

Simultaneous absorption of CO₂ and SO₂ into aqueous AMP/NH₃ solutions in binary composite absorption system

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Abstract—To enhance the absorption rate for CO₂ and SO₂, aqueous ammonia (NH₃) solution was added to an aqueous 2-amino-2-methyl-1-propanol (AMP) solution. The simultaneous absorption rates of AMP and a blend of AMP+NH₃ for CO₂ and SO₂ were measured by using a stirred-cell reactor at 303 K. The process operating parameters of interest in this study were the solvent and concentration, and the partial pressures of CO₂ and SO₂. As a result, the addition of NH₃ solution into aqueous AMP solution increased the reaction rate constants of CO₂ and SO₂ by 144 and 109%, respectively, compared to that of AMP solution alone. The simultaneous absorption rate of CO₂/SO₂ on the addition of 1 wt% NH₃ into 10 wt% AMP at a p_{A1} of 15 kPa and p_{A2} of 1 kPa was 5.50×10^{-6} kmol m⁻² s⁻¹, with an increase of 15.5% compared to 10 wt% AMP alone. Consequently, the addition of NH₃ solution into an aqueous AMP solution would be expected to be an excellent absorbent for the simultaneous removal of CO₂/SO₂ from the composition of flue gas emitted from thermoelectric power plants.

Key words: Carbon Dioxide, Sulfur Dioxide, Simultaneous Absorption, 2-Amino-2-methyl-1-propanol, Ammonia

INTRODUCTION

The atmospheric concentrations of CO₂ and other greenhouse gases have greatly increased since the Industrial Revolution. The Inter-governmental Panel on Climate Change (IPCC) has published a report that concluded that human activities result in the production of greenhouse gases (CO₂, CH₄, CFCs and N₂O), which significantly contribute to global warming. Among these gases, CO₂ has the greatest adverse impact on the observed greenhouse effect, causing approximately 55% of the observed global warming [1]. In this situation, most nations will willingly suffer economical burdens as shown by the nations signing the Kyoto Protocol, which took effect from February 2005. Therefore, innovative technology for the simultaneous removals of CO₂ and SO₂, which are discharged in great quantities from fossil fuel-fired power plants and incineration facilities, must be developed to reduce these burdens.

A number of techniques have been used for the separation of CO₂ from the flue gas of fossil fuel-fired power plants. Of these techniques, a chemical absorption process is commercially operated using aqueous alkanolamine solution. A different class of chemical absorbents, the sterically hindered amine, such as 2-amino-2-methyl-1-propanol (AMP), has been proposed as a commercially attractive new CO₂ absorbent because of their advantages in absorption capacity, absorption rate, degradation resistance, and regeneration energy [2]. Also, ammonia (NH₃) solution can be an alternative for the control of CO₂ and SO₂ emitted from flue gas due to its high absorption capacity, fast absorption rate and low corrosion potential [3]. Recently, to utilize the advantages of each amine, blended

amines have been suggested, such as blends of primary and tertiary amines [4-11].

In this study, to enhance the absorption rates of CO₂ and SO₂, NH₃ solution was added to an aqueous AMP solution. The simultaneous absorption rates of AMP and a blend of AMP+NH₃ for CO₂ and SO₂ were measured with a stirred-cell reactor at 303 K. The process operating parameters of interest in this study were the solvent and concentration, and partial pressures of CO₂ and SO₂. These operating parameters were tested to determine their effects on the absorption kinetics of the reactants in the gas-liquid phases.

THEORETICAL BACKGROUND

1. Absorption Mechanism

The reaction of CO₂ with the amino group of AMP could result in three possible reactions: the formation of carbamate, the formation of bicarbonate, and the reversion of carbamate to bicarbonate or formation of the carbonate ion [4].

Carbamate formation:



Bicarbonate formation:



The chemical reactions between CO₂ and NH₃ can be expressed by the following reactions [12,13]:



The wet method of ammonia scrubbing of the flue gas to capture CO₂ produces ammonium carbonate ((NH₄)₂CO₃) and ammonium

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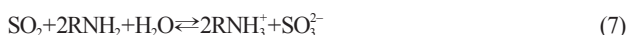
bicarbonate (NH_4HCO_3) [3,14].

The chemical reactions between SO_2 and AMP can be assuming that the SO_2 was combined with reaction occurring into water [15] and aqueous alkali solution [16].

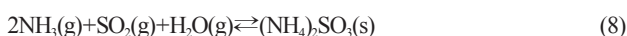
The reaction is as follows:



The overall reaction of amine- SO_2 is as follows:



The ammonium sulfite ($(\text{NH}_4)_2\text{SO}_3$) and ammonium bisulfite (NH_4HSO_3) are formed by reacting NH_3 with SO_2 at 25 °C and 1 atm [17].



