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Gas Solubility in Binary Liquid Mixtures: Carbon Dioxide in Cyclohexane + Cyclohexanone

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ABSTRACT: Experimental results are presented for the pressure above liquid mixtures of carbon dioxide and cyclohexane + cyclohexanone. The experiments were performed at a preset temperature and liquid phase composition by means of a precise high-pressure view-cell technique based on the synthetic method. The studied temperatures are: (313, 353, and 392) K. The maximum pressure was about 5.5 MPa. The gas free mole fractions of cyclohexanone are: 0, 0.3, 0.6, 0.8, and 1. From the data, Henry's law constants of carbon dioxide in the studied liquid mixtures and the pure components are calculated.

INTRODUCTION

The oxidation of cyclohexane to cyclohexanol and cyclohexanone is an important industrial reaction and a key step in the nylon production chain. Usually the reaction is carried out by contacting air with liquid cyclohexane at high temperature and pressure.[1] Because of the formation of side products, the process must be carried out at low conversion to allow for acceptable selectivity. Therefore, alternative routes are being studied. One option could be using novel octahedral molecular sieves for the heterogeneously catalyzed selective oxidation of cyclohexane. Additionally, supercritical carbon dioxide expanded liquids could be used to expand the liquid reaction mixture to enhance the mobility of the reactants and products. For a rational planning of catalytic experiments and process design, especially at elevated pressures, reliable thermodynamic data are needed.

While gas solubility or equilibrium data for the binary mixtures that are of interest in the process described above are available, equilibrium data for the relevant ternary system, namely, carbon dioxide + cyclohexane + cyclohexanone, are rare. For the binary system carbon dioxide + cyclohexane, for example, Gironi and Lavecchia,[2] Patyi et al.,[3] Anderson et al.,[4] Wilhelm and Pattino,[5] Kaminishi et al.,[6] Al-Sahhaf et al.,[7] Chang,[8] Nagarajan and Robinson,[9] and Dymond[10] measured gas solubilities at (280 to 423) K. For carbon dioxide + cyclohexanone, for example, Gallardo et al.,[11] Mellendo et al.,[12] Laugier and Richon,[13] and Chang and Chen[14] measured temperatures between (280 and 453) K. For the ternary system carbon dioxide + cyclohexane + cyclohexanone, Esmelindro et al.[15] measured equilibrium data between (293 and 343) K with high carbon dioxide mole fractions from (0.673 to 0.977) $\mathrm{mol \cdot mol^{-1}}$ for an equimolar ratio of cylohexane and cyclohexanone.

In the present work, new experimental data for the gas solubilities of carbon dioxide in mixtures of cyclohexane + cyclohexanone and in the pure substances cyclohexane and cyclohexanone are reported for T = (313.5, 352.7, and 392) K. Furthermore, the Henry's law constants were determined, and the results are compared to literature data for the pure solvents.

APPARATUS AND METHOD

The synthetic method was applied for the gas solubility experiments. The apparatus and the experimental procedure are the same as in previous investigations carried out at our laboratory.[16−20] Therefore, it is only summarized here briefly. In an experiment the pressure is determined that is required to dissolve at a constant and preset temperature a precisely known amount of the gas in a precisely known amount of the solvent. The central component of the apparatus is a cylindrical, high-pressure view cell (volume about 30 $\mathrm{cm^3}$) with two sapphire windows on each end.

In an experiment, the cell is first evacuated. Then, carbon dioxide is charged into the cell from a gas cylinder. The mass of carbon dioxide filled into the cell is determined volumetrically from the known volume of the cell and readings from temperature and pressure by applying the equation of state from Span and Wagner.[21] Then the solvent is added to the cell by a high-pressure spindle press until the gas is completely dissolved in the liquid. After equilibration, very small amounts of the solvent are withdrawn step by step until the first (very small) stable gas bubbles appear. The pressure at which the degassing starts is the solubility pressure. The mass of the solvent is calculated from the volume displacement in the calibrated spindle press and the solvent density. The solvent density was determined by separate measurements with a vibrating-tube densimeter (model DMA 4500M, Anton Paar GmbH).

Two calibrated platinum resistance thermometers in the thermostatted jacket of the view cell were used to determine the temperature. The solubility pressure was measured with two precise pressure transducers (WIKA Gmbh, full scale (2.5 and 10) MPa, respectively) in connection with a mercury barometer (Lambrecht). All pressure transducers were calibrated against a high-precision pressure balance (Desgranges and Huot).

