; Prausnitz, J. M. Solubilities of Methane, Ethane, and Carbon Dioxide in Heavy Fossil-Fuel Fractions. *Ind. Eng. Chem. Res.* **1987**, *26*, 2360–2366.

(21) Charoensombut-amon, T.; Martin, R. J.; Kobayashi, R. Application of a generalized multiproperty apparatus to measure phase equilibrium and vapor phase densities of supercritical carbon dioxide in n-hexadecane systems up to 26 MPa. *Fluid Phase Equilib.* **1986**, *31*, 89–104.

(22) Oliveira, J. V.; Uller, A. M. C. Solubility of 1,3-butadiene and methyl propene in n-methyl-2-pyrrolidone. *Fluid Phase Equilib.* **1989**, *46*, 267–280.

(23) Ohgaki, K.; Nishii, H.; Katayama, T. A Method for Gas-Solubility Measurement. *J. Chem. Eng. Jpn.* **1983**, *16*, 72–73.

(24) Reid, R. C.; Prausnitz, J. M.; Sherwood, T. K. *The Properties of Gases and Liquids*, 3rd ed.; McGraw Hill: New York, 1977.

(25) Hansen, H. K.; Coto, B.; Kuhlmann, B. *Phase Equilibria and Separation Process*; SEP-9212: Denmark, 1992.

(26) Voutsas, E. C.; Boukouvalas, C. J.; Kalospiros, N. S.; Tassios, D. P. The Performance of EoS/G^E Models in the Prediction of Vapor - Liquid Equilibria in Asymmetric Systems. *Fluid Phase Equilib.* **1996**, *116*, 480–487.

(27) Prado, M. P. Predictive model for carbon dioxide solubilities calculations in polymers. MSc. Thesis, COPPE/UFRJ: Brazil, 2004.

(28) Prausnitz, J. M.; Shair, F. H. Thermodynamic Correlation of Gas Solubilities. *AIChE J.* **1961**, *7*, 682–687.

(29) Handa, Y. P.; Benson, G. C. Partial molar volumes of gases dissolved in liquids. Part I. Selected literature data and some estimation techniques. *Fluid Phase Equilib.* **1982**, *8*, 161–180.

Received for review December 15, 2008. Accepted May 19, 2009. The authors would like to thank CNPq for the financial support.

JE800966F