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Solubility of Carbon Dioxide in Aqueous Solutions of 3-Amino-1-propanol

Lihu Dong, Jian Chen,* and Guanghua Gao

State Key Laboratory of Chemical Engineering, Department of Chemical Engineering, Tsinghua University, Beijing 100084, China

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The solubility of CO_2 in the aqueous 3-amino-1-propanol (or monoproanolamine, MPA) with amount-of-substance concentrations of (2, 4, and 5) $mol \cdot dm^{-3}$ were measured at temperatures of (313.15, 343.15, 373.15, and 393.15) K for CO_2 partial pressures from (2.5 to 700) kPa where the CO_2 loading is from (0.2 to 1.0) mol of CO_2 per mol of MPA. The results show that MPA solutions have a greater solubility of CO_2 that those in monoethanolamine (MEA) solutions. The Kent−Eisenberg model was used to correlate the experimental data with the equilibrium constants expressed as functions of temperature, amine concentration, and CO_2 loading. Satisfactory results were obtained for the correlation of solubility of CO_2 in aqueous MPA solutions.

Introduction

Recently the worldwide climate change is receiving ever-increasing attention. With the energy demand predicted to rise by 50 % and the energy-related CO_2 emissions to rise by 52 % in 2030, the global warming caused by greenhouse gases is an issue.[1,2] One of the main focuses is the reduction of greenhouse gas emissions, especially CO_2 emissions into the atmosphere. The majority of industrial CO_2 emissions comes from power plants that use fossil fuels such as coal and natural gas. CO_2 capture and sequestration (CCS) from fossil fuel power plants is gaining widespread interest as a potential method of controlling greenhouse gas emissions.

The aqueous alkanolamine solutions have to date received the greatest attention scientifically for the removal of CO_2.[3–7] Monoethanolamine (MEA) is widely used for the absorption of CO_2, especially from flue gases at low and atmospheric pressures. However, CO_2 capture technology with the MEA solution still suffers disadvantages of high energy consumption and large solvent degradation. 3-Amino-1-propanol (MPA) is a linear primary amine that is slightly higher hindered. Eirik and Hallvard[8] used quantum chemistry to study several amines and showed that MPA has a higher pK_a and a higher reaction kinetic rate with CO_2. It is suitable to deal with the low CO_2 concentration gas streams. Prachi et al.[9] pointed out that the amines with three carbon atoms have a better CO_2 absorption capacity than the others. Amr et al.[10] investigated the kinetics of MPA reacting with CO_2 using a stopped flow technique. However, the systematic solubility data for CO_2 in aqueous MPA solution is still lacking in the literature. The present work reports that the solubility of CO_2 in the MPA solution of amount-of-substance concentrations of (2, 4, and 5) $mol \cdot dm^{-3}$ were measured at temperatures of (313.15, 343.15, 373.15, and 393.15) K over a wide range of CO_2 partial pressure from (2.5 to 700) kPa.

Experimental Section

Materials. Aqueous alkanolamine solutions were prepared with deionized water. Reagent-grade MPA was obtained from Acros Organics with a mass fraction purity > 0.99 and used without any further purification. Carbon dioxide and nitrogen gases had a mass fraction purity of 0.999.

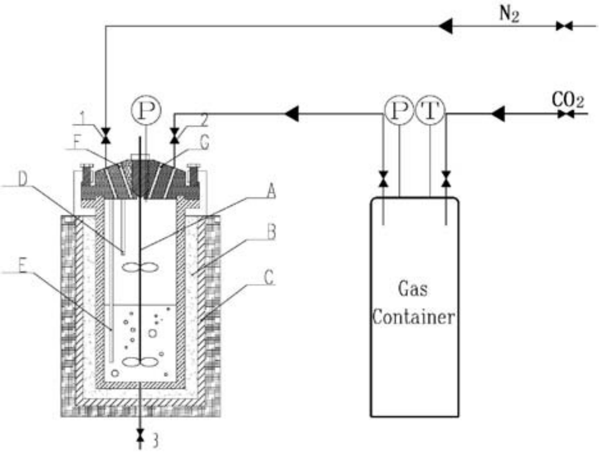

Figure 1. Schematic diagram of the experimental equipment: 1, N_2 inlet valve; 2, CO_2 inlet valve; 3, liquid outlet valve; A, magnetic stir; B, oil bath; C, heating jacket; D, gas thermodetector; E, liquid thermodetector; F, gas outlet valve; G, liquid inlet valve.