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Table 1. Solubility of Carbon Dioxide (1) in Cyclohexane (2) + Cyclohexanone (3) at T = 313.5 K[a]

$x_3'^a = 0\ \text{mol}\cdot\text{mol}^{-1}$		$x_3'^a = 0.3\ \text{mol}\cdot\text{mol}^{-1}$		$x_3'^a = 0.6\ \text{mol}\cdot\text{mol}^{-1}$		$x_3'^a = 0.8\ \text{mol}\cdot\text{mol}^{-1}$		$x_3'^a = 1\ \text{mol}\cdot\text{mol}^{-1}$	
x_1^b	p	x_1^b	p	x_1^b	p	x_1^b	p	x_1^b	p
$\text{mol}\cdot\text{mol}^{-1}$	MPa	$\text{mol}\cdot\text{mol}^{-1}$	MPa	$\text{mol}\cdot\text{mol}^{-1}$	MPa	$\text{mol}\cdot\text{mol}^{-1}$	MPa	$\text{mol}\cdot\text{mol}^{-1}$	MPa
0.051	0.86	0.149	1.56	0.142	1.27	0.185	1.67	0.142	1.23
0.096	1.45	0.169	1.77	0.175	1.58	0.208	1.87	0.155	1.33
0.173	2.39	0.195	2.03	0.177	1.59	0.241	2.13	0.174	1.49
0.200	2.76	0.217	2.23	0.214	1.93	0.264	2.36	0.223	1.91
0.221	3.00	0.247	2.54	0.244	2.21	0.304	2.69	0.225	1.95
0.255	3.38	0.276	2.84	0.274	2.49	0.334	2.94	0.235	2.07
0.278	3.61	0.319	3.23	0.315	2.85			0.268	2.37
								0.298	2.65

[a] x_3' is the gas free mole fraction of cyclohexanone. [b] x_1 is the mole fraction of carbon dioxide in the liquid phase.

Table 2. Solubility of Carbon Dioxide (1) in Cyclohexane (2) + Cyclohexanone (3) at T = 352.7 K[a]

$x_3'^a = 0\ \text{mol}\cdot\text{mol}^{-1}$		$x_3'^a = 0.3\ \text{mol}\cdot\text{mol}^{-1}$		$x_3'^a = 0.6\ \text{mol}\cdot\text{mol}^{-1}$		$x_3'^a = 0.8\ \text{mol}\cdot\text{mol}^{-1}$		$x_3'^a = 1\ \text{mol}\cdot\text{mol}^{-1}$	
x_1^b	p	x_1^b	p	x_1^b	p	x_1^b	p	x_1^b	p
$\text{mol}\cdot\text{mol}^{-1}$	MPa	$\text{mol}\cdot\text{mol}^{-1}$	MPa	$\text{mol}\cdot\text{mol}^{-1}$	MPa	$\text{mol}\cdot\text{mol}^{-1}$	MPa	$\text{mol}\cdot\text{mol}^{-1}$	MPa
0.132	2.64	0.132	2.12	0.135	1.98	0.123	1.77	0.119	1.67
0.154	3.03	0.146	2.36	0.154	2.26	0.140	2.02	0.123	1.72
0.173	3.39	0.178	2.86	0.164	2.42	0.142	2.14	0.141	1.96
0.196	3.79	0.189	3.03	0.185	2.72	0.162	2.35	0.166	2.40
0.220	4.19	0.215	3.44	0.205	3.04	0.185	2.70	0.187	2.64
0.243	4.61	0.232	3.71	0.227	3.36	0.204	2.99	0.208	2.94
0.267	4.98	0.253	4.06	0.248	3.69	0.227	3.35	0.217	3.16
		0.273	4.38					0.248	3.58

[a] x_3' is the gas free mole fraction of cyclohexanone. [b] x_1 is the mole fraction of carbon dioxide in the liquid phase.

The maximum uncertainty in the solubility pressure measurements results from the intrinsic uncertainty of the pressure transducers (0.1 % of the transducers full scale) and an additional contribution of about 0.01 MPa from a small temperature drift inside the isolated (high-pressure) tubes filled with the solvent that connects the view cell with the pressure transducer. The uncertainty of the composition mean absolute error is about $0.006\ \text{mol}\cdot\text{mol}^{-1}$.

