

Absorption of carbon dioxide into aqueous solution of 2-amino-2-methyl-1-propanol and 1, 8-diamino-p-menthane

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Abstract—Carbon dioxide was absorbed into an aqueous solution containing two reactants of 2-amino-2-methyl-1-propanol (AMP) and 1,8-diamino-p-menthane (DAM) in a stirred semi-batch tank with a planar gas-liquid interface within a range of 0-3.0 kmol/m³ of AMP, 0-0.2 kmol/m³ of DAM, and 298.15-323.15 K at 15% of CO₂ and 101.3 kPa. Diffusivity, Henry constant and mass transfer coefficient of CO₂ in the mixed solution of AMP and DAM were used to calculate the theoretical enhancement factor of CO₂, which was obtained by an approximated solution of mass balances with the instantaneous and fast regime in CO₂-AMP-DAM system. The method of the classification of the chemical regime in the heterogeneous system was used to determine the enhancement factor by adding DAM under the limited concentration of AMP.

Key words: Absorption, Carbon Dioxide, 2-Amino-2-methyl-1-propanol, 1,8-Diamino-p-menthane

INTRODUCTION

Carbon dioxide in the flue gas generated as a result of combustion of fossil fuel in thermal power plants, etc., is the main cause of global environmental problems such as global warming and acid rain. One of the conventional methods to achieve the removal and recovery of CO₂ on an industrial scale is the chemical absorption method. One important requirement involved with this method is to develop absorbents with high absorption rate and capacity. Industrially important chemical absorbents are alkanolamines [1]. Many studies have been done on the mechanisms and kinetics of the reaction of CO₂ with various alkanolamines, employing simple mass balance analysis and resulting in the zwitterion mechanism proposed by Caplow [2] and Danckwerts [3]. Some discrepancies remain according to the reaction mechanism [4] through the types of amines, gas/liquid contactor, and analysis method used for the rate data, the order of the overall reactions and the rate constants. Recently, a group of sterically hindered amines were developed [5], providing a high capacity of 1.0 mol of CO₂/mol of amine and a relatively high absorption rate, even at high CO₂ loading. One such example is 2-amino-2-methyl-1-propanol (AMP), a sterically hindered form of monoethanolamine.

Recently, mixing of a primary or secondary alkanolamine with an alkaline component has been suggested to capitalize on the advantages of each amine. Shrier and Danckwerts [6] suggested that the rate of CO₂ absorption into potash solutions could be significantly increased by addition of small amounts of amine promoters. The blended amine solvents combine the higher equilibrium capacity of the tertiary amine for CO₂ with higher CO₂ reaction rate of a primary

or secondary alkanolamine [7-11]. Amines with more than two amine groups, such as a piperazine derivative with three amine groups [12] or 1,8-diamino-p-menthane (DAM) with two amine groups [13] were used as absorbents of CO₂, and the blended amine solvents with these amines could be used to provide a high capacity of CO₂.

To obtain the absorption mechanism of CO₂ into blends of AMP and DAM, the kinetics of CO₂ into AMP or DAM have been investigated as follows. Carbon dioxide and sulfur dioxide were simultaneously absorbed into aqueous 2-amino-2-methyl-1-propanol [14]; carbon dioxide and sulfur dioxide were simultaneously absorbed into aqueous 1,8-diamino-p-menthane [15]; simultaneous absorption of carbon dioxide and nitrogen dioxide into aqueous 2-amino-2-methyl-1-propanol [16]; simultaneous absorption of carbon dioxide, sulfur dioxide, and nitrogen dioxide into aqueous 2-amino-2-methyl-1-propanol [17]; simultaneous absorption of carbon dioxide, sulfur dioxide, and nitrogen dioxide into aqueous 1,8-diamino-p-menthane [18].

In this study, the absorption rate of CO₂ into the aqueous blends of AMP and DAM was measured to analyze the experimental data using an approximated solution of the diffusion model, which is one of a series of studies containing the previous works [14-18]. DAM was added as an amine promoter to a primary reactant, AMP, where the reaction rate constant of the added reactant with CO₂ [15] is larger than that of the primary reactant [14]. This study makes a new attempt for removal of the gases emitted from power plant flues for the effective capture and utilization of carbon dioxide.

