J. Chem. Eng. Data **2002,** *47,* 855−860

Solubility of Carbon Dioxide and Methane in Aqueous Methyldiethanolamine Solutions

Jan Addicks* and Geir A. Owren

Department of Refrigeration and Air Conditioning, Norwegian University of Science and Technology,
Kolbjørn Hejes vei 1B, 7491 Trondheim, Norway

Arne O. Fredheim and Kirsti Tangvik

Statoil, Research & Development, Postuttak, 7005 Trondheim, Norway

The vapor−liquid equilibrium of the system methane, carbon dioxide, *N*-methyldiethanolamine (MDEA), and water was measured at pressures up to 200 bar and at the temperatures 313.15 and 353.15 K. The concentration of carbon dioxide in the vapor phase varied between 0.02 and 0.20 mole fraction, and the concentrations of the aqeuous MDEA solution were 0.30 and 0.50 mass fraction. The phase equilibrium was measured in a variable-volume static cell. The pressure in the cell can reach a maximum of 600 bar, and the temperature of the cell can range from 223.15 to 473.15 K.

Introduction

Aqueous solutions containing alkanolamines are widely used for the removal of acid gases from natural gas. The acid gas examined in this work is CO_2. The alkanolamine used is *N*-methyldiethanolamine (MDEA). CO_2 has to be removed from the natural gas because of transport requirements and sale gas specifications. The absorption process is classified as chemical absorption, since CO_2 and MDEA react partly to nonvolatile ionic species in the liquid phase.

Gas cleaning is currently performed at pressures around 70 bar in the absorption column. This makes it necessary to decompress the gas coming from the reservoir at a pressure of more than 200 bar to the working pressure of the absorption column. After the cleaning process, the gas must be compressed to around 200 bar to be transported through pipelines to the consumer. The energy necessary to compress the gas could be saved, if a high-pressure cleaning process were designed.

To design such a gas cleaning process, equilibrium, mass-transfer, and chemical reaction data are needed. In this work, vapor−liquid equilibrium (VLE) data for the system CO_2, MDEA, methane, and water were measured at pressures up to 200 bar and the temperatures 313 and 353 K. The concentration of the aqueous MDEA solution was 0.30 and 0.50 mass fraction.

The work of Lawson and Garst[1] was the only one found where the VLE of CO_2, CH_4, alkanolamine, and water was measured. The alkanolamine was diethanolamine.

Experimental Section

The equilibrium measurements were carried out in the cell shown in Figure 1. The cell has been designed to operate at a pressure up to 600 bar in a temperature range from 223 to 473 K. The cell volume can be varied with two movable pistons with a maximum of 450 mL. The contents of the cell can be observed through a sapphire window. The temperature in the cell is measured with a resistance thermometer with an accuracy of ±0.1 K. The pressure in the cell is measured with a high-pressure sensor up to 1000 bar, and its accuracy is ±0.1% of full scale. The equilibrium cell is placed in an air bath to keep the temperature in the cell constant. The air bath temperature fluctuated with ±0.5 °C. The pressure in the equilibrium cell is adjusted with the pistons by changing the cell volume.

The cell was filled with liquid and gas via valve 1a, 1c; 2a, 2c; or 3a, 3c. CO_2 and methane were injected in gas form from their storage bottles into the cell. The liquid MDEA−water solution was pumped from a storage vessel (volume = 1 L) into the cell. The pressure and temperature in the cell were fixed, and as long as gas was absorbing in the liquid phase, the cell volume decreased automatically to keep the pressure at its fixed value. Equilibrium was obtained when cell volume and temperature were constant. This took approximately 8 h.

After equilibrium was obtained, samples from the liquid and vapor phase were withdrawn for analysis. The samples were withdrawn from the equilibrium cell by transferring them via valves 1a, 1b; 2a, 2b; or 3a, 3b into a vessel filled with helium at atmospheric pressure and ambient temperature. This vessel is called the separator. Another vessel with variable volume is connected to the separator, it is called the gas meter. When a sample is withdrawn, the sample expands in the separator, because it is decompressed from the high pressure in the equilibrium cell to the low pressure in the separator. In addition, when a liquid phase if withdrawn, gas will be released from the liquid phase when the sample enters the separator. To avoid an increase in pressure in the separator, the volume of the gas meter is increased so that the pressure in the separator is kept at 1 atm. At the end of the sampling process, a vapor phase, and perhaps a liquid phase, will be present at 1 atm and ambient temperature. Both phases have to be analyzed to determine the composition of the sample from the equilibrium cell.