Apparatus. The solubility measurements presented here were carried out in a cell reactor shown schematically in Figure 1. The reactor (model KCFDU2-2, Tianzhouhaitai Co.) consists of an electrically heated stainless steel cylindrical tank of volume 300 cm^3 fitted with a magnetically coupled stirrer on the top. There are valves for the inlet and outlet of gas and liquid. A calibrated thermometer (PT100, Kunlunhaian Co.) was inserted into the cell to measure the temperature with an uncertainty of ± 0.1 K. A water or oil bath was used to control the temperature of the cell. A calibrated pressure transducer (model JYB-KD-HAG, Kunlunhaian Co.) was used to measure the pressures of the cylinder and gas container with an uncertainty of ± 0.5 %.

Procedure. First, the cell was completely emptied and purged with nitrogen gas to remove remaining air, and a volume of about 100 cm^3 of MPA solution of the desired concentration was injected through a valve. The temperature was adjusted to the desired value. A time of 10 h was permitted for absorption while stirring at a rate of 100 rpm during which it was assumed the vapor−liquid equilibrium had been achieved. The initial pressure while the system achieved the equilibrium was P_V. The carbon dioxide gas introduced into the cell from a cylinder, for which the initial volume, temperature, and pressure were V_{gc}, T_{gc1}, and P_{gc1}, respectively, and the subsequent volume, tem-

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* Corresponding author. E-mail: cj-dce@mail.tsinghua.edu.cn.

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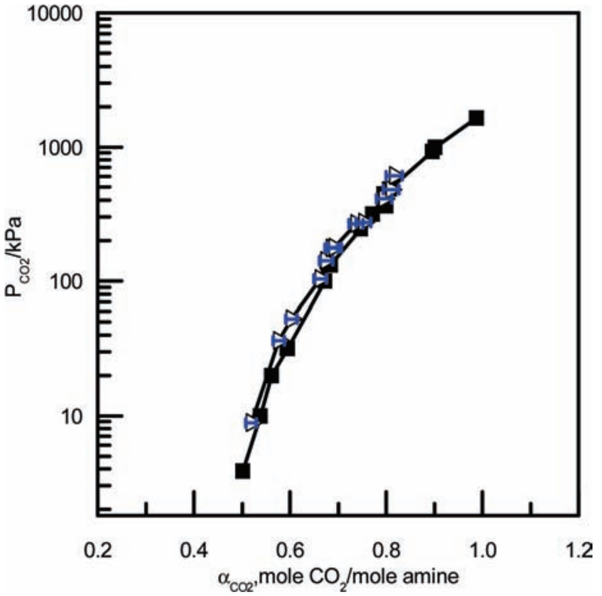

Figure 2. Solubility of CO_2 in 2.5 mol·dm^{-3} MEA solutions at $T = 313.15$ K. ■, literature data of Lee et al.;[16] ▽, this work with blue error bars for CO_2 loading.

perature and pressure were V_{gc}, T_{gc2}, and P_{gc2}, respectively. The CO_2 quantity added to the reactor, n_{co_2}, could be calculated by Peng−Robinson cubic equation.[11]

After CO_2 transfer to the reactor, the equilibrium was attained in a time of about 10 h. The equilibrium pressure, P_T, was recorded. The CO_2 partial pressure in the vapor phase was determined by assuming that the phase obeyed Dalton's law[12] and therefore simply taken as the difference between the total equilibrium pressure and the CO_2-free, aqueous MPA solution:

$$P_{co_2} = P_T - P_V \tag{1}$$

The amount of CO_2, $n_{co_2}{}^{g}$, remaining in the gas phase was determined by cubic PR cubic equation with the known pressure,P_{co_2}, and the temperature and volume of the gas phase.

