Diffusivity of Nitrous Oxide in Aqueous Alkanolamine Solutions

Jiun-Jie Ko, Tung-Chien Tsai, Chih-Yuan Lin, Hsiun-Min Wang, and Meng-Hui Li*

Department of Chemical Engineering, Chung Yuan Christian University, Chung Li,
Taiwan 32023, Republic of China

The diffusivities of nitrous oxide (N_2O) in aqueous alkanolamine solutions have been measured at (30, 35, and 40) °C. The systems studied are aqueous monoethanolamine, diethanolamine, diisopropanolamine, triethanolamine, and 2-amino-2-methyl-1-propanol solutions. The concentration of amine for monoethanolamine ranges from (1 to 5) kmol·m^{-3} and for other amines from (0.5 to 2.5) kmol·m^{-3}. The viscosities of diisopropanolamine and triethanolamine and the densities and viscosities of aqueous diisopropanolamine and triethanolamine solutions were also measured at (30, 35, and 40) °C. A simple correlation was used to correlate the diffusivity of N_2O in amine solutions. The parameters of the correlation were determined from the measured diffusivity data and the available data in the open literature. The correlation has been shown to represent reasonably the diffusivity of N_2O in six aqueous amine solutions: MEA, DEA, DIPA, MDEA, TEA, and AMP. The correlations are, in general, satisfactory for estimating the diffusivity of N_2O in amine solutions, which in turn can be used to estimate the free molecular diffusivity of CO_2 in amines.

Introduction

Solutions of alkanolamines are widely encountered in gas−liquid contact operations as absorbents for acidic gases such as CO_2 and H_2S. A wide variety of alkanolamines such as monoethanolamine (MEA), diglycolamine (DGA), diethanolamine (DEA), diisopropanolamine (DIPA), triethanolamine (TEA), *N*-methyldiethanolamine (MDEA), 2-amino-2-methyl-l-propanol (AMP), and 2-piperidineethanol (2-PE) can be used as absorbents for acid gas removal processes.[1] Aqueous alkanolamine solutions are more frequently used for gas sweetening than hot potassium carbonate solutions, since they require less energy and have lower corrosion rates.[2]

To design a process for the removal of CO_2, it is necessary to use the free molecular diffusivity of CO_2 in aqueous alkanolamines to model the reaction kinetics between CO_2 and alkanolamine. Due to the chemical reaction between CO_2 and amines, the free molecular diffusivity of CO_2 in amine solution cannot be measured directly. The N_2O analogy has been frequently used to estimate the diffusivity of CO_2 in amine solutions.[3−11]

The N_2O analogy for the free molecular diffusivity of CO_2 in amine solution has the following relation

$$D_{CO_2} = D_{N_2O}(D_{CO_2}/D_{N_2O})_{\text{in water}} \qquad (1)$$

where D_{N_2O} is the diffusivity of N_2O in amine solution.

On the basis of the available data of diffusivity of N_2O and CO_2 in water, Versteeg and van Swaaij[6] have proposed two convenient equations as follows

$$D_{N_2O}/(\mathrm{m^2{\cdot}s^{-1}}) = 5.07 \times 10^{-6} \exp\{-2371/(T/\mathrm{K})\} \qquad (2)$$

$$D_{CO_2}/(\mathrm{m^2{\cdot}s^{-1}}) = 2.35 \times 10^{-6} \exp\{-2119/(T/\mathrm{K})\} \qquad (3)$$

From eqs 1−3, the free molecular diffusivity of CO_2 in aqueous amine solution can be estimated from the measured diffusivity of N_2O in the same solution.

Although numerous diffusivity data of N_2O in aqueous amine solutions were reported in the literature such as in MEA,[3,12,13] in DEA,[12,14,15] in DIPA,[6,8,12] in MDEA,[4,6,7,14,16−18] in TEA,[9,12] and in AMP,[10,11,19] some of these data are scattered and inconsistent. For example, for the diffusivity of N_2O in aqueous DEA solution at 25 °C, Sada et al.[12] reported the value as 1.26×10^{-9} m^2·s^{-1} at the concentration of DEA 2.808 kmol·m^{-3} while Tamimi et al.[14] reported it as 0.693×10^{-9} m^2·s^{-1} at the concentration of DEA 2.853 kmol·m^{-3}. The scattered and inconsistent diffusivity data of N_2O in amine may contribute to the inconsistent results for the reaction kinetics study in the literature.[20] Accordingly, the correct diffusivity of N_2O in amines is essential to estimate the correct free molecular diffusivity of CO_2 in amine, which in turn can be used in developing the correct reaction kinetic models. This paper reports the diffusivity of N_2O in aqueous MEA, DEA, DIPA, TEA, and AMP solutions at the temperatures (30, 35, and 40) °C. The viscosities of DIPA and TEA and the densities and viscosities of DIPA + H_2O and TEA + H_2O required in the diffusivity measurement have also been measured. A simple equation has been used to correlate the diffusivity of N_2O in aqueous amine solutions. The parameters of the correlation for each system have been determined from both the diffusivity of N_2O measured in this study and the available data in the open literature. The correlation can be used to estimate the diffusivity of N_2O in aqueous amine solutions: MEA, DEA, DIPA, MDEA, TEA, and AMP over wide temperature and concentration ranges.