Liquid does not enter the gas meter but is collected in the separator, from which it is taken for analysis. A part of the gas collected in the gas meter is pumped into a glass bottle connected to the gas meter and brought to the gas chromatograph for analysis. The pressure in the equilib-

---

* Corresponding author. E-mail: jan.addicks@maskin.ntnu.no.

10.1021/je010292z CCC: $22.00

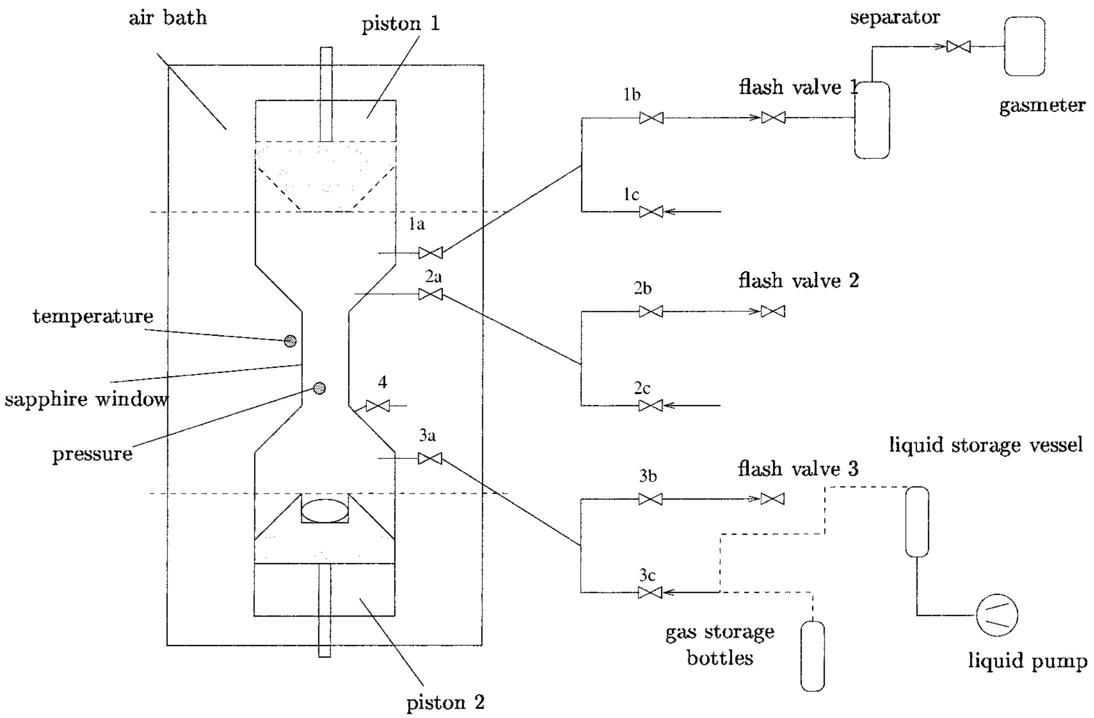

Figure 1. Equilibrium cell.

rium cell is kept constant while withdrawing a sample, by decreasing the cell volume with piston 1 or 2.

All the measuring devices are connected to a PC to store measurements and to control the functions of the equilibrium cell and the auxiliary devices. Valve 4 is used for cleaning the cell.

The vapor phase was analyzed for CO_2 and CH_4. It could not be analyzed for its water and MDEA content. It is assumed that the amount of amine in the vapor phase is negligible. The water content in the vapor phase must be calculated using adequate models, such as, for example, those in refs 2 and 3. The analysis of the vapor phase was done by gas chromatography. The liquid phase is analyzed for CO_2, methane, MDEA, and water. The content of CO_2 in the liquid phase was determined by precipitation titration and gas chromatography (GC). All the other components were directly determined by gas chromatography. Precipitation titration[4,5] was used to calibrate the gas chromatograph for analysis of CO_2 in the liquid phase.