Armed with $n_{co_2}{}^{g}$, the amount of moles of CO_2 in the MPA solution was obtained by subtracting the moles of CO_2 in the vapor phase from the total moles of CO_2 fed to the cell:

$$n^{l}_{co_2} = n_{co_2} - n^{g}_{co_2} \tag{2}$$

From the moles of CO_2 dissolved in the aqueous MPA solution and the known moles of MPA in the cell, the CO_2 loading in the liquid phase was defined as

$$\alpha_{co_2} = \frac{n^{l}_{co_2}}{n_{amine}} \tag{3}$$

where n_{amine} is the mole number of MPA in the liquid phase. The uncertainty of α is determined from the uncertainty in temperature, pressure, and volume, which are ± 0.1 %, ± 0.5 %, and ± 0.5 %, respectively, to give the estimated uncertainty in α of ± 8 %; the estimated pressure uncertainty is ± 2 %.

Thermodynamic Model

The vapor−liquid equilibrium in the system with components MPA, CO_2, and H_2O is governed by the following reactions:

$$RNH_3{}^{+} \overset{k_1}{\longleftrightarrow} H^{+} + RNH_2 \tag{4}$$

$$RNH_2 + CO_2 \overset{k_2}{\longleftrightarrow} RNHCOO^{-} + H^{+} \tag{5}$$

$$H_2O + CO_2 \overset{k_3}{\longleftrightarrow} H^{+} + HCO_3{}^{-} \tag{6}$$

$$H_2O \overset{k_4}{\longleftrightarrow} H^{+} + OH^{-} \tag{7}$$

$$HCO_3{}^{-} \overset{k_5}{\longleftrightarrow} H^{+} + CO_3{}^{2-} \tag{8}$$

In the above reactions RNH_2 represents MPA. For the convenience of calculation, the second hydrolysis reaction of

Table 1. Solubility of CO_2 in 2.0 mol·dm^{-3} MPA Solution

$T = 313.15$ K		$T = 343.15$ K		$T = 373.15$ K		$T = 393.15$ K	
	P_{co_2}		P_{co_2}		P_{co_2}		P_{co_2}
α_{co_2}	kPa	α_{co_2}	kPa	α_{co_2}	kPa	α_{co_2}	kPa
0.566 ± 0.05	4.5 ± 0.5	0.454 ± 0.04	6.7 ± 0.5	0.305 ± 0.02	3.9 ± 0.5	0.190 ± 0.02	9.2 ± 0.5
0.669 ± 0.05	20.6 ± 0.5	0.568 ± 0.05	21.6 ± 0.5	0.450 ± 0.04	31.9 ± 0.6	0.346 ± 0.03	48.0 ± 1
0.731 ± 0.06	45.1 ± 1	0.683 ± 0.05	110.0 ± 2	0.555 ± 0.04	126.6 ± 3	0.453 ± 0.04	148.5 ± 3
0.805 ± 0.06	100.7 ± 2	0.764 ± 0.06	259.7 ± 5	0.610 ± 0.05	243.6 ± 5	0.522 ± 0.04	287.3 ± 6
0.849 ± 0.07	153.9 ± 3	0.794 ± 0.06	340.8 ± 7	0.643 ± 0.05	346.1 ± 7	0.565 ± 0.05	421.0 ± 8
0.876 ± 0.07	202.0 ± 4			0.672 ± 0.05	465.3 ± 9	0.594 ± 0.05	552.5 ± 11
0.947 ± 0.08	377.6 ± 8			0.704 ± 0.06	618.0 ± 12	618.0 ± 0.05	
0.987 ± 0.08	525.6 ± 11						
1.024 ± 0.08	695.0 ± 14						

