Solubility of CO_2 in Water + Diethanolamine + 2-Amino-2-methyl-1-propanol

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This work presents new experimental results for the CO_2 solubility in aqueous blends of diethanolamine (DEA) + 2-amino-2-methyl-1-propanol (AMP) in the temperature range of (303 to 323) K and CO_2 partial pressure of (1 to 100) kPa. The mass fractions of the aqueous amine blends were 1.5 % DEA + 28.5 % AMP, 3 % DEA + 27 % AMP, 4.5 % DEA + 25.5 % AMP, and 6 % DEA + 24 % AMP. A rigorous thermodynamic model is developed to correlate and predict the vapor−liquid equilibrium (VLE) of CO_2 in aqueous blends of DEA and AMP. The modified Clegg−Pitzer equations have been used to develop a model for the quaternary system (CO_2 + AMP + DEA + H_2O) using the interaction parameters derived from the VLE data of the corresponding ternary systems (CO_2 + AMP + H_2O) and (CO_2 + DEA + H_2O). Simulated annealing has been used for parameter estimation and for the determination of equilibrium composition of various species present in the liquid phase. The model predictions are in good agreement with the experimental data of CO_2 solubility in DEA + AMP blends of this work as well as those reported in the literature.

Introduction

The removal of acid gas impurities, such as CO_2 and H_2S, from natural gas, refinery, and synthesis gas streams is a significant operation in gas processing. A wide variety of alkanolamines, such as monoethanolamine (MEA), diethanolamine (DEA), di-2-propanolamine (DIPA), and *N*-methyldiethanolamine (MDEA), has been used for industrial gas treating.[1] Primary and secondary amines react rapidly with CO_2 to form carbamate with a stoichiometric loading of 0.5 mol of CO_2/mol of amine. The CO_2 loading in the sterically hindered amine AMP approaches a value of 1.0 mol of CO_2/mol of amine as in the case of the tertiary amine MDEA, while the reaction rate constant for CO_2 in AMP is much higher than that for CO_2 in MDEA.[2] Since the sterically hindered amine AMP does not form a stable carbamate, bicarbonate and carbonate ions may be present in the solution in larger amounts. Because of relatively higher enthalpy of absorption associated with the formation of carbamate ions than in comparison to bicarbonate and carbonate ions, the regeneration energy cost for the loaded AMP solutions may be lower than that for the loaded primary and secondary amine solutions[2].

Because of the need to exploit poorer quality natural gas coupled with increasingly strict environmental regulations, highly economical and selective acid gas treating is more important today. As a result, there has been a resurgence of interest in improved alkanolamine solvents and particularly in aqueous blends of alkanolamines. A blended amine solvent, which is an aqueous blend of a primary or a secondary amine with a tertiary or a hindered amine, combines the higher equilibrium capacity of the tertiary or hindered amine with the higher reaction rate of the primary or secondary amine. Thus, the use of blended amine solvents, requiring lower circulation rates and lower regeneration energy, can bring about considerable improvement and great savings in individual gas-treating processes. The realization of such benefits depends on proper equipment design, which requires the knowledge of equilibrium solubility of the acid gases in amine blends. In addition, equilibrium solubility of the acid gases in aqueous alkanolamine solutions determines the minimum circulation rate of the solvent to treat a sour gas stream. It also determines the maximum concentration of acid gases that can be left in the regenerated solution in order to meet the product gas specifications.

(AMP + MEA + H_2O) and (AMP + DEA + H_2O) appear to be attractive new blended amine solvents in addition to (MDEA + MEA + H_2O) and (MDEA + DEA + H_2O) blends for the gas-treating processes. The lower vapor pressure of DEA and the fact that it is less corrosive than MEA makes the DEA-based blended amine solvents more attractive than MEA-based ones for gas-treating processes.

Results of CO_2 solubility measurements in aqueous mixtures of DEA with AMP are limited. Simulation studies with blends of MDEA + MEA and MDEA + DEA have indicated considerable improvements in absorption and appreciable savings in energy requirements as compared with the single amine systems.[3,4] Seo and Hong[5] reported the solubility of CO_2 in blends of DEA and AMP in the partial pressure range of (10 to 300) kPa and in the temperature range of (313 to 353) K. Murrieta-Guevera et al.[6] reported the CO_2 solubility in aqueous solutions with the following mass fractions of (25 % DEA + 5 % AMP) and (20 % DEA + 10 % AMP) in the temperature range (313.15 to 373.15) K and in the CO_2 partial pressure range of (22 to 2600) kPa. Park et al.[7] measured CO_2 solubility in aqueous solutions with the following mass fractions of (20 % DEA + 10 % AMP) and (10 % DEA + 20 % AMP) in the temperature range of (313 to 353) K and in the CO_2 partial pressure range of (0.680 to 340) kPa. In industrial gas-treating, the knowledge of vapor−liquid equilibrium (VLE) in the (1 to 100) kPa range is very important, particularly for predicting the performance toward the top of the absorption column. The major problems concerning the VLE data of aqueous (alkanolamine + acid gas) systems, in general, are the lack of systematic measurement and inconsistency in the data reported in the literature. Hence, there is always a scope to add newer data and evaluation of the quality of the data generated in comparison

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to the existing literature results. Hence, in this work, new experimental VLE data of the (CO_2 + AMP + DEA + H_2O) system with various relative amine compositions at typically absorption temperatures and CO_2 partial pressure range of (1 to 100) kPa are presented here. The measured VLE data of the (CO_2 + AMP + DEA + H_2O) system of this work, in conjunction with the VLE data for the low and moderate to high partial pressure ranges, should cover a wide range of CO_2 partial pressure for the amine concentration and temperature range studied.