If a liquid sample from the separator was analyzed by precipitation titration, 3 mL of the sample was mixed with 10 mL of 3 mol·L^{-1} NaOH. CO_2 and HCO_3^- present in the liquid sample react with NaOH to give CO_3^{2-}. When 50 mL of 0.3 mol·L^{-1} $BaCl_2$ is added, CO_3^{2-} is precipitated as $BaCO_3$. To agglomerate the $BaCO_3$ particles, the solution is heated for a few minutes. After the solution is cooled to ambient temperature, it is filtered (Millipore, Type HVLP, 0.45 μm). The filter and filtrate are put into distilled water, and the $BaCO_3$ is dissolved by the addition of 0.1 mol·L^{-1} HCl. Then the solution is boiled to remove CO_2. After the solution is cooled to ambient temperature, it is titrated with 0.1 mol·L^{-1} NaOH to find the amount of HCl not used to dissolve $BaCO_3$. From the amounts of HCl found by titration and the amount added to dissolve the $BaCO_3$ particles, the amount of CO_2 in the solution was calculated. The titration was carried out with an automatic titrator (Metrohm 785 DMP Titrino).

The measurement procedure was tested by measuring the vapor−liquid equilibrium of CO_2 and water, of CH_4 and water, and of MDEA, CO_2, and water. The deviation from experimental literature data was less than 5% in loading (moles of solute/moles of solvent).

Table 1. Solubility of CO_2 in H_2O at 20 °C

P / bar	$m_{CO_2}{}^{6}$ / mol·kg^{-1}	$m_{CO_2,exp}$ / mol·kg^{-1}
10.12	0.382	0.369
15.07	0.524	0.534
20.20	0.686	0.697
25.33	0.827	0.826

Table 2. CO_2 in MDEA−H_2O at 40 °C and $m_{MDEA} = 2.633$ mol·kg^{-1}

P / bar	$m_{CO_2}{}^{6}$ / mol·kg^{-1}	$m_{CO_2,exp}$ / mol·kg^{-1}
11.55	2.921	2.794
21.17	3.114	3.063
30.29	3.267	3.206
40.80	3.435	3.359

Results

System CO_2−H_2O. To check the accuracy of analysis of the content of CO_2 in the liquid phase by precipitation titration, the solubility of carbon dioxide in water was measured. The experimental results are compared with data from Silkenbäumer et al.[6] and are given in Table 1.

At 25 bar and 20 °C, the measuring procedure was repeated four times (equilibration, withdrawing a sample, and analyzing the liquid phase). From these measurements, the variance of the precipitation titration method is 4.2×10^{-5}, the standard deviation is 6.5×10^{-3}, and the relative standard deviation is 0.79%.

System MDEA−CO_2−H_2O. The content of CO_2 in the aqueous MDEA solution was measured by GC. The response factors for the determination of CO_2 in the liquid phase were found by precipitation titration. The response factors for the determination of H_2O and MDEA were found by analyzing MDEA−H_2O solutions of known composition. The experimental results of this work and experimental

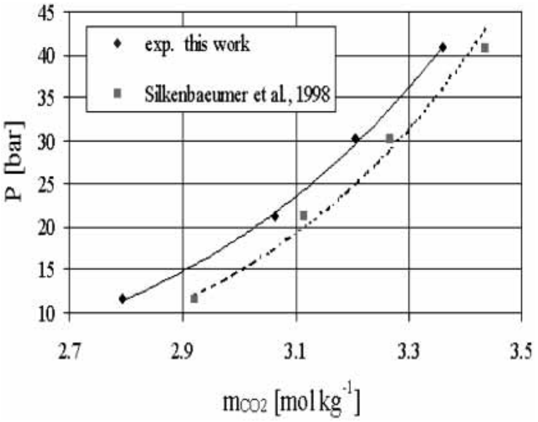

Figure 2. CO_2, MDEA, and water system, m_{MDEA} = 2.633 mol·kg^{-1}.