Table 2. Solubility of CO_2 in 4.0 mol·dm^{-3} MPA Solution

313.15 K		343.15 K		373.15 K		393.15 K	
	P_{co_2}		P_{co_2}		P_{co_2}		P_{co_2}
α_{co_2}	kPa	α_{co_2}	kPa	α_{co_2}	kPa	α_{co_2}	kPa
0.527 ± 0.04	7.8 ± 0.5	0.392 ± 0.03	2.5 ± 0.5	0.252 ± 0.02	3.2 ± 0.5	0.213 ± 0.02	15.0 ± 0.5
0.580 ± 0.05	18.2 ± 0.5	0.415 ± 0.03	7.3 ± 0.5	0.368 ± 0.03	15.3 ± 0.5	0.292 ± 0.02	33.8 ± 1
0.675 ± 0.05	76.5 ± 2	0.486 ± 0.04	10.6 ± 0.5	0.491 ± 0.04	107.7 ± 2	0.368 ± 0.03	70.8 ± 1
0.752 ± 0.06	191.2 ± 4	0.512 ± 0.04	21.4 ± 0.5	0.537 ± 0.04	235.5 ± 5	0.430 ± 0.03	153.8 ± 3
0.793 ± 0.06	293.5 ± 6	0.547 ± 0.04	34.9 ± 0.7	0.567 ± 0.05	369.3 ± 7	0.464 ± 0.04	225.8 ± 5
0.828 ± 0.07	414.8 ± 8	0.581 ± 0.05	67.8 ± 1	0.589 ± 0.05	523.9 ± 10	0.496 ± 0.04	338.8 ± 7
0.856 ± 0.07	548.6 ± 11	0.600 ± 0.05	122.6 ± 2	0.608 ± 0.05	661.5 ± 13	0.510 ± 0.04	428.8 ± 9
0.876 ± 0.07	654.8 ± 13	0.645 ± 0.05	182.4 ± 4				
		0.648 ± 0.05	261.5 ± 5				
		0.686 ± 0.05	295.7 ± 6				
		0.714 ± 0.06	393.8 ± 8				
		0.739 ± 0.06	491.8 ± 10				
		0.771 ± 0.06	645.4 ± 13				

Table 3. Solubility of CO_2 in 5.0 mol·dm^{-3} MPA Solution

$T = 313.15$ K		$T = 343.15$ K		$T = 373.15$ K		$T = 393.15$ K	
α_{co_2}	P_{co_2} kPa	α_{co_2}	P_{co_2} kPa	α_{co_2}	P_{co_2} kPa	α_{co_2}	P_{co_2} kPa
0.528 ± 0.04	5.8 ± 0.5	0.417 ± 0.03	3.7 ± 0.5	0.325 ± 0.03	8.6 ± 0.5	0.200 ± 0.02	13.5 ± 0.5
0.551 ± 0.04	11.3 ± 0.5	0.467 ± 0.04	7.3 ± 0.5	0.408 ± 0.03	26.1 ± 0.5	0.281 ± 0.02	31.6 ± 0.6
0.589 ± 0.05	26.5 ± 0.5	0.514 ± 0.04	18.5 ± 0.5	0.460 ± 0.04	65.1 ± 1	0.348 ± 0.03	63.7 ± 1
0.614 ± 0.05	46.4 ± 1	0.505 ± 0.04	24.9 ± 0.5	0.509 ± 0.04	163.1 ± 3	0.406 ± 0.03	115.3 ± 2
0.636 ± 0.05	64.4 ± 1	0.560 ± 0.04	52.4 ± 1	0.541 ± 0.04	303.1 ± 6	0.437 ± 0.03	176.2 ± 4
0.654 ± 0.05	87.4 ± 2	0.586 ± 0.04	119.2 ± 2	0.572 ± 0.05	508.3 ± 10	0.475 ± 0.04	304.4 ± 6
0.683 ± 0.05	133.6 ± 3	0.606 ± 0.05	123.4 ± 2	0.588 ± 0.05	655.3 ± 13	0.503 ± 0.04	443.6 ± 9
0.696 ± 0.06	157.2 ± 3	0.655 ± 0.05	237.8 ± 5				
0.727 ± 0.06	230.5 ± 5	0.645 ± 0.05	290.8 ± 6				
0.746 ± 0.06	289.5 ± 6	0.685 ± 0.05	461.0 ± 9				
0.776 ± 0.06	403.5 ± 8	0.712 ± 0.06	598.0 ± 12				
0.782 ± 0.06	427.7 ± 9						
0.813 ± 0.07	582.7 ± 12						
0.834 ± 0.07	704.9 ± 14						

