Solubilities of Carbon Dioxide in Water + Monoethanolamine + 2-Amino-2-methyl-1-propanol

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The solubilities of carbon dioxide in water (1) + monoethanolamine (2) + 2-amino-2-methyl-1-propanol (3) have been measured at 40, 60, 80, and 100 °C and at partial pressures of carbon dioxide ranging from 1.0 to 200 kPa. The ternary mixtures studied were $w_2 = 0$, $w_3 = 0.3$; $w_2 = 0.06$, $w_3 = 0.24$; $w_2 = 0.12$, $w_3 = 0.18$; $w_2 = 0.18$, $w_3 = 0.12$; and $w_2 = 0.24$, $w_3 = 0.06$ where w is the mass fraction. The modified Kent and Eisenberg model has been used to represent the solubility of CO_2 in the ternary solutions. The model reasonably reproduces the equilibrium partial pressure of CO_2 above the ternary solutions, not only over a temperature range from 40 to 100 °C, but also for various concentrations of components in the ternary solutions.

Introduction

Alkanolamine aqueous solutions are widely used in gas treating processes to remove acid gases, such as CO_2 and H_2S, from natural, refinery, and synthesis gas streams. Industrially important alkanolamines are monoethanolamine (MEA), diethanolamine (DEA), di-2-propanolamine (DIPA), and N-methyldiethanolamine (MDEA). Aqueous MEA solutions have been widely used due to their high reactivity, low solvent cost, ease of reclamation, and low absorption of hydrocarbons (1). When the primary (or secondary) amine reacts with CO_2, stable carbamates are usually formed (2). The maximum CO_2 loading is limited by stoichiometry to 0.5 mol of CO_2/mol of amine when carbamate formation is the only reaction. At high CO_2 partial pressures, however, carbamates may hydrolyze and generate free amines which can react with additional CO_2; thus, the CO_2 loading of MEA may exceed 0.5. For a gas stream that contains both CO_2 and H_2S, the aqueous MDEA solution is found to be an appropriate solution for the selective removal of H_2S from the gas stream (3, 4). Advantages of the use of MDEA solutions include their high loading capacity (about 1.0 mol of CO_2/mol of amine) and low enthalpy of reaction with the acid gases (5). The lower enthalpy of reaction leads to lower energy requirements for regeneration. However, a slow reaction rate of CO_2 with tertiary amines limits the use of MDEA solutions (2).

Recently the use of blended amines, a solution of two or more amines in varying compositions, shows considerable improvement in absorption and savings in energy requirements (6). Blended amines combine the absorption characteristics of the constituent amines such as a higher loading capacity and faster reaction rates. Blends of primary and tertiary amines, such as mixtures of MEA and MDEA, have been suggested for CO_2 removal (6).

Sterically hindered alkanolamines have also been proposed as commercially attractive solvents for removal of acid gases from gaseous streams (2). An example of the sterically alkanolamines is 2-amino-2-methyl-1-propanol (AMP), which is the hindered form of MEA. Due to the bulkiness of the group attached to a tertiary carbon atom of AMP, the formation of carbamates is inhibited when AMP reacts with CO_2 (2, 7). Thus, the reaction of CO_2 with AMP is mainly the formation of the bicarbonate ion as follows:

$$CO_2 + RR'NH_2 + H_2O \leftrightarrow HCO_3^- + RR'NH_3^+ \quad (1)$$
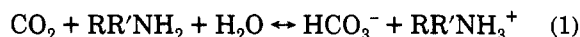
where $RR'NH_2$ denotes AMP. Therefore, the CO_2 loading of AMP can approach 1.0 mol of CO_2/mol of amine. The kinetics of the reaction of CO_2 with AMP have been shown to be first order with respect to both CO_2 and AMP, and the rate constant has the value of 1270 $m^3/(kmol\cdot s)$ at 40 °C (7). Compared to MDEA, AMP has the same high CO_2 loading capacity (about 1.0 mol of CO_2/mol of amine) but has a higher reaction rate constant for the reaction with CO_2. The reaction rate constant of CO_2 with MDEA is 3.5 $m^3/(kmol\cdot s)$ at 25 °C (8). Therefore, H_2O + MEA + AMP may be an attractive new solvent in addition to H_2O + MEA + MDEA for the acid gas treating process.