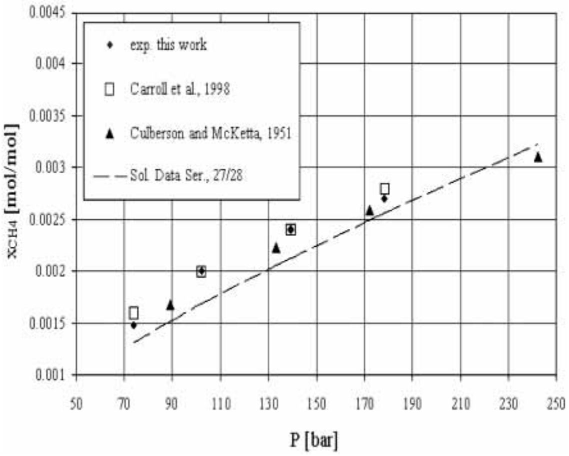

Figure 3. Solubility of CH_4 in water.

Table 3. Experimental Results for the Solubility of CH_4 in Water at 25 °C

P/bar	x_{CH_4}[7]	x_{CH_4}[8]	$x_{CH_4,exp}$
73.6	0.001 56	0.001 32	0.001 48
102.2	0.002 01	0.001 69	0.002 01
139.2	0.002 37	0.002 13	0.002 40
178.2	0.002 79	0.002 56	0.002 74

data from Silkenbäumer et al.[6] are given in Table 2 and shown in Figure 2 together with smoothing lines of the experimental data.

System H_2O-CH_4. To check if the measuring procedure is accurate enough to determine the correct amount of CH_4 in the liquid phase, the vapor−liquid equilibrium of methane and water was measured and compared with data in the literature.[7,8] In the *Solubility Data Series*[8] an equation is given to calculate the methane solubility in water based on 242 data points. This equation is used to calculate the values given in Table 3.

The maximum deviations are 5% at 73.6 bar from the data from Carroll et al.[7] and 19% at 102 bar from the data from the *Solubility Data Series*.[8]

In Figure 3 the experimental data measured in this work are plotted together with data from Carroll et al.,[7] Culberson and McKetta,[9] and the *Solubility Data Series*.[8]

Neither the vapor phase nor the liquid phase was analyzed for its composition. The vapor is supposed to consist of nearly pure CH_4, and when the liquid phase is withdrawn into the separator, it is assumed that all the dissolved CH_4 goes over in the vapor phase. The quantities measured to determine the solubility are the volume of the gas meter when withdrawing the liquid phase and the weight of the withdrawn liquid. From the weight of the liquid, the mole number of water is calculated. The mole number of CH_4 is calculated from the volume of the gas meter and the density of the methane vapor at the temperature and pressure in the separator.

Table 4. Precision (p) of the Phase Composition (comp) Measurement, Determined from the Data in Table 5

	p of the liquid-phase comp		p of the vapor-phase comp	
P	CO_2	CH_4	CO_2	CH_4
bar	mol/mol$_{MDEA}$	mol/mol$_{MDEA}$	mol/mol	mol/mol
150	±0.88%	±1.66%	±0.46%	±0.08%
200	±0.20%	±1.92%	±2.26%	±0.37%

Table 5. Liquid−Vapor Equilibrium Compositions (comp) of the System MDEA−CO_2−CH_4−H_2O at 313.15 K and w_{MDEA} = 0.30 g/g

	liquid-phase comp		vapor-phase comp	
P	CO_2	CH_4	CO_2	CH_4
bar	mol/mol$_{MDEA}$	mol/mol$_{MDEA}$	mol/mol	mol/mol
100	0.2586	0.0264	0.0011	0.9989
100	0.6203	0.0257	0.0048	0.9952
150	0.6256	0.0331	0.0039	0.9961
150	**0.9887**	**0.0256**	**0.1411**	**0.8589**
150	**1.0062**	**0.0247**	**0.1424**	**0.8576**
200	**1.0285**	**0.0295**	**0.1365**	**0.8635**
200	**1.0243**	**0.0284**	**0.1428**	**0.8572**
200	0.9086	0.0353	0.0298	0.9702
200	0.9687	0.0339	0.0544	0.9456

