

Solubility and Diffusivity of N₂O and CO₂ in (Monoethanolamine + *N*-Methyldiethanolamine + Water) and in (Monoethanolamine + 2-Amino-2-methyl-1-propanol + Water)

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The solubility and diffusivity of N₂O in (monoethanolamine + *N*-methyldiethanolamine + water) and in (monoethanolamine + 2-amino-2-methyl-1-propanol + water) were measured at 30, 35, and 40 °C and at atmospheric pressure. Six (monoethanolamine + *N*-methyldiethanolamine + water) and five (monoethanolamine + 2-amino-2-methyl-1-propanol + water) systems were studied. The total amine mass percent in all cases was 30. The solubilities were measured by a solubility apparatus similar to that of Haimour and Sandall (1984). A wetted wall column absorber was used to obtain the diffusivity of N₂O in amines. The N₂O solubilities in amine solutions have been correlated on the basis of the excess Henry constant correlation of Wang et al. (1992). The N₂O analogy was used to estimate the solubility and diffusivity of CO₂ in (monoethanolamine + *N*-methyldiethanolamine + water) and in (monoethanolamine + 2-amino-2-methyl-1-propanol + water).

Introduction

Solutions of amines are frequently used in gas-treating processes to remove acid gases, such as CO₂ and H₂S, from gas streams in the natural gas and synthetic ammonia industries and petroleum chemical plants. A wide variety of alkanolamines such as monoethanolamine (MEA), diethanolamine (DEA), di-2-propanolamine (DIPA), and *N*-methyldiethanolamine (MDEA) have been used industrially for a number of years. Due to the high reactivity, low solvent cost, ease of reclamation, and low absorption of hydrocarbons, MEA is one of the most important amines (Isaacs et al., 1980). For the selective removal of H₂S from a gas stream which contains both CO₂ and H₂S, MDEA is found to be an appropriate solution (Yu and Astarita, 1987; Srinivasan and Aiken, 1988). Advantages of the use of MDEA solutions include their high loading capacity (about 1.0 mol of CO₂/mol of amine) and low enthalpy of reaction with the acid gases (Riesenfeld and Brocoff, 1986).

Recently the use of blended amines, a solution of two or more amines in varying compositions, shows considerable improvement in absorption and savings in energy requirements (Chakravarty et al., 1985). Blended amines combine the absorption characteristics of the constituent amines such as a higher loading capacity and faster reaction rates. Blends of primary and tertiary amines, such as mixtures of MEA and MDEA, have been suggested for CO₂ removal (Chakravarty et al., 1985).

Sterically hindered alkanolamines have also been proposed as commercially attractive solvents for removal of acid gases from gaseous streams (Sartori and Savage, 1983). An example of the sterically hindered alkanolamines is 2-amino-2-methyl-1-propanol (AMP), which is the hindered form of MEA. Due to the bulkiness of the group attached to a tertiary carbon atom of AMP, the formation of carbamates is inhibited when AMP reacts with CO₂ (Sartori and Savage, 1983; Yih and Shen, 1988). The CO₂ loading of AMP can thus approach a value of 1 mol of CO₂/mol of amine (Sartori and Savage, 1983). Compared to

MDEA, AMP has the same high loading capacity but has a higher reaction rate constant for the reaction with CO₂ (Yih and Shen, 1988; Crooks and Donnellan, 1990). Therefore, MEA + AMP + H₂O may be an attractive new solvent in addition to MEA + MDEA + H₂O for the acid gases treating process.

For the rational design of the gas absorption units, physical properties such as solubility and diffusivity of acid gases in amine solutions are required to model the rate of absorption. Due to the reaction between CO₂ and amines, the directly measured solubility essentially is the total solubility, which contains both physical and chemical absorptions. However, it is the free-gas solubility (physical) that is needed to model the rate of absorption (Haimour, 1990). Because of the chemical reaction between CO₂ and amines, the free-gas solubility cannot be measured directly. It has been suggested that the solubility of CO₂ in reacting solutions can be estimated by using the N₂O analogy (Clarke, 1964). The N₂O analogy has been supported by the fact that the ratios of the solubilities of N₂O and CO₂ in different alcohols are nearly a constant value of 1.37 at 25 °C (Laddha et al., 1981). The N₂O analogy had been frequently used to predict the solubility of CO₂ in amine solutions (Haimour and Sandall, 1984; Versteeg and van Swaaij, 1988; Al-Ghawas et al., 1989; Xu et al., 1991; Saha et al., 1993).

Like the CO₂ solubility in amine, the diffusivity of CO₂ in the amine solution cannot be measured directly. Various methods to estimate the diffusivity of CO₂ in amine solutions have been proposed in the literature. The N₂O analogy has also been suggested to estimate the diffusivity of CO₂ in amine solutions (Sada et al., 1978; Haimour and Sandall, 1984; Al-Ghawas et al., 1989). Díaz et al. (1988) proved this analogy from measurements of diffusivity of CO₂ and N₂O in aqueous alcohol solutions. To estimate the diffusivity of CO₂ in aqueous AMP solutions, the N₂O analogy has been suggested as preferable to the Stokes-Einstein relation (Xu et al., 1991; Saha et al., 1993).