2. Absorption Rate

The influence of all the chemical reactions between dissolved gas and reactants in solution on the absorption kinetics is usually expressed by an "enhancement factor" E over physical absorption: [4]

$$N_A = Ek_L C_A^* = EN_{A0} \quad (10)$$

Where E is a function of the Hatta number (H_u) and the instantaneous reaction enhancement factor (E_i) is defined as follows.

$$H_u = \left(\frac{2}{m+1} D_A k_{mn} (C_A^*)^{m-1} C_B^n \right)^{1/2} / k_L \quad (11)$$

$$E_i = \left(\frac{D_A}{D_B} \right)^{1/2} + \left(\frac{D_B}{D_A} \right)^{1/2} \frac{C_B}{\nu C_A^*} \quad (12)$$

The experimental conditions were selected to ensure that the absorption of gas into the amine solutions was in a region of fast pseudo-mn-order reaction within the range of H_u between 3 and E_i . If the value of H_u is within this range, E becomes equal to H_u , and the following specific absorption rate is obtained [4,6]:

$$N_A = \left(\frac{p_A}{H_A} \right)^{(m+1)/2} \left(\frac{2}{m+1} D_A k_{mn} C_B^n \right)^{1/2} \quad (13)$$

Meanwhile, the instantaneous absorption reaction regime must satisfy $H_u \gg E_i$. In this regime the specific absorption rate is given by Eq. (14):

$$N_A = k_L C_A^* \left(1 + \frac{C_B D_B}{\nu C_A^* D_A} \right) \quad (14)$$

Also, the fast pseudo-m-order reaction regime is expressed by Eq. (15) in the case of simultaneous absorption in binary systems (CO_2/SO_2) [18].

$$H_{u,A1}/E_{i,A1} + H_{u,A2}/E_{i,A2} \ll 1 + 1/E_{i,A12} \quad (15)$$

Where, $E_{i,A12} = C_B / [\nu_{A1} C_{A1}^* D_{A1} / D_B + \nu_{A2} C_{A2}^* D_{A2} / D_B]$

The instantaneous reaction regime condition of a single gas in a mixed gas is given as follows [18]:

$$H_{u,A1} \gg E_{i,A1} \quad (16)$$

$$H_{u,A2} \gg E_{i,A2} \quad (17)$$

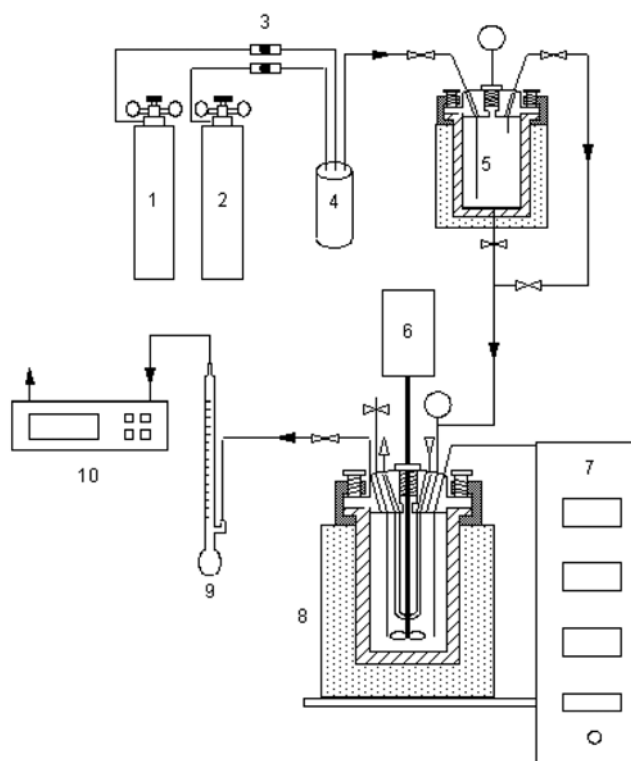


Fig. 1. Schematic of the experimental apparatus.

- | | |
|---|--|
| 1. N_2 cylinder | 7. Controller of temperature and agitation speed |
| 2. CO_2 and SO_2 cylinder | 8. Reactor (stirred-cell) |
| 3. Mass flow controller | 9. Soap bubble meter |
| 4. Mixing chamber | 10. Analyzer |
| 5. Saturator | |
| 6. Magnetic drive | |

EXPERIMENTAL METHODS

1. Materials

Analytical grade AMP and NH_3 solution, with purities of 99 and 28%, were supplied by Acros Organics (USA) and Junsei Chemical (JPN), respectively. Aqueous solutions were prepared with distilled water. The CO_2 and N_2 gases were of commercial grade, with purities of 99.99%. High purity SO_2 (99.9%) gas was also used.

