

The physical solubilities and diffusivities of N₂O and CO₂ in aqueous ammonia solutions on the additions of AMP, glycerol and ethylene glycol

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Abstract—Carbon dioxide (CO₂) is a major greenhouse gas, the emissions of which should be reduced. There are various technologies for the effective separation of CO₂. Of these, chemical absorption methods are generally accepted as the most effective. The monoethanolamine (MEA) process is an effective way to remove CO₂, but is an expensive option for the separation of CO₂ from massive gas-discharging plants. Therefore, ammonia solution, which is less expensive and more effective than MEA, was used for the removal of CO₂. In this study, the physical solubility of N₂O in (ammonia+water), (ammonia+2-amino-2-methyl-1-propanol+water), (ammonia+glycerol+water) and (ammonia+ethylene glycol+water) was measured at 293, 303, 313, 323 K. Additive concentrations of 1, 3, and 5 wt% AMP, glycerol and ethylene glycol were added for each 9 wt% ammonia solution. A solubility apparatus was used to investigate the solubility of N₂O in ammonia solutions. The diffusivity was measured with a wetted wall column absorber. The “N₂O analogy” is used to estimate the solubility and diffusivity of CO₂ in the aqueous ammonia solutions. OriginPro 7.5 was used to correlate the solubility and diffusivity of N₂O in ammonia solutions. The parameters of the correlation were determined from the measured solubility and diffusivity.

Key words: Carbon Dioxide, Ammonia, Absorption, 2-Amino-2-methyl-1-propanol, Ethylene Glycol, Glycerol

INTRODUCTION

Various technologies, such as chemical absorption, physical adsorption, cryogenic methods, membrane separation and biological fixation, are used to separate CO₂ from flue gas. Chemical absorption is generally recognized as the most effective [1]. Industrially important chemical absorbents include monoethanolamine (MEA), diethanolamine (DEA) and *N*-methyldiethanolamine (MDEA). A different class of chemical absorbents—sterically hindered amines, such as 2-amino-2-methyl-1-propanol (AMP)—have been proposed as new, commercially attractive CO₂ absorbents due to their absorption capacity, absorption rate, degradation resistance and regeneration energy [2]. Furthermore, ammonia (NH₃) solution can be an alternative for the control of the CO₂ emitted from flue gas as a result of its high absorption capacity and fast absorption rate [3,4].

Aqueous ammonia is a good example to serve the characteristics and has been used for many years at several steel works for the removal of CO₂ and sulfur compounds from coke oven gas (COG). Studies on aqueous ammonia for the absorption of greenhouse gases have been undertaken since those of Bai and Yeh [5] and Yeh et al. [6]. Moreover, Resnik et al. [7] and Diao et al. [8] have subsequently reported an aqua ammonia process and ammonia scrubbing for the capture of CO₂. Therefore, aqueous ammonia, which has a low regeneration temperature, is recommended for the process of CO₂ absorption at iron and steel works that produce much heat sensitive wastes at low and medium temperatures.

However, although a high NH₃ concentration solution enables a high CO₂ absorption efficiency, it causes ammonium ions to be lost as ammonia vapor, resulting in a reduction of CO₂ absorption by means of a lower concentration of ammonia absorbent. Their performance is likely due to the interactions between the hydroxyl groups of the additives and ammonia via hydrogen bonding [9]. AMP, glycerol and ethylene glycol are potential materials for the control of the vapor pressure of NH₃ and to improve the CO₂ absorption characteristics.

For the analysis of the experimental absorption rate and for the rational design of gas absorption units, two of the most important parameters required are the physical solubility and diffusivity of the solute gases in the solvents. Since CO₂ reacts in ammonia solutions, its physical solubility and diffusivity cannot be measured directly [10]. Therefore, the solubility and diffusivity of CO₂ in aqueous solvents were estimated by using the “N₂O analogy method” as a function of temperature (293, 303, 313 and 323 K).

EXPERIMENTAL METHODS

1. Materials

Analytical grade AMP solution, with a purity of 95 wt%, was supplied by Acros Organics. 28 wt% ammonia solution, 99 wt% glycerol and 99.5 wt% ethylene glycol were supplied by Junsei Chemical co. Ltd. All chemicals were used without further purification. Aqueous solutions were prepared with distilled water. Double-distilled water degassed by boiling was used to make the ammonia solutions. The CO₂ and N₂ gases were of commercial grade, with purities of 99.99%. High purity N₂O (99.9%) gas was also used.

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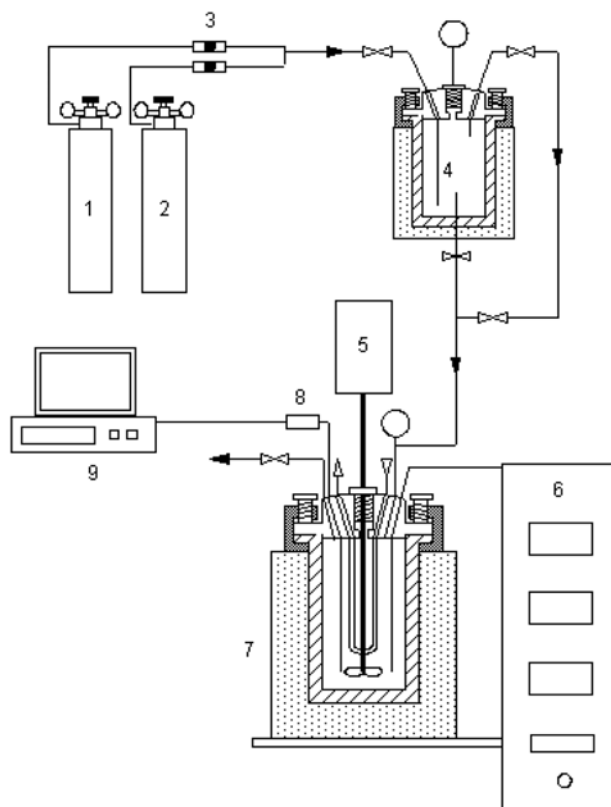


Fig. 1. Schematic diagram of experimental apparatus for measurement of physical solubility.