Some solubilities of CO_2 and H_2S in blended alkanolamine aqueous solutions are available in the literature. The solubilities of CO_2 and H_2S in H_2O + MEA + MDEA have been studied (9–12). However, the solubility of acid gases in aqueous mixtures of MEA with AMP has not been reported in the literature. Therefore, it is the purpose of this research to measure the solubility of CO_2 in aqueous MEA + AMP solutions.

Experimental Section

Alkanolamine aqueous solutions were prepared from distilled water. MEA is Riedel-de Haën reagent grade with a 99 mol % purity and AMP is Riedel-de Haën reagent grade with a 98 mol % purity. The solubility of CO_2 in aqueous MEA + AMP solutions was measured in a 1.0-L stainless steel vapor–recirculation equilibrium cell. Another stainless steel sample cylinder (0.3 L) is connected to the equilibrium cell to increase the volume of the vapor phase. Two cylinders were mounted vertically in a thermostated oil bath. The temperature of the system was controlled by an EX-810B basic immersion circulator, made by NESLAB Instruments Inc., to a stability within ±0.1 °C of the set-point temperature. The pressure of the system is indicated by an Ashcroft test gauge with the scale reading of 0.2 psi. For the low partial pressures of CO_2, nitrogen was introduced and mixed with CO_2, and the partial pressure of CO_2 was determined by on-line chromatography. The solubility (loading capacity) of CO_2 in amine solutions was determined by the titration method. When both the system pressure and the gas concentration, determined by gas chromatography, do not vary for 2 h, the equilibrium is assumed to have been reached;

Table 1. Partial Pressure P of CO_2 in $H_2O(1)$ + MEA(2) + AMP(3)[a]

$t = 40\ °C$		$t = 60\ °C$		$t = 80\ °C$		$t = 100\ °C$	
P/kPa	α	P/kPa	α	P/kPa	α	P/kPa	α
$\omega_2 = 0.0$, $\omega_3 = 0.3$							
1.05	0.396	1.43	0.200	1.68	0.102	1.12	0.039
3.14	0.497	1.67	0.230	4.83	0.163	3.62	0.091
5.54	0.566	4.27	0.331	9.82	0.248	5.79	0.104
11.5	0.647	9.84	0.451	29.6	0.382	19.7	0.188
17.2	0.666	11.3	0.452	79.5	0.518	25.0	0.200
21.2	0.749	34.2	0.582	146	0.617	71.2	0.343
74.2	0.772	67.9	0.656				
197	0.867	133	0.721				
$\omega_2 = 0.06$, $\omega_3 = 0.24$							
1.05	0.387	1.06	0.236	1.36	0.169	1.57	0.105
3.08	0.442	1.84	0.279	4.44	0.239	6.00	0.160
5.36	0.484	5.23	0.349	8.47	0.295	8.69	0.196
6.07	0.521	7.21	0.387	21.4	0.348	26.5	0.263
12.1	0.567	15.3	0.429	38.3	0.404	57.7	0.327
17.0	0.568	16.4	0.452	96.7	0.499	98.8	0.380
32.7	0.626	39.4	0.543				
94.5	0.675	47.4	0.537				
178	0.755	166	0.630				
$\omega_2 = 0.12$, $\omega_3 = 0.18$							
1.14	0.384	1.14	0.296	0.90	0.181	1.90	0.133
4.39	0.433	3.14	0.358	2.70	0.242	2.17	0.143
27.4	0.511	8.93	0.398	8.90	0.296	3.86	0.182
62.0	0.565	38.9	0.486	32.3	0.385	11.5	0.251
105	0.583	104	0.546	78	0.440	40.9	0.316
175	0.612	171	0.577	161	0.512	101	0.366
$\omega_2 = 0.18$, $\omega_3 = 0.12$							
0.86	0.379	1.15	0.352	1.05	0.236	1.15	0.151
2.18	0.393	2.18	0.359	2.94	0.254	3.16	0.199
5.47	0.413	4.65	0.390	6.05	0.319	7.75	0.268
8.25	0.457	6.64	0.392	15.1	0.360	24.8	0.323
19.4	0.458	17.4	0.436	64.8	0.443	52.2	0.359
45.7	0.498	46.1	0.465	155	0.492	96.1	0.407
108	0.546	90.7	0.486			103	0.400
199	0.592	181	0.521				
$\omega_2 = 0.24$, $\omega_3 = 0.06$							
1.00	0.403	0.89	0.333	0.95	0.237	0.74	0.121
4.20	0.433	1.98	0.348	2.00	0.282	1.46	0.157
14.2	0.460	6.27	0.391	4.35	0.314	3.25	0.208
39.2	0.482	19.5	0.408	20.9	0.376	9.07	0.260
78.8	0.510	52.1	0.448	81.8	0.434	35.3	0.328
172	0.553	97.7	0.477	165	0.466	105	0.383
		180	0.503				