2. Apparatus and Procedure

An experimental apparatus was constructed to explore the absorptions of CO_2 and SO_2 into amine solutions, as shown in Fig. 1. The reactor, with a height and ID of 160 and 95 mm, respectively, was located inside a temperature-controlled vessel, with four 5 mm wide glass plates adhering to the inner wall of the reactor as baffles. A two-blade impeller (70 mm×20 mm) was installed so as to be in the middle of the liquid level. The temperature of the reactor was measured with a K-type thermocouple, with an accuracy of ± 0.1 K. The gas flow rates were controlled by mass flow controllers (5850E, Brooks Instruments, USA). All tests were conducted with 500 mL. The stirring speed was limited to 50 rpm to keep the gas-liquid interface planar and smooth. To achieve a good gas-liquid contact, the gas was introduced on top of the reactor. The absorption rates were calculated from the difference in the amounts of gas between the inlet and outlet. A ZRF model CO_2 analyzer (Fuji Electric, JPN, 0–20 vol%) was used to measure the CO_2 gas concentration at the

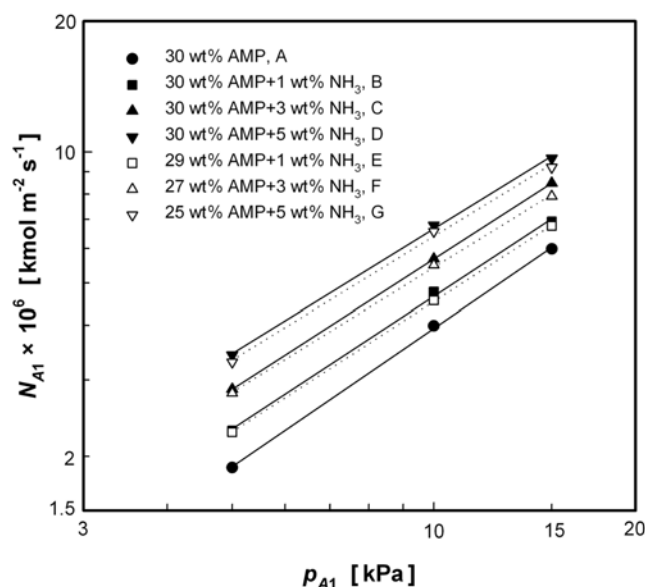


Fig. 2. The absorption rate of CO₂ with regard to the blending rate of aqueous AMP and NH₃ solutions as a function of p_{A1} at 303 K.

outlet of the reactor.

RESULTS AND DISCUSSION

1. Absorption of CO₂ into Aqueous AMP+NH₃ Solution

Fig. 2 shows the absorption rate of CO₂ with regard to the blending rate of AMP and NH₃ (wt% AMP/wt% NH₃: A, 30/0; B, 30/1; C, 30/3; D, 30/5; E, 29/1; F, 27/3; G, 25/5) as a function of CO₂ partial pressure (p_{A1} ; 5, 10, and 15 kPa) at 303 K to investigate the effect of the CO₂ absorption rate on the blending rate of aqueous AMP and NH₃ solution. The stirrer speed in all tests was 50 rpm in order to neglect the gas phase resistance and maintain a smooth, horizontal gas-liquid interface.

As shown in Fig. 2, the CO₂ absorption rate increased with increasing NH₃ concentration and p_{A1} . It was discovered that the CO₂ absorption rate on the addition of NH₃ into 30 wt% AMP solution at 15 kPa increased by 15.6 to 61.6% compared to that of the aqueous 30 wt% AMP solution without NH₃. Also, the CO₂ absorption rate of aqueous blended AMP+NH₃ solutions (E, F, and G) increased with increasing NH₃ concentration. This was due to the enhanced mass transfer because the amount of CO₂ absorbed into the gas-liquid interfacial area increased with increasing NH₃ concentration. The CO₂ absorption rates were also directly proportional to the NH₃ concentration due to the high absorption capacity for CO₂ and the additional reaction (Eqs. (3) and (4)). The slope of the straight line obtained by fitting the data to $\ln N_{A1}$ versus $\ln p_{A1}$ will give the order, m , with respect to the dissolved gas concentration C_{A1} . The plots of these data in Fig. 2 show straight lines (correlation coefficient 0.98 to 1.03) with slopes of 1.

The reaction rate constant ($k_{2,A1}$) of AMP with CO₂ at 303 K was determined to be $763.0 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$, which is different from the values of Alper ($686.6 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$ at 303 K) [5], Saha and Bandyopadhyay ($681 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$ at 301.5 K) [6] and Xu et al. ($1,059.1 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$ at 305 K) [7]. The distinction may be attributed to the

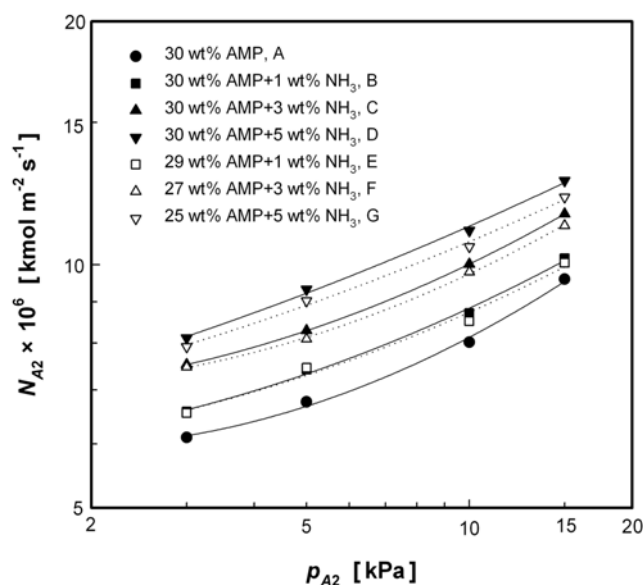


Fig. 3. The absorption rate of SO₂ with regard to the blending rate of aqueous AMP and NH₃ solutions as a function of p_{A2} at 303 K.