The N₂O analogy for the solubility and diffusivity has

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the following relations:

$$H_{\text{CO}_2} = H_{\text{N}_2\text{O}}(H_{\text{CO}_2}/H_{\text{N}_2\text{O}})_{\text{in water}} \quad (1)$$

$$D_{\text{CO}_2} = D_{\text{N}_2\text{O}}(D_{\text{CO}_2}/D_{\text{N}_2\text{O}})_{\text{in water}} \quad (2)$$

On the basis of the available data of solubility and diffusivity of N_2O and CO_2 in water, Versteeg and van Swaaij (1988) proposed four convenient equations as follows:

$$H_{\text{N}_2\text{O}}/(\text{kPa}\cdot\text{m}^3\cdot\text{kmol}^{-1}) = 8.5470 \times 10^6 \exp[-2284/(T/K)] \quad (3)$$

$$H_{\text{CO}_2}/(\text{kPa}\cdot\text{m}^3\cdot\text{kmol}^{-1}) = 2.8249 \times 10^6 \exp[-2044/(T/K)] \quad (4)$$

$$D_{\text{N}_2\text{O}}/(\text{m}^2\cdot\text{s}^{-1}) = 5.07 \times 10^{-6} \exp[-2371/(T/K)] \quad (5)$$

$$D_{\text{CO}_2}/(\text{m}^2\cdot\text{s}^{-1}) = 2.35 \times 10^{-6} \exp[-2119/(T/K)] \quad (6)$$

Some solubilities and diffusivities of N_2O and CO_2 in amine solutions were reported in the literature such as MEA (Sada et al., 1978; Wang et al., 1992), MDEA (Haimour and Sandall, 1984; Versteeg and van Swaaij, 1988; Al-Ghawas et al., 1989), and AMP (Xu et al., 1991; Saha et al., 1993). However, solubility and diffusivity of N_2O and CO_2 in (MEA + MDEA + H_2O) and in (MEA + AMP + H_2O) have not been reported in the literature. Therefore, it is the purpose of this research to measure the solubility and diffusivity of N_2O in (MEA + MDEA + H_2O) and in (MEA + AMP + H_2O). The N_2O analogy will then be applied to estimate the solubility and diffusivity of CO_2 in (MEA + MDEA + H_2O) and in (MEA + AMP + H_2O).

Experimental Section

The water was distilled and then degassed by boiling. MEA is Riedel-de Haën reagent grade with a 99 mol % purity, MDEA is Riedel-de Haën reagent grade with a 98.5 mol % purity, and AMP is Riedel-de Haën reagent grade with a 98 mol % purity.

1. Solubility Measurement. The solubility of N_2O in amine aqueous solutions was measured with an apparatus similar to that described by Weiland and Trass (1971),

Haimour and Sandall (1984), Al-Ghawas et al. (1989), and Haimour (1990). The solubility apparatus consists of a saturation flask, an absorption flask with a magnetic stirrer, and a set of burettes with branches filled with mercury. The temperature of the whole unit was kept constant within $\pm 0.5^\circ\text{C}$ by a constant-temperature water bath with water circulating through a jacket enclosing the measuring branches. The system was first purged with saturated N_2O for 5 min at the constant temperature. Then the inlet and outlet valves were closed. A known volume of amine solution was injected into the absorption flask. Mixing of the gas and amine solution was performed by the magnetic stirrer. The volume change of the system can be determined by the levels of the mercury in the burette. When the volume of the gas remains unchanged for 30 min, equilibrium is assumed to have been reached. At constant temperature and pressure, the volume of gas absorbed can be determined by the volume change of the system and the volume of the solution. The reliability of the apparatus was checked by measuring the solubility of N_2O in water and of CO_2 in water. The estimated experimental error in the measured solubility is about $\pm 2\%$.

2. Diffusivity Measurement. The diffusivity of a gas in amine solutions was measured in a short wetted wall column absorber. A schematic diagram of the apparatus is shown in Figure 1. A short, stainless steel cylinder of outside diameter 2.54 cm and a height of 10 cm was used as the wetted wall column. The liquid flowed through an annular distributor cap and distributed uniformly as a thin film on the outside of the cylinder. The liquid flow rate was varied in the range $3\text{--}6\text{ cm}^3\cdot\text{s}^{-1}$. The gas flow rate was measured by using a soap-film meter. The absorption rate was measured by the difference between the inlet and outlet gas flow rates. The system was first purged with the saturated gas. The solution was heated to the desired temperature and allowed to flow through the wetted wall column. When the solution distributed itself uniformly as a thin film on the outside of the cylinder, the liquid flow rate and the gas absorption rate were measured. The apparatus and the experimental procedure were the same as those described by Yih and Shen (1988) and Shen et al. (1991).