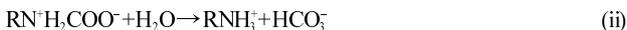
THEORY

The zwitterion mechanism originally proposed by Caplow [2] and later reintroduced by Danckwerts [3] and da Silva and Svendsen [19] is generally accepted as the reaction mechanism in the ab-

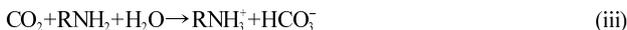
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sorption of CO₂ into aqueous AMP (RNH₂) as follows:



With overall reaction being:



And reactions in the absorption of CO₂ into aqueous DAM (H₂NRNH₂) are as follows:



Overall reaction being:



where R presents $\text{—C(H)}_2\text{—C(H)}_2\text{—}$ $\text{—C(H)}_2\text{—C(H)}_2\text{—C(H)}_2\text{—}$ $\text{—C(H)}_2\text{—C(H)}_2\text{—C(H)}_2\text{—}$.

The irreversible reactions between CO₂ and the reactant (C), as shown in reactions (iii) and (vii) may be formulated as follows:



where A presents CO₂, B₁, AMP, and B₂, DAM, respectively. ν_1 and ν_2 are 1 [14] and 0.5 [15], respectively.

The following assumptions are made to set up the mass balance of species j:

1) Henry's law holds, 2) the isothermal condition prevails, 3) species of AMP and DAM is a nonvolatile solute, and 4) the reactions between A and B₁, and between A and B₂ are first order with respect to A [14,15] and first order with respect to B₁ [14] and B₂ [15], respectively.

The mass balances of A and species j using the film theory accompanied by the chemical reaction and the boundary conditions are given as follows:

$$D_A \frac{d^2 C_A}{dz^2} = k_1 C_A C_{B1} + k_2 C_A C_{B2} \quad (\text{1})$$

$$D_{B1} \frac{d^2 C_{B1}}{dz^2} = \nu_1 k_1 C_A C_{B1} \quad (\text{2})$$

$$D_{B2} \frac{d^2 C_{B2}}{dz^2} = \nu_2 k_2 C_A C_{B2} \quad (\text{3})$$

$$z = 0; C_A = C_{A_i}, C_{B1} = C_{B1_i}, C_{B2} = C_{B2_i}, \frac{dC_{B1}}{dz} = \frac{dC_{B2}}{dz} = 0 \quad (\text{4})$$

$$z = \delta; C_A = 0, C_{B1} = C_{B1_o}, C_{B2} = C_{B2_o} \quad (\text{5})$$

The molar flux of A is defined as follows:

$$N_A = -D_A \left. \frac{dC_A}{dz} \right|_{z=0} \quad (\text{6})$$

As shown in the governing Eqs. (1)-(3) and the boundary conditions of Eqs. (4) and (5), the flux of CO₂ is influenced by physico-chemical properties (reaction rate constant, solubility, diffusivity and mass transfer coefficient) and the experimental variables (partial pressure of CO₂, concentration of reactant and temperature).

Depending on the relative rates of diffusion and reaction by using the dimensionless groups listed below, for convenience, the systems of gas absorption into one reactant may be classified into four regimes: very slow reaction, slow reaction, fast reaction, and instantaneous reaction using two groups, such as the dimensionless group (M_j), defined as the ratio of the amount of CO₂ reacting in the film to that in the bulk and the enhancement factor group (E_{ij}) based on the film theory [20].

M_j and E_{ij} are presented, respectively, as follows:

$$M_j = \frac{\sqrt{D_j k_j B_{j_o}}}{k_L} \quad (\text{7})$$

$$E_{ij} = 1 + \frac{B_{j_o} D_{Bj}}{z_j D_{A_i} D_A} \quad (\text{8})$$

In case of two reactants, it is not simple to obtain the enhancement factor (β_A) by classifying the reaction regime because of four dimensionless groups of M_1 , M_2 , E_{11} and E_{22} , and it is very tedious and complicated to solve the solutions of the simultaneous differential equations of Eqs. (1), (2) and (3) to obtain the concentration profile of CO₂ in the film.