Experimental Section

Chemicals. Some chemicals are Riedel-de Haën reagent grade with the following purities: MEA, min. 99% (water max. 0.5%); DEA, min. 99% (water max. 0.3%); TEA, min. 99% (water max. 0.2%); AMP, min. 95%. DIPA with a purity of min. 95% is from Aldrich Chemical Co. The gas

---

* Corresponding author. E-mail: mhli@cycu.edu.tw.

10.1021/je000138x CCC: $20.00

N_2O was supplied from a commercial cylinder with a purity of 99.7 mol %.

Density and Viscosity Measurements. The densities and viscosities of aqueous DIPA and TEA solutions were measured in this study. The density measurements were performed in a constant-temperature water bath, in which the temperature could be held constant to ±0.05 °C. The experimental uncertainty was estimated to be ±0.05% on the basis of comparisons with literature data. The kinematic viscosities of the solutions were measured by means of Cannon-Fenske viscometers. An electronic stopwatch with an accuracy of 0.01 s was used to measure the efflux times of the liquid samples. The absolute viscosity of the fluid can be obtained by multiplying the kinematic viscosity by the density of the fluid. The Cannon-Fenske viscometers were purchased with certificates of calibration. The end effect corrections were neglected in the calculation of the kinematic viscosity. The uncertainty of the kinematic viscosity was estimated to be ±1.0% on the basis of comparisons with literature data for aqueous DEA solutions (Hsu and Li[21]).

Diffusivity Measurement. The diffusivity of a gas in aqueous amine solution was measured in a short wetted wall column absorber. 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 apparatus and the experimental procedure are the same as those described by Li and Lai[13] and Yih and Shen.[22]

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

$$N_A = 2\left(\frac{D_A}{\pi t_c}\right)^{1/2}\left(\frac{p_A}{H_A}\right) \qquad (4)$$

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 the interface, and H_A is the solubility of the absorbed gas in the solution. The contact time t_c can be derived from wetted wall column hydrodynamics as

$$t_c = \frac{2h}{3}\left(\frac{\pi d}{L}\right)^{2/3}\left(\frac{3\eta}{\rho g}\right)^{1/3} \qquad (5)$$

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, η is the viscosity of the fluid, ρ is the density of the fluid, and g is the gravitational acceleration. Rearranging eq 4, one has

$$D_A = (N_A H_A)^2 \pi t_c/(2p_A)^2 \qquad (6)$$

The diffusivities of the gas in solution were calculated from the specific gas absorption rate, the solubility of the gas in solution, the contact time, and the partial pressure of the gas at the interface. The density and viscosity of the solution were required in the calculation of the contact time. The viscosities and densities of aqueous MEA, DEA, and AMP solutions were estimated using the correlations of Hsu and Li.[21,23] The deviations (average absolute percentage deviation, AAD %) for the density for MEA + H_2O (30−80 °C, up to 4.91 $kmol\cdot m^{-3}$, 14 data points), DEA + H_2O (20−100 °C, up to 2.85 $kmol\cdot m^{-3}$, 27 data points), and AMP + H_2O (20−90.7 °C, up to 9.90 $kmol\cdot m^{-3}$, 47 data points) are 0.054%, 0.053%, 0.068%, respectively. The deviations (AAD %) for the viscosity for MEA + H_2O (30−80 °C, up to 4.91 $kmol\cdot m^{-3}$, 22 data points), DEA + H_2O (20−100 °C, up to 2.85 $kmol\cdot m^{-3}$, 30 data points), and AMP + H_2O (30−80 °C, up to 3.37 $kmol\cdot m^{-3}$, 12 data points) are 0.42%, 1.51%, 1.0%, respectively. The amine concentrations studied in this work were (0.5−5) $kmol\cdot m^{-3}$ for MEA and (0.5−2.5) $kmol\cdot m^{-3}$ for DEA and AMP. Thus, the correlations for viscosity and density for MEA + H_2O, DEA + H_2O, and AMP + H_2O[21,23] were used. For DIPA + H_2O and TEA + H_2O, the densities and viscosities of the aqueous solutions were measured in this study. The solubilities of N_2O in five aqueous (MEA, DEA, DIPA, TEA, and AMP) solutions were taken directly from our previous work (Tsai et al.[24]). To calibrate the wetted wall column absorber, the diffusivities of N_2O in water were measured. The estimated experimental uncertainty in the measured diffusivity was ±2% on the basis of comparisons with literature data for the diffusivities of N_2O in water.

Table 1. Viscosities of DIPA and TEA

	η/(mPa·s)	
t, °C	DIPA	TEA
30		402.8
35		279.4
40		197.5
45		140.2
50	125.2	104.8
55	82.02	79.52
60	55.91	61.24
65	39.39	47.84
70	28.08	38.16
75	20.75	30.79
80	15.57	25.30
	Parameters of Eq 7	
a_1	−5.1873	−3.33
a_2	1.1658×10^3	1.0949×10^3
a_3	-2.069×10^2	-1.8545×10^2
AAD %[a]	0.2%	0.3%

[a] Average absolute percentage deviation.

Results and Discussion

Viscosities of DIPA and TEA. The viscosity measurements of DIPA and TEA are presented in Table 1. The viscosity of amine was represented by the following expression

$$\ln\{\nu/(mm^2\cdot s^{-1})\} = a_1 + \frac{a_2}{(T/K) + a_3} \qquad (7)$$

where ν is the kinematic viscosity, η/ρ, where η is the viscosity and ρ the density. Since the melting point of DIPA is 45.5 °C, the viscosity measurement for DIPA starts at 50 °C. The densities of pure DIPA and TEA were taken from ref 2. The parameters a_i in eq 7 and the results of the viscosity calculation are presented in Table 1.