[a] CO_2 loading α = (mol of CO_2/total mol of amine), ω = mass fraction.

it usually takes 6–8 h for the system to reach equilibrium. At equilibrium, a liquid sample was withdrawn from the equilibrium cell into a vessel containing excess 1.0 M NaOH solution, thus converting free dissolved CO_2 into the nonvolatile ionic species. An excess amount of $BaCl_2$ solution is then added to the solution. The solution is shaken well to permit all absorbed (physically and chemically) CO_2 to precipitate the carbonate as $BaCO_3$. The excess NaOH was titrated with HCl solution using phenolphthalein as the indicator. Using methyl orange as the indicator, a solution of HCl is then added. The amount of HCl added can be used to calculate the solubility of CO_2 in terms of the moles of CO_2 per mole of amine. The experimental error in the solution CO_2 loading (mol of CO_2/mol of total amine) is estimated to be ±2–3%. The materials, vapor–liquid equilibrium apparatus, and methods of analysis are essentially the same as those used in our previous work on the solubility of acid gases in H_2O + MEA + MDEA (10–12).
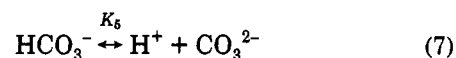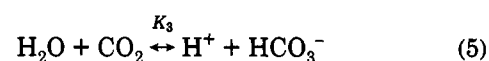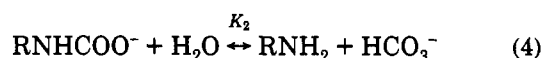
Results and Discussion

For the measurements of solubility of CO_2 in H_2O (1) + MEA (2) + AMP (3), the systems studied are $w_2 = 0$, $w_3 =$ 0.3; $w_2 = 0.06$; $w_3 = 0.24$; $w_2 = 0.12$, $w_3 = 0.18$; $w_2 = 0.18$, $w_3 = 0.12$; and $w_2 = 0.24$, $w_3 = 0.06$ where w is the mass fraction. The CO_2 solubility data in aqueous MEA + AMP solutions at 40, 60, 80, and 100 °C are presented in Table 1.

Owing to its simplicity, the model of Kent of Eisenberg (13) has often been used to represent the solubility of acid gases in aqueous alkanolamine systems (14–18). With the exception of the equilibrium constants for reactions involving amines, literature values for all the ionization constants and Henry's law constants were used in the model of Kent and Eisenberg (13). Jou et al. (15) pointed out that the equilibrium constants involving amines are essentially functions of temperature, acid gas loading, and amine concentration, rather than a function of temperature alone, as used in the model of Kent of Eisenberg. On the basis of the model of Kent and Eisenberg, Hu and Chakma (17, 18) proposed a modified expression for the equilibrium constants, governing the main amine reactions, as functions not only of temperature but also of acid gas partial pressure and amine concentration. In a similar manner, Li and Shen (19) allowed the chemical equilibrium constants involving alkanolamines to be functions of temperature, amine concentration, and carbon dioxide loading for the calculation of CO_2 solubilities in H_2O + MEA + MDEA. In this study, the method of Li and Shen (19) will be applied to correlate the solubility data of CO_2 in aqueous MEA + AMP solutions. The constants in the model will be determined by fitting to the CO_2 solubility data in H_2O + MEA + AMP.

