Effect of Blending Ratio and Temperature on CO₂ Solubility in Blended Aqueous Solution of Monoethanolamine and 2-Amino-2-methyl-propanol: Experimental and Modeling Study Using the Electrolyte Nonrandom Two-Liquid Model

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ABSTRACT: This paper reports the newly measured experimental data for CO₂ solubility in a blended aqueous solution of monoethanolamine (MEA) and 2-amino-2-methyl-propanol (AMP) at different amine mixing ratios (MEA/AMP/H₂O = 9:21:70, 15:15:70, and 21:9:70 wt %) and working temperatures (323.15, 373.15, and 383.15 K). The successive substitution method was used for calculating the mole fractions of all molecules (four molecules) and electrolytes (three cations and four anions) from the equilibrium along with the material and charge balance equations (11 equations). The electrolyte nonrandom two-liquid (e-NRTL) model was used to investigate nonideality in the liquid phase. Using the abovementioned thermodynamic models, the partial pressures of CO₂ in the gas phase, mole fractions of all components in the liquid phase, pH variations, heats of absorption, and cyclic capacities of CO₂ according to the absorption/desorption temperature and the blending ratio of MEA/AMP were estimated.

INTRODUCTION

Carbon capture and storage (CCS) technology is considered as the most practical method to reduce greenhouse gas emissions. Depending on the CO₂ capture method, CCS technologies can be categorized into absorption, adsorption, membrane, and other hybrid processes. Among these, wet absorption is the most widely used for the CCS method.

The amines that can be used in the commercial CCS technology must have high cyclic capacity, high absorption rate, and excellent chemical stability. Commercial amines for the CCS technology can be classified into four types: (i) primary amine: monoethanolamine (MEA); (ii) secondary amine: diisopropanolamine (DIPA) and diethanolamine (DEA); (iii) tertiary amine: methyltriethanolamine (MDEA), triethanolamine (TEA), and triethylamine (TEA); and (iv) steric hindrance amine: 2-amino-2-methyl propanol (AMP). MEA is the most widely used amine because of its high reactivity and reasonable cost.¹⁻³ AMP has also attracted attention owing to its high CO₂ absorption capacity and chemical stability.¹⁻⁵ Recently, CO₂ absorption solvents have been developed by mixing two or more amines to take advantage of the favorable properties of each amine.¹⁻⁸ For instance, blended amine solutions (1) can utilize both physical and chemical properties, (2) can improve equilibrium amounts and kinetic characteristics, and (3) can control heat of absorption, pH, corrosion, and solvent degradation.⁹⁻¹⁰ In a previous study of Mahmoodi et al.,¹¹ MEA was used as an activator to enhance the CO₂ solubility of AMP. In this study, the characteristics of CO₂ absorption in a blended aqueous solution...
solution of MEA and AMP were examined using experiments and thermodynamic models.

To predict CO₂ solubility in a blended aqueous solution of MEA and AMP, 11 equations (six equilibrium equations, four mass balance equations, and one charge balance equation) for 11 variables (four molecules, three cations, and four anions) must be solved simultaneously with extremely low relative tolerances (under 10⁻⁷ in this study). In an aqueous amine solution, bicarbonate ions (HCO₃⁻) are formed when CO₂ reacts with H₂O. In primary and secondary amine aqueous solutions, CO₂ reacts with amines and forms both carbamate and bicarbonate ions. However, Saha et al. and Mandal and Bandypadhyay reported that sterically hindered amine does not form carbamate stably. Therefore, in the blended solution of primary amine and sterically hindered amine, such as MEA and AMP, it is highly useful to investigate the concentration behavior of both carbamate (MEACOO⁻ in this study) and bicarbonate ions.

