# Solubility of Nitrous Oxide in Alkanolamine Aqueous Solutions

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

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

The solubility of nitrous oxide (N<sub>2</sub>O) in alkanolamine aqueous solutions has been measured at (30, 35, and 40) °C. The systems studied are monoethanolamine, diethanolamine, diisopropanolamine, triethanolamine, and 2-amino-2-methyl-1-propanol aqueous solutions. The concentration of amine for monoethanolamine ranges from (1 to 6) kmol·m<sup>-3</sup> and for other amines from (0.5 to 3) kmol·m<sup>-3</sup>. The accuracy of the measurement is estimated to be  $\pm$  2%. A semiempirical model of the excess Henry's constant proposed by Wang et al. (1992) was used to correlate the solubility of N<sub>2</sub>O in amine solutions. The parameters of the correlation were determined from the measured solubility data and the available data in the open literature. For a wide temperature range from (15 to 75) °C, the obtained correlation has been shown to represent reasonably the solubility of N<sub>2</sub>O in six amine aqueous solutions: MEA, DEA, DIPA, TEA, MDEA, and AMP. For the purpose of process design, the obtained correlations are, in general, satisfactory for estimating the solubility of N<sub>2</sub>O in amine solutions, which in turn can be used to estimate the correct free-gas solubility of CO<sub>2</sub> in amines.

## Introduction

Solutions of alkanolamines are widely encountered in gas-liquid contact operations as absorbents of 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-1-propanol (AMP), and 2-piperidineethanol (2-PE) can be used as absorbents for acid gas removal processes (Kohl and Riesenfeld, 1985). Aqueous alkanolamine solutions are more widely used than hot potassium carbonate solutions, since they require less energy and have lower corrosion rates (Kohl and Riesenfeld, 1985).

To design a process for the removal of  $CO_2$ , it is necessary to use the free-gas solubility of  $CO_2$  in aqueous alkanolamines at various amine concentrations and temperatures to model the reaction kinetics between  $CO_2$  and alkanolamine. Due to the chemical reaction between  $CO_2$  and amines, the free-gas solubility of  $CO_2$  in amine solutions cannot be measured directly. The N<sub>2</sub>O analogy has been frequently used to estimate the solubility of  $CO_2$  in amine solutions (Clarke, 1964; Haimour and Sandall, 1984; Versteeg and van Swaaij, 1988; Al-Ghawas et al., 1989; Xu et al., 1991; Saha et al., 1993).

The  $N_2O$  analogy for the solubility of  $CO_2$  in amine solution has the following relation

$$H_{\rm CO_2} = H_{\rm N_2O} (H_{\rm CO_2} / H_{\rm N_2O})_{\rm in \ water}$$
(1)

where  $H_{N_2O}$  is the solubility of  $N_2O$  in amine solution.

Versteeg and van Swaaij (1988) proposed, on the basis of the available data of solubility of  $N_2O$  and  $CO_2$  in water, two convenient equations as follows

 $H_{\rm N_{s}O}/({\rm kPa}\cdot{\rm m}^3\cdot{\rm kmol}^{-1}) =$ 

$$8.5470 \times 10^{\circ} \exp(-2284/(T/K))$$
 (2)

 $H_{\rm CO_o}/({\rm kPa}\cdot{\rm m}^3\cdot{\rm kmol}^{-1}) =$ 

 $2.8249 \times 10^{6} \exp(-2284/(T/K))$  (3)

From eqs 1-3, the free-gas solubility of  $CO_2$  in amine aqueous solution can be estimated from the measured solubility of  $N_2O$  in the amine aqueous solution.