Table 6. Liquid−Vapor Equilibrium Compositions (comp) of the System MDEA−CO_2−CH_4−H_2O at 353.15 K and w_{MDEA} = 0.30 g/g

	liquid-phase comp		vapor-phase comp	
P	CO_2	CH_4	CO_2	CH_4
bar	mol/mol$_{MDEA}$	mol/mol$_{MDEA}$	mol/mol	mol/mol
100	0.6895	0.0234	0.0778	0.9222
100	0.7713	0.0222	0.1056	0.8944
100	0.8347	0.0206	0.1441	0.8559
150	0.7365	0.0323	0.0631	0.9369
150	0.8736	0.0289	0.1343	0.8657
200	0.7593	0.0367	0.0594	0.9406
150	0.8122	0.0311	0.0928	0.9072
200	0.8288	0.0378	0.0875	0.9125
200	0.9690	0.0275	0.1455	0.8545

Table 7. Liquid−Vapor Equilibrium Compositions (comp) of the System MDEA−CO_2−CH_4−H_2O at 313.15 K and w_{MDEA} = 0.50 g/g

	liquid-phase comp		vapor-phase comp	
P	CO_2	CH_4	CO_2	CH_4
bar	mol/mol$_{MDEA}$	mol/mol$_{MDEA}$	mol/mol	mol/mol
100	0.5815	0.0114	0.0124	0.9876
200	0.8128	0.0158	0.0488	0.9512
100	0.9232	0.0074	0.2627	0.7373
200	0.9426	0.0116	0.2533	0.7457

System CH_4, CO_2, H_2O, and MDEA. The liquid−vapor equilibrium of the system MDEA, H_2O, CO_2, and CH_4 was measured at 100, 150, and 200 bar and at the temperatures 313.15 K and 353.15 K. The concentration of the aqueous MDEA solution was 0.30 and 0.50 g per g. The amount of MDEA and water in the vapor phase could not be measured, and the values given in Table 5−8 for the amount of CO_2 and CH_4 in the vapor phase are therefore given on a water and MDEA free basis.

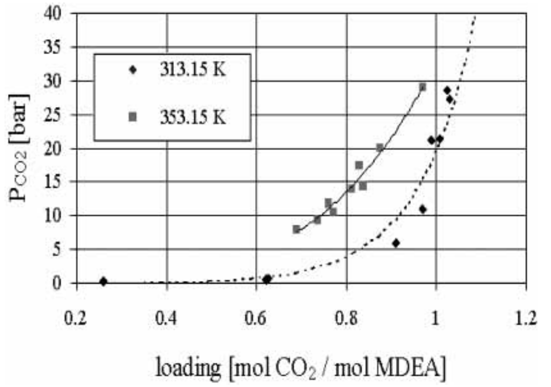

Figure 4. CO_2, CH_4, MDEA, and water system, w_{MDEA} = 0.30 g/g.

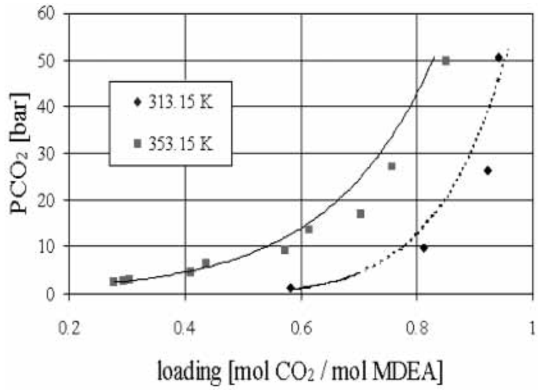

Figure 5. CO_2, CH_4, MDEA, and water system, w_{MDEA} = 0.50 g/g.

Table 8. Liquid−Vapor Equilibrium Compositions (comp) of the System MDEA−CO_2−CH_4−H_2O at 353.15 K and w_{MDEA} = 0.50 g/g

	liquid-phase comp		vapor-phase comp	
P	CO_2	CH_4	CO_2	CH_4
bar	mol/mol$_{MDEA}$	mol/mol$_{MDEA}$	mol/mol	mol/mol
100	0.2762	0.0188	0.0243	0.9757
150	0.2935	0.0257	0.0187	0.9813
200	0.3037	0.0315	0.0159	0.9841
100	0.4091	0.0187	0.0467	0.9533
200	0.4364	0.0270	0.0314	0.9686
100	0.5713	0.0112	0.0948	0.9052
200	0.6134	0.0204	0.0687	0.9313
100	0.7026	0.0114	0.1712	0.8288
200	0.7569	0.0209	0.1364	0.8636

It is assumed that these measurements are done with the same accuracy as that of the reference measurements. To get an idea about the precision of the measurements, two measurement points were done twice (bold face in Table 5). The precision *p* in percent is calculated with the following equation

$$p = \pm \left| \frac{x_i - \bar{x}}{\bar{x}} \right| \times 100 \qquad (1)$$

and is given in Table 4. $\bar{x}$ is the average of two measurements, and x_i is the value of one measurement.