Equations describing the CO_2 + H_2O + MEA + AMP in the mixture equilibrium, with RNH_2 and $RR'NH_2$ representing MEA and AMP, respectively, are as follows:

$$RNH_3^+ \overset{K_{11}}{\leftrightarrow} H^+ + RNH_2 \tag{2}$$

$$RR'NH_3^+ \overset{K_{12}}{\leftrightarrow} H^+ + RR'NH_2 \tag{3}$$

$$RNHCOO^- + H_2O \overset{K_2}{\leftrightarrow} RNH_2 + HCO_3^- \tag{4}$$

$$H_2O + CO_2 \overset{K_3}{\leftrightarrow} H^+ + HCO_3^- \tag{5}$$

$$H_2O \overset{K_4}{\leftrightarrow} H^+ + OH^- \tag{6}$$

$$HCO_3^- \overset{K_5}{\leftrightarrow} H^+ + CO_3^{2-} \tag{7}$$

Note that the carbamate ion, $RNHCOO^-$, is formed only by the reaction of CO_2 with MEA and not by CO_2 with AMP. The expressions for the apparent equilibrium constants are

$$K_{11} = [H^+][RNH_2]/[RNH_3^+] \tag{8}$$

$$K_{12} = [H^+][RR'NH_2]/[RR'NH_3^+] \tag{9}$$

$$K_2 = [RNH_2][HCO_3^-]/[RNHCOO^-] \tag{10}$$

$$K_3 = [H^+][HCO_3^-]/[CO_2] \tag{11}$$

$$K_4 = [H^+][OH^-] \tag{12}$$

$$K_5 = [H^+][CO_3^{2-}]/[HCO_3^-] \tag{13}$$

Henry's law relates the CO_2 partial pressure to the concentration of the physically dissolved CO_2 in the solvent

according to

$$P_{CO_2} = H_{CO_2}[CO_2] \tag{14}$$

The following mass balance equations for the reacting species hold:

$$m_1 = [RNH_2] + [RNH_3^+] + [RNHCOO^-] \tag{15}$$

$$m_2 = [RR'NH_2] + [RR'NH_3^+] \tag{16}$$

$$(m_1 + m_2)\alpha = [CO_2] + [CO_3^{2-}] + [HCO_3^-] + [RNHCOO^-] \tag{17}$$

$$[RNH_3^+] + [RR'NH_3^+] + [H^+] = [OH^-] + [RNHCOO^-] + [HCO_3^-] + 2[CO_3^{2-}] \tag{18}$$

where m_1 and m_2 are the molarity of MEA and AMP, respectively, and α is the loading capacity of CO_2 of the solvent.

In this study, literature values of the equilibrium constants for eqs 5–7 and the Henry's law constant for CO_2 are used in the calculation, as in the model of Kent and Eisenberg (13). These equations are given as follows:

$$K_3/(kmol\ m^{-3}) = \exp[-241.828 + 29.8253 \times 10^4/(T/K) - 1.48528 \times 10^8/(T/K)^2 + 0.332647 \times 10^{11}/(T/K)^3 - 0.282393 \times 10^{13}/(T/K)^4] \tag{19}$$

$$K_4/(kmol\ m^{-3})^2 = \exp[-39.5554 - 9.879 \times 10^4/(T/K) + 0.568827 \times 10^8/(T/K)^2 - 0.146451 \times 10^{11}/(T/K)^3 + 0.136145 \times 10^{13}/(T/K)^4] \tag{20}$$

$$K_5/(kmol\ m^{-3}) = \exp[-294.74 + 36.4385 \times 10^4/(T/K) - 1.84157 \times 10^8/(T/K)^2 + 0.415792 \times 10^{11}/(T/K)^3 - 0.354291 \times 10^{13}/(T/K)^4] \tag{21}$$

$$H_{CO_2}/\{kPa\ (kmol\ m^{-3})^{-1}\} = \exp[20.2669 - 1.38306 \times 10^4/(T/K) + 0.06913 \times 10^8/(T/K)^2 - 0.015589 \times 10^{11}/(T/K)^3 + 0.01200 \times 10^{13}/(T/K)^4] \tag{22}$$

The equilibrium constants which govern the main amine reactions, i.e., eqs 2–4, are assumed to be functions of temperature, amine concentration, and CO_2 loading. Using the least-squares fit to the equilibrium partial pressure data of CO_2 in aqueous MEA + AMP solutions, the equilibrium constants were determined as follows:

$$K_{11} = \exp[-33.118 + 777.64/(T/K) - 1.12718 \times 10^8/(T/K)^3 + 18.6673\alpha + 3.83728/\alpha - 0.231423/\alpha^2 - 0.711864\ \ln\{m_1(kmol\ m^{-3})\}] \tag{23}$$

$$K_{12} = \exp[-20.6346 + 1027.78/(T/K) - 2.7166 \times 10^8/(T/K)^3 + 5.55063\alpha + 0.885938/\alpha - 0.048573/\alpha^2 - 0.188085\ \ln\{m_2/(kmol\ m^{-3})\}] \tag{24}$$

$$K_2 = \exp[1.67709 - 2961.26/(T/K) - 1.07644 \times 10^8/(T/K)^3 + 18.7857\alpha + 0.992151/\alpha - 0.056525/\alpha^2 - 1.5674\ \ln\{m_1/(kmol\ m^{-3})\}] \tag{25}$$