For physical absorption of a sparingly soluble gas at short contact time and with an initial gas-free liquid, the Higbie penetration theory gives the specific absorption rate as

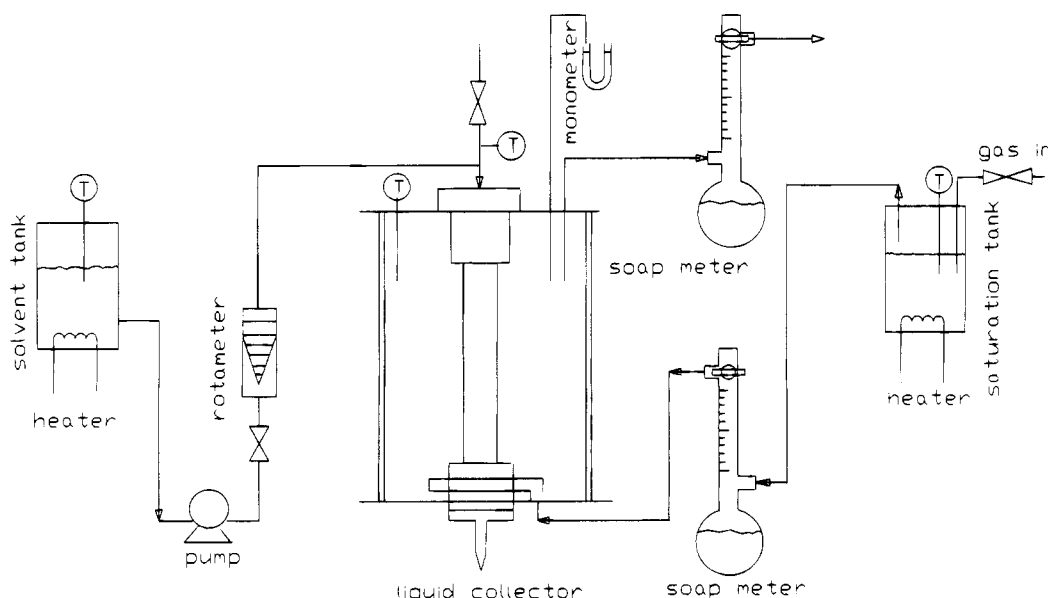


Figure 1. Wetted wall column absorber apparatus.

Table 1. Solubility of CO₂ and N₂O in Water

	<i>t</i> /°C	<i>H</i> / (kPa·m ³ ·kmol ⁻¹)	reference
CO ₂	30	3394	Al-Ghawas et al., 1989
		3332 ^a	Versteeg and van Swaaij, 1988
		3382	this study
	40	4250	Al-Ghawas et al., 1989
		4133 ^a	Versteeg and van Swaaij, 1988
		4227	this study
	50	5167	Al-Ghawas et al., 1989
		5058 ^a	Versteeg and van Swaaij, 1988
		5136	this study
N ₂ O	30	4350	Al-Ghawas et al., 1989
		4568 ^b	Versteeg and van Swaaij, 1988
		4406	this study
	40	5021	Al-Ghawas et al., 1989
		5811 ^b	Versteeg and van Swaaij, 1988
		5725	this study
	50	5369	Al-Ghawas et al., 1989
		7282 ^b	Versteeg and van Swaaij, 1988
		7254	Xu et al., 1991
	7264	this study	

^a Calculated using eq 4. ^b Calculated using eq 3.

(Danckwerts, 1970)

$$N_A = 2(D_A/\pi t_c)^{1/2}(P_A/H_A) \quad (7)$$

where D_A is the diffusivity of the absorbed gas in the solution, t_c is the contact time, P_A is the partial pressure of the gas at interface, H_A is the Henry constant of the absorbed gas. The contact time can be derived from wetted wall column hydrodynamics as (Danckwerts, 1970)

$$t_c = (2h/3)(\pi d/L)^{2/3}(3\mu/\rho g)^{1/3} \quad (8)$$

where h is the height of the wetted wall column, d is the diameter of the wetted wall column, L is the liquid flow rate, μ is the viscosity of the fluid, ρ is the density of the fluid, and g is the gravitational acceleration. Equations 7 and 8 only apply so long as the flow is nonturbulent and free from ripples (Danckwerts, 1970).

From eq 7, one has

$$D_A = (N_A H_A)^2 \pi t_c / (2 P_A)^2 \quad (9)$$

The diffusivity of the gas in solution can be calculated by the Henry constant, specific gas absorption rate, and contact time, density, and viscosity of the solution. The density and viscosity of the amine solutions will be adopted from our previous work (Li and Lie, 1994). To calibrate the wetted wall column absorber, the diffusivities of N₂O in water and CO₂ in water were measured. The estimated experimental error in the measured diffusivity is estimated as $\pm 2\%$.