Density and Viscosity of DIPA + H_2O and TEA + H_2O. The results of density and viscosity measurements for DIPA + H_2O and TEA + H_2O are presented in Table 2. The concentrations of aqueous DIPA and TEA solutions are (0.5−3.0) $kmol\cdot m^{-3}$, and the temperatures are (30, 35, and 40) °C.

Density. A Redlich−Kister type equation for the excess molar volume was to represent the density of aqueous alkanolamine solutions as follows

$$V^E_{12}/(cm^3\cdot mol^{-1}) = x_1 x_2 \sum_{i=0}^{n} A_i (x_1 - x_2)^i \qquad (8)$$

Table 2. Densities and Viscosities of DIPA + H_2O and TEA + H_2O

	c_{amine}	DIPA + H_2O		TEA + H_2O	
t, °C	kmol·m^{-3}	$\rho/(\text{g·cm}^{-3})$	$\eta/(\text{mPa·s})$	$\rho/(\text{g·cm}^{-3})$	$\eta/(\text{mPa·s})$
30	0.5	0.9986	1.047	1.0067	0.9933
	1.0	1.0028	1.410	1.0182	1.2659
	1.5	1.0071	1.962	1.0294	1.6496
	2.0	1.0119	2.783	1.0406	2.2154
	2.5	1.0160	4.061	1.0517	2.9640
	3.0	1.0205	6.032	1.0631	4.2281
35	0.5	0.9969	0.941	1.0051	0.8942
	1.0	1.0009	1.237	1.0164	1.1242
	1.5	1.0052	1.693	1.0273	1.4503
	2.0	1.0094	2.346	1.0387	1.9240
	2.5	1.0134	3.353	1.0497	2.5735
	3.0	1.0174	4.863	1.0609	3.5789
40	0.5	0.9951	0.836	1.0027	0.8020
	1.0	0.9988	1.090	1.0136	1.0048
	1.5	1.0029	1.472	1.0242	1.2712
	2.0	1.0069	2.009	1.0356	1.6879
	2.5	1.0107	2.810	1.0461	2.2226
	3.0	1.0139	3.945	1.0571	3.0622

where A_i is assumed to have the following temperature dependence

$$A_i = a + b(T/\text{K}) + c(T/\text{K})^2 \tag{9}$$

The density of H_2O was from ref 25. The parameters in eq 9 and the deviations (AAD%) of the density calculation for DIPA + H_2O and TEA + H_2O are presented in Table 3.

Viscosity. To correlate the viscosity of aqueous alkanolamine solutions, the viscosity deviation expression proposed by Pikkarainen[26] was applied as follows

$$\delta\nu_{12} = \ln \nu_{\text{m}} - \sum_{i=1}^{2} x_i \ln \nu_i \tag{10}$$

where ν is the kinematic viscosity. The subscripts m and i represent the mixture and the ith pure fluid, respectively. $\delta\nu_{12}$ is a function of temperature and mole fraction and assumed to have the Redlich−Kister type expression

$$\delta\nu_{12} = x_1 x_2 \sum_{i=0}^{m} A_i (x_1 - x_2)^i \tag{11}$$

where A_i are pair parameters and assumed to have the following temperature dependence

$$A_i = a + \frac{b}{(T/\text{K}) + c} \tag{12}$$

where a, b, and c are parameters. The parameters in eq 12 and the results of viscosity calculations for DIPA + H_2O and TEA + H_2O are also presented in Table 3.

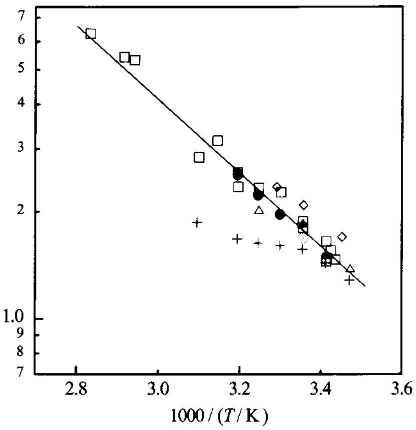

Figure 1. Diffusivity of N_2O in water as a function of temperature: △, Haimour and Sandall;[4] □, Versteeg and van Swaaij;[6] +, Al-Ghawas et al.;[7] ◇, Davidson and Cullen;[27] ⊕, Duda and Vrentas;[28] ●, this study; line, calculated using eq 2.

Diffusivity of N_2O in Aqueous Alkanolamine Solutions. To validate the diffusivity apparatus and the experimental procedure of the measurement, the diffusivities of N_2O in water were measured. The measured diffusivities of N_2O in water are (2.00, 2.27, and 2.58) × 10^{-9} $m^2·s^{-1}$ for (30, 35, and 40) °C, respectively. In Figure 1, the comparisons between the literature values[4,6,7,27,28] and values obtained in this study are shown. Also in Figure 1, the solid line is calculated by eq 2.[6] Figure 1 shows that our measured diffusivities of N_2O in water are in good agreement with literature values. Also, eq 2 appears to be an accurate correlation for the diffusivity of N_2O in water as a function of temperature. The diffusivities of N_2O in water reported by Al-Ghawas et al.[7] are lower than other literature values, especially at high temperatures.