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|----------------------------------|--|
| 1. N_2O cylinder | 6. Controller of temp. and agitation speed |
| 2. CO_2 cylinder | 7. Reactor (Agitated vessel) |
| 3. Mass flow controller | 8. Pressure transducer |
| 4. Feeder | 9. Computer |
| 5. Magnetic drive | |

2. Physical Solubility Measurement

The experimental apparatus for measuring the physical solubilities is shown in Fig. 1. The reactor, with a height of 160 mm and i.d. of 95 mm, was located inside a temperature-controlled vessel, with four 5-mm-wide glass plates adhered to the inner wall of the reactor as baffles. The total volume of the reactor was about 1,134 cm^3 , with an active interface area (A_s) of 70.88 cm^2 . A two-blade impeller (70 mm $\times$ 20 mm) was installed in the middle of the liquid level. The reactor temperature was measured with a K-type thermocouple, with an accuracy of 0.1 K. A pressure transducer (MGI/MGAMP series, accuracy of ± 0.1 kPa) was installed in the reactor, with a feeder used to measure the pressure. The gas flow rates were controlled using mass flow controllers (5850E, Brooks Instruments).

After the temperature of the reactor had stabilized, the reactor was purged with pure N_2O for one hour to remove the remaining air from the reactor. After the reactor had reached atmospheric pressure, 200 mL of an amine solution was injected, using a syringe, and then agitated. The reactor pressure decreased as the amine solution absorbed N_2O ; therefore, N_2O was continuously injected to maintain the reactor pressure at 1 atm. When the reactor pressure had stabilized, without the need for further N_2O injection, the solubility was calculated by the difference in the feeder pressures before and after the absorption. The experimental uncertainty in the measured solubility was estimated at about $\pm 2\%$.

3. Diffusivity Measurement

A cylindrical wetted wall column absorber was constructed of 316 stainless steel, with an outside diameter of 2.54 cm and a height of 8.6 cm, to estimate the diffusivities of CO_2 in water and the amine solutions using the N_2O analogy method. The apparatus and experimental procedure used were the same as those described by Li and Lai [10]. A thin solution film was maintained on the outside wall of the column by adding 0.03-0.05 vol% of Tween 80 to each

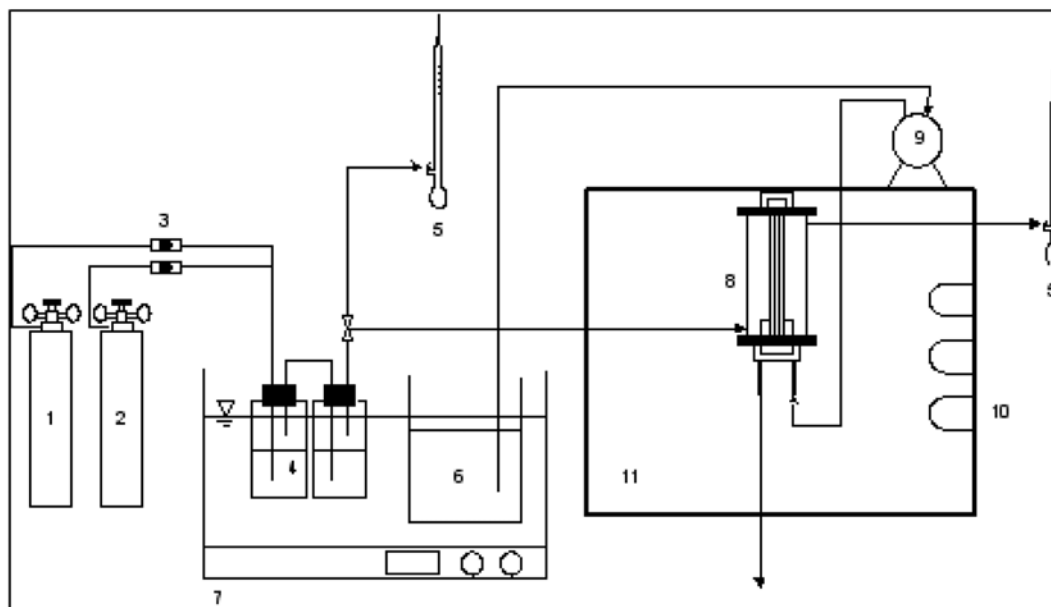


Fig. 2. Schematic diagram of experimental apparatus for measurement of diffusivity.

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|----------------------------------|----------------------|-----------------------|-----------------------------|
| 1. N_2O cylinder | 4. Saturator | 7. Water bath | 10. Refrigerator and heater |
| 2. CO_2 cylinder | 5. Soap bubble meter | 8. Wetted wall column | 11. Air bath |
| 3. Mass flow controller | 6. Solution storage | 9. Liquid pump | |

amine solution. When the solution was uniformly distributed, the diffusivity was determined by the difference between the inlet and outlet gas flow rates. The estimated experimental uncertainty in the measured diffusivity was $\pm 2\%$ on the basis of comparisons with literature data for the diffusivities of N_2O in water.