Thermodynamic modeling for the vapor–liquid equilibrium (VLE) of the CO₂–aqueous amine solution system is one of the most important factors for designing amine-based chemical absorption processes for CCS, and it can be categorized into three approaches. The first approach involves finding new chemical equilibrium constants for amines, such as amine protonation or carbamate formation, from the experimentally obtained VLE data. Kent and Eisenberg proposed a very simple and useful model by neglecting the activity coefficients. The second and third approaches use excess Gibbs energy models, namely, the Deshmukh–Mather model and the electrolyte models, respectively. Deshmukh and Mather suggested a rigorous thermodynamic model and calculated the activity coefficients using the Guggenheim equation. The second and third approaches use excess Gibbs energy models, namely, the Deshmukh–Mather model and the electrolyte models, respectively. Deshmukh and Mather suggested a rigorous thermodynamic model and calculated the activity coefficients using the Guggenheim equation. The second and third approaches use excess Gibbs energy models, namely, the Deshmukh–Mather model and the electrolyte models, respectively. Deshmukh and Mather suggested a rigorous thermodynamic model and calculated the activity coefficients using the Guggenheim equation.

新材料。MEA (≥99%, Sigma-Aldrich), AMP (≥99% Acros Organics), and distilled water were mixed to prepare the absorption solutions. The total mass fraction of the blended amine solution was 30 wt %, and the mass ratios of MEA and AMP were 3:7, 5:5, and 7:3 (70 wt % water balanced). That is, the mass fractions of MEA and AMP were 9:21, 15:15, and 21:9 wt % and that of water was 70 wt %. Pure CO₂ (99.999%) and pure N₂ (99.999%) were introduced sequentially into the experimental apparatus.

Experimental Procedure. The solubility of CO₂ in the MEA and AMP blended amines was measured in the temperature of 323.15 K (absorption condition) and 373.15–383.15 K (stripping condition) using the static method. In the cleaning and preparation steps, the reactor was washed with distilled water and nitrogen three times. Then, the gases in the reactor were analyzed by gas chromatography to confirm the presence of other gaseous molecules. In the CO₂ absorption step, the mixed gas (CO₂/N₂) was injected into the reactor five times for converting nitrogen conditions to reaction gas conditions while keeping the reaction temperature constant. Then, approximately 150 mL of the absorption solution was introduced into the reactor with a high-pressure liquid syringe pump. When the temperature of the injected absorption solution became constant, the pressure was measured. The vapor pressure of the solution was calculated by measuring the pressure change of the reactor, the amount of the injected solution, and the volume of the reactor. The obtained vapor pressure was used to calculate the partial pressure of the absorbed gas at the equilibrium pressure. After the solution injection was completed, the mixed gas was introduced into the reactor. To improve the contact efficiency between the absorbent and the gas molecules, a magnetic drive with an impeller-type stirrer was used.

When the pressure in the absorption reactor decreased owing to CO₂ absorption in the solution and no further pressure changes were observed, the feed gas was reintroduced into the reactor from the gas reservoir. If the pressure did not change, it was regarded that the absorption equilibrium had been reached, and the pressure was then measured. Next, the mixed gas in the gas reservoir was reinjected into the absorption reactor. Experimental solubility results were represented by the CO₂ loading ratio, the mole ratio of CO₂ per total amine in the liquid mixture. The equilibrium loading ratio, αₐ₃, (mol CO₂/mol total amine), was calculated using the partial pressure and the absorbed quantity (mole ratio) at each equilibrium point.

\[ \alpha_{CO_2} = \frac{n_{CO_2,liq}}{\sum n_{amine,liq}} \]  

where \( n_{CO_2,liq} \) is the mole of CO₂ in the liquid phase and \( \sum n_{amine,liq} \) is the total moles of blended amine in the liquid phase. The amount of the absorbed CO₂ was first calculated by the ideal gas law. Then, the Redlich–Kwong equation was applied to determine the nonideality of the gas mixtures. Detailed information about the device has been provided in the Supporting Information.