The derived diffusivities of N_2O in aqueous alkanolamine solutions at the temperatures (30, 35, and 40) °C are presented in Table 4. The concentrations of amine are from (0.5 to 2.5) kmol·m^{-3} while for MEA they are from (0.5 to 5) kmol·m^{-3}. The results indicate that the diffusivity increases as temperature increases and decreases with increasing amine concentration.

The diffusivities of N_2O in aqueous amine solutions were correlated by the following equation:

$$D_{\text{A}}/(\text{m}^2\text{·s}^{-1}) = \{b_0 + b_1 c_{\text{amine}}/(\text{kmol·m}^{-3}) + b_2 c_{\text{amine}}^2\} \times \exp\{(b_3 + b_4 c_{\text{amine}}/(\text{kmol·m}^{-3}))/(T/\text{K})\} \tag{13}$$

where D_{A} is the diffusivity of N_2O in the solvent and c_{amine} is the concentration of amine in solution. When the

Table 3. Parameters of the Redlich−Kister Equations for the Density and Viscosity Calculations

		DIPA + H_2O		TEA + H_2O	
parameters		density[a]	viscosity[b]	density[a]	viscosity[b]
A_0	a	−353.3	−11.76	−4.478	1.926
	b	1.128	964.8	1.99 × 10^{-2}	−4127
	c	7.236 × 10^{-6}	−236.5	−3.415 × 10^{-5}	1582
A_1	a	−408	3.433 × 10^{-2}	15.92	−5.255
	b	1.34	−841	−2.46 × 10^{-2}	−1.508 × 10^4
	c	−1.876 × 10^{-5}	−14.07	−4.314 × 10^{-5}	1064
A_2	a		15.48		−18.75
	b		1863		2610
	c		253.1		−168.8
AAD %[c]		0.08%	3.0%	0.02%	0.5%

[a] Density parameters in eq 8. [b] Viscosity parameters in eq 11. [c] Average absolute percentage deviation.

Table 4. Diffusivity of N_2O in Aqueous Alkanolamine Solutions

c_{amine}	diffusivity/(10^9 $m^2 \cdot s^{-1}$)				
$kmol \cdot m^{-3}$	MEA	DEA	DIPA	TEA	AMP
		30 °C			
0.5	1.98	1.64	1.74	1.89	1.76
1.0	1.86	1.41	1.42	1.69	1.62
1.5	1.75	1.20	1.22	1.57	1.44
2.0	1.70	1.12	0.97	1.30	1.17
2.5	1.67	0.91	0.75	1.04	1.01
3.0	1.63				
5.0	1.41				
		35 °C			
0.5	2.16	1.94	2.15	2.20	1.99
1.0	2.09	1.67	1.70	1.80	1.81
1.5	2.01	1.46	1.53	1.76	1.60
2.0	1.95	1.30	1.18	1.42	1.36
2.5	1.89	1.05	0.89	1.12	1.17
3.0	1.86				
5.0	1.61				
		40 °C			
0.5	2.46	2.25	2.44	2.35	2.28
1.0	2.38	1.98	1.93	2.15	2.06
1.5	2.30	1.79	1.63	1.84	1.76
2.0	2.24	1.53	1.27	1.65	1.61
2.5	2.22	1.21	0.97	1.35	1.32
3.0	2.16				
5.0	1.91				

Table 5. Parameters in the Diffusivity Equation (Eq 13) for Aqueous Alkanolamine Solutions

system	b_1	b_2	b_4
MEA + H_2O	8.65×10^{-7}	2.78×10^{-7}	-9.34×10^{1}
DEA + H_2O	2.17×10^{-6}	2.29×10^{-6}	-2.92×10^{2}
DIPA + H_2O	1.05×10^{-6}	-3.73×10^{-7}	-1.28×10^{2}
MDEA + H_2O	1.31×10^{-6}	8.73×10^{-8}	-1.50×10^{2}
TEA + H_2O	-1.70×10^{-6}	-1.21×10^{-7}	-5.99×10^{1}
AMP + H_2O	-3.85×10^{-7}	5.81×10^{-8}	-4.70×10^{-1}

concentration of the amine solution approaches zero, that is, pure water, eq 13 should reduce to the diffusivity of N_2O in water, that is, eq 2. Thus, in this study, we retain b_0 and b_3 of eq 13 as the same values as in eq 2; that is, b_0 is 5.07×10^{-6} and b_3 is -2371. Consequently, only values of b_1, b_2, and b_4 are determined from the diffusivity of N_2O in aqueous amine solutions. It should be noted that the relation between the diffusivity and the reciprocal temperature, in eq 13, depends on the solution parameter b_4; the slope of the diffusivity–reciprocal temperature relation will be different for water and aqueous alkanolamine solutions.

The parameters b_1, b_2, and b_4 of eq 13 were determined from both the measured diffusivity of N_2O in amine solutions and the available data in the open literature. The parameters of the diffusivity of N_2O in six aqueous (MEA, DEA, DIPA, MDEA, TEA, and AMP) solutions are presented in Table 5. The deviations (AAD %) of the calculations of the diffusivity of N_2O in aqueous alkanolamine solutions are presented in Table 6.

MEA + H_2O. For MEA + H_2O, a plot of diffusivity of N_2O in the solution versus the concentration of MEA is shown in Figure 2. The calculated diffusivities are in good agreement with the data of Clarke[3] and of Sada et al.[12] except for the data of Sada et al. at 25 °C and concentrations of MEA beyond 2.766 $kmol \cdot m^{-3}$. At 25 °C, the diffusivity data of Sada et al.[12] are lower than those of Clarke[3] for concentration of MEA beyond 2.766 $kmol \cdot m^{-3}$. The diffusivity data of Li and Lai[13] at (30, 35, and 40) °C are in good agreement with the present study. As shown in Figure 2, eq 13 yields a reasonable representation for the diffusivity of N_2O in MEA + H_2O systems for concentrations of MEA up to 5.0 $kmol \cdot m^{-3}$ and temperatures up to 45 °C.