For 313.15 and 353.15 K and w_{MDEA} = 0.30, the measurements are shown as plots of partial pressure of CO_2 versus loading of CO_2 in Figure 4. The partial pressure of CO_2 is not corrected for the partial pressure of water. Figure 5 shows results at 313.15 and 353.15 K for w_{MDEA} = 0.5. In both figures, smoothing curves of the experimental data are added to demonstrate the consistency of the data.

Discussion

Figures 4 and 5 show the expected curve of partial pressure of CO_2 versus loading of CO_2. The partial pressure increases with the loading, since more and more MDEA is used up in the reaction. The isotherms at 353.15 K are above the isotherm at 313.15 K, because more CO_2 can be absorbed at low temperatures.

For the system CO_2, CH_4, MDEA, and water equilibrium was obtained after 8 h. Monitoring of the cell volume, the temperature, and the pressure ensured that the sample was taken at equilibrium. The volume was always the last quantity to become constant. Stirring is very important while the system is approaching the equilibrium state. The mass transport over the small vapor−liquid surface in the equilibrium cell is so slow that, without stirring, pressure, volume, and temperature can become constant without equilibrium being reached.

The equilibrium points were measured at 100, 150, and 200 bar. Because the pressure difference between the single measuring points is comparatively large, it was not unimportant if the pressure was decreased or increased by 50 bar to adjust to a new equilibrium point. When the pressure was decreased, foaming occurred in the equilibrium cell and this foam did not vanish completely but stayed at the vapor−liquid interface and on the sapphire window. Therefore, the pressure was always increased to adjust to a new equilibrium point.

The reference measurements were the check of the measuring procedure used. They agree well with the literature data chosen for comparison. However, for the system CH_4 and water, the maximum relative deviation was found to be 20% of the data from the *Solubility Data Series*[8] while the measurements agree very well with the data from Carroll et al.[7]

In contrast to the measurements on the system CO_2, CH_4, MDEA, and water where the vapor phase was analyzed for its CO_2 and CH_4 content, it was not necessary to analyze the vapor phase for any of the reference measurements. In the reference measurements only the liquid-phase composition and pressure were determined and compared to literature data. This together with the small number of test measurements might be the largest uncertainty when assuming the same accuracy for the high-pressure measurements as for the reference measurements. Another effect which was not checked by the reference measurements is the solubility of CH_4 in aqueous MDEA solutions. More CH_4 dissolves in aqueous MDEA solutions than in water.[10] Thus, if the collected liquid phase in the separator is not analyzed for CH_4, the error might be larger than that in the reference measurements done on the system CH_4 and water. But also the MDEA−water solution withdrawn into the separator was regarded to be free of CH_4, because no CH_4 peak was detected on the chromatogram when analyzing this liquid.

The data obtained in this work were measured at pressures up to 200 bar. To investigate the influence of the high pressure and the presence of CH_4, the measurements are compared with literature data obtained at lower pressures but at the same temperature and a similar MDEA concentration. In Figure 6 this comparison is made for the partial pressure of CO_2 at 313.15 K and 0.30 g/g. The high-pressure measurements are compared with data from Kuranov et al.[11] and Jou et al.[4] Kuranov et al.[11] measured the VLE of CO_2, water, and MDEA at pressures

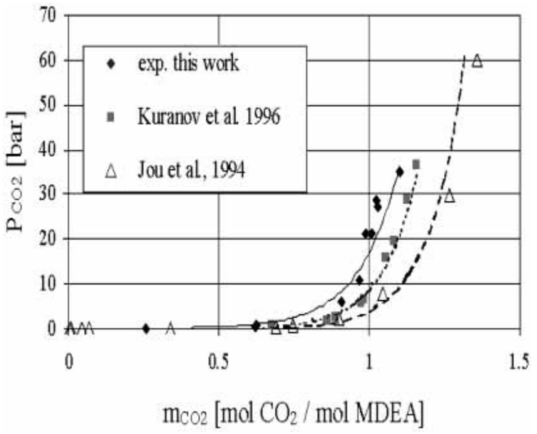

Figure 6. Comparison between high-pressure and low-pressure measurements, $T = 313.15$ K, $w_{MDEA} = 0.30$ g/g.