THEORETICAL BACKGROUND

1. Physical Properties

Determining the physical solubility and diffusivity of CO_2 in the amine solutions is very important in understanding their reaction kinetics and for effective process design. However, the physical solubility and diffusivity are not easily determined for the chemical reactions between CO_2 and amine solutions. As a result, the N_2O analogy method was used to estimate the solubility and diffusivity of CO_2 in the amine solutions, as the use of a nonreacting gas, such as N_2O , with similar mass, molecular structure and molecular interactions is required [11–13]. The N_2O analogy method for measuring the physical solubility and diffusivity of CO_2 in amine solutions is as follows:

$$(H_{CO_2})_{amine} = (H_{N_2O})_{amine} \times (H_{CO_2}/H_{N_2O})_{water} \quad (1)$$

$$(D_{CO_2})_{amine} = (D_{N_2O})_{amine} \times (D_{CO_2}/D_{N_2O})_{water} \quad (2)$$

The solubility and diffusivity of CO_2 in ammonia solution were calculated using the N_2O analogy method employing Eqs. (1) and (2) as the structure and properties of ammonia solution are similar to those of amines.

2. Physical Solubility

The physical solubility was calculated in terms of Henry's law constant, as follows:

$$P = H_A C_A^+ \quad (3)$$

The concentration of the gas in the bulk liquid at gas-liquid equilibrium (C_A^+) was calculated from the following mass balance:

$$C_A^+ = \frac{(P_f V_f) - [(P_c - P_{vap}) V_f]}{RT V_L} \quad (4)$$

3. Diffusivity

Higbie penetration theory gives the specific absorption rate (N_A), as follows:

$$N_A = 2(D_A/\pi d/L)^{1/2} (p_A/H_A) \quad (5)$$

The contact time (t_c) can be derived from a wetted wall column, using the following equation:

$$t_c = (2h/3)(\pi d/L)^{2/3} (3\eta/\rho g)^{1/3} \quad (6)$$

The diffusivity (D_A) of the gas absorbed via Eqs. (5) and (6) is expressed as follows:

$$D_A = (N_A H_A)^2 \pi d / (2p_A)^2 \quad (7)$$

4. Calculation of the Average Absolute Deviation

Henry's constant and the diffusivity of the target absorbent were studied experimentally. The experimental data were fitted to a single equation of concentration and temperature from the general model. The average deviation between the calculated and experimental values was analyzed by using Eq. (8).

$$\sigma = \frac{1}{n} \sum_{i=1}^n \frac{|H_{m,calc} - H_{m,exp}|}{H_{m,exp}} \quad (8)$$

RESULTS AND DISCUSSION

1. Solubility

To predict of the solubility of CO_2 in the amine, the N_2O analogy method, which uses N_2O gas, was used because it is impossible to measure the solubility of CO_2 in NH_3 solution. The solubility of N_2O in NH_3 solution (5, 7, 9 and 11 wt%) was measured at various temperatures (293, 303, 313 and 323 K) before measuring the solubility of CO_2 in the NH_3 solution containing additive in order to compare the results.

Table 1 shows the measured and inferred solubilities using the N_2O analogy method (Eq. (1)). Figs. 3 and 4 show Henry's constants for CO_2 in NH_3 solution as functions of the temperature and

Table 1. Henry's constants for N_2O and CO_2 in aqueous NH_3 solutions as a function of temperature

T/K	$H_{N_2O}/kPam^3kmol^{-1}$				
	Water	5 wt% NH_3	7 wt% NH_3	9 wt% NH_3	11 wt% NH_3
293	3504	3496	3667	3858	4101
303	4745	4102	4282	4480	4750
313	5882	4638	4858	5100	5416
323	7035	5102	5329	5564	5879

T/K	$H_{CO_2}/kPam^3kmol^{-1}$				
	Water	5 wt% NH_3	7 wt% NH_3	9 wt% NH_3	11 wt% NH_3
293	2625	2619	2747	2890	3072
303	3491	3018	3151	3296	3494
313	4172	3290	3446	3617	3842
323	4875	3535	3693	3856	4074

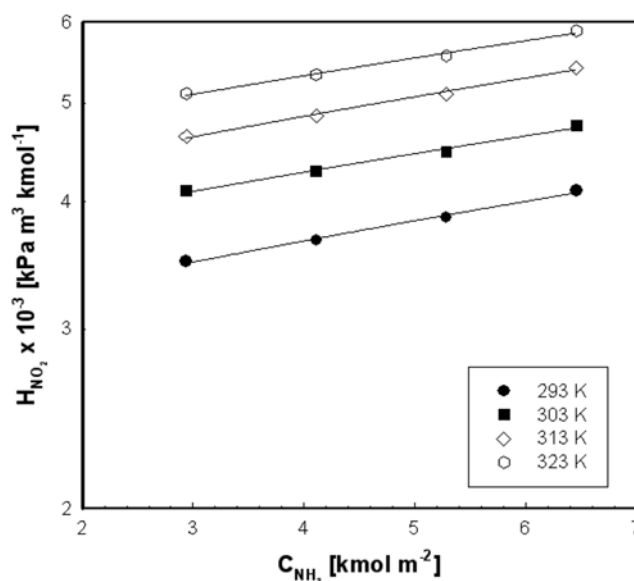


Fig. 3. Henry's constant for N_2O in aqueous NH_3 solutions as a function of temperature.