Uncertainty Measurement. The uncertainty was estimated using the equation presented by NIST44 and Kim et
−blendedsoluti ons, is governed by the following expressions.

$$K_j = x_{\text{H}_2\text{O}}/x_{\text{H}^+ \cdot \text{H}_2\text{O}} = \gamma_{\text{H}_2\text{O}}/\gamma_{\text{H}^+ \cdot \text{H}_2\text{O}}$$ (13)

$$K_5 = x_{\text{H}_2\text{O}}/x_{\text{OH}^-} = \gamma_{\text{H}_2\text{O}}/\gamma_{\text{OH}^-}$$ (14)

$$K_6 = x_{\text{H}_{2}\text{O}}^2/x_{\text{MEACOO}^-} \gamma_{\text{MEACOO}^-}/\gamma_{\text{H}_{2}\text{O}}$$ (15)

Table 1. Temperature-Dependent Coefficients ($A_1$–$A_4$) for Reaction Equilibrium Constants ($K_i$)

| constant | $A_1$ | $A_2$ | $A_3$ | $A_4$ | reference |
|----------|-------|-------|-------|-------|-----------|
| $K_1$ | 2.1211 | −8189.38 | 0 | −0.007484 | Bates and Pinching$^{42}$ |
| $K_2$ | 3.918 | −7658.3 | 0 | 0 | Teng et al.$^{33}$ |
| $K_3$ | 231.465 | −12092.1 | −367816 | 0 | Edwards et al.$^{34}$ |
| $K_4$ | 216.049 | −12431.7 | −354819 | 0 | Edwards et al.$^{34}$ |
| $K_5$ | 132.899 | −13445.9 | −224773 | 0 | Edwards et al.$^{34}$ |
| $K_6$ | 2.8898 | −3635.09 | 0 | 0 | Austgen et al.$^{22}$ |

al.$^{43}$ Before calculating the uncertainty, the mean values of the experimental data were obtained.

$$\bar{W}_i = \frac{1}{n} \sum_{k=1}^{n} W_{i,k}$$ (2)

where $\bar{W}_i$ is the average value of the experimental data obtained under the same conditions. The standard uncertainty can be described as follows:

$$u(w_i) = \left( \frac{1}{n(n-1)} \sum_{k=1}^{n} (w_{i,k} - \bar{W}_i)^2 \right)^{1/2}$$ (3)

where $u(w_i)$ is the uncertainty and $n$ is the number of data.

## MODEL AND OPTIMIZATION

The reaction equilibrium of CO$_2$ in the MEA and AMP blended amine solutions is governed by the following expressions.

alkanolamine (MEA): MEAH$^+$ + H$_2$O ↔ MEA + H$_3$O$^+$

(4)

alkanolamine (AMP): AMPH$^+$ + H$_2$O ↔ AMP + H$_3$O$^+$

(5)

carbon dioxide: CO$_2$ + 2H$_2$O ↔ H$_2$O$^+$ + HCO$_3^-$

(6)

carbonate: HCO$_3^-$ + H$_2$O ↔ H$_3$O$^+$ + CO$_3^{2-}$

(7)

glycerine: 2H$_2$O ↔ H$_2$O$^+$ + OH$^-$

(8)

carbamate: MEACOO$^-$ + H$_2$O ↔ MEA + HCO$_3^-$

(9)

Ciftja et al.$^{46}$ experimentally demonstrated that carbamate is formed in the CO$_2$–AMP–water system under low CO$_2$ loading conditions using $^{13}$C NMR. However, some researchers reported that carbamate is not stably formed in AMP, the sterically hindered amine, and the amount formed is very small compared to MEA, the primary amine.$^{12,13}$ Therefore, in this study, it was assumed that the formation of AMPCOO$^-$ does not occur. Thermodynamic frameworks, the equilibrium constants, $K_i$ (–), for the above expressions can be described as follows:

$$K_1 = \frac{x_{\text{MEA}^+} x_{\text{H}_2\text{O}}}{x_{\text{MEAH}^+} x_{\text{H}_2\text{O}}^2} \gamma_{\text{MEA}^+}/\gamma_{\text{MEAH}^+}$$ (10)

$$K_2 = \frac{x_{\text{AMP}^+} x_{\text{H}_2\text{O}}}{x_{\text{AMPH}^+} x_{\text{H}_2\text{O}}^2} \gamma_{\text{AMP}^+}/\gamma_{\text{AMPH}^+}$$ (11)