Although numerous solubility data of N<sub>2</sub>O in amine aqueous solutions were reported in the literature such as MEA (Sada and Kito, 1972; Littel et al., 1992; Li and Lai, 1995), DEA (Sada et al., 1977; Versteeg and van Swaaij, 1988; Versteeg and Oyevaar, 1989; Haimour, 1990; Littel et al., 1992; Li and Lee, 1996), DIPA (Sada et al., 1978; Versteeg and van Swaaij, 1988), MDEA (Haimour and Sandall, 1984; Versteeg and van Swaaij, 1988; Al-Ghawas et al., 1989), TEA (Sada et al., 1977; Littel et al., 1992), and AMP (Bosch et al., 1990; Xu et al., 1991; Littel et al., 1992; Saha et al., 1993), some of these data are scattered and inconsistent. For example, for the solubility of N<sub>2</sub>O in DEA aqueous solution at 30 °C, Haimour (1990) reported the value as 7101 kPa·m<sup>3</sup>·kmol<sup>-1</sup> at the concentration of DEA 3.12 kmol·m<sup>-3</sup> while Littel et al. (1992) reported it as 5699 kPa·m<sup>3</sup>·kmol<sup>-1</sup> at the concentration of DEA 3.64 kmol·m<sup>-3</sup>. The scattered and inconsistent data of the Henry's constant of N<sub>2</sub>O in an amine may contribute to the inconsistent results for the reaction kinetics study in the literature (Blauwhoff et al., 1984). Accordingly, the correct solubility of N<sub>2</sub>O in amines is essential to estimate the correct solubility of CO2 in an amine, which in turn can be used in developing the correct reaction kinetic models. Therefore, it was the objective of this research to measure the solubility of N<sub>2</sub>O in MEA, DEA, DIPA, TEA, and AMP aqueous solutions for the temperatures (30, 35, and 40) °C. A semiempirical model proposed by Wang et al. (1992) will be used to correlate the solubility of N<sub>2</sub>O in amine solutions. The parameters of the correlation for each

<sup>\*</sup> Corresponding author. E-mail: mhli@cycu.edu.tw.



Figure 1. Solubility apparatus.

system will be determined from both the solubility of  $N_2O$  measured in this study and the available data in the open literature. The correlation can be used to estimate the solubility of  $N_2O$  in amine aqueous solutions: MEA, DEA, DIPA, TEA, MDEA, and AMP for wide temperature and concentration ranges.

### **Experimental Section**

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 purity of min. 95% is from Aldrich Chemical Co. The gas  $N_2O$  was supplied from commercial cylinders with a purity of 99.7 mol %.

The solubility of N<sub>2</sub>O in amine aqueous solutions was measured by using a solubility apparatus, similar to those presented by Al-Ghawas et al. (1989) and Haimour (1990). The solubility apparatus is shown schematically in Figure 1. The whole apparatus is kept at constant temperature inside a temperature-controller water bath. The accuracy of the temperature of the system is estimated to be  $\pm 0.5$  °C. The reproducibility between the various experiments is always within 3%. The estimated experimental error in the measured solubility is about  $\pm 2\%$ . The experimental procedure and methods of analysis are essentially the same as those used in our previous work (Li and Lai, 1995).

### **Results and Discussion**

To validate the solubility apparatus and the experimental procedure of the measurement, the solubilities of N<sub>2</sub>O in water were measured. The measured solubilities of N<sub>2</sub>O in water are (4660, 5140, and 5637)  $kPa\cdot m^3\cdot kmol^{-1}$  for (30, 35, and 40) °C, respectively. In Figure 2, the comparison between the literature values (Duda and Vrentas, 1968; Versteeg and van Swaaij, 1988; Al-Ghawas et al., 1989; Haimour, 1990; Xu et al., 1991) for the solubility of N<sub>2</sub>O in water and the values obtained in this study is shown. Also in Figure 2, the solid line is calculated by using eq 2 (Versteeg and van Swaaij, 1988). As shown in Figure 2, it is seen that the measured solubilities of N<sub>2</sub>O in water are in good agreement with the literature values. Equation 2 appears to be an accurate correlation for the solubility of N<sub>2</sub>O in water as a function of temperature, as shown in Figure 2. Compared to the literature values, the Henry's constants of N<sub>2</sub>O in water reported by Al-Ghawas et al. (1989) are smaller than those of the calculated values using eq 2.



Figure 2. Solubility of N<sub>2</sub>O in water as a function of temperature.