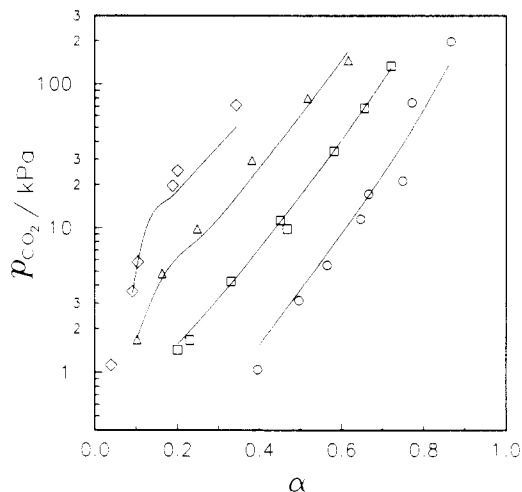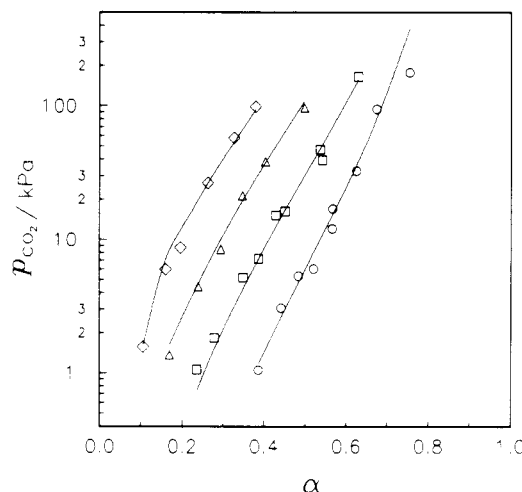where α is the ratio of the moles of CO_2 to the total moles of amine and K is in kmol m^{-3}.

Figure 1. Partial pressure P of CO_2 in $w_2 = 0$, $w_3 = 0.3$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α (mol of CO_2/total mol of amine): O, 40 °C; □, 60 °C; △, 80 °C; ◇, 100 °C. Lines are calculated by the method of Kent and Eisenberg.
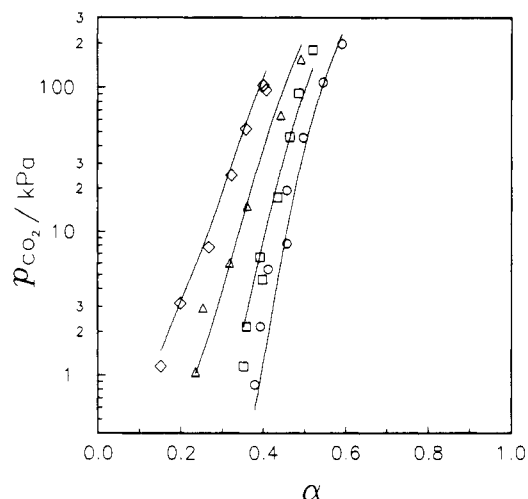