$$K_3 = \frac{x_{\text{H}_2\text{O}} x_{\text{HCO}_3^-}}{x_{\text{CO}_3^{2-}} x_{\text{H}_2\text{O}}^2} \gamma_{\text{H}_2\text{O}}/\gamma_{\text{CO}_3^{2-}}$$ (12)
The electrolyte NRTL model considers nonidealities of liquid phases in an aqueous electrolyte system. Using binary and pair parameters, this model can calculate the activity coefficients for ionic species and molecular species. The electrolyte NRTL is an excess Gibbs energy model that follows two assumptions: (i) The local composition of cations (anions) around cations (anions) is zero. (ii) For the central molecule, the local net ionic charge of cations and anions is zero. The excess Gibbs free energy is expressed as follows:

\[ G^{\text{ex}} = G^{\text{PDH}} + G^{\text{Bos}} + G^{\text{LC}} \]

where \( G^{\text{ex}} \) is the excess Gibbs energy of the mixed solvent electrolyte system, \( G^{\text{PDH}} \) is the excess Gibbs energy calculated using the Pitzer–Debye–Hückel formula, \( G^{\text{Bos}} \) is the excess Gibbs energy calculated using the Born expression, and \( G^{\text{LC}} \) is the excess Gibbs energy calculated using the NRTL hypothesis. The activity coefficients for species \( i \) are obtained using eqs 27–29.\(^{26–31}\) The interactive binary parameter of \( \gamma_{ij}^{LC} \) is expressed in eq 30.

\[ \ln \gamma_i = \left[ \frac{\partial (n_i G^{\text{ex}}/RT)}{\partial n_i} \right]_{T,P,n_{j\neq i}} \]

\[ \ln \gamma'_i = \ln \gamma_{i\text{PDH}} + \ln \gamma_{i\text{Bos}} + \ln \gamma_{i\text{LC}} \]

\[ \tau_{ij} = a_{ij} + b_{ij}T \]

The symmetric mean absolute percentage error (SMAPE) method was used to compare the experimental data with the electrolyte NRTL model.

\[ \text{SMAPE} = \frac{100}{n} \sum_{i=1}^{n} \left| \frac{A_i - C_i}{(A_i + C_i)/2} \right| \]

Table 2. Temperature-Dependent Coefficients (B\(_1\)–B\(_4\)) for Henry’s Constants (\(H_{i,H_2O}\))

| constant | \(B_1\) | \(B_2\) | \(B_3\) | \(B_4\) | temperature range (°C) | reference |
|---------|------|------|------|------|----------------------|---------|
| \(H_{CO_2,H_2O}\) | 94.4914 | -6789.04 | 11.4519 | -0.0010454 | 0–250 | Edwards et al.\(^{44}\) |

The partial molar volume of \(CO_2\), \(\bar{\nu}^{\infty}_i\), in \(H_2O\) at infinite dilution: \(^{49}\)

\[ \ln p^0 = 16.3872 - 3885.70/(230.170 + T) \]

\[ \bar{\nu}^{\infty}_i = 0.000375 - 0.177362 - 52.1675 \]

The equilibrium constants \(K_i\) for the \(j\)th reaction and Henry’s constants \(H_{i,H_2O}\) for component \(i\) were used in the equation of temperature dependency as follows:

\[ \ln (K_i) = A_1 + A_2/T + A_3 \ln T + A_4T \]

Henry’s constants: \(H_{i,H_2O}\)

\[ = B_1 + B_2/T + B_3 \ln T + B_4T \]

Temperature-dependent coefficients for \(K_j\) \((A_1–A_4)\) and \(H_{i,H_2O}\) \((B_1–B_4)\) are listed in Tables 1 and 2, respectively.