Results and Discussion

1. Solubility. To examine the applicability of the present experimental solubility measurement, the solubilities of N₂O in water and CO₂ in water were measured with the solubility apparatus. The measured solubilities of N₂O in water and CO₂ in water at 30, 40, and 50 °C along with literature results are presented in Table 1. In Figure 2, the comparison between the literature values (Duda and Vrentas, 1968; Versteeg and van Swaaij, 1988; Al-Ghawas et al., 1989; Xu et al., 1991) and values obtained in this study for the solubility of N₂O in water are shown. Also in Figure 3, the comparison between the values obtained

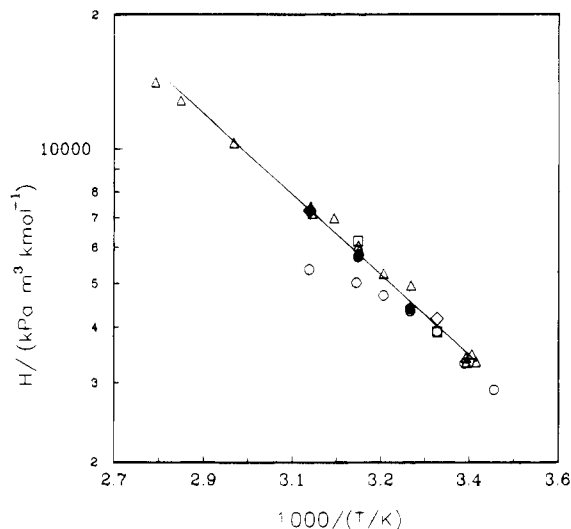


Figure 2. Solubility of N₂O in water as a function of temperature: ●, this study; ○, data of Al-Ghawas et al. (1989); □, data of Duda and Vrentas (1968); △, data of Versteeg and van Swaaij (1988); ◇, data of Xu et al. (1991); —, calculated using eq 3.

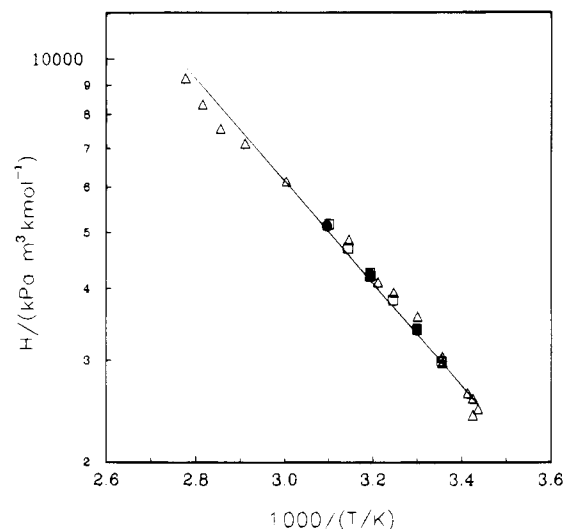


Figure 3. Solubility of CO₂ in water as a function of temperature: ●, this study; □, data of Al-Ghawas et al. (1989); △, data of Versteeg and van Swaaij (1988); —, calculated using eq 4.

in this study and literature values (Versteeg and van Swaaij, 1988; Al-Ghawas et al., 1989) for the solubility of CO₂ in water are shown. In Figures 2 and 3, the solid lines are calculated values using eqs 3 and 4 (Versteeg and van Swaaij, 1988). As can be seen from Figures 2 and 3, there is a good agreement between literature results and those of the present study. The data for the solubility of N₂O in (MEA + MDEA + H₂O) and in (MEA + AMP + H₂O) at 30, 35, and 40 °C are presented in Tables 2 and 3 and plotted in Figures 4 and 5. As shown in Figure 4, the solubility of N₂O in (MEA + MDEA + H₂O) increases systematically from 30 mass % MEA to 30 mass % MDEA at constant temperature. Also, the solubilities of N₂O in 30 mass % AMP are higher than those in 30 mass % MEA as shown in Figure 5.

To correlate the solubility of N₂O in amine solutions, a semiempirical model proposed by Wang et al. (1992) was used. In this method, the excess Henry coefficient for the mixed solvent system has the form

$$R = \ln H_{1,m} - \sum_{i=2}^n \Phi_i \ln H_{1,i} \quad (10)$$

Table 2. Estimated Solubility of CO₂ in MEA + MDEA + H₂O Using the N₂O Analogy

<i>t</i> /°C	in water		in MEA + MDEA + H ₂ O			
	<i>H</i> _{CO₂} ^a /(kPa·m ³ ·kmol ⁻¹)	<i>H</i> _{N₂O} ^b /(kPa·m ³ ·kmol ⁻¹)	mass % MEA +	mass % MDEA	<i>H</i> _{N₂O} ^b /(kPa·m ³ ·kmol ⁻¹)	<i>H</i> _{CO₂} ^a /(kPa·m ³ ·kmol ⁻¹)
30	3332.40	4568.17	30 + 0		4361.9	3181.9
			24 + 6		4481.2	3269.0
			18 + 12		4579.6	3340.7
			12 + 18		4696.2	3425.8
			6 + 24		4867.6	3550.8
			0 + 30		5168.9	3770.6
35	3717.66	5162.19	30 + 0		4696.4	3382.2
			24 + 6		4859.6	3499.7
			18 + 12		4957.3	3570.1
			12 + 18		5155.9	3713.1
			6 + 24		5391.6	3882.9
			0 + 30		5585.0	4022.2
40	4133.01	5810.73	30 + 0		5126.9	3646.6
			24 + 6		5276.8	3753.2
			18 + 12		5396.3	3838.2
			12 + 18		5624.0	4000.2
			6 + 24		5934.8	4221.3
			0 + 30		6188.0	4401.4

^a Calculated using eq 4. ^b Calculated using eq 3.