Figure 2. Partial pressure P of CO_2 in $w_2 = 0.06$, $w_3 = 0.24$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α (mol of CO_2/total mol of amine): O, 40 °C; □, 60 °C; △, 80 °C; ◇, 100 °C. Lines are calculated by the method of Kent and Eisenberg.
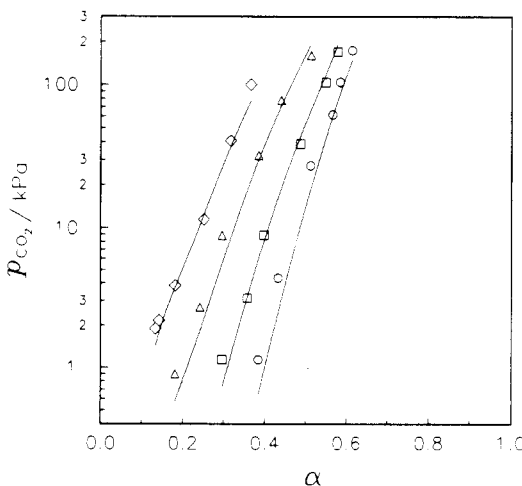
Figure 3. Partial pressure P of CO_2 in $w_2 = 0.12$, $w_3 = 0.18$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α (mol of CO_2/total mol of amine): O, 40 °C; □, 60 °C; △, 80 °C; ◇, 100 °C. Lines are calculated by the method of Kent and Eisenberg.
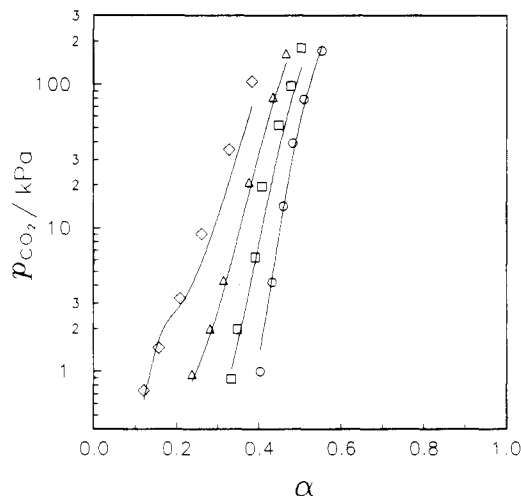
Figure 4. Partial pressure P of CO_2 in $w_2 = 0.18$, $w_3 = 0.12$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α (mol of CO_2/total mol of amine): O, 40 °C; □, 60 °C; △, 80 °C; ◇, 100 °C. Lines are calculated by the method of Kent and Eisenberg.

Figure 6. Partial pressure P of CO_2 in H_2O (1) + MEA (2) + AMP (3) at 60 °C for various loadings α (mol of CO_2/total mol of amine): O, $w_2 = 0$, $w_3 = 0.3$; □, $w_2 = 0.06$, $w_3 = 0.24$; △, $w_2 = 0.12$, $w_3 = 0.18$, ◇, $w_2 = 0.18$, $w_3 = 0.12$; ▽, $w_2 = 0.24$, $w_3 = 0.06$. Lines are calculated by the method of Kent and Eisenberg.

Figure 5. Partial pressure P of CO_2 in $w_2 = 0.24$, $w_3 = 0.06$ H_2O (1) + MEA (2) + AMP (3) at various temperatures for various loadings α (mol of CO_2/total mol of amine): O, 40 °C; □, 60 °C; △, 80 °C; ◇, 100 °C. Lines are calculated by the method of Kent and Eisenberg.

Figures 1–5 show comparisons between calculated and experimental results for the five blended amine aqueous systems. The calculated partial pressures of CO_2 over aqueous MEA + AMP systems are in good agreement with experimental data for temperatures ranging from 40 to 100 °C and for systems of various concentrations, as shown in Figures 1–5. The model reasonably reproduces the equilibrium partial pressure of CO_2 over aqueous MEA + AMP systems. To investigate the effects of amine concentrations on CO_2 solubility, plots of CO_2 solubilities in H_2O + MEA + AMP at 60 °C for solutions with different concentrations are shown in Figure 6. As can be seen in Figure 6, CO_2 solubilities vary systematically from $w_2 = 0$, $w_3 = 0.3$; $w_2 = 0.06$, $w_3 = 0.24$; $w_2 = 0.12$, $w_3 = 0.18$; $w_2 = 0.18$, $w_3 = 0.12$; to $w_2 = 0.24$, $w_3 = 0.06$. The solubilities of CO_2 in higher AMP mass fraction solutions are higher than those of solutions with a high MEA mass fraction. To test the applicability of the model, calculations of the solubility for data not included in the database have also been performed. Figures 7 and 8 give comparisons between the results of calculated and experimental CO_2 solubilities in 2.0 and 3.0 M (kmol m^{-3}) AMP aqueous solutions for temperatures ranging from 20 to 80 °C (data of Tonti-