If the reaction between A and B₂ of the added reactant is instantaneous compared to the diffusion rates of A and B₂, and the reaction between A and B₁ of the primary reactant is a fast reaction compared to the diffusion rates of A and B₁, the conditions for the instantaneous reaction and fast reaction are given as follows, respectively:

$$M_2 \gg E_{22} \quad (\text{9})$$

$$M_1 \ll E_{11} \quad (\text{10})$$

And, β_A is derived as follows [20]:

$$\beta_A = \frac{M_1}{\tanh\left(M_1 \frac{\lambda}{\delta}\right)} \quad (\text{11})$$

where, λ is defined by the reaction plane location at $C_A=0$ and $C_{B2}=0$ and δ the thickness of diffusion film based on the film theory. This λ/δ is obtained by the following equation:

$$\frac{1 - \frac{\lambda}{\delta}}{\sinh\left(M_1 \frac{\lambda}{\delta}\right)} = \frac{E_{22} - 1}{M_1} \quad (\text{12})$$

As shown in Eqs. (11) and (12), only two groups of M_1 and E_{22} in case of the conditions mentioned above are required to classify the reaction regime.

Using Eq. (11), the flux of CO₂ is obtained as follows:

$$N_A = k_L C_{A_i} \beta_A \quad (\text{13})$$

EXPERIMENTAL

1. Chemicals

The chemicals, AMP and DAM, were of reagent grade more than 99% (Aldrich Chem. Co.) and used without further purification, Distilled water was used and the purity of N₂, and CO₂ was more than 99.9%.

2. Absorption of CO₂

Absorption experiments were carried out in an agitated vessel [16,21]. The absorption vessel was constructed of glass with an inside diameter of 0.073 m and a height of 0.151 m. Four equally-spaced vertical baffles, each one-tenth of the vessel diameter in width, were attached to the internal wall of the vessel. The gas and liquid phase were agitated with an agitator driven by a 1/4 Hp variable speed motor. A straight impeller 0.034 m in length and 0.05 m in width was used as the liquid phase agitator and located at the middle position of the liquid phase. The surface area was calculated as a ratio of the volume of added water to the measured height of water in the absorber, and its value was 40.947 cm². The gas and liquid in the vessel were agitated at a speed of 50 rpm. The value of the cumulative volume of the soap bubble was measured by a soap bubble for the change of absorption time to obtain the absorption rate of CO₂. Each experiment was duplicated at least once under identical conditions. It was assumed that the volumetric rising rate of the soap bubble in the soap bubbler attached to the absorption vessel was equal to the value of the absorption rate of gases. The gaseous compositions of CO₂ at the inlet of the absorber were measured by gas chromatography (PTFE, 6 feet x 1/8 inch OD, Chromosorb 107, 80/100; Detector: TCD at 100 °C; He: 18 cm³/min; retention time of N₂: 2.05 min; CO₂: 6.13 min). The absorption experiments were carried out in a range of 0-3.0 kmol/m³ of AMP, 0-0.2 kmol/m³ of DAM, and 298.15-323.15 K at 15% mole fraction of CO₂ and 101.3 kPa to measure the molar flux of CO₂ in a solution containing AMP and DAM.

A sketch of the experimental set up and a typical experimental run were presented in the previous work [16].

3. Physicochemical Properties

The physicochemical properties, required to obtain M₁ and E₁₂ in Eqs. (11) and (12), are obtained as follows. Both the solubility and diffusivity of CO₂ in the mixed solution of AMP and DAM are obtained using an approximate method of the nitrous oxide analogy as follows:

The Henry constants of N₂O and CO₂ in water are obtained from the following empirical equations [22]:

$$H_{N_2O}^o = 8.547 \times 10^6 \exp\left(-\frac{2284}{T}\right) \tag{14}$$

$$H_{Aw} = 2.8249 \times 10^6 \exp\left(-\frac{2044}{T}\right) \tag{15}$$

The Henry constant of N₂O in the solution was estimated accordingly [23]:

$$H_{N_2O} = (5.52 + 0.7C_{Co}) \times 10^6 \exp\left(-\frac{2166}{T}\right) \tag{16}$$

where, C_{Co} is the total concentration of AMP and DAM.