Table 3. Estimated Solubility of CO₂ in MEA + AMP + H₂O Using the N₂O Analogy

<i>t</i> /°C	in water		in MEA + AMP + H ₂ O			
	<i>H</i> _{CO₂} ^a	<i>H</i> _{N₂O} ^b /(kPa·m ³ ·kmol ⁻¹)	mass % MEA +	mass % AMP	<i>H</i> _{N₂O} ^b /(kPa·m ³ ·kmol ⁻¹)	<i>H</i> _{CO₂} ^a /(kPa·m ³ ·kmol ⁻¹)
30	3332.40	4568.17	30 + 0		4361.9	3181.9
			24 + 6		4547.5	3317.3
			18 + 12		4910.8	3582.3
			12 + 18		5414.5	3949.8
			6 + 24		5597.1	4083.0
			0 + 30		5855.5	4271.5
35	3717.66	5162.19	30 + 0		4696.4	3382.2
			24 + 6		5000.4	3601.1
			18 + 12		5656.4	4073.6
			12 + 18		5960.8	4292.8
			6 + 24		6303.6	4539.7
			0 + 30		6545.2	4713.7
40	4133.01	5810.73	30 + 0		5126.9	3646.6
			24 + 6		5544.7	3943.8
			18 + 12		6130.8	4360.7
			12 + 18		6813.3	4846.1
			6 + 24		7144.5	5081.7
			0 + 30		7530.6	5356.3

^a Calculated using eq 4. ^b Calculated using eq 3.

where *H*_{1,m} is the Henry constant of the absorbed gas in the mixed solvent, *H*_{1,i} is the Henry constant of the absorbed gas in pure solvent *i*, and Φ_{*i*} is the volume fraction of solvent *i*. The volume fraction is calculated as

$$\Phi_i = x_i v_i / \sum_{i=1}^n x_i v_i \quad (11)$$

where *v*_{*i*} is the molar volume of pure solvent *i* and *x*_{*i*} is the mole fraction of solvent *i*. In the method of Wang et al., the Henry constants of N₂O in pure solvents such as MEA, MDEA, and AMP have been measured and correlated as an exponential function of temperature as

$$H_{N_2O\text{-amine}} = b_1 \exp(b_2/T) \quad (12)$$

where *b*₁ and *b*₂ are parameters for pure amine. The densities of pure amines were also correlated as functions of temperature in the method of Wang. The Henry constant of N₂O in water was calculated using eq 3. By the use of eq 10, the excess Henry constant of N₂O in mixed solvent can be calculated from the measured *H*_{1,m}.

For binary solvent systems, the excess Henry constant was correlated as

$$R_{ij} = \Phi_i \Phi_j \alpha_{ij} \quad (13)$$

where α_{*ij*} is temperature dependent and is assumed in this study to be the expression

$$\alpha_{ij} = a_1 + a_2/T \quad (14)$$

where *T* is in Kelvins and *a*₁ and *a*₂ are constants and were determined using the N₂O solubility in (MEA + H₂O), (MDEA + H₂O), and (AMP + H₂O).

For ternary solvent systems, the excess Henry constant has the form

$$R_{ijk} = \Phi_i \Phi_j \alpha_{ij} + \Phi_i \Phi_k \alpha_{ik} + \Phi_j \Phi_k \alpha_{jk} + \Phi_i \Phi_j \Phi_k \alpha_{ijk} \quad (15)$$

In this study, α_{*ij*} for MEA + MDEA and MEA + AMP and α_{*ijk*} were determined from the solubility of N₂O in (MEA + MDEA + H₂O) and in (MEA + AMP + H₂O) and are presented in Table 4. α_{*ijk*} in eq 15 is set to be a constant. The comparisons of the calculated and experi-

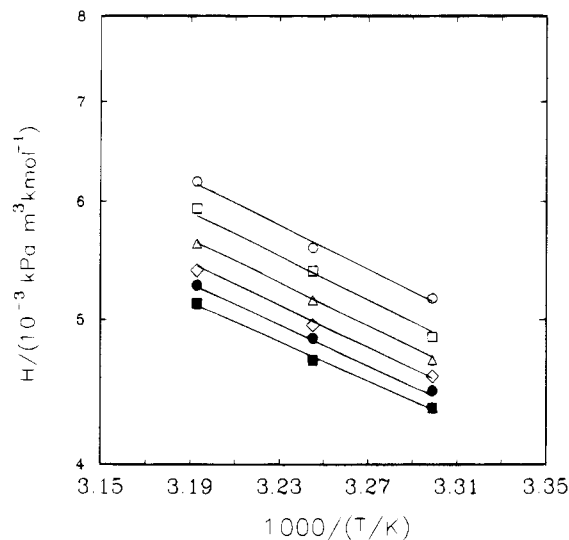


Figure 4. Solubility of N_2O in MEA + MDEA + H_2O : \circ , 30 mass % MDEA; \square , 6 mass % MEA + 24 mass % MDEA; \triangle , 12 mass % MEA + 18 mass % MDEA; \diamond , 18 mass % MEA + 12 mass % MDEA; \bullet , 24 mass % MEA + 6 mass % MDEA; \blacksquare , 30 mass % MEA; $-$, calculated using eq 10.