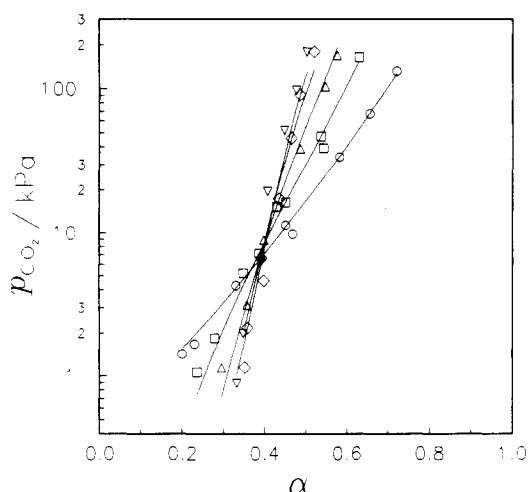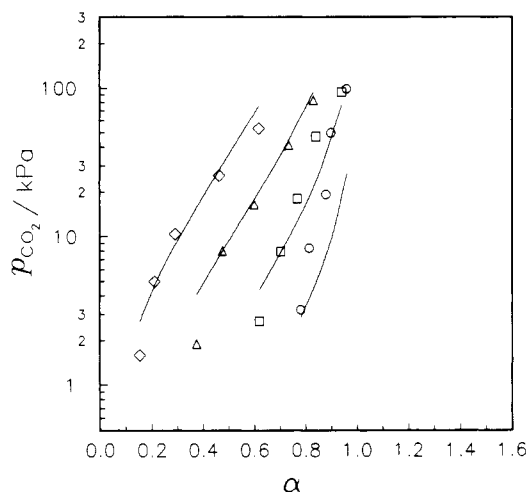**Figure 7.** Comparison of calculated and experimental results for the solubility of CO_2 in 2.0 kmol m^{-3} AMP aqueous solution at 20–80 °C: O, 20 °C; □, 40 °C; △, 60 °C; ◇, 80 °C. Data were reported by Tontiwachwuthikui (20). Lines are calculated by the method of Kent and Eisenberg.

wachwuthikui et al. (20)). The model predicts the solubility of CO_2 in 2.0 and 3.0 kmol m^{-3} AMP aqueous solutions reasonably well except for temperatures below 40 °C (see Figures 7 and 8).

Conclusion

The solubilities of CO_2 in H_2O + MEA + AMP have been reported from 40 to 100 °C and at CO_2 partial pressures up to 200 kPa. The systems studied are $w_2 = 0$, $w_3 = 0.3$; $w_2 = 0.06$, $w_3 = 0.24$; $w_2 = 0.12$, $w_3 = 0.18$; $w_2 = 0.18$, $w_3 = 0.12$; and $w_2 = 0.24$, $w_3 = 0.06$ where w is the mass fraction. The model of Kent and Eisenberg for calculating the solubilities of acid gases in alkanolamine solutions has been extended to represent the solubility of CO_2 in aqueous mixtures of MEA with AMP. The equilibrium constants of chemical reactions involving amines are expressed as functions of temperature, amine concentration, and CO_2 loading. The model reasonably reproduces the equilibrium partial pressure of CO_2 over aqueous MEA + AMP solutions, not only over a temperature range of 40–100 °C, but also for systems of various concentrations. The model has also been tested for systems not
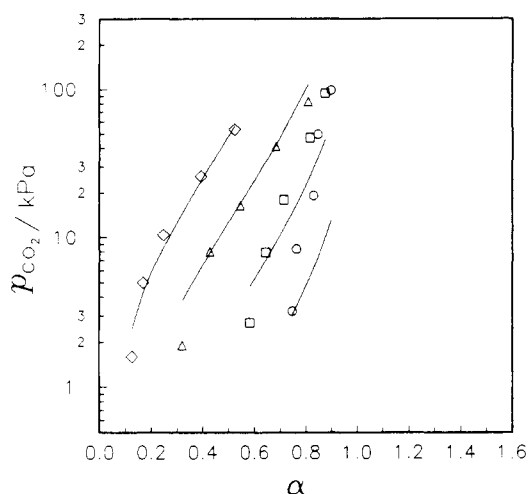
Figure 8. Comparison of calculated and experimental results for the solubility of CO_2 in 3.0 kmol m^{-3} AMP aqueous solution at 20–80 °C: O, 20 °C; □, 40 °C; △, 60 °C; ◇, 80 °C. Data were reported by Tontiwachwuthikui (20). Lines are calculated by the method of Kent and Eisenberg.

included in the database. Satisfactory results were obtained for the calculation of the solubility of CO_2 in AMP aqueous solutions.

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