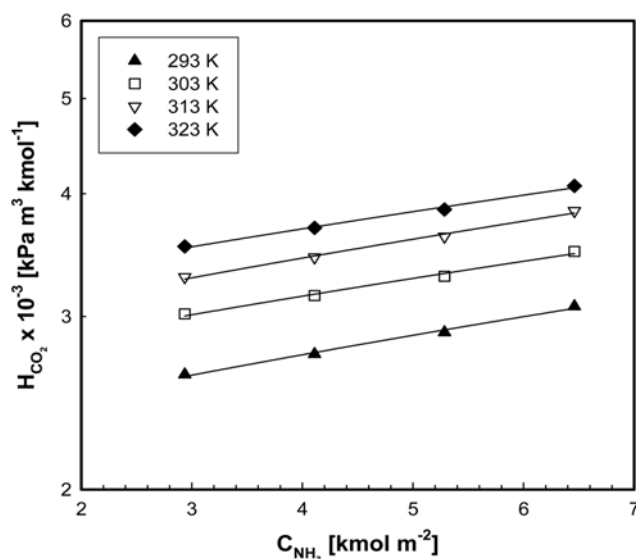


Fig. 4. Henry's constant for CO₂ in aqueous NH₃ solution as a function of temperature.

concentration. As shown in Figs. 3 and 4, Henry's constant increased with increasing concentration, which means the solubility of CO₂ decreased with increasing NH₃ solution concentration. Also, Henry's constant increased with increasing NH₃ concentration, so the solubility of CO₂ increased with decreasing solution concentration. Concentration increase of aqueous ammonia makes an NO₂ dissolution decreasing in reactor. From Eq. (4), solubility decreased as temperature increased, so Henry's constant increased.

To predict Henry's constants as functions of the temperature and concentration of the NH₃ solution, a regression analysis was conducted by using Eq. (9).

$$H/10^{-6} \text{ kPa m}^3 \text{ kmol}^{-1} = (a_1 + a_2 M_1) \times 10^6 \exp(c/T) \quad (9)$$

Where a_1 , a_2 , c is a required parameter, the molarity of AMP and T

Table 2. Solubility correlation parameters and AAD% of aqueous NH₃ solution

	N ₂ O			AAD%
	a_1	a_2	c	
NH ₃	1.552×10^{-1}	8.170×10^{-3}	-1.148×10^3	1.20
	CO ₂			AAD%
	a_1	a_2	c	
NH ₃	4.908×10^{-2}	2.590×10^{-3}	-8.953×10^2	1.42

the absolute temperature. Table 2 shows the results of the regression analysis using OriginPro 7.5, with a dependent variable (H) and independent variables (M_1 , T). The solid lines in Figs. 3 and 4 are from the experimental data and the data calculated using Eq. (9), which show that the modeled equation simulated by the calculation program corresponded with the experimental values.

The N₂O analogy method was also used to measure the CO₂ solubility in 9 wt% NH₃ solution containing additives (1, 3 and 5 wt% AMP, glycerol and ethylene glycol) at 303, 313 and 323 K. Tables 3 and 4 show the solubilities of N₂O and CO₂ as a function of temperature. Figs. 5 and 6 show Henry's constants for N₂O and CO₂ at 313 K, which is an ideal condition for CO₂ absorption into NH₃ solution. Henry's constants for 9 wt% NH₃ solution containing additives (AMP, glycerol and ethylene glycol) were higher than that for NH₃ 9 wt% solution alone, and the rate of increase was similar for each NH₃ solution irrespective of the additive. This was because the difference between the partial pressure of the gas and the vapor pressure slowly influences the vapor pressure in the supply vessel. Henry's constant as functions of the solution temperature and concentration, with additive, was predicted using the above results and Eq. (10).

$$H/10^{-6} \text{ kPa m}^3 \text{ kmol}^{-1} = (b_1 + b_2 M_1 + b_3 M_2 + b_4 M_1 M_2) \exp(-c/T) \quad (10)$$

b_1 , b_2 , b_3 , b_4 , c are parameters for obtaining, M_1 and M_2 the molarities of each 9 wt% NH₃ solution and additives. A regression analysis