Representation of the experimental data with a thermodynamically rigorous model is essential, so that one can systematically correlate and predict the VLE of these systems. Austgen et al.[8,9] utilized the NRTL theory to model acid gas (CO_2 and H_2S) VLE in aqueous MDEA and in MDEA + MEA and MDEA + DEA blends. Li and Mather[10] simplified the Clegg−Pitzer equations and applied them to model the system (CO_2 + MDEA + MEA + H_2O). Li and Mather[11,12] also have correlated and predicted the solubility of CO_2 and H_2S in aqueous solutions of triethanolamine and MDEA, respectively. Qian et al.[13] correlated and predicted the solubility of CO_2 and H_2S in aqueous solutions of MDEA + sulfolane. Jane and Li[14] correlated the solubility of mixed gases (CO_2 and H_2S) in an aqueous blend of DEA + AMP using the model of Deshmukh and Mather.[15] Park et al.[7] correlated the solubility of CO_2 into aqueous blends of DEA + AMP with a modified Kent−Eisenberg approach. In this paper, a rigorous thermodynamic model to predict the solubility of CO_2 in aqueous blends of DEA + AMP is presented.

Experimental Section

Materials. 2-Amino-2-methyl-1-propanol (AMP) and diethanolamine (DEA) were supplied by E. Merck, Germany, and had a mole fraction purity of 98 %. Amine concentration was determined by titration with standard HCl using methyl orange indicator. Double-distilled water, degassed by boiling, was used to prepare the aqueous amine solutions. Pure carbon dioxide, obtained from Hydrogas India, has a certified mole fraction purity of 99.995 %. The mole fraction purity of nitrogen gas, obtained from BOC India, was better than 99 %.

Apparatus and Procedure. For the VLE measurements, a stirred glass equilibrium cell was used. A detailed description of the experimental setup and procedure is presented by Kundu et al.[16,22] The temperature of the equilibrium cell was controlled within ± 0.2 K of the desired level, and all VLE measurements were done at atmospheric pressure. Double-distilled water degassed by prolonged boiling and cooled to ambient temperature under airtight conditions was used to prepare aqueous amine solutions. For each equilibrium run, 10 mL of amine solution was taken from freshly prepared 100 mL of blended amine stock solution of desired relative amine composition (by mass fraction). Determination of the CO_2 concentration in the gas phase in the cell at equilibrium was made for each equilibrium run by the "absolute calibration method" using gas chromatography. The results of the gas analysis for CO_2 mol fraction was used to determine the equilibrium CO_2 partial pressure after correction for the vapor pressures of water and amine at the temperature of the equilibrium run. The equilibrium CO_2 loading in the liquid phase was determined by acidulating a known volume of the loaded liquid sample in a glass cell placed in a thermostated bath and measuring the volume of the evolved gas by a precisely graduated gas buret. Initially the temperature of the room as well as the cell and buret assembly was brought within ± 0.5 K of 293 K. A measured volume of the liquid sample (1.0×10^{-6} m^3) diluted to (10×10^{-6} m^3) with double-distilled water was then introduced into the cell, and the cell was sealed. Then 0.6 mol % HCl solutions were added to it in excess and thoroughly mixed with the help of a magnetic stirrer. The system was then allowed to attain equilibrium with respect to temperature, and the volume of evolved gas was measured with the gas buret. The measured volume of evolved gas was corrected for the vapor pressure of the liquid at the test temperature.

Uncertainty in Measured Quantities and Reproducibility of Results. The uncertainty of the data is influenced by the uncertainty of the following measurements: temperature of the thermostated bath and the equilibrium cell, total system pressure, vapor phase CO_2 mol fraction, and CO_2 loading of the liquid sample.

The temperature of the thermostated bath and the equilibrium cell was measured with Pt-100 sensors with an uncertainty of ± 0.2 K. The total pressure for each run was measured to an uncertainty of within ± 0.2 kPa. The overall uncertainty in the determination of vapor-phase CO_2 mol fraction by the gas chromatograph is 3 %. Replicated measurements indicated that the overall uncertainty in the CO_2 solubility is 3 %.

Model Development

Chemical Equilibria. In the aqueous phase for the (CO_2 + DEA + AMP + H_2O) system, the following equilibrium chemical reactions are involved:

$$CO_2 + 2H_2O \overset{K_1}{\longleftrightarrow} H_3O^+ + HCO_3^- \qquad (1)$$

$$H_2O + DEAH^+ \overset{K_2}{\longleftrightarrow} H_3O^+ + DEA \qquad (2)$$

$$H_2O + AMPH^+ \overset{K_3}{\longleftrightarrow} H_3O^+ + AMP \qquad (3)$$

$$DEACOO^- + H_2O \overset{K_4}{\longleftrightarrow} DEA + HCO_3^- \qquad (4)$$

where K_1, K_2, K_3, and K_4 are the thermodynamic chemical equilibrium constants. Since the present work is limited to the low to moderate partial pressure range, the fugacity of CO_2 is assumed to be its partial pressure, and solubility of CO_2 is identical to Henry's constant (H_{CO_2}) of CO_2. The VLE is given by

$$p_{CO_2} = H_{CO_2} x_{CO_2} \gamma_{CO_2} \qquad (5)$$

Mandal et al.[17] has reported the physical solubility of CO_2 in aqueous single amines and blends of alkanolamines using the "N_2O analogy" over a wide range of temperature and amine composition. The Henry's constant of CO_2 in aqueous single alkanolamines, aqueous amine blends, and H_{CO_2} in water have been taken from Mandal et al.[17] and Austgen et al.,[9] respectively. In view of the unavailability of the Henry's constant of CO_2 in pure alkanolamines, due to the immediate reaction of amines with CO_2, the reference state chosen for the molecular solute CO_2 is the ideal infinitely dilute aqueous solution at the system temperature and pressure. For the determination of interaction parameters in CO_2−DEA−H_2O and CO_2−AMP−H_2O systems, the H_{CO_2} in aqueous single alkanolamine solutions has been assumed to be equal to its value in pure water at system temperature and pressure, following a similar assumption of several previous workers for acid gas + aqueous alkanolamine systems.[8−10] However, some of the predictions of CO_2 partial pressure have also been checked with the value of Henry's

Table 1. Solubility of CO_2 in Aqueous Solutions of Mass Fractions (1.5 % DEA + 28.5 % AMP) in the Temperature Range of (303 to 323) K