The measured solubilities of  $N_2O$  in alkanolamine aqueous solutions for the temperatures (30, 35, and 40)°C are presented in Table 1. The concentrations of amine are from (0.5 to 3) kmol·m<sup>-3</sup> while for MEA they are from (1 to 6) kmol·m<sup>-3</sup>.

A semiempirical model proposed by Wang et al. (1992) was used to correlate the solubility of  $N_2O$  in amine solutions. In this method, the excess Henry's coefficient for the binary system has the following form

$$R = \ln H_{N_2O,m} - \sum_{i=2}^{3} \Phi_i \ln H_{N_2O,i}$$
(4)

where  $H_{N_2O,m}$  is Henry's constant of  $N_2O$  in the solvent,  $H_{N_2O,i}$  is Henry's constant of  $N_2O$  in pure solvent *i*, and  $\Phi_i$  is the volume fraction of solvent *i*. From eq 4, the excess Henry's quantity *R* can be calculated from the measured  $H_{N_2O,m}$  and the estimated  $H_{N_2O,i}$ .

The calculated excess Henry's quantity is then correlated as a function of volume fraction as follows

$$R_{ij} = \Phi_i \Phi_j \alpha_{ij} \tag{5}$$

where the two-body interaction parameter,  $\alpha_{ij}$ , is temperature dependent and has assumed the expression

$$\alpha_{ij} = k_1 + k_2 (T/K) + k_3 (T/K)^2 + k_4 \Phi_3$$
 (6)

where  $\Phi_3$  is the volume fraction of water ( $\Phi_2$  for amine) and  $k_1$ ,  $k_2$ ,  $k_3$ , and  $k_4$  are parameters for each binary system and to be determined from corresponding solubility data of N<sub>2</sub>O in amine aqueous solutions.

Wang et al. (1992) proposed the solubility of  $N_2O$  in pure amine solvent as follows

$$H_{\rm N_2O,amine} = b_1 \exp\left(\frac{b_2}{(77\rm K)}\right) \tag{7}$$

The parameters  $b_1$  and  $b_2$  for six pure amines (MEA, DEA, DIPA, MDEA, TEA, and AMP) were reported. As pointed out by Wang et al. (1992), for temperatures between 20 and 85 °C, the average regression deviations between the calculated solubilities of N<sub>2</sub>O in pure amines and experimental data are <2.5%, which is satisfactory for estimating the solubilities of N<sub>2</sub>O in pure amines. Thus, eq 7 with the parameters  $b_i$  of Wang et al. (1992) was adopted directly

# Table 1. Solubility of $N_2 O \mbox{ in Alkanolamine Aqueous Solutions} \label{eq:solution}$

I	MEA	DEA		
$C_{\text{amine}}$	$H_{ m N_2O}$	$C_{\text{amine}}$	$H_{ m N_2O}$	
kmol⋅m <sup>-3</sup>	kPa•m <sup>3</sup> •kmol <sup>−1</sup>	kmol⋅m <sup>-3</sup>	kPa•m <sup>3</sup> •kmol <sup>-1</sup>	
	30	°C		
1.0	4685	0.5	4691	
2.0	4722	1.0	4723	
3.0	4790	1.5	4760	
4.0	4820	2.0	4840	
5.0	4924	2.5	4955	
6.0	5062	2.0	5002	
0.0	5002	5.0	5002	
	35	°C		
1.0	5163	0.5	5165	
2.0	5245	1.0	5178	
3.0	5233	1.5	5149	
4.0	5216	2.0	5237	
5.0	5262	2.5	5388	
6.0	5422	3.0	5400	
	40	°C		
1.0	5669	0.5	5707	
2.0	5750	1.0	5690	
3.0	5757	1.5	5704	
4.0	5761	2.0	5827	
5.0	5766	2.5	5844	
6.0	5799	3.0	5922	
C	ц		$mol^{-1}$	
Camine				
kmol·m <sup>-3</sup>	DIPA	TEA	AMP	
	30	°C	1080	
0.5	4566	4651	4650	
1.0	4702	4704	4791	
1.5	4994	4910	4827	
2.0	5314	5032	5012	
2.5	5490	5174	5213	
3.0	5686	5288	5364	
	35	°C		
0.5	5217	5276	5220	
1.0	5312	5306	5253	
1.5	5445	5367	5401	
2.0	5810	5581	5708	
2.0	5075	5761	5974	
2.5	6230	5032	6020	
5.0	0233	995£	0020	
0.5	5077	L = 700	E7E0	
0.5	58//	5/26	5/50	
1.0	5981	5826	5766	
1.5	6142	5987	5808	
2.0	6342	6191	6082	
2.5	6512	6355	6354	
3.0	6697	6563	6421	