\[ \begin{align*}
\text{Experimental Data of CO}_2 \text{ Solubility.} & \quad \text{To validate the experimental method and data, the solubility of CO}_2 \text{ in aqueous solution of MEA and AMP at 313.15–393.15 K was measured and the results were compared with published data in the literature.} \quad \text{Figure 1 shows the comparison of experimental CO}_2 \text{ solubility data in 30 wt % AMP and 70 wt % H}_2\text{O at 313.15–323.15 K between results of this work and the literature.}^{1,2,9,18,30,34,49–55} \quad \text{The experimental CO}_2 \text{ solubility data measured in this study were very close to the published data reported by Kundu et al.,}^{50} \quad \text{Seo and Hong,}^{51} \quad \text{Li and Chang,}^{1} \quad \text{Tong et al.,}^{49} \quad \text{Maun et al.,}^{59} \quad \text{and Shen and Li.}^{57} \quad \text{As can also be seen in Figure 2, the experimental CO}_2 \text{ solubility data measured in this study were also very close to the published data reported by Jou et al.}^{56} \quad \text{Teng et al.}^{59} \quad \text{Ma’mun et al.}^{59} \quad \text{and Shen and Li.}^{57} \quad \text{The experimental data of CO}_2 \text{ solubility with uncertainty in single MEA and single AMP aqueous solutions are in the Supporting Information.} \quad \text{In this study, the newly measured CO}_2 \text{ solubility in the blended amine aqueous solution of MEA and AMP was presented according to the blending ratio over a wide temperature range and compared with previously published literatures.}^{1,11} \quad \text{As can be seen in Figure 3, the experimental CO}_2 \text{ solubility data at 323.15 and 373.15 K obtained in this work were in good agreement with the literature of Li and} \end{align*} \]
Table 3. The parity plot, depicted in Figure 5, shows the SMAPE between the experimental and estimated values of the CO₂ partial pressure. In most cases, the calculated partial pressures of CO₂ seem to be in good agreement with the experimental data. As can be seen in Figure 4, the electrolyte NRTL model describes the experimental data well at low concentration of AMP rather than at high concentration of AMP. The obtained SMAPE value was 43.90% in various blending ratios (9:21, 15:15, and 21:9 wt % MEA and AMP) and a wide range of temperatures (323.15−383.15 K) and partial pressures (0−350 kPa).

Mole Fraction and pH Change in the Liquid Phase. Figure 6 shows the liquid-phase mole fraction of 10 components (three molecules, three cations, and four anions) except for the H₂O molecule in the 15 wt % MEA and 15 wt % AMP blended amine system at 323.15 K. To calculate the mole fractions or molar concentrations, all the equilibrium equations, material balances, and charge balance must be solved simultaneously with extremely high accuracies. As can be seen in Figure 6, the components that predominantly affect the CO₂ solubility behavior are amine molecules (MEA, AMP), protonated amine cations (MEA⁺H, AMPH⁺), bicarbonate (HCO₃⁻), and carbamate (MEACOO⁻) anions. In the loading ratio range 0−0.5, the mole fractions of MEA and AMP decrease, whereas those of carbamate, MEAH⁺, and AMPH⁺ increase. In the case of MEACOO⁻, the mole fraction decreases slightly in the loading ratio of 0.3−0.5. If the loading ratio is about 0.5 or more, the carbamate concentration decreases, whereas the CO₂ concentration starts to increase. Also, the mole fraction of AMPH⁺ increases slowly, whereas the concentrations of bicarbonate and MEAH⁺ increase more sharply than AMPH⁺. Before the absorption of CO₂, the OH⁻ concentration is much higher than the H₃O⁺ concentration because of the basicity of the amines. When the CO₂ loading ratio increases, both the H₂O⁺ concentration and pH increase. A precise calculation of the H₂O⁺ concentration is key to predicting the reactor pH and to designing the CO₂ absorption process.

Figure 7a,b shows changes in pH with the CO₂ loading ratio at each temperature (323.15, 373.15, and 383.15 K) and amine mass fraction (9:15, 15:15, and 9:21 wt % MEA and AMP), respectively. Equation 32 was used to calculate the pH by using the obtained activity coefficient and mole fraction of H₂O⁺.

\[
\text{pH} = -\log_{10}\left(x_{\text{H}_3\text{O}^+} \gamma_{\text{H}_3\text{O}^+}\right)
\]  

In most cases, the higher the temperature and mass fraction of MEA, the stronger the acidity of the blended amine aqueous solution. The pH decreases rapidly when the CO₂ loading ratio is below 0.1 (Figure 7). Then, it decreases even more sharply when the CO₂ loading ratio is over 0.5 rather than at low temperatures (323.15 K) and when the low MEA mass fraction is excluded (Figure 7a,b, respectively). When the amine mass fraction is 15:15 wt % MEA and AMP, the pH range is approximately 9−12.5 at low temperatures and 8.8−11 at high temperatures (Figure 7a). If the temperature is 323.15 K, the pH range is approximately 9.2−12.3 at 9:21 wt % MEA and AMP and 8.9−12.4 for other values of MEA and AMP mass fractions (Figure 7b).