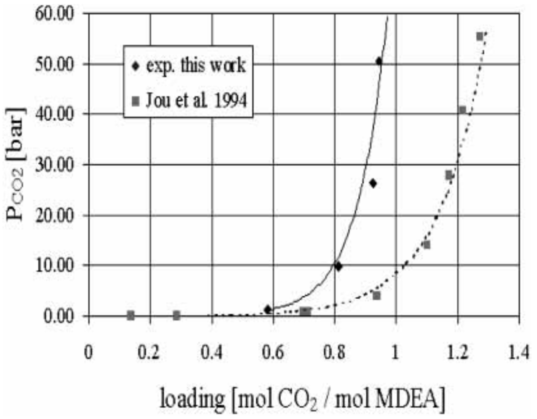

Figure 7. Comparison between high-pressure and low-pressure measurements, $T = 313.15$ K, $w_{MDEA} = 0.50$ g/g.

up to 50 bar and a concentration of the aqueous MDEA solution of 0.32 g/g. Jou et al.[4] measured the VLE of CO_2, water, and MDEA at pressures up to 150 bar and a concentration of the aqueous MDEA solution of 0.30 g/g.

The partial pressure curve from this work shown in Figure 6 increases sharply at a lower CO_2 concentration than that for the literature data.

The comparison at 313.15 K and 0.50 g/g shown in Figure 7 shows the same picture as that discussed for Figure 6.

The amount of water in the vapor phase could not be measured. However, the vapor pressure of water at 353.15 K is only 0.474 bar, and at 313.15 it is 0.074 bar. Therefore, the error introduced in the measured partial pressure of CO_2 and CH_4 seems to be negligible. The vapor pressure of MDEA[12] is even smaller than the vapor pressure of water and can therefore also be neglected.

Further discussion of the experimental data presented here should include model calculations. With model calculations it was possible to examine the influence of pressure and methane by comparing fugacities of CO_2 instead of partial pressures of CO_2. If the fugacity of CO_2 were the same for the high-pressure and the low-pressure measurements, it could be concluded that the high pressure and the presence of methane had no influence on the liquid phase. With model calculations also the amount of water in the vapor phase could be determined and the influence of MDEA and dissolved CO_2 on the solubility of methane could be investigated. These calculations are presented in ref 15. In ref 15 the electrolyte NRTL[16] equation and the extended UNIQUAC equation[17] are used to model the data presented here.

No investigation of the salting in or salting out of methane in loaded amine solutions was presented here. But there are two trends present in loaded amine solutions. CH_4 is salted in into aqeous alkanolamine solutions[7,13,14] because of the presence of the alkanolamine, and salted out from loaded aqueous alkanolamine solutions[1] because of the presence of ions and molecular CO_2. Plotting the partial pressure of CH_4 versus the loading of CH_4 shows a larger scatter of the experimental data than that for CO_2. It is assumed that this is due to the influence of MDEA and dissolved CO_2 in the liquid phase on the solubility of CH_4.[15]

Conclusion

The simultaneous absorption of CO_2 and CH_4 was measured at pressures between 100 and 200 bar and at two different temperatures and concentrations of aqueous MDEA solutions. The accuracy of the experimental method used was demonstrated by comparing measurements of the systems CO_2−water, CH_4−water and CO_2, and MDEA and water with data from the literature. The accuracy of the new data points obtained was estimated from repetitive measurements. The new high-pressure data are compared with experimental data obtained at lower pressures and with CO_2 as the only gas present. This comparison shows that the partial pressure curve of CO_2 for the high-pressure measurements increases steeply at lower loadings of CO_2 than those for the low-pressure measurements.

Literature Cited

(1) Lawson, J. D.; Garst, A. W. Hydrocarbon Gas Solubility in Sweetening Solutions: Methane and Ethane in Aqueous Monoethanolamine and Diethanolamine. *J. Chem. Eng. Data* **1976**, *21*, 30−32.