Table 6. Result for the Calculation of the Diffusivity of N_2O in Alkanolamine (1) + Water (2)

t/°C	c_1/($kmol \cdot m^{-3}$)	no. of data points	ref	AAD % calcd by eq 13
		MEA (1) + H_2O (2)		
25	1.64−4.88	3	3	0.9
25	0−3.36	6	12	7.6
30−40	4.91	3	13	5.2
30−40	0−5	21	this study	0.9
		DEA (1) + H_2O (2		
25	0.5−3.46	5	12	27
20−95	0.95−2.85	21	14	10
30−40	2.85	3	15	27
30−40	0−2.5	18	this study	3.8
		DIPA (1) + H_2O (2)		
20	0.33−2.22	7	6	36
20−40	0.005−0.5	21	8	2.3
25	0.40−1.66	5	12	13
30−40	0−2.5	18	this study	2.8
		MDEA (1) + H_2O (2)		
15−35	0−1.70	16	4	7.9
20−60	0.23−2.87	37	6	6.8
15−50	0−4.20	42	7	19
20−95	0.84−4.20	35	14	28
25−75	1.68−3.36	6	16	15
30−50	2.52−3.36	6	17	13
30−40	0−2.5	15	18	2.1
		TEA (1) + H_2O (2)		
20−40	0.01−0.5	15	9	1.5
25	0.49−1.93	4	12	9.8
30−40	0−2.5	18	this study	2.1
		AMP (1) + H_2O (2)		
25−76	2.0−3.0	17	10	21
21−45	0−2.0	20	11	2.7
25	0−2.37	12	19	3.6
30−40	0−2.5	18	this study	1.8

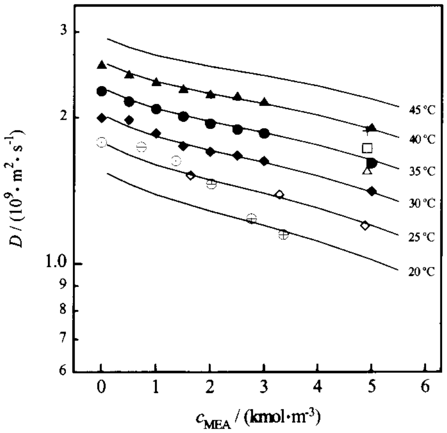

Figure 2. Diffusivity of N_2O in MEA + H_2O as a function of MEA concentration for temperatures from 20 to 45 °C: ◇, 25 °C, Clarke;[3] ⊕, 25 °C, Sada et al.;[12] △, 30 °C, □, 35 °C, +, 40 °C, Li and Lai;[13] ◆, 30 °C, ●, 35 °C, ▲, 40 °C, this study; lines, calculated using eq 13.

DEA + H_2O. For the diffusivity of N_2O in DEA + H_2O, the correlation is compared with literature values and shown in Figure 3. At 25 °C, the data of Sada et al.[12] and Tamimi et al.[14] are not quite consistent. The data of Sada et al.[12] at 25 °C are greater than those of Tamimi et al.[14] and the calculated values using eq 13. The calculated values are in good agreement with the data of Tamimi et al.[14] for temperatures from (20 to 80) °C. At 95 °C, the calculated value at the DEA concentration 2.948 kmol m^{-3} is in good agreement with the data of Tamimi et al.[14] The

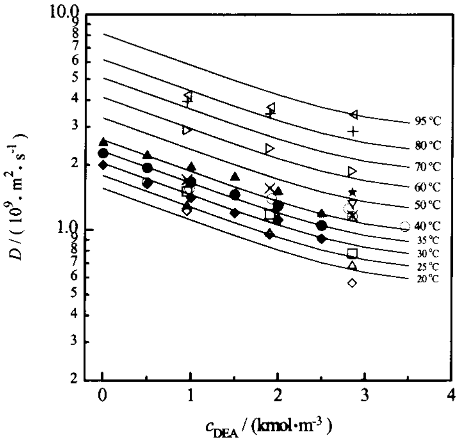

Figure 3. Diffusivity of N_2O in DEA + H_2O as a function of DEA concentration for temperatures from 20 to 95 °C: ○, 25 °C, Sada et al.;[12] ◇, 20 °C, △, 25 °C, □, 30 °C, ×, 40 °C, open triangle pointing right, 60 °C, +, 80 °C, open triangle pointing left, 95 °C, Tamimi et al.;[14] ⊕, 30 °C, ▽, 35 °C, ★, 40 °C, Li and Lee;[15] ◆, 30 °C, ●, 35 °C, ▲, 40 °C, this study; lines, calculated using eq 13.