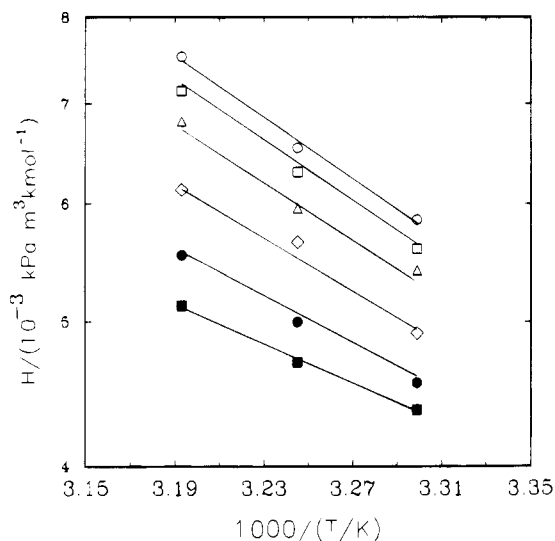


Figure 5. Solubility of N_2O in MEA + AMP + H_2O : \circ , 30 mass % AMP; \square , 6 mass % MEA + 24 mass % AMP; \triangle , 12 mass % MEA + 18 mass % AMP; \diamond , 18 mass % MEA + 12 mass % AMP; \bullet , 24 mass % MEA + 6 mass % AMP; \blacksquare , 30 mass % MEA; $-$, calculated using eq 10.

Table 4. Parameters in Excess Henry Constant for Binary and Ternary Solvent Systems

system	a_1	a_2	a_{234}	AAD% ^a
MEA + H_2O	-5.9535	1938.24		0.37
MDEA + H_2O	-2.8999	1405.43		0.63
AMP + XXXXX	9.4188	-2056.59		0.74
MEA + MDEA	36.8828	-1637.43		
MEA + AMP	151.822	-5972.13		
MEA + MDEA + H_2O			-45.8808	0.46
MEA + AMP + H_2O			-189.666	0.91

^a AAD%, average absolute percent deviation.

mental solubilities of N_2O in amine solutions are shown in Figures 4 and 5. The overall average absolute percent deviation for the calculation of solubility of N_2O in amine solutions is 0.66%. The results are satisfactory. Thus, the N_2O solubilities in amine solutions are well correlated using the method of Wang et al. (1992).

Table 5. Diffusivity of CO_2 and N_2O in Water

	$t/^\circ C$	$10^9 D/(m^2 s^{-1})$	reference
CO_2	30	2.160	Thomas and Adams, 1965
		2.165 ^a	Versteeg and van Swaaij, 1988
		2.120	this study
	35	2.420	Thomas and Adams, 1965
		2.425 ^a	Versteeg and van Swaaij, 1988
		2.462	this study
	40	2.710	Thomas and Adams, 1965
		2.706 ^a	Versteeg and van Swaaij, 1988
		2.775	this study
N_2O	30	1.607	Al-Ghawas et al., 1989
		2.034 ^b	Versteeg and van Swaaij, 1988
		2.009	this study
	35	1.634	Al-Ghawas et al., 1989
		2.309 ^b	Versteeg and van Swaaij, 1988
		2.302	this study
	40	1.679	Al-Ghawas et al., 1989
		2.611 ^b	Versteeg and van Swaaij, 1988
		2.648	this study

^a Calculated using eq 6. ^b Calculated using eq 5.

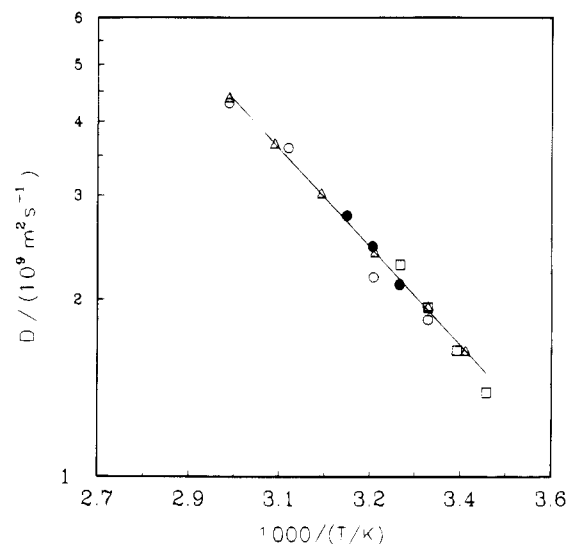


Figure 6. Diffusivity of CO_2 in water as a function of temperature: \bullet , this study; \square , data of Davidson and Cullen (1957); \circ , data of Unver and Himmelblau (1964); \triangle , data of Thomas and Adams (1965); $-$, calculated using eq 6.