The studied concentration range of carbon dioxide was chosen to be similar for all studied systems so that, at the same time, the gas solubility pressure was almost always above 1 MPa.

SUBSTANCES AND SAMPLE PRETREATMENT

Carbon dioxide (4.5, volume fraction 0.99995) was purchased from Messer Griesheim GmbH, Krefeld, Germany. Cyclohexane (purity ≥ 99.0 %) and cyclohexanone (purity ≥ 99.0 %) were purchased from Sigma-Aldrich and degassed under vacuum. The solvent mixtures were prepared gravimetrically.

EXPERIMENTAL RESULTS

The solubility of carbon dioxide (1) in mixtures of cyclohexane (2) and cyclohexanone (3) was measured at cyclohexanone mole fractions of the gas-free solvent mixtures (x_3') of about (0, 0.3, 0.6, 0.8, and 1), at temperatures T = (313.5, 352.7, and 392) K, and total pressures up to 5.5 MPa. The results are given in Tables 1 to 3. The gas solubility is expressed in terms of the mole fraction, that is, the amount of substance (the number of moles) of the gas per amount of (the number of moles) solvent mixture and gas. The total pressure above those solutions is plotted against the mole fraction of carbon dioxide in the liquid phase in Figure 1.

As shown in Figure 1, a purely physical gas solubility behavior is observed. For all amounts of the gas in the liquid, the solubility pressure increases almost linearly with an increasing amount of dissolved gas. The gas solubility of carbon dioxide is better in cyclohexane than it is in cyclohexanone. Hence, the solubility of carbon dioxide is higher in the cyclohexanone-rich solutions than it is in the cyclohexane-rich solutions. It decreases strongly upon adding small amounts of cyclohexanone to cyclohexane, whereas adding small amounts of cyclohexane to cyclohexanone only has a minor effect on the solubility of carbon dioxide. For all solvent mixture compositions, the solubility of carbon dioxide in (cyclohexane + cyclohexanone) decreases with rising temperature.

HENRY'S LAW CONSTANT OF CARBON DIOXIDE IN (CYCLOHEXANE + CYCLOHEXANONE)

The (mole fraction based) Henry's law constant of carbon dioxide (1) in solvent mixtures of (cyclohexane (2) + cyclohexanone (3)) $H_{1,2+3}(T, x_3')$ (at the vapor pressure of the gas-free solvent mixture) was determined from the experimental data taken in the present work

Table 3. Solubility of Carbon Dioxide (1) in Cyclohexane (2) + Cyclohexanone (3) at T = 392 K[a]

$x_3'^a$ = 0 mol·mol^{-1}		$x_3'^a$ = 0.3 mol·mol^{-1}		$x_3'^a$ = 0.6 mol·mol^{-1}		$x_3'^a$ = 0.8 mol·mol^{-1}		$x_3'^a$ = 1 mol·mol^{-1}	
x_1^b	p	x_1^b	p	x_1^b	p	x_1^b	p	x_1^b	p
mol·mol^{-1}	MPa	mol·mol^{-1}	MPa	mol·mol^{-1}	MPa	mol·mol^{-1}	MPa	mol·mol^{-1}	MPa
0.120	3.09	0.118	2.65	0.119	2.51	0.111	2.34	0.109	2.19
0.140	3.53	0.134	2.99	0.142	2.97	0.132	2.78	0.128	2.55
0.161	3.99	0.153	3.39	0.153	3.20	0.148	3.08	0.147	2.97
0.181	4.44	0.171	3.78	0.169	3.52			0.165	3.37
0.200	4.88	0.197	4.31	0.188	3.94			0.179	3.72
0.207	5.01	0.209	4.58	0.202	4.24			0.196	4.03
0.218	5.33	0.227	4.96	0.217	4.54			0.215	4.46
0.236	5.64								

[a] x_3' is the gas free mole fraction of cyclohexanone. [b] $x_1$is the mole fraction of carbon dioxide in the liquid phase.