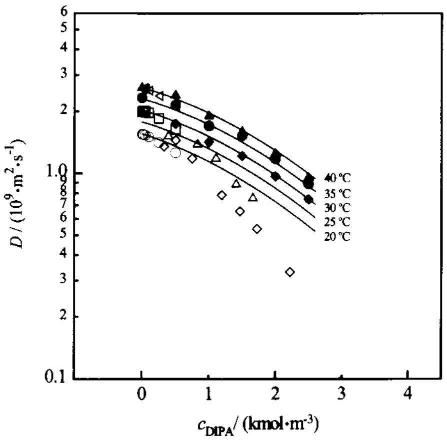

Figure 4. Diffusivity of N_2O in DIPA + H_2O as a function of DIPA concentration: ◇, 20 °C, Versteeg and van Swaaij;[6] ○, 20 °C, □, 30 °C, open triangle pointing left, 40 °C, Sotelo et al.;[8] △, 25 °C, Sada et al.;[12] ◆, 30 °C, ●, 35 °C, ▲, 40 °C, this study; lines, calculated using eq 13.

composition dependence of the diffusivity data of Tamimi et al.[14] at (80 and 95) °C shows a weaker dependence than those of the calculated values. The data of Li and Lee[15] at (30, 35, and 40) °C at 2.948 kmol m^{-3} appear to be higher than those of the present study. For the DEA + H_2O system, the following N_2O diffusivity data are recommended: (20, 25, 30, 40, 60, 80) °C of Tamimi et al.;[14] (30, 35, 40) °C of the present study. More data are required to confirm the diffusivity of N_2O in DEA + H_2O at concentrations less than 1.0 kmol m^{-3} and temperatures above 50 °C.

DIPA + H_2O. For the diffusivity of N_2O in DIPA + H_2O, the correlation and the literature data are shown in Figure 4. Using the N_2O analogy, Sotelo et al.[8] reported the diffusivity of CO_2 in aqueous DIPA solutions for temperatures from (20 to 40) °C. The values of the diffusivity of N_2O in DIPA + H_2O of Sotelo et al.[8] in Figure 4 were calculated from the CO_2 data of Sotelo et al.[8] by using eqs 1−3. Equation 13 fits well the values of Sotelo et al.,[8] the data of Versteeg and van Swaaij[6] at 20 °C up to 0.751 kmol·m^{-3}, and the data of Sada et al.[12] at 25 °C up to 1.107 kmol·m^{-3}. Both the data of Sada et al.[12] at 25 °C and the data of Versteeg and van Swaaij[6] at 20 °C show a stronger composition dependence than those of the present study.

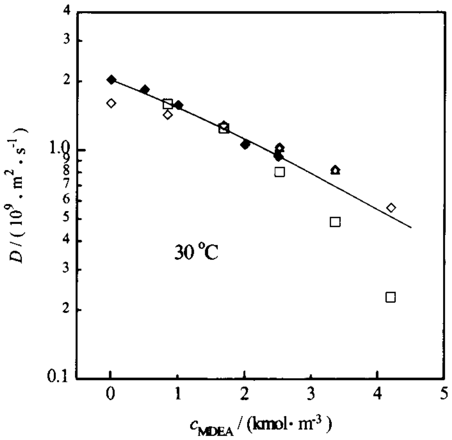

Figure 5. Diffusivity of N_2O in MDEA + H_2O as a function of MDEA concentration at 30 °C: ◇, Al-Ghawas et al.;[7] □, Tamimi et al.;[14] △, Hagewiesche et al.;[17] ◆, Ko and Li;[18] line, calculated using eq 13.

Figure 4 shows that the calculated diffusivities of N_2O in DIPA + H_2O are reasonable for temperatures from (20 to 40) °C and for concentrations of DIPA up to 2.5 kmol·m^{-3}. More data are required for the diffusivity of N_2O in DIPA + H_2O for temperatures above 40 °C.

MDEA + H_2O. For MDEA + H_2O, the N_2O diffusivity correlation is compared with literature values in Table 6. Equation 13 fits the data of Haimour and Sandall[4] (7.9%), Versteeg and van Swaaij[6] (6.8%), and Ko and Li[18] (2.1%) within the experimental uncertainty. Fair results were obtained from the diffusivity correlation for the data of Al-Ghawas et al.[7] (19%), Tomcej and Otto[16] (15%), and Hagewiesche et al.[17] (13%). The largest deviation (AAD %) for diffusivity is for the data of Tamimi et al.[14] (28%). A plot of the diffusivity of N_2O in aqueous MDEA solutions versus the concentration of MDEA at 30 °C is shown in Figure 5. Compared to the literature values, the data of Tamimi et al.[14] show lower values for concentrations of MDEA beyond 3.0 kmol·m^{-3} at the temperatures (20, 25, 30, and 40) °C, as shown in Figure 5 for 30 °C. Probably due to smaller values of N_2O in water than those of the literature values as shown in Figure 1, most of the data of N_2O diffusivity in MDEA + H_2O of Al-Ghawas et al.[7] are lower than other literature values for the low concentration region as shown in Figure 5. As shown in Figure 5 (at 30 °C), eq 13 yields satisfactory results for the data of Al-Ghawas et al.[7] beyond 1.0 kmol·m^{-3}, the data of Tamimi et al.[14] up to 2.517 kmol·m^{-3}, and the data of Ko and Li.[18] The data of Hagewiesche et al.[17] agree reasonably well with the values of Al-Ghawas et al.[7] For MDEA + H_2O, the following N_2O diffusivity data are recommended: Haimour and Sandall[4] (15, 20, 25, and 35) °C; Versteeg and van Swaaij[6] (20, 25, 35, 45, and 60) °C; Al-Ghawas et al.[7] (15, 20, 25, 30, and 40) °C; Tamimi et al.[14] (20, 25, 30, and 40) °C; Tomcej and Otto[16] (25 and 49.6) °C; for MDEA concentrations up to 2.517 kmol·m^{-3} and (60 and 80) °C; Hagewiesche et al.[17] (30 and 40) °C; Ko and Li[18] (30, 35, and 40) °C.