Heat of Absorption. It is important to accurately calculate the heats of CO₂ absorption for estimating the regeneration energy in amine wet scrubbing processes. The heats of CO₂ absorption are strongly dependent on the temperature and composition of the amine aqueous solution.
absorption can be estimated using the Gibbs–Helmholtz relationship.

\[
\Delta H_{\text{abs,CO}_2} = \left( \frac{\Delta H_{\text{abs,CO}_2}}{R} \right) \left( \frac{\text{d}P}{\text{d}(1/T)} \right)_{\text{CO}_2} 
\]

(33)

Figure 8a shows the heats of CO2 absorption with the CO2 loading ratio at each temperature (323.15 K, 373.15 K, and 383.15 K), while Figure 8b shows those at each amine mass fraction (9:21, 15:15, and 21:9 wt % MEA and AMP). The range of heat of absorption is approximately 30–120 kJ/mol CO2. Kim et al.\textsuperscript{20,35,60} also reported the behavior of heat of absorption in aqueous solutions of various amine solutions including MEA and AMP. The range of heat of absorption in single MEA aqueous solutions was 80–120 kJ/mol CO2 at low CO2 loading ratio and decreased sharply when CO2 loading ratio was about 0.5. The enthalpy behavior according to temperature and CO2 loading was very similar to those obtained in this study. Heat of absorption changes sharply when the range of CO2 loading ratio is 0.4–0.6. When the solution has a higher temperature and higher mass fraction of AMP, heat of absorption changes in a wider range of CO2 loading ratios (0.3–0.7).

Cyclic Capacity of CO2. Sensible heat loss occurs when the rich and lean solvent exchange the heat in the CCS process. To reduce this heat loss, the cyclic capacity must be high. In general, cyclic capacity \((g \text{ CO}_2/kg \text{ solution})\) is defined as the difference between the CO2 concentration in the rich solution and the lean solution as follows.\textsuperscript{21}

\[
\Delta \alpha = \alpha_{\text{rich}} - \alpha_{\text{lean}} 
\]

(34)

However, the meaning of cyclic capacity is slightly different for wet CO2 absorption and dry CO2 absorption at coal gas plants. Also, because different temperature and pressure conditions were used in the CCS process, the cyclic capacity will also be different in each condition. In the present study, the cyclic capacity of wet CO2 absorption–desorption, \(\Delta \alpha_{\text{CO}_2}\) (mol CO2/total mol amine), is defined as follows

\[
\Delta \alpha_{\text{CO}_2} = \alpha_{\text{CO}_2}(\text{T}_{\text{abs}}, P_{\text{CO}_2} = 15 \text{ kPa}) - \alpha_{\text{CO}_2}(\text{T}_{\text{des}}, P_{\text{CO}_2} = 100 \text{ kPa}) 
\]

(35)

where \(T_{\text{abs}}\) is the temperature in the absorption condition (313.15–333.15 K) and \(T_{\text{des}}\) is the temperature in the desorption condition (373.15–393.15 K). The value of \(\Delta \alpha_{\text{CO}_2}\) was evaluated using the absorption temperature, desorption temperature, and MEA and AMP mixing ratios (0–30:30–0 wt % MEA and AMP, fixed 70 wt % H2O) under various conditions.

Figure 9 shows the cyclic capacity, \(\Delta \alpha_{\text{CO}_2}\), based on the absorption temperature condition, 313.15–333.15 K, and the desorption temperature condition, 363.15–393.15 K, in 09:21, 15:15, 21:09 wt % MEA and AMP blended amine solutions. The cyclic capacity increases as the absorption temperature decreases or the desorption temperature increases. When the mass ratio of AMP is high and/or the mass ratio of MEA is low, the cyclic capacity increases steeply and changes rapidly.