different reactor configurations, reaction conditions and factors ($D_A^{0.5}/H_A$) mentioned by researchers [19]. The $k_{2,A1}$ for a blend of 30 wt% AMP solution and NH₃ with CO₂ at 303 K was determined to be $1,526.7 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$, which was 2-fold higher than that of AMP without NH₃.

2. Absorption of SO₂ into Aqueous AMP+NH₃ Solution

Fig. 3 shows the absorption rate of SO₂ with regard to the blending rate of AMP and NH₃ (wt% AMP/wt% NH₃: A, 30/0; B, 30/1; C, 30/3; D, 30/5; E, 29/1; F, 27/3; G, 25/5) as a function of SO₂ partial pressure (p_{A2} ; 3, 5, 10, and 15 kPa) at 303 K to investigate the effect of the SO₂ absorption rate on the blending rate of aqueous AMP and NH₃ solution. As shown in Fig. 3, the SO₂ absorption rates on the addition of NH₃ to the aqueous 30 wt% AMP solution at 15 kPa were 11.15 to $13.42 \times 10^{-6} \text{ kmol m}^{-2} \text{ s}^{-1}$; 15.6 to 61.6% higher than that of the aqueous 30 wt% AMP solution alone. Also, the SO₂ absorption rate of aqueous blended AMP+NH₃ solutions (E, F, and G) increased with increasing NH₃ concentration in the same tale with result of CO₂ absorption rate. This was due to the fast reactions of $(\text{NH}_4)_2\text{SO}_3$ and NH_4HSO_3 formation and the high absorption capacity for SO₂.

The SO₂ absorption rates were also exponentially proportional to the NH₃ concentration, although the CO₂ absorption rates increased linearly with increasing NH₃ concentration. The SO₂ absorption rates with respect to the concentration of aqueous AMP solution and partial pressure were 1.3 to 3.6-fold higher than those for CO₂, but the increased rates were lower. Typically, on the addition of NH₃ into 30 wt% AMP at 15 kPa, the increases in the CO₂ and SO₂ absorption rates were 15.6 to 61.6% and 6.1 to 32.5%, respectively. Also, as with the partial pressure, the increased SO₂ absorption rates at 5 kPa (9.6 to 37.8%) were higher than those at 15 kPa (6.1 to 32.5%), which was caused by the fast instantaneous reaction of SO₂ due to hydration reaction, unlike CO₂ [20]. Therefore, most of the reaction initially occurred with SO₂.

In addition, the reaction rate constants ($k_{2,A2}$) of AMP and the

blend of 30 wt% AMP+NH₃ with SO₂ at 303 K were determined to be 1492.0 and 2823.0 m³ kmol⁻¹ s⁻¹, respectively. The k_{2,A2} of the blend of 30 wt% AMP+NH₃ was 2-fold higher than that of AMP without NH₃.

3. Classification of Reaction Regime in CO₂/SO₂ Simultaneous Absorption

The reaction regime was classified before the simultaneous CO₂/SO₂ absorption rates were investigated with the addition of NH₃ to the aqueous AMP solution. These experiments were performed by using 3, 5 and 10 wt% AMP and 10 wt% AMP+1 wt% NH₃ solutions at 303 K, at p_{A1} of 5, 10 and 15 kPa and p_{A2} of 1, 3 and 5 kPa.

For the simultaneous absorption in binary system (CO₂/SO₂), the conditions under the fast pseudo-first-order reaction regime are shown in Eq. (15). For a single gas absorption in the binary system, the conditions under the fast pseudo-first-order reaction regime are 3 << H_a << E_i, and those under the instantaneous reaction regime are shown in Eqs. (16) and (17) [18].

Initially, in order to classify the reaction regime of the amine solution for the binary system (CO₂/SO₂), the reaction of AMP with CO₂ and SO₂ in the binary system in terms of the gas partial pressure and amine concentration was assumed to be first-order. The dimensionless group was calculated by substituting the reaction rate constants (k_{2,A1} and k_{2,A2}) for the aqueous AMP and AMP+NH₃ solution with CO₂ and SO₂, as mentioned in paragraphs 1 and 2, into Eqs. (11) and (12). The values of D_A, C_A^{*} and k_L were obtained from the literature [21], with the reaction regime then confirmed by substituting these values into Eqs. (15) to (17).