The Henry constants of CO₂ in the solution were estimated by the N₂O analogy as follows:

$$H_A = H_{Aw} \frac{H_{N_2O}}{H_{N_2O}^o} \tag{17}$$

The solubility (C_{Ai}) of CO₂ at a partial pressure of P_A was estimated as follows:

$$P_A = H_A C_{Ai} \tag{18}$$

The diffusivities of CO₂ in water were obtained from the following empirical equations [22]:

$$D_{Aw} = 2.35 \times 10^{-6} \exp\left(-\frac{2119}{T}\right) \tag{19}$$

The diffusivities of CO₂ in the solution were estimated as follows [24]:

$$D_A = D_{Aw} \left(\frac{\mu_w}{\mu}\right)^{2/3} \tag{20}$$

The diffusivity (D_{B1} and D_{B2}) of AMP and DAM in the mixed solution of AMP and DAM was estimated by the method of Wilke [25].

Viscosity of the aqueous solution was measured with a Brookfield viscometer (Brookfield Eng. Lab. Inc., USA).

The liquid-side mass transfer coefficient (k_{Lw}) of CO₂ in water was measured by using the absorption rates of pure CO₂ at a given temperature and 50 rpm, and 4.846, 4.497, 4.742 and 4.889×10⁻⁵ m/s at 25, 30, 40, and 50 °C, respectively.

The mass transfer coefficients (k_L) of CO₂ in the solution were calculated from the relationship between the mass transfer coefficient in water and viscosity in reference as follows [26]:

$$k_L = k_{Lw} \left(\frac{\mu_w}{\mu}\right)^{2/3} \tag{21}$$

The reaction rate constant (k₁) between CO₂ and AMP was obtained from the following equation [14]:

$$k_1 = 1.2 \times 10^{10} \exp(-5096.4/T) \tag{22}$$

The reaction rate constant (k₂) between CO₂ and DAM was obtained from the following equation [15]:

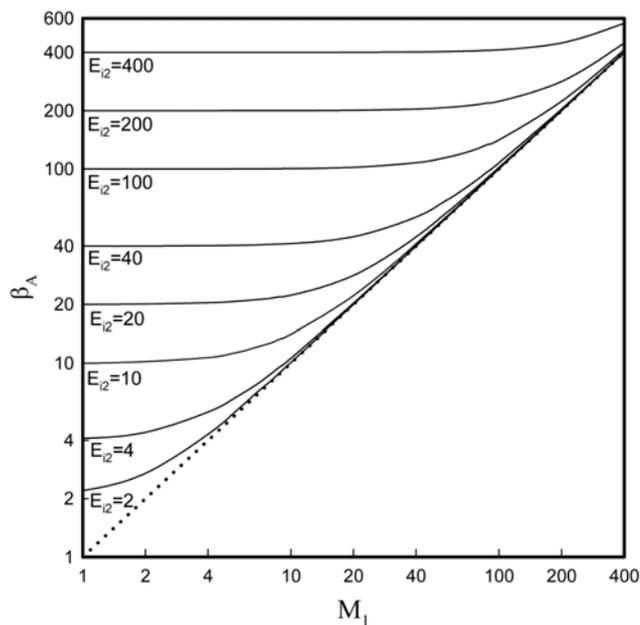


Fig. 1. Enhancement factor of CO₂ in two reactants: one instantaneous reaction and one fast reaction (solid line: the instantaneous reaction between CO₂ and DAM, and fast reaction between CO₂ and AMP, dotted line: fast reaction between CO₂ and DAM and fast reaction between CO₂ and AMP in the system of CO₂-AMP-DAM, respectively).

$$k_2 = 1.72 \times 10^{12} \text{ Exp}(-5747/T) \quad (23)$$

RESULTS AND DISCUSSION

In case that the reaction between CO_2 and the added reactant is classified as instantaneous reaction, β_A is influenced by only M_1 and E_{i2} . To observe the effect of M_1 and E_{i2} on β_A , β_A is estimated against various M_1 with a parameter of E_{i2} using Eq. (7) and (8), and then, plotted in Fig. 1.

As shown in Fig. 1, β_A increases with increasing M_1 and E_{i2} , and it approaches an asymptote presented as a dotted line, which is equivalent to the fast reaction regime, that is, the added reactant does not exist in the film region. Because β_A exists at the upper region of the asymptote, β_A can be increased by the added reactant with the instantaneous reaction regime. Also, this dimensional analysis gives a shifting condition to the instantaneous reaction regime from the fast reaction regime, that is, the minimum E_{i2} at a given M_1 .