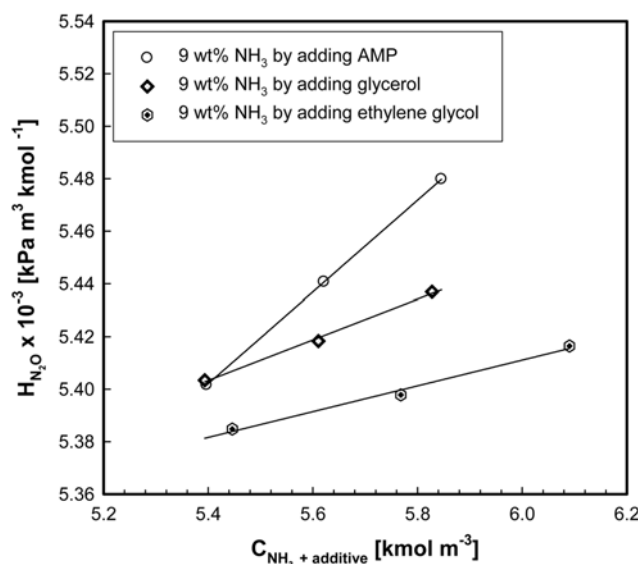
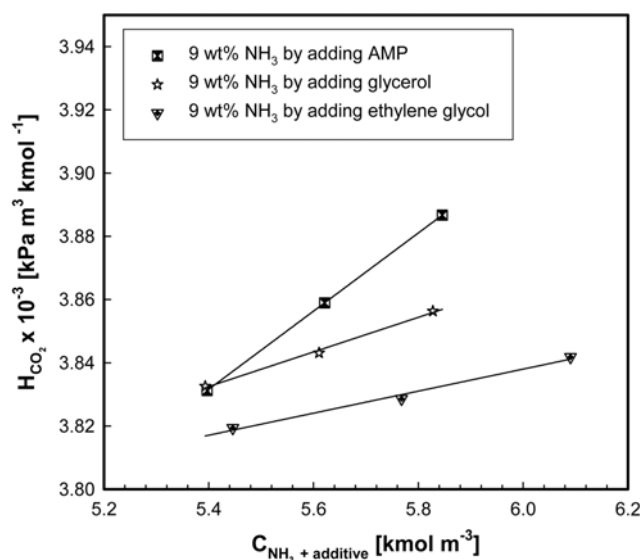
Table 3. Henry's constants for N₂O and CO₂ in aqueous 9 wt% NH₃ solution containing additives as a function of temperature

T/K	$H_{N_2O}/\text{kPa m}^3 \text{ kmol}^{-1}$								
	9 wt% NH ₃								
	1 wt% AMP	3 wt% AMP	5 wt% AMP	1 wt% Glycerol	3 wt% Glycerol	5 wt% Glycerol	1 wt% Ethylene glycol	3 wt% Ethylene glycol	5 wt% Ethylene glycol
293	3872	3900	3928	3874	3884	3898	3860	3870	3883
303	4531	4564	4597	4533	4545	4561	4517	4528	4544
313	5402	5441	5480	5403	5418	5437	5385	5398	5416
323	6354	6400	6446	6356	6373	6395	6334	6349	6371
T/K	$H_{CO_2}/\text{kPa m}^3 \text{ kmol}^{-1}$								
	9 wt% NH ₃								
	1 wt% AMP	3 wt% AMP	5 wt% AMP	1 wt% Glycerol	3 wt% Glycerol	5 wt% Glycerol	1 wt% Ethylene glycol	3 wt% Ethylene glycol	5 wt% Ethylene glycol
293	2901	2922	2943	2902	2910	2920	2892	2899	2909
303	3334	3358	3382	3335	3344	3356	3323	3332	3343
313	3831	3859	3887	3833	3843	3856	3819	3829	3842
323	4403	4435	4467	4404	4417	4432	4389	4400	4415

Table 4. Solubility correlation parameters and AAD% of aqueous 9 wt% NH₃ solution+additives

	N ₂ O					AAD%
	b ₁	b ₂	b ₃	b ₄	c	
9 wt% NH ₃ +AMP	4.304×10^{-1}	8.144×10^{-2}	1.393×10^{-2}	2.640×10^{-3}	1.587×10^3	0.67
9 wt% NH ₃ +glycerol	4.305×10^{-1}	8.146×10^{-2}	6.160×10^{-3}	1.170×10^{-3}	1.587×10^3	0.66
9 wt% NH ₃ +ethylene glycol	4.292×10^{-1}	8.121×10^{-2}	3.910×10^{-3}	7.400×10^{-4}	1.587×10^3	0.67

	CO ₂					AAD%
	b ₁	b ₂	b ₃	b ₄	c	
9 wt% NH ₃ +AMP	1.323×10^{-1}	2.503×10^{-2}	4.290×10^{-3}	8.100×10^{-4}	1.325×10^3	0.45
9 wt% NH ₃ +glycerol	1.322×10^{-1}	2.513×10^{-2}	9.187×10^{-1}	-1.731×10^{-1}	1.325×10^3	0.45
9 wt% NH ₃ +ethylene glycol	1.320×10^{-1}	2.498×10^{-2}	1.220×10^{-3}	2.300×10^{-4}	1.324×10^3	0.45

**Fig. 5. Henry's constants for N₂O in aqueous 9 wt% NH₃+AMP, glycerol and ethylene glycol solution as a function of the additive concentration at 313 K.****Fig. 6. Henry's constant for CO₂ in aqueous 9 wt% NH₃+AMP, glycerol and ethylene glycol solution as a function of the additive concentration at 313 K.**

was conducted with a set of independent variables (M_1 , M_2 , T) and a dependent variable (H), the results of which are shown in Table 4.

The solid lines in Figs. 4 and 5 were obtained using Eq. (10) and the parameters in Table 4. From the results, the AAD% of N₂O and CO₂ were below 1%; therefore, the prediction equation corresponded with the experiment data.