T = 303 K		T = 313 K		T = 323 K	
p_{CO_2}/kPa	loading (α_{CO_2})	p_{CO_2}/kPa	loading (α_{CO_2})	p_{CO_2}/kPa	loading (α_{CO_2})
1.925	0.524	7.25	0.595	7.30	0.455
6.25	0.648	22.80	0.690	11.49	0.503
24.15	0.718	41.43	0.764	18.01	0.550
43.0	0.820	82.79	0.798	28.90	0.610
88.09	0.864			45.05	0.673
				74.28	0.756
				89.02	0.785

Table 2. Solubility of CO_2 in Aqueous Solutions of Mass Fractions (3.0 % DEA + 27.0 % AMP) in the Temperature Range of (303 to 323) K

T = 303 K		T = 313 K		T = 323 K	
p_{CO_2}/kPa	loading (α_{CO_2})	p_{CO_2}/kPa	loading (α_{CO_2})	p_{CO_2}/kPa	loading (α_{CO_2})
3.26	0.513	2.00	0.420	4.80	0.370
5.00	0.602	4.38	0.500	16.30	0.528
19.65	0.642	6.80	0.571	40.63	0.636
45.21	0.759	18.47	0.612	90.00	0.755
90.95	0.806	44.07	0.725		
		90.60	0.782		

Table 3. Solubility of CO_2 in Aqueous Solutions of Mass Fractions (4.5 % DEA + 25.5 % AMP) in the Temperature Range of (303 to 323) K

T = 303 K		T = 313 K		T = 323 K	
p_{CO_2}/kPa	loading (α_{CO_2})	p_{CO_2}/kPa	loading (α_{CO_2})	p_{CO_2}/kPa	loading (α_{CO_2})
2.58	0.447	2.22	0.422	5.175	0.370
6.70	0.577	8.00	0.558	16.00	0.500
20.0	0.647	35.0	0.696	55.14	0.632
37.0	0.717	62.0	0.740	86.00	0.706
90.0	0.788				

Table 4. Solubility of CO_2 in Aqueous Solutions of Mass Fractions (6.0 % DEA + 24.0 % AMP) in the Temperature Range of (303 to 323) K

T = 303 K		T = 313 K		T = 323 K	
p_{CO_2}/kPa	loading (α_{CO_2})	p_{CO_2}/kPa	loading (α_{CO_2})	p_{CO_2}/kPa	loading (α_{CO_2})
2.2	0.425	3.35	0.450	10.57	0.425
6.0	0.558	5.10	0.500	29.00	0.541
10.0	0.593	7.60	0.541	57.70	0.613
30.0	0.699	30.0	0.678	92.77	0.698
84.0	0.778	35.68	0.696		
		60.82	0.743		

constant measured by Mandal et al.[17]in aqueous single amine solutions using the N_2O analogy. It has been found that there is a negligible change in the predicted CO_2 partial pressures. Chemical equilibrium constants are taken from Austgen et al.,[9] Li and Shen,[18] and Silkenbäumer et al.[19] and used after appropriate conversion. The equilibrium constants (K_1, K_2, and K_4) based on the mole fraction scale are presented in Table 5. The chemical equilibrium constants in this work should adopt the same reference states for each component in equilibrium. The protonated amine dissociation constants (reactions 2 and 3) were corrected to the pure amine reference state. The correction to the equilibrium constant is related to the infinite dilution activity coefficient of the amine in water. The equilibrium constant K_3 has been found by mathematical manipulation of the following two reactions as described below:[16]

$$\mathrm{AMP} + \mathrm{H_2O} \overset{K_6}{\longleftrightarrow} \mathrm{OH^-} + \mathrm{AMPH^+} \tag{6}$$

$$\mathrm{H_2O} \overset{K_7}{\longleftrightarrow} \mathrm{H^+} + \mathrm{OH^-} \tag{7}$$

where K_6 and K_7 are the thermodynamic chemical equilibrium constants based on a molarity scale.

Thermodynamic Framework. For the (CO_2 + DEA + AMP + H_2O) system, the equilibrated liquid phase is assumed to contain three molecular species (H_2O, DEA, and AMP) and five ionic species ($AMPH^+$, HCO_3^-, $DEAH^+$, $DEACOO^-$, and H_3O^+). Species such as free molecular CO_2, OH^-, and CO_3^{2-} ions will have little effect on the observed equilibria.[15] Several previous workers have observed that for CO_2 loading below 1.0, neglecting the concentrations of free molecular CO_2 and OH^- and CO_3^{2-} ions in the liquid phase does not result in significant error in the VLE predictions.[10−12,20,21] In our calculation of activity coefficients of the components in the aqueous phase, only the activity coefficients of DEA, AMP, H_2O, $DEAH^+$, $AMPH^+$, $DEACOO^-$, and HCO_3^- are included to account for the nonideality of the liquid phase. As the free molecular CO_2 concentration in the liquid phase is negligible below the loading of 1.0, the value of the activity coefficient of CO_2 (γ_{CO_2}) will be close to unity following the unsymmetric normalization of activity coefficient. Thus, we can calculate liquid-phase mole fractions based on true molecular or ionic species.

Standard States. In this work water, AMP, and DEA are treated as solvents. The standard state associated with each solvent is the pure liquid at the system temperature and pressure. The adopted standard state for ionic solutes is the ideal, infinitely dilute aqueous solution (infinitely dilute in solutes and alkanolamines) at the system temperature and pressure. The reference state chosen for molecular solute CO_2 is the ideal, infinitely dilute aqueous solution at the system temperature and pressure. This leads to the following unsymmetric normalization of activity coefficients.

solvents:

$$\gamma_s \rightarrow 1 \text{ as } x_s \rightarrow 1$$

where the subscripts s and i refer to any nonaqueous solvent ionic and neutral solutes:

$$\gamma_i \rightarrow 1 \text{ as } x_i \rightarrow 0$$

and ionic or neutral solute, respectively. Activity coefficients of all species are assumed to be independent of pressure.