To classify the chemical regime, M_j and E_{ij} are obtained by Eqs. (7) and (8) for various values of B_{1o} , B_{2o} and T using the physico-chemical properties, such as C_{Aib} , D_A , D_{B1} , D_{B2} , k_L , through Eqs. (14)-(23). The four groups for the various B_{2o} at the typical conditions of 0.5 kmol/m^3 of AMP and 303.15 K are plotted in Fig. 2.

As shown in Fig. 2, Eqs. (9) and (10) are satisfied for the system of CO_2 and AMP, and CO_2 and DAM, respectively. This means that the reaction of CO_2 with DAM is instantaneous and the reaction with AMP is a fast reaction. These results are similar for other concentrations of AMP and temperatures. From these results of Fig. 2, β_A can be obtained by Eq. (11) for various DAM concentrations at typical temperature of 303.15 K with a parameter of AMP concentration and presented as a solid line in Fig. 3. The various symbols represent the various concentrations of AMP. β_A at B_{2o} of zero is equivalent to the value in case of CO_2 absorption into only AMP solution. To obtain this value, the governing equations and boundary equations based on the film theory are as follows:

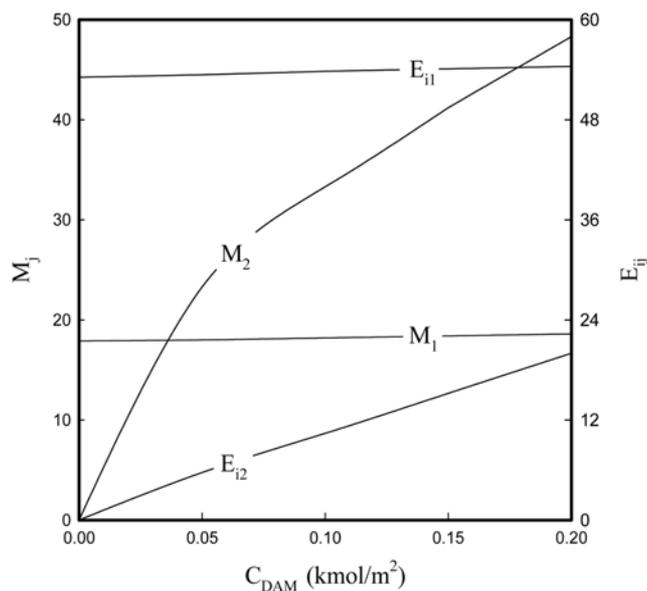


Fig. 2. M_j vs. E_{ij} for determination of the regime for validity of reactions at $C_{B1o} = 0.5 \text{ kmol/m}^3$ and 303.15 K .

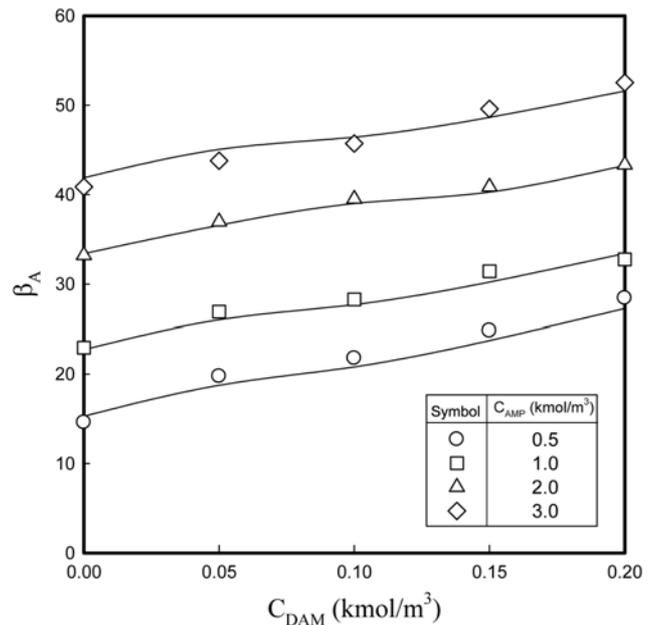


Fig. 3. Enhancement factor of CO_2 for various concentrations of DAM with parameter of AMP concentration at 303.15 K .