The classification of the reaction regime for the aqueous solution concentration with p_{A2} and p_{A1} set at p_{A1}=15 and p_{A2}=1 kPa is presented in Tables 1 and 2, respectively. The reaction regime of the amine solution for the binary system was classified by applying each dimensionless group (H_{a,A1}, H_{a,A2}, E_{i,A1}, E_{i,A2} and E_{i,A12}), as

Table 1. Confirmation of the reaction regime with respect to p_{A1}=15 kPa for the CO₂/SO₂ system

p _{A2} [kPa]	C _B [kmol m ⁻³]	H _{a,A1} [-]	E _{i,A1} [-]	H _{a,A2} [-]	E _{i,A2} [-]	E _{i,A12} [-]	LT [-]	RT [-]
1	AMP							
	0.33	39.4	83.4	49.6	21.2	16.9	2.81	1.06
	0.56	50.8	142.0	63.0	35.6	28.5	2.13	1.04
	1.11	62.8	285.0	84.1	53.0	44.7	1.81	1.02
	AMP+NH ₃							
	1.11+0.53	119.5	412.3	140.2	60.5	52.8	2.6	1.02
3	AMP							
	0.33	39.4	83.4	49.6	13.8	11.8	4.07	1.08
	0.56	50.8	142.0	63.0	22.2	19.2	3.20	1.05
	1.11	62.8	285.0	84.1	37.2	32.9	2.48	1.03
	AMP+NH ₃							
	1.11+0.53	119.5	412.3	140.2	43.8	39.6	3.49	1.03
5	AMP							
	0.33	39.4	83.4	49.6	8.5	7.7	6.31	1.13
	0.56	50.8	142.0	63.0	13.7	12.5	4.96	1.08
	1.11	62.8	285.0	84.1	22.8	21.1	3.91	1.05
	AMP+NH ₃							
	1.11+0.53	119.5	412.3	140.2	26.9	25.3	5.50	1.04

Table 2. Confirmation of the reaction regime with respect to p_{A2}=1 kPa for the CO₂/SO₂ system

p _{A1} [kPa]	C _B [kmol m ⁻³]	H _{a,A1} [-]	E _{i,A1} [-]	H _{a,A2} [-]	E _{i,A2} [-]	E _{i,A12} [-]	LT [-]	RT [-]
5	AMP							
	0.33	39.4	248.3	49.6	21.2	19.5	2.50	1.05
	0.56	50.8	424.1	63.0	35.6	32.8	1.89	1.03
	1.11	62.8	853.2	84.1	53.0	49.9	1.66	1.02
	AMP+NH ₃							
	1.11+0.53	119.5	1235.0	140.2	60.5	57.7	2.41	1.02
10	AMP							
	0.33	39.4	124.6	49.6	21.2	18.1	2.66	1.06
	0.56	50.8	212.5	63.0	35.6	30.5	2.01	1.03
	1.11	62.8	427.1	84.1	53.0	47.1	1.73	1.02
	AMP+NH ₃							
	1.11+0.53	119.5	618.0	140.2	60.5	55.1	2.51	1.02
15	AMP							
	0.33	39.4	83.4	49.6	21.2	16.9	2.81	1.06
	0.56	50.8	142.0	63.0	35.6	28.5	2.13	1.04
	1.11	62.8	285.0	84.1	53.0	44.7	1.81	1.02
	AMP+NH ₃							
	1.11+0.53	119.5	412.3	140.2	60.5	52.8	2.61	1.02

shown in Tables 1 and 2, to Eq. (15). As a result, as the left term (LT) of Eq. (15) has a higher value than right term (RT) under all experimental conditions, it was not applicable to the fast pseudo-first-order reaction regime. Moreover, in the binary system (CO₂/SO₂), the applicability of CO₂ and SO₂ to fast pseudo-first-order and instantaneous reaction regimes can be obtained, as they were satisfied by 3 < H_a << E_i and Eq. (17), respectively.

4. Effect of the SO₂ Partial Pressure on the Simultaneous Absorption of CO₂/SO₂

The simultaneous absorption rates of the binary system (CO₂/SO₂) into aqueous AMP and AMP+NH₃ solutions were investigated

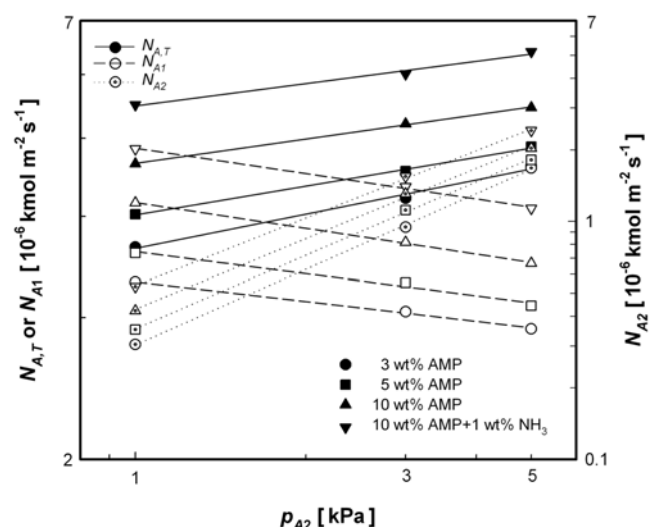


Fig. 4. The absorption rates of CO₂/SO₂ into aqueous AMP and AMP+NH₃ solutions as a function p_{A2} at different C_B, p_{A1}=15 kPa and 303 K.