in this study. Also, the solubility of  $N_2O$  in pure water,  $H_{N_2O}, H_{H_2O}$ , is calculated using eq 2 (Versteeg and van Swaaij, 1988). The densities of amine aqueous solutions, required in the calculation of volume fraction, are estimated on the basis of the correlation of Hsu and Li (1997).

Using the available solubility data in the literature and the solubility data obtained in this study, that is, Table 1, the parameters,  $k_1$ ,  $k_2$ ,  $k_3$ , and  $k_4$  in eq 6 are determined for each binary system and the results are presented in Table 2. Due to the significant difference from the correlation, the data of Versteeg and Oyevaar (1989) for DEA +  $H_2O$  at 25 °C and concentrations beyond 6.0 kmol·m<sup>-3</sup> were not included in the database. Except for AMP +  $H_2O$ , the parameter  $k_3$  for all the systems was found to be an extremely small value, and thus  $k_3$  is set equal to zero. The results of solubility calculations from eq 4 along with the calculation from the correlation of Wang et al. (1992) are presented in Tables 3–8. It is noted that the temperature in eq 6 is, in this study, in Kelvin (K) while degree

### Table 2. Parameters in Excess Henry's Constant for Binary Systems

0 0				
system	$k_1$	$k_2$	$k_3$	$k_4$
$MEA + H_2O$	4.793	$-7.446 imes10^{-3}$	0	-2.201
$DEA + H_{2O}$	11.94	$-3.315 imes10^{-2}$	0	-1.381
$DIPA + H_2O$	10.55	$-1.753 imes10^{-2}$	0	-4.073
$MDEA + H_2O$	6.85	$9.99 imes10^{-3}$	0	2.89
$TEA + H_2O$	6.106	$-1.11 imes10^{-2}$	0	-1.983
$AMP + H_2O$	32.77	$-1.74 imes10^{-1}$	$2.80 imes10^{-4}$	-4.82

Table 3. Results for the Calculation of the  $N_2O$  Solubility in MEA +  $H_2O$ 

				AAD %	
°C	$\frac{C_{\rm MEA}}{\rm kmol \cdot m^{-3}}$	no. of data points	data source <sup>a</sup>	Wang et al., 1992	calc by eq 4
15-25	0-5.86	15	1	1.1	2.6
30-75	0.18-3.91	30	2	31.3	6.6
30-40	4.95	3	3	8.2	13.8
30-40	0–6	21	4	5.8	1.5
01	verall	69		16.0	4.5

 $^{a}\left( 1\right)$  Sada and Kito, 1972. (2) Littel et al., 1992. (3) Li and Lai, 1995. (4) This study.

Table 4. Results for the Calculation of the  $N_2O$  Solubility in DEA +  $H_2O$ 

				AAD %		
°C	$\frac{C_{\rm DEA}}{\rm kmol \cdot m^{-3}}$	no. of data points	data source <sup>a</sup>	Wang et al., 1992	calc by eq 4	
25	0-3.08	6	1	2.2	1.8	
25	0.45 - 3.08	10	2	1.9	1.7	
25	0.19 - 8.55	17	3	6.3	6.5	
15 - 30	0 - 3.12	16	4	2.1	6.0	
30 - 60	0.24 - 3.64	23	5	1452.7	9.3	
30 - 40	2.94	3	6	20.5	43.2	
30-40	0-3	21	7	63.8	1.0	
01	/erall	96		364.4	6.2	

 $^a$  (1) Sada et al., 1977. (2) Versteeg and van Swaaij, 1988. (3) Versteeg and Oyevaar, 1989. (4) Haimour, 1990. (5) Littel et al., 1992. (6) Li and Lee, 1996. (7) This study.