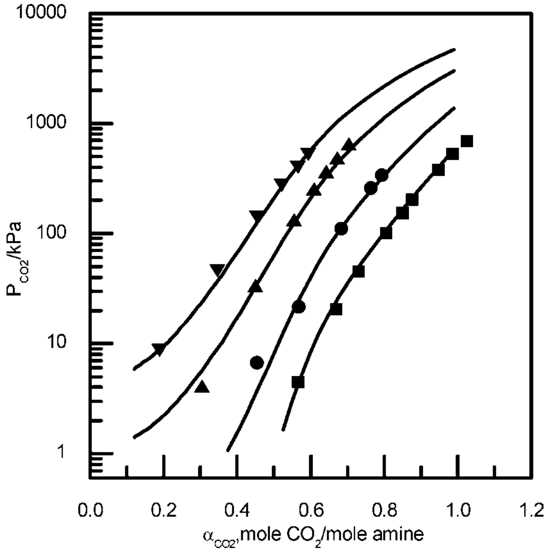

Figure 3. Solubility of CO_2 in 2 mol·dm^{-3} MPA solutions. The points show the experimental data. The lines show the KE model predicted results. ■, $T = 313.15$ K; ●, $T = 343.15$ K; ▲, $T = 373.15$ K; ▼, $T = 393.15$ K.

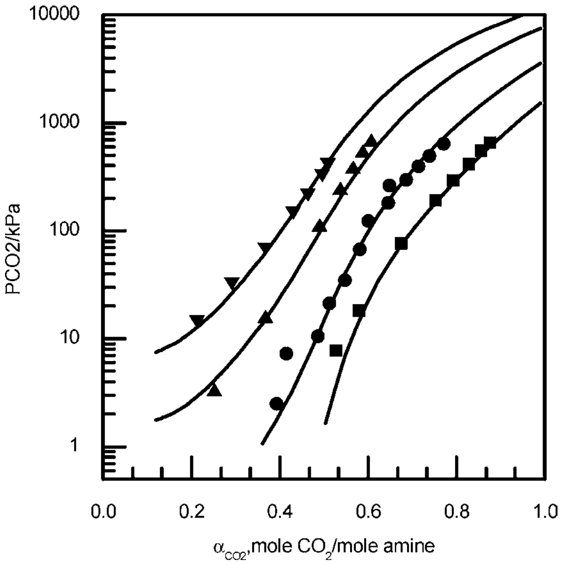

Figure 4. Solubility of CO_2 in 4 mol·dm^{-3} MPA solutions. The points show the experimental data. The lines show the KE model predicted results. ■, $T = 313.15$ K; ●, $T = 343.15$ K; ▲, $T = 373.15$ K; ▼, $T = 393.15$ K.