$$D_A \frac{d^2 C_A}{dz^2} = k_1 C_A C_{B1} \quad (24)$$

$$D_{B1} \frac{d^2 C_{B1}}{dz^2} = \nu_1 k_1 C_A C_{B1} \quad (25)$$

$$z=0; C_A = C_{Aib}, C_{B1} = C_{B1ib}, \frac{dC_{B1}}{dz} = 0 \quad (26)$$

$$z=\delta; C_A = 0, C_{B1} = C_{B1o} \quad (27)$$

β_A is estimated from the solutions of the differential equations of

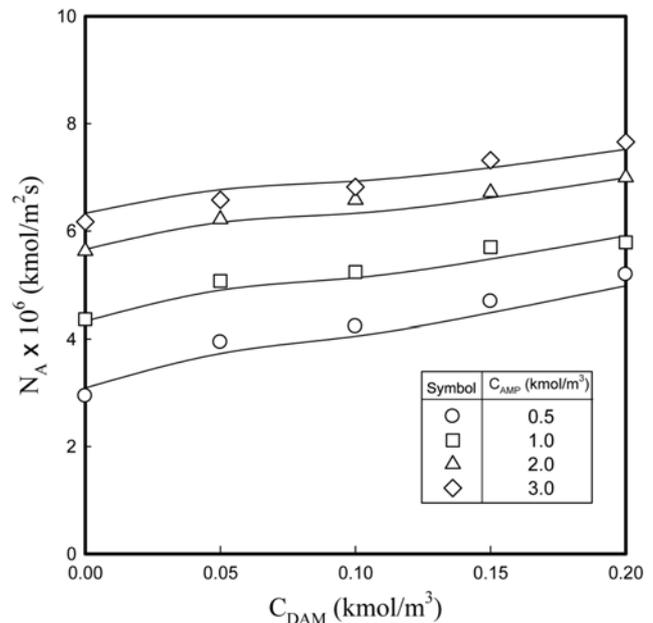


Fig. 4. The flux of CO_2 for various concentrations of DAM with parameter of AMP concentration at 303.15 K .

Eqs. (24) and (25) using a numerical method of FEMLAB®.

As shown in Fig. 3, the increase of not only B_{2o} , but B_{1o} makes β_A increase, and the experimental values approach the calculated ones very well. These results are similar for other temperatures. The results in Fig. 3 coincide with those in Fig. 1, which β_A increases with increasing of M_1 and E_2 . Also, because the reaction rate constant [15] of CO_2 with DAM is larger than that [14] of CO_2 with AMP, DAM could be used as an amine promoter to a primary reactant, AMP.

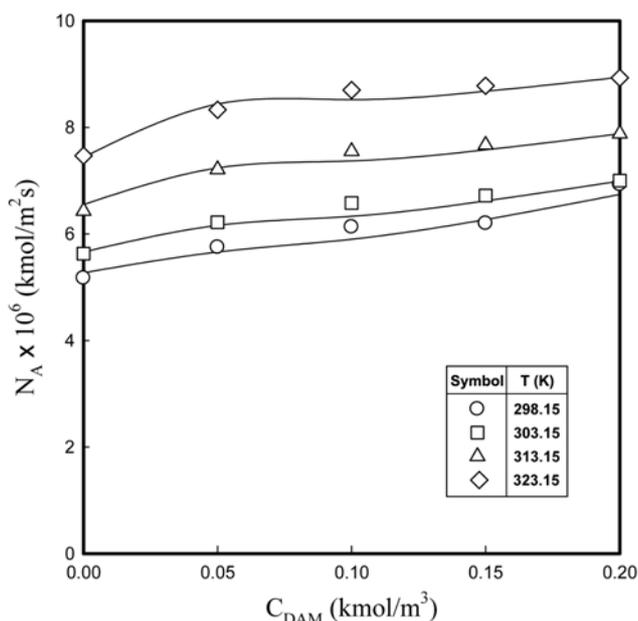


Fig. 5. The flux of CO_2 for various concentrations of DAM with parameter of temperature at AMP concentration of 2.0 kmol/m^3 .