TEA + H_2O. For the diffusivity of N_2O in TEA + H_2O, the comparison of the correlation with literature values is shown in Figure 6. The deviations (AAD %) for the calculation of the diffusivity of N_2O in TEA + H_2O using eq 13 are 1.5%, 9.8%, and 2.1% for the data of Sotelo et al.,[9] Sada et al.,[12] and the present study, respectively. As shown in Figure 6, eq 13 fits the diffusivity data quite well except for the value of Sada et al.[12] at the TEA concentration 1.921 kmol·m^{-3} at 25 °C. As shown in Figure 6, the

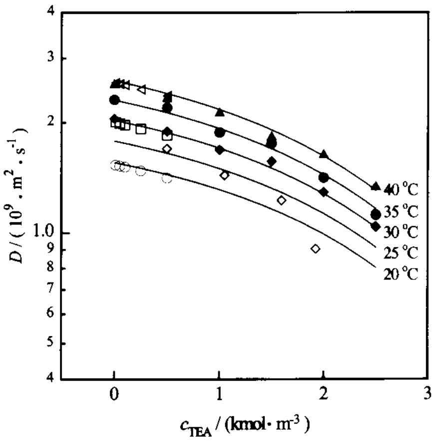

Figure 6. Diffusivity of N_2O in TEA + H_2O as a function of TEA concentration: ○, 20 °C, □, 30 °C, open triangle pointing left, 40 °C, Sotelo et al.;[9] ◇, 25 °C, Sada et al.;[12] ◆, 30 °C, ●, 35 °C, ▲, 40 °C, this study; lines, calculated using eq 13.

calculated diffusivities of N_2O in TEA + H_2O are reasonable to represent the data from (20 to 40) °C and for concentrations of TEA up to 2.5 kmol·m^{-3}.

AMP + H_2O. For AMP + H_2O, the deviations (AAD %) for the N_2O diffusivity are presented in Table 6. The deviations (AAD %) for N_2O diffusivity calculations using eq 13 are (2.7, 3.6, and 1.8) % for the data of Saha et al.,[11] Bosch et al.,[19] and the present study, respectively. Equation 13 also fits the data of Xu et al.[10] (34.9 °C) reasonably well. The calculation of the diffusivity of N_2O in AMP + H_2O from eq 13 is satisfactory to represent the N_2O diffusivity data for temperatures from (20 to 50) °C and for AMP concentrations up to 2.5 kmol·m^{-3}.

Conclusions

The diffusivities of nitrous oxide (N_2O) in aqueous alkanolamine solutions have been measured at (30, 35, and 40)°C. The systems studied are aqueous MEA, DEA, DIPA, TEA, and AMP solutions. The concentration of amine for MEA ranges from (0.5 to 5) kmol·m^{-3} and for other amines from (0.5 to 2.5) kmol·m^{-3}. The uncertainty of the measurement is estimated to be ±2%. The viscosities of diisopropanolamine and triethanolamine and the densities and viscosities of aqueous diisopropanolamine and triethanolamine solutions were also measured at (30, 35, and 40) °C. A simple model for the diffusivity of nitrous oxide in amine was used to correlate the diffusivity of N_2O in amine solutions. The parameters of the correlation were determined from the measured diffusivity data and the available data in the open literature. The correlation has been shown to represent reasonably the diffusivity of N_2O in six aqueous amine solutions: MEA, DEA, DIPA, MDEA, TEA, and AMP. For the purpose of process design, the correlations can be used to estimate the correct free molecular diffusivity of CO_2 in amines.

Acknowledgment

Thanks are extended to Mr. Shyh-Yun Horng and Mr. Chen-Hung Liao for performing the viscosity measurements for diisopropanolamine and triethanolamine.