By applying the N_2O analogy and the solubilities of N_2O in water and CO_2 in water, calculated by eqs 3 and 4, the solubilities of CO_2 in (MEA + MDEA + H_2O) and in (MEA + AMP + H_2O) were estimated and are presented in Tables 2 and 3, respectively.

2. Diffusivity. The diffusivity of N_2O in amine solution was measured by the wetted wall column absorber. To confirm the apparatus and the experimental procedure, the diffusivities of N_2O and CO_2 in water were measured at 30, 35, and 40 $^\circ C$. The results are presented in Table 5. The comparisons between the literature values (Davidson and Cullen, 1957; Unver and Himmelblau, 1964; Thomas and Adams, 1965; Versteeg and van Swaaij, 1988) and the values obtained in this study for the diffusivity of CO_2 in water are also shown in Figure 6. The solid line in Figure 6 is composed of values calculated using eq 6. As shown in Figure 6, the values obtained in this study are in good agreement with the literature values. The diffusivities of N_2O in amine solutions are presented in Tables 6 and 7 for (MEA + MDEA + H_2O) and (MEA + AMP + H_2O), respectively. At the same temperature, the diffusivity of

Table 6. Estimated Diffusivity of CO₂ in MEA + MDEA + H₂O Using the N₂O Analogy

t/°C	in water		in MEA + MDEA + H ₂ O		
	$D_{CO_2^a}/(10^9 m^2 s^{-1})$	$D_{N_2O^b}/(10^9 m^2 s^{-1})$	mass %	$D_{N_2O}/(10^9 m^2 s^{-1})$	$D_{CO_2}/(10^9 m^2 s^{-1})$
			MEA + mass % MDEA		
30	2.165	2.034	30 + 0	1.559	1.659
			24 + 6	1.491	1.587
			18 + 12	1.396	1.486
			12 + 18	1.336	1.422
			6 + 24	1.244	1.324
			0 + 30	1.084	1.154
35	2.425	2.309	30 + 0	1.731	1.818
			24 + 6	1.665	1.749
			18 + 12	1.546	1.624
			12 + 18	1.464	1.538
			6 + 24	1.351	1.419
			0 + 30	1.180	1.239
40	2.706	2.611	30 + 0	1.879	1.947
			24 + 6	1.807	1.873
			18 + 12	1.676	1.737
			12 + 18	1.572	1.629
			6 + 24	1.465	1.518
			0 + 30	1.285	1.332

^a Calculated using eq 6. ^b Calculated using eq 5.

Table 7. Estimated Diffusivity of CO₂ in MEA + AMP + H₂O Using the N₂O Analogy

t/°C	in water		in MEA + AMP + H ₂ O		
	$D_{CO_2^a}/(10^9 m^2 s^{-1})$	$D_{N_2O^b}/(10^9 m^2 s^{-1})$	mass %	$D_{N_2O}/(10^9 m^2 s^{-1})$	$D_{CO_2}/(10^9 m^2 s^{-1})$
			MEA + mass % AMP		
30	2.165	2.034	30 + 0	1.559	1.659
			24 + 6	1.509	1.606
			18 + 12	1.420	1.512
			12 + 18	1.324	1.409
			6 + 24	1.214	1.292
			0 + 30	1.047	1.114
35	2.425	2.309	30 + 0	1.731	1.818
			24 + 6	1.628	1.710
			18 + 12	1.567	1.646
			12 + 18	1.458	1.531
			6 + 24	1.353	1.421
			0 + 30	1.157	1.215
40	2.706	2.611	30 + 0	1.879	1.947
			24 + 6	1.804	1.870
			18 + 12	1.726	1.789
			12 + 18	1.599	1.657
			6 + 24	1.490	1.544
			0 + 30	1.303	1.350

^a Calculated using eq 6. ^b Calculated using eq 5.

N₂O in 30 mass % MEA is higher than that in either 30 mass % MDEA or 30 mass % AMP, as shown in Tables 6 and 7. At 30 °C, the viscosity of 30 mass % MEA solution is 2.109 mPa·s, which is smaller than that of either 30 mass % MDEA (2.626 mPa·s) or 30 mass % AMP (3.053 mPa·s) (Li and Lie, 1994). This may explain why diffusivities of N₂O in 30 mass % MEA are higher than that in either 30 mass % MDEA or 30 mass % AMP solutions.

To correlate the diffusivity of N₂O in a single-amine aqueous solution, the following form is assumed:

$$D = (b_0 + b_{i1}M_i + b_{i2}M_i^2) \exp(-c/T) \quad (16)$$

M_i is the molarity of the amine solution and b_i and c are parameters. For the blended amine solutions, the following form is assumed:

$$D = (b_0 + b_{11}M_1 + b_{12}M_1^2 + b_{21}M_2 + b_{22}M_2^2 + c_{12}M_1M_2) \exp(-c/T) \quad (17)$$

M_1 is the molarity of MEA and M_2 is the molarity of MDEA or AMP. The parameters in eq 17 were determined using