2. Diffusivity

The measured diffusivity, like the solubility, was calculated by using the N₂O analogy method. Before the diffusivity measurements for the additions of AMP, glycerol and ethylene glycol to NH₃ solution, the diffusivity of N₂O was obtained as a function of the aqueous NH₃ solution concentration at 293, 303, 313 and 323 K. The diffusivity of CO₂ using the N₂O analogy method is shown in Figs. 7 and 8. As seen, the overall diffusivities of N₂O and CO₂ decreased as concentration of aqueous NH₃ solution increases, due to the diffusivity of the solution decreasing with increasing viscosity. When a small molecule in solvent was diffused in the solution, according to t_c increase, the diffusivity increased from Eq. (6). Meanwhile, based on the results shown in Figs. 7 and 8, Eq. (11) was used to predict the diffusivity as functions of the temperature and concen-

tration of the AMP solution.

$$D/10^9 \text{ m}^2 \text{ s}^{-1} = (p_1 + p_2 M_1 + p_3 M_1^2) \exp\left(-\frac{Z}{T}\right) \quad (11)$$

a_1 , a_2 , c are required parameters, M_1 is the molarity of AMP and T is absolute temperature. Table 6 shows the results of a regression analysis using OriginPro 7.5, with a dependent variable (H) and independent variables (M_1 , T), and the AAD(%) of N₂O and CO₂ were 2.22 and 2.93, respectively.

The experimental diffusivities of N₂O and CO₂ with AMP, glycerol and ethylene glycol added to the NH₃ solution are shown in Table 7; the diffusivities of N₂O and CO₂ are shown in Figs. 9 and 10 at 313 K, which is an ideal condition for CO₂ absorption into NH₃ solution. The diffusivities of 9 wt% NH₃ solution on the additions of AMP, glycerol and ethylene glycol were higher than that for aqueous NH₃ solution alone, but according to the blended additives the diffusivity tended to decrease, which was caused by a decrease in the viscosity with increasing NH₃ concentration, a basic characteristic of diffusivity. Meanwhile, Eq. (12) was used to analyze the diffusivity as functions of the temperature and concentration.

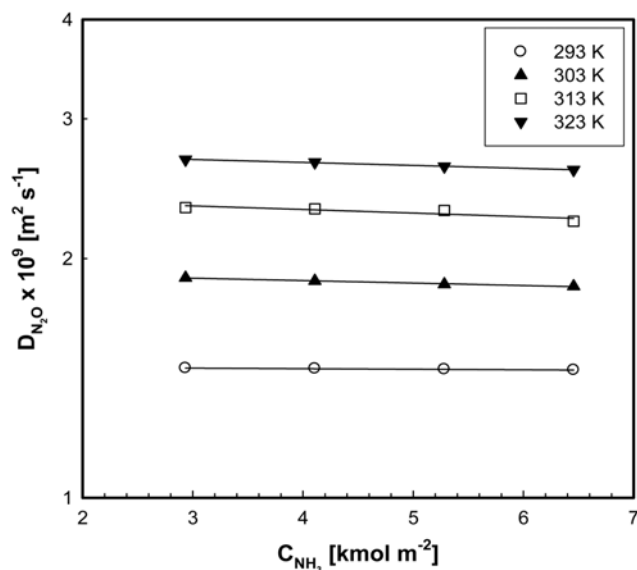


Fig. 7. Diffusivities of N₂O in aqueous NH₃ solution as a function of temperature.

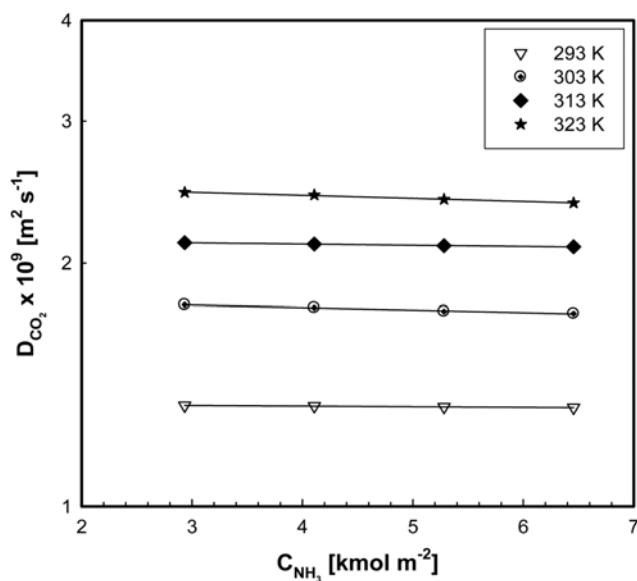


Fig. 8. Diffusivities of CO₂ in aqueous NH₃ solution as a function of temperature.

$$D/10^9 \text{ m}^2 \text{ s}^{-1} = (q_1 + q_2 M_1 + q_3 M_1^2 + q_4 M_2 + q_5 M_2^2 + q_6 M_1 M_2) \exp(-z/T) \quad (12)$$

$q_1, q_2, q_3, q_4, q_5, q_6$ and z are parameters that need to be obtained and M_1 and M_2 are the molarities of each additive in the 9 wt% NH₃ solution (AMP, glycerol, ethylene glycol). The regression analysis obtained from a set of independent variables (M_1, M_2, T) and a dependent variable (diffusivity, D) and $q_1, q_2, q_3, q_4, q_5, q_6$ and z are shown in Table 8. The solid lines in Figs. 9 and 10 were obtained using Eq. (10) and the parameters in Table 8. From the results, the AAD% of N₂O with AMP, glycerol and ethylene glycol were 3.16, 1.91 and 1.93, respectively, and those of CO₂ were 2.55, 1.27 and 1.42; therefore, the prediction equation corresponded with the experiment data.