Literature Cited

(1) Kohl, A. L.; Riesenfeld, F. C. *Gas Purification,* 4th ed.; Gulf: Houston, TX, 1985.
(2) Wang, Y. W.; Xu, S.; Otto, F. D.; Mather, A. E. Solubility of N_2O in Alkanolamines and in Mixed Solvents. *Chem. Eng. J.* **1992**, *48*, 31−40.
(3) Clarke, J. K. A. Kinetics of Absorption of Carbon Dioxide in Monoethanolamine Solutions at Short Contact Times. *Ind. Eng. Chem. Fundam.* **1964**, *3*, 239−245.
(4) Haimour, N.; Sandall, O. C. Absorption of Carbon Dioxide into Aqueous Methyldiethanolamine. *Chem. Eng. Sci.* **1984**, *39*, 1791−1796.
(5) Diaz, J. M.; Vega, A.; Coca, J. Diffusivities of CO_2 and N_2O in Aqueous Alcohol Solutions. *J. Chem. Eng. Data* **1988**, *33*, 10−12.
(6) Versteeg, G. F.; van Swaaij, W. P. M. Solubility and Diffusivity of Acid Gases (CO_2, N_2O) in Aqueous Alkanolamine Solutions. *J. Chem. Eng. Data* **1988**, *33*, 29−34.
(7) Al-Ghawas, H. A.; Hagewiesche, D. P.; Ruiz-Ibanez, G.; Sandall, O. C. Physicochemical Properties Important for Carbon Dioxide Absorption in Aqueous Methyldiethanolamine. *J. Chem. Eng. Data* **1989**, *34*, 385−391.
(8) Sotelo, J. L.; Benitez, F. J.; Beltran-Heredia, J. B.; Rodriguez, C. Kinetics of Carbon Dioxide Absorption in Aqueous Solutions of Diisopropanolamine. *Chem. Eng. Technol.* **1992**, *15*, 114−118.
(9) Sotelo, J. L.; Benitez, F. J.; Beltran-Heredia, J.; Rodriguez, C. Absorption of Carbon Dioxide into Aqueous Solutions of Triethanolamine. *AIChE J.* **1990**, *36*, 1263−1266.
(10) Xu, S.; Otto, F. D.; Mather, A. E. Physical Properties of Aqueous AMP Solutions. *J. Chem. Eng. Data* **1991**, *36*, 71−75.
(11) Saha, A. K.; Bandyopadhyay, S. S.; Biswas, A. K. Solubility and Diffusivity of N_2O and CO_2 in Aqueous Solutions of 2-Amino-2-methyl-1-propanol. *J. Chem. Eng. Data* **1993**, *38*, 78−82.
(12) Sada, E.; Kumazawa, H.; Butt, M. A. Solubility and Diffusivity of Gases in Aqueous Solutions of Amines. *J. Chem. Eng. Data* **1978**, *23*, 161−163.
(13) Li, M. H.; Lai, M. D. Solubility and diffusivity of N_2O and CO_2 in (Monoethanolamine + *N*-Methyldiethanolamine + Water) and in (Monoethanolamine + 2-Amino-2-methyl-1-propanol + Water). *J. Chem. Eng. Data* **1995,** *40*, 486-492.
(14) Tamimi, A.; Rinker, E. B.; Sandall, O. C. Diffusivity of Nitrous Oxide in Aqueous Solutions of *N*-Methyldiethanolamine and Diethanolamine from 293 to 368 K. *J. Chem. Eng. Data* **1994**, *39*, 396−398.
(15) Li, M. H.; Lee, W. C. Solubility and Diffusivity of N_2O and CO_2 in (Diethanolamine + *N*-Methyldiethanolamine + Water) and in (Diethanolamine + 2-Amino-2-methyl-l-propanol + Water). *J. Chem. Eng. Data* **1996**, *41*, 551−556.
(16) Tomcej, R. A.; Otto, F. D. Absorption of CO_2 and N_2O into Aqueous Solutions of Methyldiethanolamine. *AIChE J.* **1989**, *35*, 861−864.
(17) Hagewiesche, D. P.; Ashour, S. S.; Sandall, O. C. Solubility and Diffusivity of Nitrous Oxide in Ternary Mixtures of Water, Monoethanolamine, and *N*-Methyldiethanolamine and Solution Densities and Viscosities. *J. Chem. Eng. Data* **1995**, *40*, 627−629.
(18) Ko, J. J.; Li, M. H. Kinetics of Absorption of Carbon Dioxide into Solutions of *N*-Methyldiethanolamine + Water. *Chem. Eng. Sci.* **2000**, *55*, 4139−4147.
(19) Bosch, H.; Versteeg, G. F.; van Swaaij, W. P. M. Kinetics of the Reaction of CO_2 with the Sterically Hindered Amine 2-Amino-2-methyl-1-propanol at 298 K. *Chem. Eng. Sci.* **1990**, *45*, 1167−1173.
(20) Blauwhoff, P. M. M.; Versteeg, G. F.; van Swaaij, W. P. M. A Study of the Reaction Between CO_2 and Alkanolamines in Aqueous Solutions. *Chem. Eng. Sci.* **1984**, *39*, 207−225.
(21) Hsu, C. H.; Li, M. H. Viscosities of Aqueous Blended Amines. *J. Chem. Eng. Data* **1997**, *42*, 2**,** 714-720**.**
(22) Yih, S. M.; Shen, K. P. Kinetics of Carbon Dioxide Reaction with Sterically Hindered 2-Amino-2-methyl-1-propanol Aqueous Solutions. *Ind. Eng. Chem. Res.* **1988**, *27*, 2237−2241.
(23) Hsu, C. H.; Li, M. H. Densities of Aqueous Blended Amines. *J. Chem. Eng. Data* **1997,** *42***,** 502−507.
(24) Tsai, T. C.; Ko, J. J.; Wang, H. M.; Lin, C. Y.; Li, M. H. Solubility of Nitrous Oxide in Alkanolamine Aqueous Solutions. *J. Chem. Eng. Data* **2000**, *45*, 341−347.
(25) Haar, L.; Gallagher, J. S.; Kell, G. S., *NBS/NRC Steam Tables*; Hemisphere Publishing Corp.: Bristol, PA, 1984.
(26) Pikkarainen, L. Densities and Viscosities of Binary Solvent Mixtures of *N*-Methylacetamide with Aliphatic Alcohols. *J. Chem. Eng. Data* **1983**, *28*, 381−383.
(27) Davidson, J. F.; Cullen, E. J. The Determination of Diffusion Coefficients for Sparingly Soluble Gases in Liquids. *Trans. 1nstn. Chem. Eng.* **1957**, *35*, 51−60.
(28) Duda, J. F.; Vrentas, J. C. Laminar Liquid Jet Diffusion Studies. *AIChE J.* **1968**, *14*, 286−294.

Received for review May 4, 2000. Accepted October 26, 2000. This research was supported by a grant, NSC 89-2214-E-033-011, of the National Science Council of the Republic of China.

JE000138X