Table 8. Determined Parameters of the Diffusivity of N₂O in Amine Solutions Using Eq 17

parameter	MEA + MDEA + H ₂ O	MEA + AMP + H ₂ O
b_0	-1.89645×10^{-7}	1.61801×10^{-6}
b_{11}	2.44679×10^{-7}	-2.43516×10^{-7}
b_{12}	-2.71690×10^{-8}	-8.52221×10^{-9}
b_{21}	3.98653×10^{-7}	-3.99462×10^{-7}
b_{22}	-8.96763×10^{-8}	-1.51765×10^{-8}
c_{12}	-8.75280×10^{-8}	3.22860×10^{-8}
c	-1644.03	-1496.75
AAD%	0.6	0.5

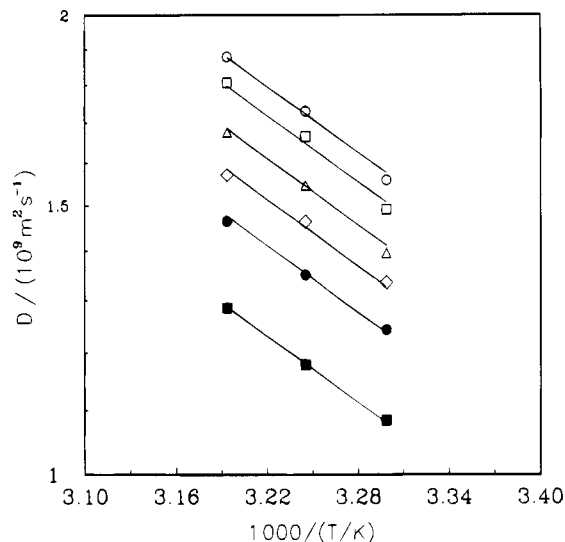


Figure 7. Diffusivity of N₂O in MEA + MDEA + H₂O: ○, 30 mass % MEA; □, 24 mass % MEA + 6 mass % MDEA; △, 18 mass % MEA + 12 mass % MDEA; ◇, 12 mass % MEA + 18 mass % MDEA; ●, 6 mass % MEA + 24 mass % MDEA; ■, 30 mass % MDEA; —, calculated using eq 17.

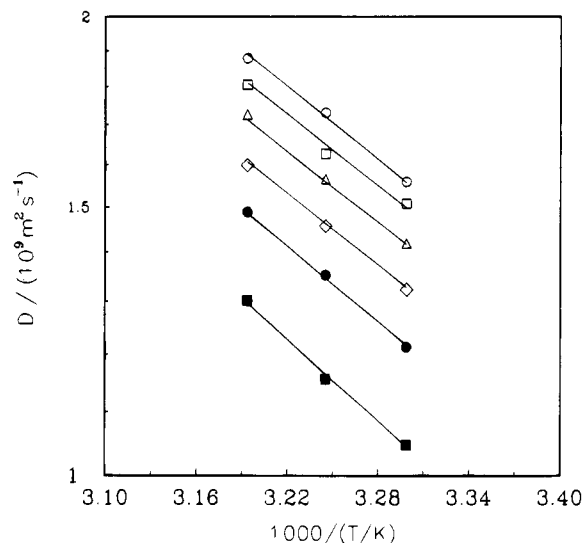


Figure 8. Diffusivity of N₂O in MEA + AMP + H₂O: ○, 30 mass % MEA; □, 24 mass % MEA + 6 mass % AMP; △, 18 mass % MEA + 12 mass % AMP; ◇, 12 mass % MEA + 18 mass % AMP; ●, 6 mass % MEA + 24 mass % AMP; ■, 30 mass % AMP; —, calculated using eq 17.

the diffusivity data in Tables 6 and 7, and are presented in Table 8. The overall average absolute percentage deviations for the calculations of the diffusivity of N₂O in amine solutions are 0.6% and 0.5% for (MEA + MDEA + H₂O) and (MEA + AMP + H₂O), respectively. Figures 7 and 8 show the comparison between the experimental values and calculated values using eq 17 for the diffusivity of N₂O in amine solutions. As shown in Figures 7 and 8,

the calculations of the diffusivity of N₂O in amine solutions are satisfactory. Applying the N₂O analogy and eqs 5 and 6, the diffusivities of CO₂ in amine solutions are estimated and presented in Tables 6 and 7 for (MEA + MDEA + H₂O) and (MEA + AMP + H₂O), respectively.

Conclusion. The solubility and diffusivity of N₂O in (MEA + MDEA + H₂O) and (MEA + AMP + H₂O) were measured at 30, 35, and 40 °C and at atmospheric pressure. Six (MEA + MDEA + H₂O) and five (MEA + AMP + H₂O) systems were studied. The total amine mass percent in all cases was 30. The solubility data were measured by a solubility apparatus similar to that of Haimour and Sandall (1984). A wetted wall column absorber was used to obtain the diffusivity of N₂O in amine solutions. The N₂O solubilities in amine solutions have been correlated based on the excess Henry constant correlation of Wang et al. (1988). The N₂O analogy was used to estimate the solubility and diffusivity of CO₂ in (MEA + MDEA + H₂O) and (MEA + AMP + H₂O).

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