at p_{A1} between 5 and 15 kPa, p_{A2} between 1 and 5 kPa and 303 K. The stirrer speed in all tests was 50 rpm in order to neglect the gas-phase resistance and maintain a smooth, horizontal gas-liquid interface. First, in order to examine the effect of p_{A2} on the absorption rate, the absorption rate of CO₂/SO₂ into aqueous AMP (3, 5 and 10 wt%) and 10 wt% AMP+1 wt% NH₃ solutions as a function p_{A2} and p_{A1} set at 15 kPa and 303 K is shown in Fig. 4.

It was observed that the simultaneous absorption rates of CO₂/SO₂ with AMP concentration were 3.66 to $4.65 \times 10^{-6} \text{ kmol m}^{-2} \text{ s}^{-1}$ and 4.62 to $5.46 \times 10^{-6} \text{ kmol m}^{-2} \text{ s}^{-1}$ at p_{A2} of 1 and 5 kPa, respectively, and increased with an increasing AMP concentration and p_{A2} . This could have been caused by promotion of the mass transfer with increasing amount of SO₂ absorbed at the gas-liquid interface, and was in good agreement with the results in paragraphs 1 and 2. The simultaneous absorption rates of CO₂/SO₂ by the addition of 1 wt% NH₃ to 10 wt% AMP with p_{A2} between 1 and 5 kPa and p_{A1} set at 15 kPa were between 5.50 and $6.40 \times 10^{-6} \text{ kmol m}^{-2} \text{ s}^{-1}$; increases of 15.5 to 14.7% compared to 10 wt% AMP as a result of the high loading ratio and fast additional reaction (Eqs. (8) and (9)) due to the addition of NH₃. The absorption rate of SO₂ increased with p_{A2} and p_{A1} set at 15 kPa, but the absorption rate of CO₂ decreased. These tendencies were caused by the reaction rate of SO₂ being approximately 2-fold higher than that of CO₂, and was selectively fast absorbed by the hydration reaction in water, unlike CO₂ [20].

Fig. 5 shows the absorption rates of CO₂/SO₂ into aqueous AMP and AMP+NH₃ solutions as a function C_B at different p_{A2} from the result of Fig. 4. It was observed that due to the strong influence of the hydration reaction, the increased SO₂ absorption rate with concentration of aqueous AMP solution was lower than that of CO₂. Overall, the increased simultaneous absorption rate of CO₂/SO₂ on the addition of 1 wt% NH₃ into 10 wt% AMP was higher than with AMP alone. Therefore, the addition of NH₃ to an aqueous AMP solution could be an effective way of enhancing the absorption rate of CO₂/SO₂.

5. Effect of the CO₂ Partial Pressure on the Simultaneous Ab-

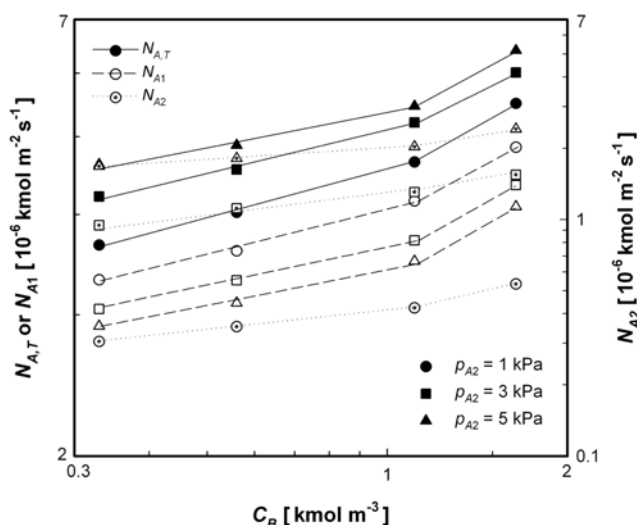


Fig. 5. The absorption rates of CO₂/SO₂ into aqueous AMP and AMP+NH₃ solutions as a function C_B at different p_{A2} , p_{A1} = 15 kPa and 303 K.