H_2CO_3 as eq 8 is not considered in the calculation as suggested by Jostein et al.[13] Then the corresponding expressions for the apparent equilibrium constants are:

$$k_1 = [H^+][RNH_2]/[RNH_3^{\ +}] \tag{9}$$

$$k_2 = [RNHCOO^-][H^+]/[CO_2][RNH_2] \tag{10}$$

$$k_3 = [H^+][HCO_3^{\ -}]/[CO_2][H_2O] \tag{11}$$

$$k_4 = [H^+][OH^-] \tag{12}$$

To satisfy the material balance and charge balance in the system, the following equations can be formed:

$$[MPA]^\circ = [RNH_3^{\ +}] + [RNH_2] + [RNHCOO^-] \tag{13}$$

$$[MPA]^\circ\alpha_{co_2} = [CO_2] + [HCO_3^{\ -}] + [RNHCOO^-] \tag{14}$$

$$[RNH_3^{\ +}] + [H^+] = [HCO_3^{\ -}] + [OH^-] + [RNHCOO^-] \tag{15}$$

For an MPA solution with a known total concentration $[MPA]^\circ$ and α_{co_2} in the solution, concentrations of seven

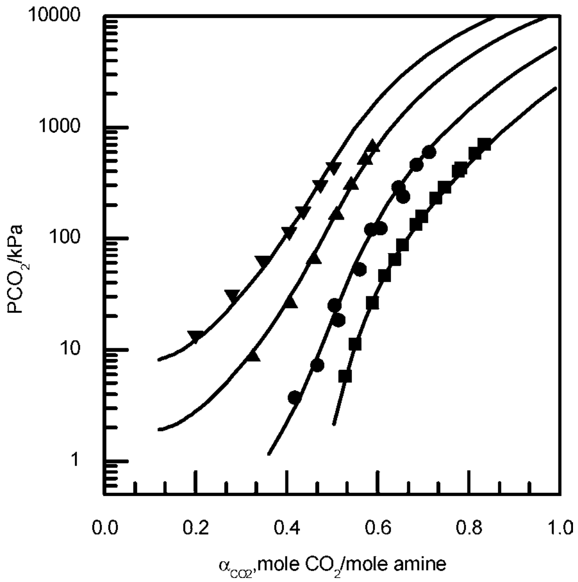

Figure 5. Solubility of CO_2 in 5 mol·dm^{-3} MPA solutions. The points show the experimental data. The lines show the KE model predicted results. ■, $T = 313.15$ K; ●, $T = 343.15$ K; ▲, $T = 373.15$ K; ▼, $T = 393.15$ K.

Table 4. Fitted Values of Parameters in the KE Model for the CO_2 + MPA + H_2O System

equilibrium constants	a_1	a_2	a_3	b_1	b_2	b_3	b_4
k_1	−6.5270	−6795.5	59797.0	−0.48360	0.54556	−0.042698	−0.44447
k_2	−21.729	3282.2	75274.0	−0.45030	0.19489	−0.043821	−0.5996

components $[RNH_2]$, $[RNH_3^+]$, $[RNHCOO^-]$, $[CO_2]$, $[HCO_3^-]$, $[H^+]$, and $[OH^-]$ can be solved with seven eqs 9 to 15. Then the partial pressure of CO_2 is related to the CO_2 concentration physically dissolved in the solvent by means of Henry's law:

$$p_{co_2} = H_{co_2}[CO_2] \qquad (16)$$

in which H_{co_2} is the Henry's constant for CO_2 in pure water.

Kent and Eisenberg[14] developed a simple thermodynamic model (KE) for representing equilibrium data in amine + CO_2 + H_2O using apparent equilibrium constants. In the studies of Li and Shen and Hu and Chakma[15,16] the apparent equilibrium constants of the main amine controlling reaction, k_1 and k_2, are correlated as a function of temperature, CO_2 loading, and amine concentration, with parameters determined by fitting the experimental CO_2 solubility data. The nonidealities of the solution have been lumped into k_1 and k_2. The expressions of equilibrium constants k_3, k_4, and H_{co_2} are obtained from Edwards et al.[17] The equations for equilibrium constants used are as follows:

$$k_1 = \exp(a_1 + a_2T^{-1} + a_3T^{-2} + b_1\alpha_{co_2} + b_2\alpha_{co_2}^{-1} + b_3\alpha_{co_2}^{-2} + b_4 \ln m) \qquad (25)$$