Table 5. Results for the Calculation of the  $N_2O$  Solubility in DIPA +  $H_2O$ 

				AAD 9	%
$\frac{t}{^{\circ}C}$	$\frac{C_{\rm DIPA}}{\rm kmol \cdot m^{-3}}$	no. of data points	data source <sup>a</sup>	Wang et al., 1992	calc by eq 4
25	0.25 - 2.92	10	1	8.7	6.1
20 - 60	0.28 - 4.06	55	2	4.5	3.2
30 - 40	0-3	21	3	2.3	2.2
03	verall	86		4.5	3.3

 $^a\left(1\right)$ Sada et al., 1978. (2) Versteeg and van Swaaij, 1988. (3) This study.

centigrade (°C) was used in the correlation of Wang et al. (1992).

For the MEA +  $H_2O$  system, the results of the solubility calculation using eq 4 are presented in Table 3 and also shown in Figures 3 and 4. Using only the solubility data at 15 and 25 °C (data of Sada and Kito, 1972), the correlation of Wang yields poor results for the solubility calculations for temperatures above 40 °C. The broken line in Figure 3 shows the calculated values from the correlation of Wang et al. (1992) at 40 °C. The obtained data at 30 °C are in good agreement with those of Littel et al. (1992). As shown in Figure 3, the data of Li and Lai (1995) at (30, 35, and 40) °C are slightly lower than those values obtained in this study. As shown in Figure 4, the data of Littel et al. (1992) at 45 °C are approaching the calculated values

Table 6. Results for the Calculation of the  $N_2O$  Solubility in MDEA +  $H_2O$ 

				AAD %	
°C	$\frac{C_{\rm MDEA}}{\rm kmol \cdot m^{-3}}$	no. of data points	data source <sup>a</sup>	Wang et al., 1992	calc by eq 4
15-35	0 - 3.46	24	1	2.6	2.6
20 - 60	0.35 - 2.75	50	2	3.5	3.1
15 - 50	0 - 4.36	42	3	7.1	7.7
30 - 40	0 - 3.0	21	4	2.1	2.4
0	verall	137		4.3	4.4

 $^a$  (1) Haimour and Sandall, 1984. (2) Versteeg and van Swaaij, 1988. (3) Al-Ghawas et al., 1989. (4) Ko and Li, 1999.

Table 7. Results for the Calculation of the  $N_2O$  Solubility in TEA +  $H_2O$ 

− t °C	$\frac{C_{\text{TEA}}}{\text{kmol} \cdot \text{m}^{-3}}$	no. of data points	data source <sup>a</sup>	AAD% calc by eq 4
25	0.62 - 2.92	5	1	17
20 - 60	0.12 - 3.20	31	2	2.3
30 - 40	0-3.0	21	3	1.8
0	verall	57		3.0

<sup>a</sup> (1) Sada et al., 1977. (2) Littel et al., 1992. (3) This study.

Table 8. Results for the Calculation of the  $N_2O$  Solubility in  $AMP \,+\, H_2O$ 

				AAD 9	%
<u>t</u> °C	$\frac{C_{\rm AMP}}{\rm kmol \cdot m^{-3}}$	no. of data points	data source <sup>a</sup>	Wang et al., 1992	calc by eq 4
25	0-2.5	13	1	1.1	1.2
10.2 - 75.2	2.0 - 3.0	14	2	1.7	1.5
30	0.19 - 3.25	8	3	5.0	4.8
15.5 - 30	0.5 - 2.0	16	4	5.9	5.6
30 - 40	0 - 3.0	21	5	1.4	1.3
ove	rall	72		2.8	2.7

<sup>a</sup> (1) Bosch et al., 1990. (2) Xu et al., 1991. (3) Littel et al., 1992.
(4) Saha et al., 1993. (5) This study.