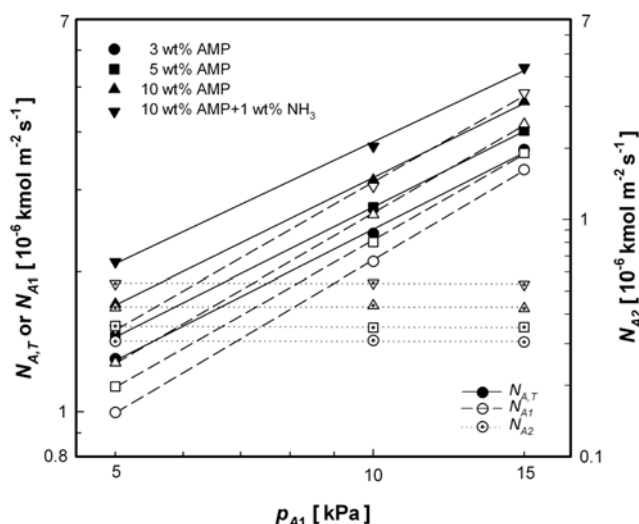


Fig. 6. The absorption rates of CO₂/SO₂ into aqueous AMP and AMP+NH₃ solutions as a function p_{A1} at different C_B , p_{A2} = 1 kPa and 303 K.

sorption of CO₂/SO₂

Fig. 6 shows absorption rate of CO₂/SO₂ into aqueous AMP (3, 5 and 10 wt%) and 10 wt% AMP+1 wt% NH₃ solutions as a function of p_{A1} (5, 10 and 15 kPa) and a p_{A2} of 1 kPa and 303 K to examine the effect on the absorption rate with p_{A1} .

As shown in Fig. 6, the simultaneous absorption rates of CO₂/SO₂ with AMP concentration were 1.30 to $1.71 \times 10^{-6} \text{ kmol m}^{-2} \text{ s}^{-1}$ and 3.66 to $4.65 \times 10^{-6} \text{ kmol m}^{-2} \text{ s}^{-1}$ at p_{A1} of 5 and 15 kPa, respectively, and increased with increasing AMP concentration and p_{A1} . Also, the simultaneous absorption rates of CO₂/SO₂ on the addition of 1 wt% NH₃ to 10 wt% AMP with p_{A1} between 5 and 15 kPa and a p_{A2} of 1 kPa were 2.10 to $5.50 \times 10^{-6} \text{ kmol m}^{-2} \text{ s}^{-1}$; increases of more than 18.6 to 15.5% compared to 10 wt% AMP alone. The increases in the rates with p_{A1} were higher than those with p_{A2} presented in

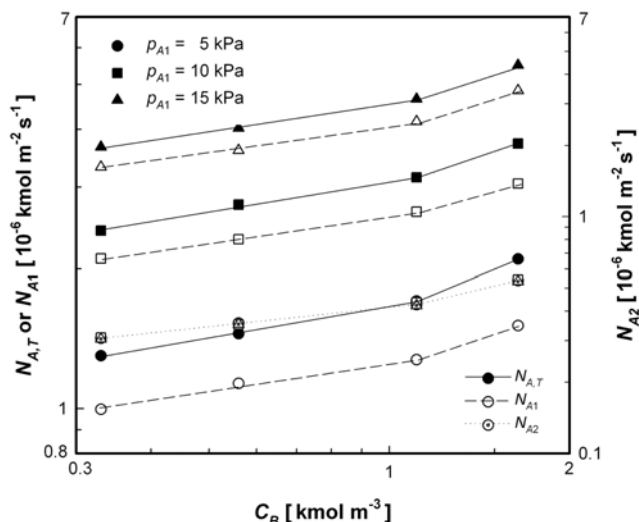


Fig. 7. The absorption rates of CO₂/SO₂ into aqueous AMP and AMP+NH₃ solutions as a function C_B at different p_{A1} , p_{A2} = 1 kPa and 303 K.

paragraph 4. This is because the aqueous NH_3 solution predominantly reacts with CO_2 at a low p_{A2} of 1 kPa. Therefore, the CO_2 absorption rate at each NH_3 concentration with p_{A1} increased with a p_{A2} of 1 kPa, but those of SO_2 remained almost constant.

Fig. 7 shows the absorption rates of CO_2/SO_2 into aqueous AMP and $\text{AMP}+\text{NH}_3$ solutions as a function C_B at different p_{A1} from the results of Fig. 6. As shown in Fig. 7, the absorption rate increased with increasing p_{A1} , AMP concentration and on the addition of NH_3 . Overall, the increasing absorption rate due to the addition of 1 wt% NH_3 into 10 wt% AMP was higher than the increase with AMP alone, and this result was in agreement with that shown in Fig. 5.

CONCLUSIONS

The addition of NH_3 solution to an aqueous AMP solution increased the reaction rate constants for CO_2 and SO_2 by 144 and 109%, respectively, compared to AMP solution alone. The simultaneous absorption rates of the binary system (CO_2/SO_2) into aqueous AMP and $\text{AMP}+\text{NH}_3$ solutions did not satisfy the fast pseudo-first-order reaction regime. Each reaction with CO_2 and SO_2 was classified as a fast pseudo-first-order reaction regime and an instantaneous reaction regime; the SO_2 gas was selectively absorbed faster than CO_2 . The simultaneous absorption rates of CO_2/SO_2 on the addition of 1 wt% NH_3 to 10 wt% AMP, with p_{A2} between 1 and 5 kPa and a p_{A1} of 15 kPa were 5.50 to $6.40 \times 10^{-6} \text{ kmol m}^{-2} \text{ s}^{-1}$; increases of 15.5 to 14.7% compared to 10 wt% AMP alone as a result of the high loading ratio and fast additional reaction (Eqs. (3) and (4)) on the addition of NH_3 .