**Table 3. Partial Pressure \(P\) of CO2 in MEA + AMP + H2O**

| \(\alpha\) | \(P\) (kPa) | uncertainty | \(\alpha\) | \(P\) (kPa) | uncertainty |
|----------|-------------|-------------|----------|-------------|-------------|
| 0.122    | 1.503       | 0.00275     | 0.115    | 3.291       | 0.00349     |
| 0.267    | 2.520       | 0.00700     | 0.223    | 15.819      | 0.00529     |
| 0.403    | 5.674       | 0.00448     | 0.314    | 43.204      | 0.00506     |
| 0.544    | 18.998      | 0.00679     | 0.393    | 85.695      | 0.00358     |
| 0.655    | 78.580      | 0.01068     | 0.441    | 135.429     | 0.00645     |
| 0.707    | 165.705     | 0.01930     | 0.488    | 210.491     | 0.00574     |
| 0.738    | 249.113     | 0.01550     | 0.520    | 278.752     | 0.00646     |
| 0.782    | 333.458     | 0.00234     | 0.582    | 326.037     | 0.00478     |

\(\alpha\) CO2 loading ratio defined as the mol of CO2/total mol of amine, H2O balanced (70 wt % H2O).

\(\Delta \alpha_{\text{CO}_2} = \alpha_{\text{CO}_2}(\text{T}_{\text{abs}}, P_{\text{CO}_2} = 15 \text{ kPa}) - \alpha_{\text{CO}_2}(\text{T}_{\text{des}}, P_{\text{CO}_2} = 100 \text{ kPa})\)
Namely, the line intervals of contour become shorter when the mass ratios of MEA become lower. Based on these results, cyclic capacity was estimated with the blending ratio of blended amine of MEA and AMP. Figure 10 shows the cyclic capacity behaviors of the MEA and AMP blended amine solutions (70 wt % H2O) at (a) three absorption temperatures (313.15, 323.15, and 333.15 K) and one fixed desorption temperature (383.15 K) and (b) one fixed absorption temperature (323.15 K) and three desorption temperatures (373.15, 383.15, and 393.15 K). To compare the absorption and desorption temperature changes, the same condition (T_{abs} = 323.15 K and T_{des} = 383.15 K) is used, as indicated by the thick line in Figure 10a,b. Under typical absorption and desorption (stripping) conditions, the cyclic capacity of CO2 in the MEA and AMP blended aqueous solutions is in the range of 0.07−0.53 mol CO2/total mol amine. The cyclic capacity is high in low-concentration MEA and high-concentration AMP. When the MEA mass concentration is 0−10 wt % (AMP mass concentration: 30−20 wt %), the cyclic capacity decreases sharply. When the mass fraction of AMP is high, the increase of cyclic capacity is consistent with the results of Figure 9. In contrast, in the other range of MEA mass fraction, the cyclic temperature (383.15 K) and (b) one fixed absorption temperature (323.15 K) and three desorption temperatures (373.15, 383.15, and 393.15 K).

Figure 4. Experimental data (point) and estimated CO2 partial pressure (line) at different temperatures: (a) 9 wt % MEA:21 wt % AMP; (b) 15 wt % MEA:15 wt % AMP; (c) 21 wt % MEA:09 wt % AMP.

Figure 5. Parity plots for experimental data and simulated results of the partial pressure of CO2 in MEA and AMP blended amine solutions over the entire temperature range (circle closed: 9 wt % MEA, 21 wt % AMP; square closed, gray: 15 wt % MEA, 15 wt % AMP; diamond open: 21 wt % MEA, 9 wt % AMP).

Figure 6. Model-predicted liquid-phase concentration of each species of i for 15 wt % MEA, 15 wt % AMP blended amine at 323.15 K.  

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capacity is decreased slightly. When only the cyclic capacity based on the mixing ratio of MEA and AMP is considered, it is more advantageous to use a single AMP aqueous solution than MEA and AMP mixed amine aqueous solutions (Figure 10a,b). The above results were obtained because the cyclic capacity was calculated only in terms of equilibrium. For a more accurate calculation of the cyclic capacity, kinetic studies, experiments, and modeling should also be considered. To design the wet CO2 absorption processes, the properties of blended amines, such as equilibrium, kinetics, mass transfer, operability, material and operating costs, and corrosiveness, must be considered simultaneously.