Figure 3. Solubility of  $N_2O$  in MEA +  $H_2O$  as a function of MEA concentration for temperatures from 15 to 40  $^\circ C.$ 

at 50 °C while at 75 °C the data of Littel et al. are closely approaching the calculated values at 70 °C. As shown in Figures 3 and 4, the calculated Henry's constants for N<sub>2</sub>O in MEA + H<sub>2</sub>O are reasonable to represent the data for a wide temperature range from (15 to 75) °C and for concentrations of MEA up to 6 kmol·m<sup>-3</sup>. For the MEA +



Figure 4. Solubility of  $N_2O$  in MEA +  $H_2O$  as a function of MEA concentration for temperatures from 45 to 75  $^\circ C.$ 



Figure 5. Solubility of  $N_2O$  in DEA +  $H_2O$  as a function of DEA concentration for temperatures up to 25 °C.

 $H_2O$  system, the following solubility of  $N_2O$  data are recommended: (15 and 25) °C of Sada and Kito (1972); (30 and 60) °C of Littel et al. (1992); and (30, 35, 40) °C of the present study.

For the solubility of  $N_2O$  in DEA +  $H_2O$ , the results of calculation are presented in Table 4 and also shown in Figures 5 and 6. As mentioned earlier, the data of Versteeg and Oyevaar (1989) at 25 °C beyond 6.0 kmol·m<sup>-3</sup> were not used in the database to determine the parameters. Probably due to using only the low-temperature solubility data (15, 20, 25, and 30) °C, the correlation of Wang gives poor predictions for temperatures above 40 °C. As can be seen in Figure 6, the broken lines show the calculated values from the correlation of Wang et al. (1992) at 30 and 40 °C. At 15 °C, eq 4 fits the data of Haimour (1990) quite well (1.7%), as shown in Figure 5. The solubility data of Haimour (1990) at 20 °C are approaching the data at 15 °C and result in a 4.3% deviation from the calculated values using eq 4. At 25 °C, the data of Versteeg and Oyevaar (1989) cover a wide concentration range up to 8.55 kmol $\cdot$ m<sup>-3</sup>. Compared to the literature values at 25 °C, the results of the solubility calculation are satisfactory for DEA concentrations up to 6.0 kmol·m<sup>-3</sup>. For temperatures above 30



Figure 6. Solubility of  $N_2O$  in DEA +  $H_2O$  as a function of DEA concentration for temperatures from 30 to 70 °C.

°C, the data in the open literature are not quite consistent, as shown in Figure 6. Compared to the data of Littel et al. (1992) and this study, the solubility data of Haimour (1990) at 30 °C exhibit quite large values as the concentration of DEA goes to 3.12 kmol·m<sup>-3</sup>. The data of Littel et al. (1992) at 30 °C also show slightly higher values when compared with the values obtained in this study. The data of Li and Lee (1996) at (30, 35, and 40) °C also appear to have too much higher values of  $N_2O$  solubility in DEA +  $H_2O$ . At 45 °C, the data of Littel et al. (1992) appear to have higher values than the calculated values. At 60 °C, the calculated Henry's values are consistent with the data of Littel et al. (1992) at low concentrations of DEA, while at high concentration of DEA, the solubility data of Littel et al. (1992) are higher than the calculated values. The data of the Henry's constant of  $N_2O$  in DEA +  $H_2O$  are quite scattered, which may contribute to the inconsistent results for the reaction kinetics study in the literature (Blauwhoff et al., 1984). The reaction rate expressions found in the literature vary widely in dependence on the DEA and OH- concentrations (Blauwhoff et al., 1984). For the DEA + H<sub>2</sub>O system, the following solubility of N<sub>2</sub>O data are recommended: (15, 20, 25) °C of Haimour (1990); 25 °C of Sada et al. (1977); 25 °C of Versteeg and van Swaaij (1988); 25 °C of Versteeg and Ovevaar (1989); (30, 35, 40) °C of the present study; and 60 °C of Littel et al. (1992).