Table 5. Diffusivities of N₂O and CO₂ in aqueous AMP solution as a function of temperature

T/K	$D_{N_2O}/10^9 \text{ m}^2 \text{ s}^{-1}$				
	Water	5 wt% NH ₃	7 wt% NH ₃	9 wt% NH ₃	11 wt% NH ₃
293	1.5321	1.456	1.454	1.451	1.448
303	2.0084	1.892	1.875	1.857	1.846
313	2.5549	2.319	2.311	2.300	2.229
323	2.9238	2.665	2.645	2.613	2.587

T/K	$D_{CO_2}/10^9 \text{ m}^2 \text{ s}^{-1}$				
	Water	5 wt% NH ₃	7 wt% NH ₃	9 wt% NH ₃	11 wt% NH ₃
293	1.6683	1.334	1.332	1.329	1.326
303	2.1434	1.777	1.762	1.744	1.734
313	2.7868	2.120	2.112	2.102	2.095
323	3.1814	2.447	2.429	2.399	2.375

Table 6. Diffusivity correlation parameters and AAD% of the aqueous NH₃ solutions

	N ₂ O				
	p_1	p_2	p_3	z	AAD%
NH ₃	7.422×10^2	4.513	-1.108	1.815×10^3	2.22

	CO ₂				
	p_1	p_2	p_3	z	AAD%
NH ₃	6.346×10^2	-3.333	-4.117×10^{-2}	1.785×10^3	2.93

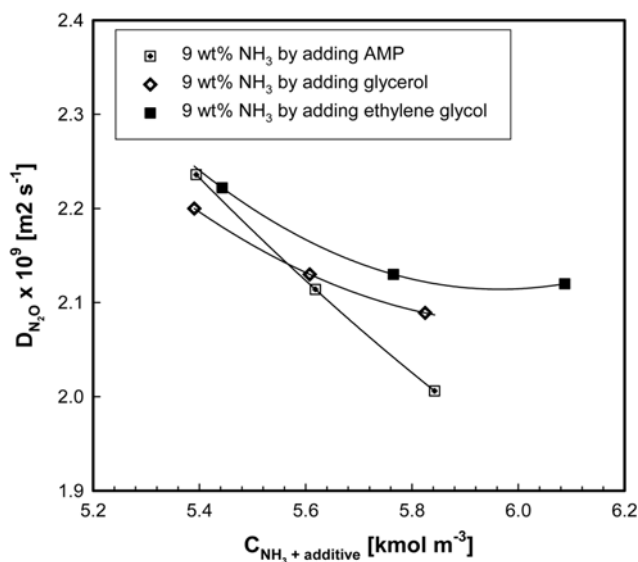


Fig. 9. Diffusivities of N₂O in aqueous 9 wt% NH₃+AMP, glycerol and ethylene glycol solutions as a function of the additive concentration at 313 K.

CONCLUSIONS

The physical solubilities and diffusivities of N₂O and CO₂ in aqueous ammonia solutions on the additions of AMP, ethylene glycol

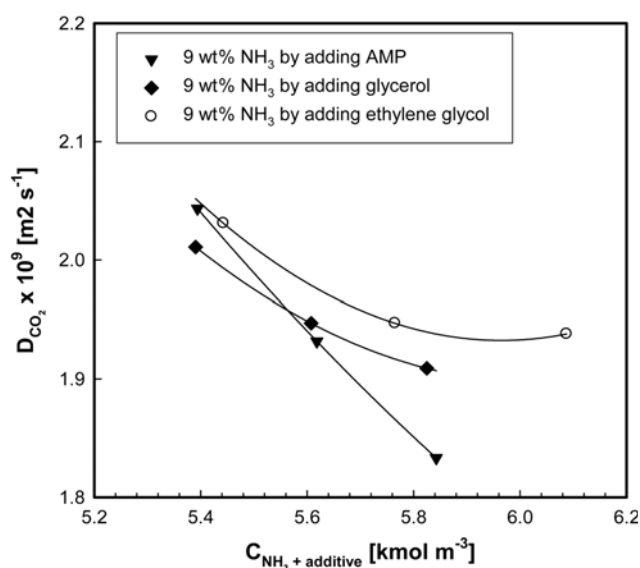
Table 7. Diffusivities of N₂O and CO₂ in aqueous 9 wt% NH₃+additive solutions as a function of temperature

T/K	$D_{N_2O}/10^9 \text{ m}^2\text{s}^{-1}$								
	9 wt% NH ₃								
	1 wt% AMP	3 wt% AMP	5 wt% AMP	1 wt% glycerol	3 wt% glycerol	5 wt% glycerol	1 wt% ethylene glycol	3 wt% ethylene glycol	5 wt% ethylene glycol
293	1.320	1.265	1.219	1.606	1.521	1.468	1.591	1.514	1.467
303	1.801	1.621	1.579	1.810	1.705	1.695	1.815	1.725	1.703
313	2.236	2.114	2.006	2.200	2.130	2.089	2.222	2.130	2.120
323	2.532	2.463	2.262	2.468	2.428	2.295	2.434	2.415	2.333

T/K	$D_{CO_2}/10^9 \text{ m}^2\text{s}^{-1}$								
	9 wt% NH ₃								
	1 wt% AMP	3 wt% AMP	5 wt% AMP	1 wt% glycerol	3 wt% glycerol	5 wt% glycerol	1 wt% ethylene glycol	3 wt% ethylene glycol	5 wt% ethylene glycol
293	1.209	1.159	1.117	1.471	1.393	1.345	1.458	1.387	1.344
303	1.692	1.523	1.483	1.700	1.601	1.592	1.704	1.620	1.599
313	2.044	1.932	1.833	2.011	1.947	1.909	2.031	1.947	1.938
323	2.325	2.261	2.077	2.266	2.230	2.107	2.235	2.218	2.142