Thermodynamic Expression of Equilibrium Partial Pressure. From eqs 1 to 4, we get the expression of partial pressure of CO_2 over aqueous DEA + AMP solutions as follows:

$$p_{CO_2} = \frac{H_{CO_2} K_2^{\,2} (x^2_{DEAH^+} \gamma^2_{DEAH^+} x_{AMP} \gamma_{AMP} x^2_{HCO_3^-} \gamma^2_{HCO_3^-})}{(x_{AMPH^+} \gamma_{AMPH^+} x_{DEA} \gamma_{DEA} x_{DEACOO^-} \gamma_{DEACOO^-} x^2_{H_2O} \gamma^2_{H_2O})(K_1 K_3 K_4)} \tag{8}$$

where p_{CO_2} is the partial pressure of CO_2 and x_n' is the liquid-phase mole fraction in equilibriated liquid phase, based on true molecular or ionic species. Since an unsymmetric normalization convention is used in this work, compositions and activity coefficients are expressed in terms of the mole fraction scale. Activity coefficient γ_i in eq 8 for different species present in the liquid phase, are calculated by the "activity coefficient model" using the modified Clegg−Pitzer equations.

Table 5. Temperature Dependence of the Equilibrium Constants and Henry's Constant[a]

$$K_i = \exp[A + B/(T/\mathrm{K}) + C\ln(T/\mathrm{K}) + D(T/\mathrm{K})] \text{ where } i = 1, 2, 4$$
$$K_6 = \exp[A/(T/\mathrm{K}) + B\ln(T/\mathrm{K}) + C(T/\mathrm{K}) + D]$$
$$K_7 = \exp[A + B/(\mathrm{T}/\mathrm{K}) + C/(T/\mathrm{K})^2 + D/(T/\mathrm{K})^3 + E/(T/K)^4]$$
$$H = \exp[A + (B/(T/\mathrm{K})) + C\ln(T/\mathrm{K}) + D(T/\mathrm{K})]$$

reaction	A	B	C	D	E	ref
1	231.465	−12092.1	−36.7816	0.0		9
2	−13.3373	−4218.71	0	0.009872		9
4	16.5027	−4068.76	−1.5027	0.0		9
6	−7261.78	−22.4773	0	142.58612		19
7	39.5554	−9.879e4	0.568827e8	−0.14645e11	0.136145e13	18
Henry's constant	170.7126	−8477.711	−21.95743	0.005781		9

[a] For $i = 1, 2$, and 4, A is in kmol ^0K/kmol ^0K, B is in kmol ^0K/kmol, C is in kmol/kmol ^0K, D is in kmol/kmol ^0K. For $i = 6$, A is in kmol·m^{-3} ^0K, B is in kmol·m^{-3}/^0K, C is in kmol·m^{-3}/^0K, D is in kmol·m^{-3} ^0K/^0K. For $i = 7$, A is in (kmol·m^{-3})^2 ^0K/^0K, B is in (kmol·m^{-3})^2 (^0K), C is in (kmol·m^{-3})^2 (^0K)^2, D is in (kmol·m^{-3})^2 (^0K)^3, E is in (kmol·m^{-3})^2 (^0K)^4. For H, A is in Pa ^0K/^0K, B is in Pa ^0K, C is in Pa/^0K, D is in Pa/^0K.

Equilibrium Liquid-Phase Composition. From eqs 1 to 4, we can write the following equilibrium relations among the reacting species:

$$K_c^{\,1} = \frac{c_{\mathrm{H_3O^+}}\gamma_{\mathrm{H_3O^+}} c_{\mathrm{HCO_3^-}}\gamma_{\mathrm{HCO_3^-}}}{c_{\mathrm{CO_2}}\gamma_{\mathrm{CO_2}} a^2_{\mathrm{H_2O}}} \tag{9}$$

$$K_c^{\,2} = \frac{c_{\mathrm{H_3O^+}}\gamma_{\mathrm{H_3O^+}} c_{\mathrm{DEA}}\gamma_{\mathrm{DEA}}}{c_{\mathrm{H_2O}}\gamma_{\mathrm{H_2O}} c_{\mathrm{DEAH^+}}\gamma_{\mathrm{DEAH^+}}} \tag{10}$$

$$K_c^{\,3} = \frac{c_{\mathrm{H_3O^+}}\gamma_{\mathrm{H_3O^+}} c_{\mathrm{AMP}}\gamma_{\mathrm{AMP}}}{c_{\mathrm{H_2O}}\gamma_{\mathrm{H_2O}} c_{\mathrm{AMPH^+}}\gamma_{\mathrm{AMPH^+}}} \tag{11}$$

$$K_c^{\,4} = \frac{c_{\mathrm{DEA}}\gamma_{\mathrm{DEA}} c_{\mathrm{HCO_3^-}}\gamma_{\mathrm{HCO_3^-}}}{c_{\mathrm{H_2O}}\gamma_{\mathrm{H_2O}} c_{\mathrm{DEACOO^-}}\gamma_{\mathrm{DEACOO^-}}} \tag{12}$$

where c stands for amount concentration of ions and molecular species. K_c^1 to K_c^4 are equilibrium constants based on molarity scale. The following balance equation for the reacting species can be formed:

CO_2 balance:

$$c_{\mathrm{CO_2}} + c_{\mathrm{HCO_3^-}} + c_{\mathrm{DEACOO^-}} = \alpha(c^0_{\mathrm{AMP}} + c^0_{\mathrm{DEA}}) \tag{13}$$

where c^0 and α stand for the initial amount concentration of

DEA balance:

$$c_{\mathrm{DEA}} + c_{\mathrm{DEAH^+}} + c_{\mathrm{DEAHCOO^-}} = c^0_{\mathrm{DEA}} \tag{14}$$

AMP balance:

$$c_{\mathrm{AMP}} + c_{\mathrm{AMPH^+}} = c^0_{\mathrm{AMP}} \tag{15}$$

Electroneutrality:

$$c_{\mathrm{DEAH^+}} + c_{\mathrm{AMPH^+}} + c_{\mathrm{H_3O^+}} = c_{\mathrm{HCO_3^-}} + c_{\mathrm{DEAHCOO^-}} \tag{16}$$

molecular species and equilibrium loading of CO_2 in the liquid phase, respectively. In this work, activity of water has been assumed to be its mole fraction in accordance with the work of Deshmukh and Mather.[15] Solving the system of nonlinear equations (eqs 9 to 16), we get the equilibrium amount concentration of the species and the mole fraction of water present in the equilibrated liquid phase, by setting all the activity coefficients of seven other species as unity.