Consequently, the addition of NH_3 solution to an aqueous AMP solution would be expected to be an excellent absorbent for the simultaneous removals of CO_2/SO_2 from the composition of flue gas emitted from thermoelectric power plants, as SO_2 has a fast reaction with NH_3 and hydration reaction with water.

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NOMENCLATURE

A	: gaseous species absorbed into the liquid B
C_A^*	: concentration of dissolved gas A at the gas-liquid interface [kmol m^{-3}]
C_B	: concentration of reactant B [kmol m^{-3}]
D_A	: diffusivity of dissolved gas A in the liquid phase [$\text{m}^2 \text{ s}^{-1}$]
D_B	: diffusivity of reactant B [$\text{m}^2 \text{ s}^{-1}$]
E	: enhancement factor [-]
E_i	: value of E in the instantaneous reaction regime [-]
H_a	: Hatta number [-]
H_A	: Henry's constant [$\text{kPa m}^3 \text{ kmol}^{-1}$]
k_L	: mass transfer coefficient of A in absorbent [m s^{-1}]
k_{mn}	: reaction rate constant, m th order in A and n th order in B [m^3

	$\text{kmol}^{-1})^{m+n-1} \text{ s}^{-1}$]
k_{ov}	: overall pseudo-first order reaction rate constant [s^{-1}]
k_2	: second-order reaction rate constant [$\text{m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$]
N_A	: absorption rate of species A at the gas-liquid interface with chemical reaction [$\text{kmol m}^{-2} \text{ s}^{-1}$]
N_{A0}	: absorption rate of species A at the gas-liquid interface with physical reaction [$\text{kmol m}^{-2} \text{ s}^{-1}$]
P_A	: partial pressure of species A [kPa]
T	: temperature [K]
ν	: stoichiometric coefficient [-]

Subscripts

A1	: CO_2
A2	: SO_2

REFERENCES

1. IPCC, *Policymaker's summary of the scientific assessment of climate change*, Report to IPCC from working group, Branknell, UK: Meteorological Office (1990).
2. B. P. Mandal, A. K. Biswas and S. S. Bandyopadhyay, *Chem. Eng. Sci.*, **58**, 4137 (2003).
3. A. C. Yeh and H. Bai, *Sci. Total Environ.*, **228**, 121 (1999).
4. S. M. Yih and K. P. Shen, *Ind. Eng. Chem. Res.*, **27**, 2237 (1988).
5. E. Alper, *Ind. Eng. Chem. Res.*, **29**, 1725 (1990).
6. A. K. Saha, S. S. Bandyopadhyay and A. K. Biswas, *Chem. Eng. Sci.*, **50**, 3587 (1995).
7. S. Xu, Y. W. Wang, F. D. Otto and A. E. Mather, *Chem. Eng. Sci.*, **51**, 841 (1996).
8. A. Aroonwilas and A. Veawab, *Ind. Eng. Chem. Res.*, **43**, 2228 (2004).
9. S. W. Park, J. W. Lee, B. S. Choi and J. W. Lee, *Korean J. Chem. Eng.*, **23**, 806 (2006).
10. W. J. Choi, K. C. Cho, S. S. Lee, J. G. Shim, H. R. Hwang, S. W. Park and K. J. Oh, *Green Chemistry*, **9**, 594 (2007).
11. D. H. Lee, W. J. Choi, K. C. Cho, S. J. Moon, S. Y. Jang and K. J. Oh, *Korean J. Chem. Eng.*, **25**, 279 (2008).
12. Y. F. Diao, X. Y. Zheng, B. S. He, C. H. Chen and X. C. Xu, *Energy Con. Manage.*, **45**, 2283 (2004).
13. H. L. Bai and A. C. Yeh, *Ind. Chem. Eng. Res.*, **36**, 2490 (1997).
14. J. T. Yeh, K. P. Renik, K. Rygle and H. W. Pennline, *Fuel Process. Technol.*, **86**, 1533 (2005).
15. H. Hikita, S. Asai and T. Tsufi, *AIChE J.*, **23**, 538 (1977).
16. H. Hikita, S. Asai and H. Nose, *AIChE J.*, **24**, 147 (1978).
17. B. He, X. Zheng, Y. Wen, H. Tong, M. Chem and C. Chen, *Energy Con. Manage.*, **44**, 2175 (2003).
18. L. K. Daraiswany and M. M. Sharma, *Heterogeneous reaction: Analysis, example and reactor design*, John Wiley & Sons, New York, 17-26 (1984).
19. B. Messaoudi and E. Sada, *J. Chem. Eng. Japan*, **29**, 193 (1996).
20. H. Hikita, S. Asai and H. Nose, *AIChE J.*, **24**, 147 (1978).
21. W. J. Choi, J. B. Seo, J. W. Kim, S. J. Moon, S. W. Park and K. J. Oh, *1st ASCON-IEEChE*, 751 (2008).