$$k_2 = \exp(a_1' + a_2'T^{-1} + a_3'T^{-2} + b_1'\alpha_{co_2} + b_2'\alpha_{co_2}^{-1} + b_3'\alpha_{co_2}^{-2} + b_4' \ln m) \qquad (26)$$

$$k_3 = \exp(231.465 - 12092 \cdot 10^2 T^{-1} - 36.7816 \log(T)) \qquad (27)$$

$$k_4 = \exp(132.899 - 13445.9T^{-1} - 22.4773 \log(T)) \qquad (28)$$

$$H_{co_2} = \exp(110.035 - 6789.04T^{-1} - 11.452 \log(T) - 0.01045T) \qquad (30)$$

In the k_1 and k_2 equations, a_1, a_2, a_3, b_1, b_2, b_3, and b_4 are KE parameters obtained by regression with the experimental solubility data.

Results and Discussion

To verify the uncertainty of the experiment, the solubility of CO_2 in 2.5 mol·dm^{-3} MEA solution was determined, and the results were compared with those reported by Lee et al.[18] in Figure 2 at T = 313.15 K. Tables 1, 2, and 3 list the partial pressure of CO_2 above CO_2-loaded MPA solutions, and the results are also shown in Figures 3, 4, and 5 along with the KE models with parameters obtained in this work that are listed in Table 4 for the (MPA + CO_2 + H_2O).

Low concentrations of MPA have a higher CO_2 loading at the same temperature. Comparing the data of this work with that of Lee et al.,[18] it is clear that with the same concentration of amines, MPA has higher CO_2 loading than MEA at T = 313.15 K, while at T = 398 K this difference is smaller. Meanwhile, it is also found in our experiment that MPA is more stable than MEA. The normal boiling point of MPA is between (184 and 187) °C, while that of MEA is 170 °C. There is less degradation and loss of MPA during absorption and desorption operations.

Compared with the work of Lee et al.[19] for aqueous diethanolamine (DEA) solution, the solubility of CO_2 in 2.0 mol·dm^{-3} aqueous MPA solution is slightly higher at T = 313.15 K but is higher at T = 398 K. Considering the difference in molecular weights of two amines, the DEA solution is still slightly better than MPA solution.

Table 5. Correlation Deviations of Solubility in the CO_2 + MPA + H_2O System by the KE Model

C(MPA) mol·dm^{-3}	T K	no. of points	AAD[a] %
2.0	313.15	8	10.5
2.0	343.15	5	20.2
2.0	373.15	7	19.8
2.0	393.15	6	12.5
4.0	313.15	7	14.7
4.0	343.15	13	20.4
4.0	373.15	7	20.3
4.0	393.15	7	21.8
5.0	313.15	14	11.5
5.0	343.15	11	20.4
5.0	373.15	7	20.8
5.0	393.15	7	19.7

[a] Average absolute deviation (AAD) % = $\sum_n[|p_{cal} - p_{exp}| \cdot 100/p_{exp}]/n$.

The average deviations for solubility predicted from the KE model for MPA + CO_2 + H_2O are listed in Table 5. The minimum deviation is 10.5 % at T = 313.15 K on 2.0 mol·dm^{-3}, and the maximum deviation is 21.8 % at T = 393.15 K on 4.0 mol·dm^{-3}. In all of the cases, the KE model correlates the solubility of CO_2 in aqueous MPA.

Conclusion

The solubility of CO_2 in the aqueous MPA solution has been measured at temperatures from (313.15 to 393.15) K. The Kent−Eisenberg model has been used to correlate the solubility of CO_2 in the MPA solutions, and results show that the KE model performs well in the CO_2 solubility correlation. Compared with other alkanolamine solutions for CO_2 absorption, the aqueous MPA solution is better than the aqueous MEA solution but still slightly worse than the aqueous DEA solution.

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