Activity Coefficient Model. The modified Clegg−Pitzer equations have been used to derive the activity coefficients of different species present in the equilibrated liquid phase.[16] Differentiation of the expressions for the short-range and long-range force contributions to the excess Gibbs energy yields activity coefficients for the ions and the neutral species. The expression for the activity coefficient for solvent N and ion M^+ in blended amine solvent are as follows:

$$\begin{aligned}\ln\gamma_{\mathrm{N}} = {} & 2A_xI_x^{3/2}/(1+\rho I_x^{1/2}) - \sum_c\sum_a x_cx_aB_{ca}\exp(-\alpha_1I_x^{1/2}) + \\ & x_{\mathrm{I}}(1-x_{\mathrm{N}})\sum_c\sum_a F_cF_aW_{\mathrm{N}ca} - x_{\mathrm{I}}\sum_n{}' x_n\sum_c\sum_a F_cF_aW_{nca} + \\ & \sum_n{}' x_n[A_{\mathrm{N}n}x_n(1-2x_{\mathrm{N}}) + 2A_{n\mathrm{N}}x_{\mathrm{N}}(1-x_{\mathrm{N}})] - \\ & 2\sum_n{}'\sum_{n'}{}' x_nx_{n'}(A_{nn'}x_{n'} + A_{n'n}x_n)\end{aligned} \tag{17}$$

$$\begin{aligned}\ln\gamma_{\mathrm{M^+}} = {} & -z_{\mathrm{M}}^2A_x\left[\frac{2}{\rho}\ln(1+\rho I_x^{1/2}) + \frac{I_x^{1/2}(1-2I_x/z_{\mathrm{M}}^2)}{1+\rho I_x^{1/2}}\right] + \\ & \sum_a x_aB_{\mathrm{M}a}\,g(\alpha_1I_x^{1/2}) - \sum_c\sum_a x_cx_aB_{ca}[z_{\mathrm{M}}^2g(\alpha_1I_x^{1/2})/2I_x + \\ & (1-z_{\mathrm{M}}^2/2I_x)\exp(-\alpha_1I_x^{1/2})] + 2\sum_n x_n\sum_a F_aW_{n\mathrm{M}a} - \\ & \sum_n x_n(1+x_{\mathrm{I}})\sum_c\sum_a F_cF_aW_{nca} - 2\sum_a F_aW_{1\mathrm{M}a} + \\ & \sum_c\sum_a F_cF_aW_{1ca} - 2\sum_n\sum_{n'}(A_{nn'}x_{n'} + A_{n'n}x_n)\end{aligned} \tag{18}$$

where I_x is the ionic strength on mole fraction basis, A_x is the Debye−Hückel parameter on mole fraction basis, $A_{n'n}$ and $A_{nn'}$ are solvent−solvent interaction parameters, ρ is the density of mixed amine solution, W is the interaction parameter between neutral and ionic species, Z_i is the valency of an ion, F is the ionic fraction, α_1 is the Pitzer universal constant in eq 17, and B_{ca} is the ion−ion interaction parameter. The subscript 1 in eq 18 stands for water. The function $g(x)$ in eq 18 is expressed by

$$g(x) = 2[1 - (1 + x)\exp(-x)]/x^2 \tag{19}$$

where

$$x = (\alpha_1I_x^{1/2}) = 2I^{1/2} \tag{20}$$

and I is the ionic strength on concentration basis.

Data Regression: Estimation of Interaction Parameter

The solubility data of the systems (CO_2 + DEA + H_2O) and (CO_2 + AMP + H_2O) over a wide range of temperature, gas loading (below 1.0), and amine compositions covering the usual operating conditions of gas-treating processes are used to

Table 6. Effect of Errors in the Parameters W_{1MY}, W_{2MY}, W_{2NX}, W_{2NY}, W_{3MX}, W_{3MY}, B_{MY}, A_{23}, and A_{32} on the Predicted Equilibrium Partial Pressure of CO_2 in (H_2O + DEA + AMP) System[a]

temperature	error introduced	deviation in predicted partial pressure
W_{1MY}	−50 % to +50 %	−10.6 % to +11.8 %
W_{2MY}	−50 % to +50 %	+2.9 % to −2.9 %
W_{2NX}	−50 % to +50 %	−5.6 % to +5.9 %
W_{2NY}	−50 % to +50 %	−0.65 % to +0.66 %
W_{3MX}	−50 % to +50 %	+3.02 % to −2.9 %
W_{3MY}	−50 % to +50 %	+0.9 % to −0.9 %
A_{23}	−50 % to +50 %	+0.013 % to 0.000 %
A_{32}	−50 % to +50 %	+0.03 % to −0.03 %
B_{MY}	−50 % to +50 %	+0.47 % to −0.46 %

[a] Subscripts: 1, H_2O; 2, AMP; 3, DEA; M, $AMPH^+$; X, HCO_3^-; N, $DEAH^+$; Y, $DEACOO^-$.

Table 7. Fitted Values of Interaction Parameters for (CO_2 + AMP + H_2O) System: B (or W or A) = a + b/(T/K)[a]

B or *W* or *A*	*a*	*b*
W_{1MX}	19.87250	−5358.036
W_{2MX}	12.12244	249.8551
B_{MX}	26.71344	2E−2
A_{12}	26.33370	51.24970
A_{21}	−31.31813	7362.850

[a] Subscripts: 1, AMP; 2, H_2O; M, $AMPH^+$; X, HCO_3^-.