Table 8. Diffusivity correlation parameters and AAD% of aqueous 9 wt% NH₃+additive solutions

	N ₂ O							
	q ₁	q ₂	q ₃	q ₄	q ₅	q ₆	z	AAD%
9 wt% NH ₃ +AMP	4.014×10 ²	7.601×10 ¹	1.439×10 ¹	−1.233×10 ²	−4.368×10 ¹	−2.335×10 ¹	1.974×10 ³	3.16
9 wt% NH ₃ +glycerol	8.023×10 ¹	1.519×10 ¹	2.876	−2.058×10 ¹	8.395	−3.896	1.469×10 ³	1.91
9 wt% NH ₃ +ethylene glycol	8.042×10 ¹	1.522×10 ¹	2.883	−1.592×10 ¹	1.310×10 ¹	−3.014	1.470×10 ³	1.93
	CO ₂							
	q ₁	q ₂	q ₃	q ₄	q ₅	q ₆	z	AAD%
9 wt% NH ₃ +AMP	3.336×10 ²	6.316×10 ¹	1.195×10 ¹	−1.046×10 ²	−3.044×10 ¹	−1.981×10 ¹	1.941×10 ³	2.55
9 wt% NH ₃ +glycerol	6.762×10 ¹	1.280×10 ¹	2.424	−1.744×10 ¹	7.336	−3.303	1.442 ×10 ³	1.27
9 wt% NH ₃ +ethylene glycol	6.773×10 ¹	1.282×10 ¹	2.428	−1.338×10 ¹	1.095×10 ¹	−2.534	1.442×10 ³	1.42

**Fig. 10. Diffusivities of CO₂ in aqueous 9 wt% NH₃+AMP, glycerol and ethylene glycol solutions as a function of the additive concentration at 313 K.**

and glycerol have been calculated at 293, 303, 313, 323 K. The N₂O analogy method was used to estimate the solubility and diffusivity of CO₂ in the amine solutions as the use of a nonreacting gas, such as N₂O. The solubility and diffusivity of CO₂ in ammonia solution were calculated by using the N₂O analogy method as the structure and properties of ammonia solution are similar to those of amines. New correlations are developed for the solubilities and diffusivities of N₂O and CO₂ in aqueous ammonia and additive solutions.

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NOMENCLATURE

AAD% : average absolute percent deviation [%]

C_A^{*} : dissolved gas concentration in equilibrium at the gas-liq-

liquid interface [kmol m⁻³]

- C_B : total ammonia concentration [kmol m⁻³]
 d : diameter of wetted wall column [m]
 D_A : diffusion coefficient of gas liquid [m² s⁻¹]
 h : height of wetted wall column [m]
 L : liquid flow rate [m³ s⁻¹]
 M_1 : concentration of ammonia [kmol m⁻³]
 M_2 : concentration of added absorbent [kmol m⁻³]
 N_A : specific absorption rate
 P : pressure
 p_A : CO₂ partial pressure [kPa]
 R : gas constant [J mol⁻¹ K⁻¹]
 T : temperature
 t_c : contact time [s]
 V : volume in the stirred cell [m³]

Greek Letters

- η : viscosity of aqueous ammonia [cP, mPas]
 ρ : density of liquid [g cm⁻³]

Subscripts

- A : gaseous species that is being absorbed into liquid B
 calc : calculation
 exp : experimental
 f : feeder
 L : liquid phase
 vap : vapor

REFERENCES

1. C. Hendriks, Kluwer Academic Publishers, Dordrecht, The Netherlands (1994).
2. G. Sartori and D. W. Savage, *Ind. Eng. Chem. Fundam.*, **22**, 293 (1983).
3. J. T. Yeh, K. P. Resnik, K. Rygle and H. W. Pennline, *Fuel Process. Technol.*, **86**, 1533 (2005).
4. W. J. Choi, J. B. Seo, S. W. Park and K. J. Oh, *Korean J. Chem. Eng.*, **26**, 705 (2009).
5. H. Bai and A. C. Yeh, *Ind. Eng. Chem. Res.*, **36**, 2490 (1997).
6. J. T. Yeh, K. P. Resnik and H. W. Pennline, *Prepr. Am. Chem. Soc., Fuel Chem.*, **49** (2004).
7. K. P. Resnik, J. T. Yeh and H. W. Pennline, *Int. J. Environ. Technol. Manage.*, **4**, 1 (2004).
8. Y. Diao, X. Y. Zheng, B. S. He, C. H. Chen and X. C. Xu, *Energy Convers. Manage.*, **45**, 2283 (2004).
9. J. K. You, H. S. Park, S. H. Yang, W. H. Hong, W. Shin, J. K. Kang, K. B. Yi and J. N. Kim, *J. Phys. Chem.*, **112**, 4323 (2008).
10. B. P. Mandal, M. Kundu and S. S. Bandyopadhyay, *J. Chem. Eng. Data*, **50**, 352 (2005).
11. G. F. Versteeg and W. P. M. van Swaaij, *J. Chem. Eng. Data*, **33**, 29 (1988).
12. M. H. Li and M. D. Lai, *J. Chem. Eng. Data*, **40**, 486 (1995).
13. B. P. Mandal, M. Kundu, N. U. Padhiyar and S. S. Bandyopadhyay, *J. Chem. Eng. Data*, **49**, 264 (2004).