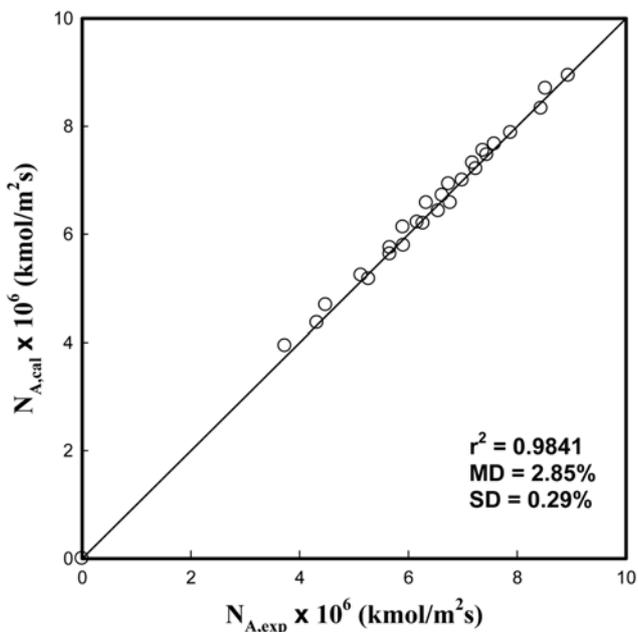


Fig. 6. Comparison of the calculated and measured molar flux of CO_2 in the system of CO_2 -AMP-DAM.

The flux of CO_2 is obtained by Eq. (13) using β_A is in Fig. 3. They are plotted against various concentrations of DAM with parameters of AMP concentration and temperature in Fig. 4 and 5, respectively.

As shown in these figures, the flux increases with increasing DAM, AMP concentration and temperature. The experimental values approach the calculated ones very well. From the results in Fig. 4, the added reactant could be used to increase the flux of CO_2 in the system of CO_2 and AMP.

$N_{A,cal}$ was plotted vs. $N_{A,exp}$ in Fig. 6 under all experimental conditions of 35 mentioned above.

As shown in Fig. 6, the observed values of the molar flux agree well with the calculated values, where the classification method to select the reaction regime in the heterogeneous system becomes a very convenient tool.

CONCLUSIONS

Carbon dioxide was absorbed into an aqueous solution containing two reactants of 2-amino-2-methyl-1-propanol (AMP) and 1,8-diamino-p-menthane (DAM) in a stirred semi-batch tank with a planar gas-liquid interface within a range of $0\text{--}3.0 \text{ kmol/m}^3$ of AMP, $0\text{--}0.2 \text{ kmol/m}^3$ of DAM, and $298.15\text{--}323.15 \text{ K}$ at the fixed 15% of CO_2 and 101.3 kPa . Physicochemical properties, used to calculate the theoretical the enhancement factor of CO_2 , were obtained from the reference data measured by N_2O analogy. The theoretical molar fluxes of CO_2 were calculated by an approximated solution of mass balances with the instantaneous reaction regime with respect to both CO_2 and DAM and fast reaction regime with respect to both CO_2 and AMP in CO_2 -AMP-DAM system. From the viewpoint of the absorption capacity of the absorbent, the method of the classification of the chemical regime could be used to select the added reactant of DAM in CO_2 absorption of AMP solution, i.e., to increase the enhancement factor by using the added reactant under the limited concentration of the primary reactant.

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NOMENCLATURE

- C_j : concentration of species, j [kmol/m^3]
- C_{Ai} : solubility of CO_2 [kmol/m^3]
- D_j : diffusivity of species, j [m^2/s]
- H_A : henry constant of CO_2 [$\text{atm}\cdot\text{m}^3/\text{kmol}$]
- k_j : reaction rate constant of species, j [$\text{m}^3/\text{kmol}\cdot\text{s}$]
- k_L : liquid-side mass transfer coefficient of species, j [m/s]
- MD : mean deviation
- N_A : molar flux of CO_2 [$\text{kg mol/m}^2\cdot\text{s}$]
- P_A : partial pressure of CO_2 [atm]
- r^2 : correlation coefficient
- SD : standard deviation
- T : temperature [$^\circ\text{K}$]
- z : diffusion coordinate of gas [m]

z_L : film thickness [m]

Greek Letters

β_j : enhancement factor of species, j

μ : viscosity of liquid [$N \cdot s/m^2$]

ν_j : stoichiometric coefficients of species, j

Subscripts

A : CO₂

B₁ : AMP

B₂ : DAM

j : species

o : feed

W : water

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