Table 8. Fitted Values of Interaction Parameters for (CO_2 + DEA + H_2O) System: B (or W or A) = a + b/(T/K)[a]

B or *W* or *A*	*a*	*b*
W_{1MX}	88.5765	−22577.12
W_{1MY}	1818.47	−698723.11
W_{2MX}	−1001.406	279705.525
W_{2MY}	−4545.2886	1.4435e6
B_{MX}	−4531.727	1.399e6
B_{MY}	1135.67	−427437.18
A_{12}	−3.035082	−45.182890
A_{21}	1.706795	−321.523

[a] Subscripts: 1, H_2O; 2, DEA; M, $DEAH^+$; X, HCO_3^-; Y,$=RR'NCOO^-$.

determine the interaction parameters. The objective function (ψ) used for optimization is given by

$$\psi = \sum |\{(p_{CO_2})_{cal} - (p_{CO_2})_{exp}\}^2/(p_{CO_2})_{exp}{}^2| \qquad (21)$$

We seek the numerical values of interaction parameters that will minimize the difference between the measured values of equilibrium partial pressure of CO_2 over aqueous (DEA + AMP) solutions and the values calculated from the model. The objective function chosen in this work takes care of rendering uniform weightage throughout the entire range of partial pressure (from low to high), provided the data scatter throughout the entire range of partial pressure (from low to high) is, as claimed by the previous workers, more or less uniform.[22]

The 11 parameters determined from the ternary systems (CO_2 + DEA + H_2O) and (CO_2 + AMP + H_2O) and two solvent−solvent interaction parameters determined from binary system (DEA + water) have been used to predict the VLE of the quaternary system (CO_2 + DEA + AMP + H_2O) without any additional parameter. AMP−water and water−AMP interaction parameters are determined from regression analysis of ternary (CO_2 + AMP + H_2O) VLE data.

For the determination of DEA−water and water−DEA parameters, the Margules interaction parameters in DEA + H_2O system are obtained by regression analysis of the water activity coefficient using NRTL binary interaction parameters based on freezing point depressions and total pressure data.[23] The DEA−AMP and AMP−DEA interaction parameters have been neglected because of the little influence on the uncertainty in the VLE prediction due to the relatively lower mol fractions of DEA and AMP in comparison to that of water. Six solvent−ion pair interactions (H_2O−$AMPH^+$ $DEACOO^-$, AMP−$AMPH^+$ $DEACOO^-$, DEA−$AMPH^+$ $DEACOO^-$, AMP−$DEAH^+$ $DEACOO^-$, AMP−$DEAH^+$ HCO_3^-, and DEA−$AMPH^+$ HCO_3^-) and one ion−ion interaction ($AMPH^+$−$DEACOO^-$) have been assumed to be zero as they cannot be derived from the ternary system. Similar assumptions have been made by Li and Mather[10] for modeling the VLE of CO_2 in MEA + MDEA blends. The validity of this assumption is further verified from the results of a parametric sensitivity analysis using the developed model. The parametric sensitivity analyses of the aforesaid parameters have been done over mass fractions of 6 % DEA + 24 % AMP aqueous solutions at a particular liquid-phase CO_2 loading of 0.541, at a temperature of 313 K. The results are summarized in Table 6. It has been observed that these parameters have negligible influences on the VLE of CO_2 in aqueous solutions of DEA + AMP.

For (CO_2 + AMP + H_2O) system the experimental solubility data from the literature for acid gas loadings in the range (0.03 to 1.0) and the CO_2 partial pressure range of (0.300 to 1000) kPa have been used to estimate the interaction parameters.[16] The interaction parameters for (CO_2 + AMP + H_2O) system with their temperature coefficients are listed in Table 7.

For the (CO_2 + DEA + H_2O) system, the experimental solubility data for solutions of mass fractions of 21 %, 25 %, and 29.96 % DEA solutions in the temperature range (311 to 394) K, CO_2 partial pressure in the range (0.2 to 5000) kPa, and below a CO_2 loading of 1.0 are used to estimate the interaction parameters.[5,24−27] The parameters are listed in Table 8.

Method of Solution

Owing to the presence of multiple solutions while solving the system of nonlinear equations, some approaches were unable to obtain the global solution because they could not jump over the local minima. Simulated annealing (SA) has been used in this work for the estimation of liquid-phase composition at equilibrium and the interaction parameters of the VLE model in an effort to predict the VLE of CO_2 in the aqueous amine blends with good accuracy.[16]

Results and Discussion

The solubility of CO_2 in aqueous solutions of the following mass fractions (1.5 % DEA + 28.5 % AMP), (3 % DEA + 27 % AMP), (4.5 % DEA + 25.5 % AMP), and (6 % DEA + 24 % AMP) are presented at (303 to 323) K, keeping the total amine mass fraction at 30 %. As the temperature increases the CO_2 solubility decreases at a particular amine composition. With the increasing DEA concentration in the amine blend, the CO_2 solubility decreases at a particular temperature.

The interaction parameters presented for the (CO_2 + AMP + H_2O) and (CO_2 + DEA + H_2O) systems in Tables 7 and 8 are used to predict the VLE of (CO_2 + AMP + DEA + H_2O) without any additional parameter. The model has been validated with the experimental results of this work for various relative amine compositions in the temperature range (303 to 323) K. The predictions of CO_2 solubility in aqueous DEA + AMP solutions of various relative amine compositions are in good agreement as shown in Figures 1 to 3. Above a certain CO_2 partial pressure, corresponding to a CO_2 loading of 0.5 mol of CO_2/mol of amine, the CO_2 solubility in aqueous AMP of mass

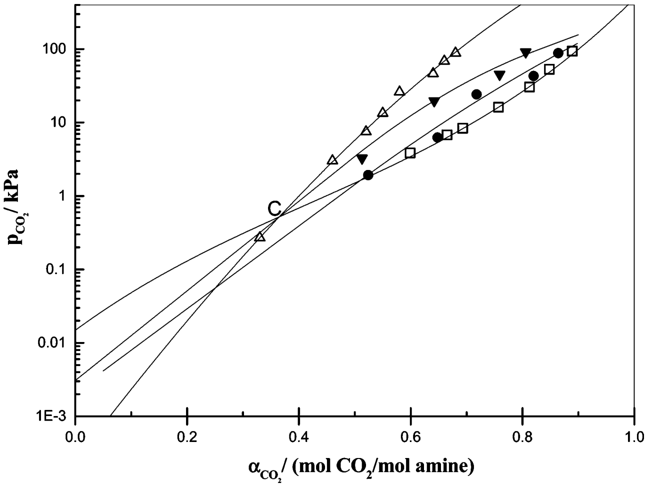

Figure 1. Solubility of CO_2 in (DEA + AMP) solutions with the following mass fractions as a function of loading (α) at 303 K: —, model predicted; □, 0 % DEA + 30 % AMP, Kundu et al.;[16] ●, 1.5 % DEA + 28.5 % AMP, this study; ▼, 3 % DEA + 27 % AMP, this study; △, 30 % DEA + 0 % AMP, Kundu and Bandyopadhyay.[27]