**CONCLUSIONS**

CO2 solubility in MEA and AMP blended amine aqueous solution was experimentally and theoretically investigated at different amine mixing ratios (MEA/AMP/H2O = 9:21:70, 15:15:70, and 21:9:70 wt %) and working temperature conditions (323.15, 373.15, and 383.15 K). The successive substitution method was used for calculating the mole fractions of all species (four molecules, three cations, and four anions). To predict CO2 solubility in a blended aqueous solution of MEA and AMP, 11 equations (six equilibrium equations, four mass balances, and one charge balance) were solved simultaneously with extremely low relative tolerances (under 10−7). The electrolyte NRTL model was used to investigate the liquid-phase nonideality. The binary parameter of the electrolyte NRTL model was extracted from the entire experimental range. The obtained SMAPE value was 43.90%. Since those parameters were obtained by applying a wide range of temperature and mixing ratio conditions, the thermodynamic models developed in this study can be used for process simulation. Using the abovementioned models with the obtained binary parameters, the CO2 partial pressure in the gas phase and liquid-phase concentrations for all components including carbamates, pH, and heats of CO2 absorption were estimated for the CO2 loading ratio ranging from 0 to 0.8. Notably, the concentration behavior of chemical species in the liquid phase rapidly changes from the point where the CO2 loading ratio is near 0.5. The pH was in the range of 8.8−12.5 in the given CO2 loading range, but it rapidly decreased after the CO2 loading ratio becomes 0.5, especially in the low-temperature region. Similarly, in the region of low-CO2 loading ratio, the heat of absorption is in the range of 110−120 kJ/mol, but in the high-CO2 loading ratio region, after a CO2 loading ratio of 0.5 is reached, the heat of absorption rapidly lowered...
Finally, the cyclic capacity of CO$_2$ using the temperature and mixing ratio of MEA and AMP was examined. Cyclic capacity was high at low absorption temperature and high desorption temperature. In the region with the high AMP ratio, high cyclic capacity of 0.28−0.52 was observed, while in the region with the high ratio of MEA from the region of 15:15 wt % MEA and AMP, relatively low cyclic capacity of 0.05−0.1 was obtained. For a more accurate calculation of cyclic capacity, equilibrium and kinetic aspects should be considered simultaneously. If the results of the CO$_2$ solubility model in the blended amine aqueous solution obtained through this study are utilized, the accuracy of the process simulation can be improved. In addition, it will be possible to provide insights into CO$_2$ solubility, cyclic capacity, heat of absorption, pH, etc. according to the blending ratios.

**ASSOCIATED CONTENT**

*Supporting Information*

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c04046.

Detailed experimental setup and equipment, experimental CO$_2$ solubility data of single amine aqueous solution, successive substitution method, electrolyte NRTL model, and binary parameters of the electrolyte NRTL model (PDF)

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Figure 9. Cyclic capacity (Δα$_{CO_2}$) according to the absorption temperature (T$_{abs}$ 313.15−333.15 K) and the desorption temperature (T$_{des}$ 363.15−393.15 K) in (a) 09:21 wt %, (b) 15:15 wt %, and (c) 21:09 wt % MEA and AMP blended amine solutions.

Figure 10. Cyclic capacity (Δα$_{CO_2}$) of amine mass fraction (a) in absorption temperatures (313.15−333.15 K) at a constant desorption temperature of 383.15 K and (b) in desorption temperatures (373.15−393.15 K) at a constant absorption temperature of 323.15 K.
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Notes
The authors declare no competing financial interest.

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■ ABBREVIATIONS
AMP 2-amino-2-methyl-propanol
CCS carbon capture and storage
DEA diethanolamine
DIPA diisopropanolamine
MDEA methyl diethanolamine
MEA monoethanolamine
NRTL nonrandom two liquid
SMAPE symmetric mean absolute percentage error
TEA triethylamine
VLE vapor–liquid equilibrium.

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