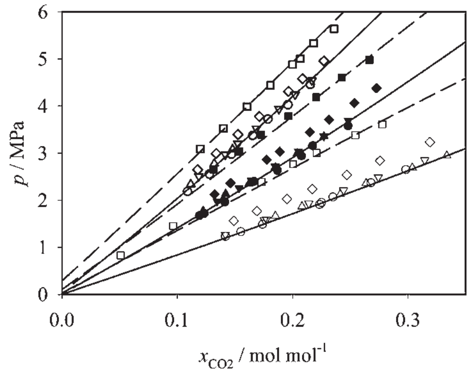

Figure 1. Total pressure p above solutions of carbon dioxide (1) + cyclohexane (2) + cyclohexanone (3) plotted against the mole fraction x_1 of carbon dioxide at different temperatures; normal symbols, T = 313.5 K; filled symbols, T = 352.7 K; bold symbols, T = 392 K, and different solvent compositions; □, x_3' = 0; ◇, x_3' = 0.3; ▽, x_3' = 0.6; △, x_3' = 0.8; ○, x_3' = 1. Full lines: PR EOS for carbon dioxide (1) + cyclohexanone (3). Dashed lines: PR EOS for carbon dioxide (1) + cyclohexane (2).

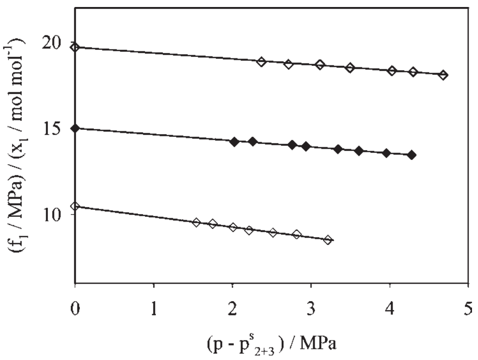

Figure 2. Influence of the pressure p on the ratio of estimated carbon dioxide fugacity (in gaseous phase) to the carbon dioxide liquid phase mole fraction for a solvent composition of x_3' = 0.3 at the temperatures: diamond, T = 315.5 K; filled diamond, T = 352.7 K; and bold diamond, T = 392 K. The points on the left are the results of the Henry's law constants found from the extrapolation according to eq 1.

presented in the previous section using an extrapolation procedure (at constant temperature and solvent composition):

$$H_{1,2+3}(T, x_3') = \lim_{p \to p_{23}^s} \left[\frac{f_1(T, p, \underline{y})}{x_1}\right] \qquad (1)$$

where

$$f_1(T, p, \underline{y}) = p y_1 \varphi_1(T, p, \underline{y}) \qquad (2)$$

f_1 and φ_1 are the fugacity and the fugacity coefficient of carbon dioxide, respectively. x_1 and y_1 are the mole fractions of carbon dioxide in the liquid and vapor phase, respectively. $\underline{y}$ is the vector specifying the composition of the vapor phase.

The vapor pressure of the gas-free (cyclohexane + cyclohexanone) mixture p_{23}^s was calculated with the Peng−Robinson equation of state (PR EOS).[22] The vapor pressures of pure cyclohexane p_2^s and pure cyclohexanone p_3^s were taken from a DIPPR equation.[23] The vapor phase composition y_1 and vapor phase fugacity coefficient of carbon dioxide φ_1 is also calculated with the PR EOS. The binary interaction parameter k_{ij} is adjusted to the experimental data from the present work for each data set at a constant temperature and solvent composition.

Table 4. Henry's Law Constant of Carbon Dioxide (1) in Cyclohexane (2) + Cyclohexanone (3)[a]

x_3'	T = 298.7 K	T = 315.5 K	T = 352.7 K	T = 392 K
mol·mol^{-1}	$H_{1,2+3}$/MPa	$H_{1,2+3}$/MPa	$H_{1,2+3}$/MPa	$H_{1,2+3}$/MPa
0.0	14.4 (3)	16.2 (3)	20.1 (3)	22.8 (3)
0.3		10.5 (3)	15.0 (3)	19.7 (3)
0.6		8.7 (3)	13.8 (3)	18.8 (3)
0.8		9.0 (3)	13.3 (3)	18 (3)
1.0	6.6 (5)	8.5 (3)	13.5 (3)	18.7 (3)

[a] The number in parentheses indicates the uncertainty in the last digit.