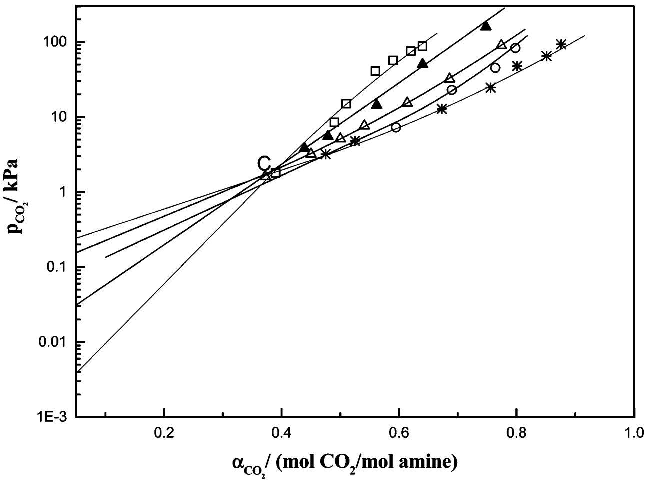

Figure 2. Solubility of CO_2 in (DEA + AMP) solutions with the following mass fractions as a function of loading (α) at 313 K: —, model predicted; □, 30 % DEA + 0 % AMP, Kundu and Bandyopadhyay;[27] ○, 1.5 % DEA + 28.5 % AMP, this study; △, 6 % DEA + 24 % AMP, this study; ▲, 18 % DEA + 12 % AMP, Seo and Hong;[5] *, 0 % DEA + 30 % AMP, Kundu et al.[16]

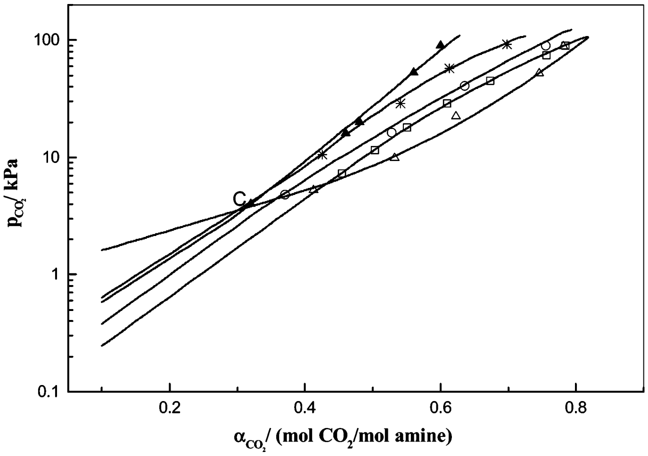

Figure 3. Solubility of CO_2 in (DEA + AMP) solutions with the following mass fractions as a function of loading (α) at 323 K: —, model predicted; ▲, 30 % DEA + 0 % AMP, Kundu and Bandyopadhyay;[27] *, 4.5 % DEA + 25.5 % AMP, this study; ○, 3 % DEA + 27 % AMP, this study; □, 1.5 % DEA + 28.5 % AMP, this study; △, 0 % DEA + 30 % AMP, Kundu et al.[16]

fraction 30 % is greater than that in aqueous DEA solution having mass fraction 30 %, which is also evident from Figures 1 to 3. A crossover point (indicated by **C**) of the solubility curves occur in Figures 1 to 3. Similar crossover in solubility curves were reported earlier by Li and Shen[18] for the aqueous MEA + MDEA blends, by Park et al.[7] for aqueous MEA + AMP and DEA + AMP blends, by Li and Chang[28] for aqueous MEA + AMP blend, and by Seo and Hong[5] for aqueous DEA + AMP blend. Moreover, with increasing temperature the crossover point shifts toward a higher equilibrium CO_2 partial pressure. This crossover may result from the fact that AMP forms a very unstable carbamate with CO_2 and the stoichiometric loading of AMP reaches 1.0 mol of CO_2/mol of amine. However, DEA forms a very stable carbamate with CO_2. In this reaction, two DEA molecules react with each CO_2 molecule. Hence, for an aqueous solution of DEA, it is reasonable to assume that the amine molecules are completely converted to the product at CO_2 loading greater than 0.5 mol of CO_2/mol of amine, and the sudden change in equilibrium partial pressure occurs because of the physical absorption rather than chemical absorption. If the solution mixture of DEA and AMP at low CO_2 loading is considered, DEA can react better than AMP with CO_2 because of the higher reactivity of DEA toward CO_2. However, as CO_2 loading increases, the unreacted DEA concentration in the solvent decreases and results in an increased ratio of AMP to DEA. Therefore, both DEA and AMP affect the solubility of CO_2 at moderate loadings. If the CO_2 loading is considerably high, the equilibrium is closer to that of AMP than to that of DEA, and the CO_2 + DEA equilibrium curve crosses the CO_2 + AMP curve.

The addition of DEA to an aqueous AMP solution reduces the equilibrium partial pressure at low to moderate loadings. However, this reduction is not as significant as the reduction, which results when MEA is added to the aqueous solutions of AMP or MDEA. This is expected since for the same CO_2 loading, the equilibrium partial pressure of CO_2 over aqueous solutions of DEA is higher than that over aqueous MEA.

The developed model can predict the VLE for the following mass fractions of solutions (12 % DEA + 18 % AMP) and (18 % DEA + 12 % AMP) in the temperature range (313 to 353) K with reasonable accuracy when compared with the experimental results of Seo and Hong.[5] The model predictions in the CO_2 partial pressure range of (150 to 2600) kPa at 373.15 K are in very good agreement with the experimental results of Murrieta-Guevera et al.[6] The accuracy of the predicted CO_2 partial pressures from the model in comparison with the various literature results as well as the experimental results of this work is summarized in Table 9.