(2) Chunxi, L.; Fürst, W. Representation of CO_2 and H_2S Solubility in Aqueous MDEA Solutions using an Electrolyte Equation of State. *Chem. Eng. Sci.* **2000**, *55*, 2975−2988.

(3) Posey, M. L. Thermodynamic Model for Acid Gas Loaded Aqueous Alkanolamine Solutions. Ph.D. Dissertation of University of Texas at Austin, 1996.

(4) Jou, F. Y.; Otto, F. D.; Mather, A. E. Vapor-Liquid Equilibrium of Carbon Dioxide in Aqueous Mixtures of Monoethanolamine and Methyldiethanolamine. *Ind. Eng. Chem. Res.* **1994**, *33*, 2002−2005.

(5) Jou, F. Y.; Mather, A. E.; Otto, F. D. The Solubility of CO_2 in a 30 Mass Percent Monoethanolamine Solution. *Can. J. Chem. Eng.* **1995**, *73*, 140−147.

(6) Silkenbäumer, D.; Rumpf, B.; Lichtenthaler, R. N. Solubility of Carbon Dioxide in Aqueous Solutions of 2-Amino-2-methyl-1-propanol and *N*-Methyldiethanolamine and their Mixtures in the Temperature Range from 313 to 353 K and Pressures up to 2.7 MPa. *Ind. Eng. Chem. Res.* **1998**, *37*, 3133−3141.

(7) Carroll, J. J.; Jou, F.; Mather, A. E.; Otto, F. The Solubility of Methane in Aqueous Solutions of Monoethanolamine, Diethanolamine and Triethanolamine. *Can. J. Chem. Eng.* **1998**, *76*, 945−951.

(8) Battino, R. *Solubility Data Series IUPAC, Methane*; Pergamon Press: 1987; Vol. 27/28.

(9) Culberson, O. L.; McKetta, J. J. Phase Equilibria in Hydrocarbon−Water Systems III − The Solubility of Methane in Water at Pressures to 10,000 PSIA. *Pet. Transactions, AIME* **1951**, *192*, 223−226.

(10) Carroll, J. J.; Jou, F.; Mather, A. E.; Otto, F. D. The Solubility of Methane in Aqueous Solutions of Monoethanolamine, Diethanolamine and Triethanolamine. *Can. J. Chem. Eng.* **1998**, *76*, 945−951.

(11) Kuranov, G.; Rumpf, B.; Smirnova, N. A.; Maurer, G. Solubility of Single Gases Carbon Dioxide and Hydrogen Sulfide in Aqueous Solutions of *N*-methyldiethanolamine in the Temperature Range 313−413 K at Pressures up to 5 MPa. *Ind. Eng. Chem. Res.* **1996**, *35*, 1959−1966.

(12) Xu, S.; Qing, S.; Zhen, Z.; Zhang, C. Vapor Pressure Measurements of Aqueous *N*-methyldiethanolamine. *Fluid Phase Equilib.* **1991**, *67*, 197−201.

(13) Jou, F. Y.; Carroll, J. J.; Mather, A. E.; Otto, F. D. Solubility of Methane and Ethane in Aqueous Solutions of Methyldiethanolamine. *J. Chem. Eng. Data* **1998**, *43*, 781−784.

(14) Carroll, J. J.; Mather, A. E. A model of the Solubility of Light Hydrocarbons in Water and Aqueous Solutions of Alkanolamines. *Chem. Eng. Sci.* **1997**, *52*, 545−552.

(15) Addicks, J. Solubility of Carbon Dioxide and Methane in Aqueous *N*-Methyldiethanolamine Solutions at Pressures between 100 and 200 bar. Ph.D. Dissertation of the Norwegian University of Science and Technology, to be published April 2002, download at www.kkt.ntnu.no/kkt2/research/Multiphase.

(16) Chen, C. C.; Evans, L. B. A Local Composition Model for the Excess Gibbs Energy of Aqueous Electrolyte Systems. *AIChE J.* **1986**, *32*, 444−454.

(17) Thomsen, K. Aqueous Electrolytes: Model Parameters and Process Simulation. Ph.D. Dissertation, Technical University Denmark, 1997.

Received for review November 5, 2001. Accepted March 23, 2002.

JE010292Z