The calculated values for f_1/x_1 (for preset temperature and solvent mixture composition) are plotted in Figure 2, for one solvent composition, against the difference between the total pressure above (carbon dioxide + cyclohexane + cyclohexanone) and the vapor pressure of the (cyclohexane + cyclohexanone) solvent mixture. The extrapolation was done by linear regression. The resulting numerical values for the Henry's law constant of carbon dioxide in solvent mixtures of (cyclohexane + cyclohexanone) are given in Table 4. Henry's law constant is plotted in Figure 3. The relatively

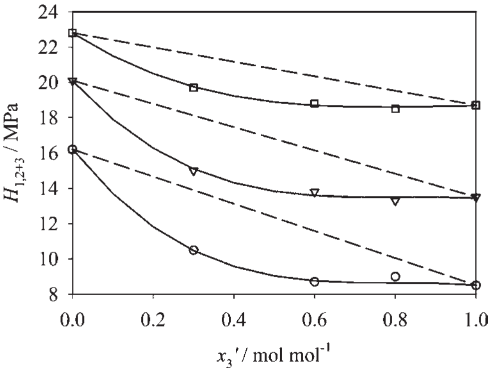

Figure 3. Henry's law constant $H_{1,2+3}$ for carbon dioxide (1) in cyclohexane (2) + cyclohexanone (3) plotted against the mole fraction x_3' of cyclohexanone (3) (on a carbon dioxide-free basis) at different temperatures: ○, T = 315.5 K; ▽, T = 352.7 K; □, T = 392 K. Full lines: guide for the eye. Dashed lines: linear interpolation.

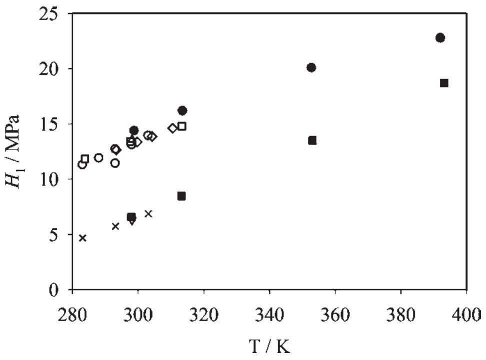

Figure 4. Experimental Henry's law constant H_1 plotted against the temperature of carbon dioxide in pure cyclohexane and pure cyclohexanone, respectively. Cyclohexane: ●, this work; ○, Gironi and Lavecchia;[2] △, Patyi et al.;[3] □, Wilhelm and Battino;[5] ◇, Dymond.[10] Cyclohexanone: ■, this work; ×, Gallardo et al.;[11] and ▽, Mellendo et al.[12]

large uncertainty for the Henry's law constant at T = 392 K and x_3' = 0.8 is due to fact that the extrapolation for that Henry's law constant is only based on three experimental data points. As shown in Figure 3, and according to the observations for the gas solubility, the Henry's law constant decreases strongly upon adding small amounts of cyclohexanone to cyclohexane, whereas adding small amounts of cyclohexane to cyclohexanone has only a minor effect on the Henry's law constant.

COMPARISON WITH LITERATURE DATA

The experimental results from the present study for the solubility of carbon dioxide in cyclohexane at (353 and 313) K can directly be compared to those of Anderson et al.[4] and Al-Sahhaf et al.,[7] respectively. The agreement in the overlapping concentration range is very good. Deviations are within the scattering of the data sets. The same holds for the results at 313 K for the solubility of carbon dioxide in cyclohexanone compared to the data of Chang and Chen.[14] For the ternary system, a direct comparison with literature data is not possible. However, an extrapolation of our data at 313 K and x_3' = 0.6 at low carbon dioxide concentrations fits with the data of Esmelindro et al.[15] at the same temperature and x_3' = 0.5 at high carbon dioxide concentrations within the uncertainty of the extrapolation.

The Henry's law constant of carbon dioxide determined in the present work is compared to experimental results from the literature for carbon dioxide in cyclohexane and in cyclohexanone in Figure 4. The literature data were taken from the Dortmund Data Bank.[24] The agreement between the present experimental data and the literature data are very good for cyclohexanone. For cyclohexane, the present experimental results are slightly above the literature data. Note that Table 4 also contains two Henry's law constants in the pure solvents at 298 K that were only measured to have more data for the comparison with the literature.

CONCLUSIONS

Experimental results are presented for the solubility of carbon dioxide in pure liquid cyclohexane and cyclohexanone and in liquid mixtures of (cyclohexane + cyclohexanone). From the data, the Henry's law constant of carbon dioxide was determined.

A purely physical gas solubility behavior is observed. The gas solubility of carbon dioxide in cyclohexane + cyclohexanone decreases strongly upon adding small amounts of cyclohexanone to cyclohexane, whereas adding small amounts of cyclohexane to cyclohexanone only has a minor effect on the solubility of carbon dioxide.

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