An important result from rigorous equilibrium modeling is the speciation, that is, the capability to predict equilibrium liquid species concentration as a function of CO_2 loading. The equilibrium concentration profiles for CO_2-loaded aqueous blends having mass fractions of (4.5 % DEA + 25.5 % AMP) at 323 K and (18 % DEA + 12 % AMP) at 313 K are presented in Figures 4 and 5, respectively. Figures 4 and 5 indicate that DEA disappears at a slow rate in DEA/AMP mixture while AMP decreases monotonically with increasing CO_2 loading. Since molecular DEA is present in the solution up to the CO_2 loading of about 0.8 mol of CO_2/mol of amine, it appears that DEA will continue to enhance the absorption rate of CO_2 in an AMP based solvent mixture even when the solution is highly loaded. It is evident from these figures that the curve for the carbamate ion is sharper in blends with higher DEA concentra-

Table 9. Average Deviation for Prediction of Vapor−Liquid Equilibrium of CO_2 in (H_2O + DEA + AMP) System

ref	mass % DEA	mass % AMP	T/K	data points	p_{CO_2}/kPa	AAD %[a]
Seo and Hong[5]	6.0	24.0	313, 333, 353	48	1.0−400	6.03 %
	12.0	18.0				
	18.0	12.0				
Murrieta-Guevera et al.[6]	20.0	10.0	313, 373	9	10−3000	2.90 %
Murrieta-Guevera et al.[6]	25.0	5.0	313, 373	4	10−3000	2.90 %
	1.5	28.5	303, 313, 323	58	1−100	8.15 %
this work	3.0	27.0				
	4.5	25.5				
	6.0	24.0				

[a] AAD, average absolute deviation between predicted and experimental results; 100 % AAD = $((\sum_n|(p_{cal} - p_{exp})|/p_{exp})/(n)) \times 100$.

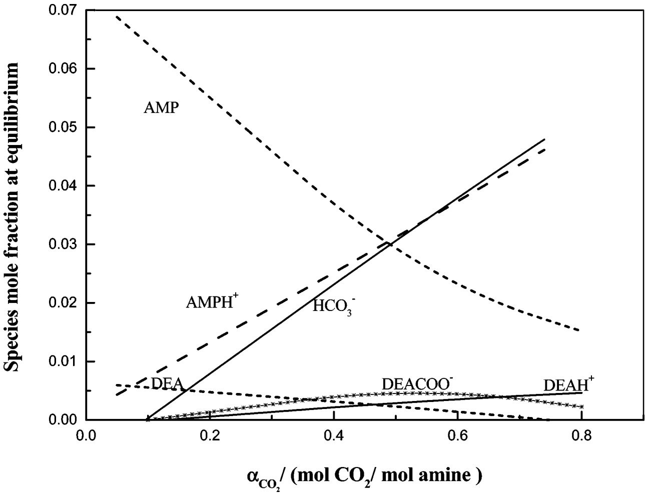

Figure 4. Equilibrium liquid-phase concentration of a CO_2-loaded aqueous solution having mass fractions of (4.5 % DEA + 25.5 % AMP) at 323 K.

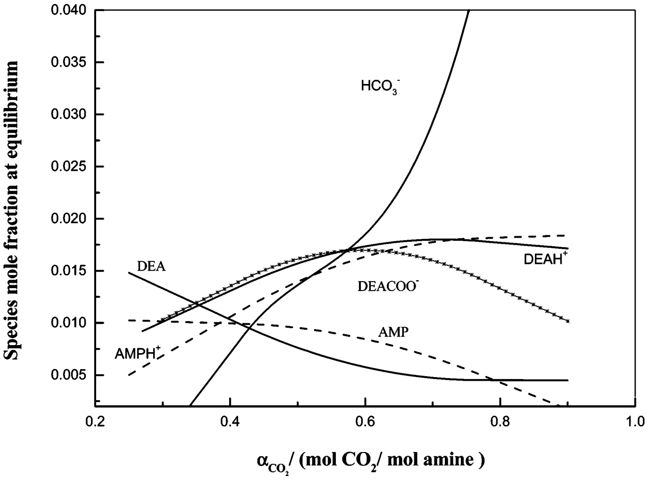

Figure 5. Equilibrium liquid-phase concentration of a CO_2-loaded aqueous solution having mass fractions of (18 % DEA + 12 % AMP) at 313 K.

tion and flatter in blends with lower DEA concentration. As shown in Figures 4 and 5, in the aqueous mixtures of DEA + AMP, CO_2 is mainly converted to carbamate at lower CO_2 loadings. At moderate CO_2 loadings, the carbamate concentration does not vary significantly while excess absorbed CO_2 is converted primarily to bicarbonate. At higher CO_2 loadings, the carbamate reverts to bicarbonate, and the bicarbonate concentration further increases with CO_2 loading. However, even at a loading approaching 1.0 mol of CO_2/mol of amine, a significant fraction of CO_2 is present as carbamate in the blended amine solution. It appears that the reversion of carbamate to bicarbonate plays an important role in determining the equilibrium behavior of CO_2 even at loadings approaching unity.

Conclusions

In this work a rigorous thermodynamic model has been developed to correlate and predict the VLE of CO_2 in aqueous solutions of DEA +AMP. For CO_2 loading lower than 1.0, the model predicted partial pressures of CO_2 over aqueous blends of DEA + AMP have been found to be in good agreement with the experimental results of this study as well as results obtained from the open literature over a wide range of temperature and CO_2 partial pressure. In this paper, a global algorithm, simulated annealing (SA), has been adopted to obtain equilibrium liquid-phase compositions and interaction parameters. New experimental data on the CO_2 solubility in the aqueous blends of DEA + AMP of relative amine mass fractions (1.5 % DEA + 28.5 % AMP), (3 % DEA + 27 % AMP), (4.5 % DEA + 25.5 % AMP), and (6 % DEA + 24 % AMP) in the CO_2 partial pressure range of (1 to 100) kPa and temperature range of (303 to 323) K are also reported in this paper.

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