For the solubility of  $N_2O$  in DIPA +  $H_2O$ , the calculation results are presented in Table 5 and also shown in Figure 7. The overall AAD % values for the solubility calculations are 4.5% and 3.3% for the correlation of Wang et al. (1992) and eq 4 of this study, respectively. Compared to the solubility data of Versteeg and van Swaaij (1988) at 25 °C, the solubility data of Sada et al. (1978) at 2.918 kmol $\cdot$ m<sup>-3</sup> yield a quite large value, 7701 kPa·m<sup>3</sup>·kmol<sup>-1</sup>. Except for concentrations above 2.5 kmol $\cdot$ m<sup>-3</sup>, the solubility data of Sada et al. (1978) at 25 °C are in good agreement with those of Versteeg and van Swaaij (1988). At 35 °C, the solubility data of Versteeg and van Swaaij (1988) are in good agreement with values obtained in this study except for concentrations of DIPA <0.5 kmol·m<sup>-3</sup>. At 45 °C, eq 4 fits the data of Versteeg and van Swaaij (1988) well except at the concentration 1.0 kmol·m<sup>-3</sup>. As shown in Figure 7, the calculated Henry's constants for  $N_2O$  in DIPA +  $H_2O$  are reasonable for temperatures from (20 to 60) °C and for concentrations of DIPA up to 4 kmol·m<sup>-3</sup>.



Figure 7. Solubility of  $N_2O$  in  $DIPA+H_2O$  as a function of DIPA concentration.



Figure 8. Solubility of  $N_2O$  in MDEA +  $H_2O$  as a function of MDEA concentration for temperatures up to 30  $^\circ C.$ 

For the MDEA + H<sub>2</sub>O system, the results of the N<sub>2</sub>O solubility calculation are presented in Table 6 and also shown in Figures 8 and 9. The results of the calculations of the two methods are quite similar; the overall AAD % values are 4.3% and 4.4% for the method of Wang et al. (1992) and eq 4 of this study, respectively. Compared to the solubility data of Haimour and Sandall (1984) and Al-Ghawas et al. (1989), the value of Versteeg and van Swaaij (1988) at 25 °C and the MDEA concentration 2.75 kmol·m<sup>-3</sup> appears to be too large. At 35 °C, the values of Al-Ghawas et al. (1989) are lower than those of Haimour and Sandall (1984), those of Versteeg and van Swaaij (1988), and the values obtained in this study. Probably due to smaller values of N<sub>2</sub>O in water as shown in Figure 2, most of the data of Al-Ghawas et al. (1989) are lower than the literature values. The obtained data in this study at 35 °C are in good agreement with those of Versteeg and van Swaaij (1988). The data of Al-Ghawas et al. at 50 °C are lower than those of Versteeg and van Swaaij (1988) at 45 °C and the values obtained in this study at 40 °C. The largest deviation (AAD % = 25.7%) of the calculations obtained is found for the data of Al-Ghawas et al. (1989) at 50 °C. At 60 °C, eq 4 fits the data of Versteeg and van



Figure 9. Solubility of  $N_2O$  in MDEA +  $H_2O$  as a function of MDEA concentration for temperatures from 35 to 60 °C.



Figure 10. Solubility of  $N_2O$  in  $TEA+H_2O$  as a function of TEA concentration.

Swaaij (1988) pretty well. As shown in Figures 8 and 9, the calculated Henry's constants for  $N_2O$  in MDEA +  $H_2O$  are reasonable to represent the data from (15 to 60) °C and for concentrations of MDEA up to 4 kmol·m<sup>-3</sup>.

For the solubility of  $N_2O$  in TEA +  $H_2O$ , the calculated results are presented in Table 7 and also shown in Figure 10. Wang et al. (1992) does not provide the parameters for the solubility of  $N_2O$  in TEA +  $H_2O$ . The overall AAD % for the calculation of the solubility of  $N_2O$  in TEA +  $H_2O$  using eq 4 is 3.0%. The Henry's constants of  $N_2O$  in TEA +  $H_2O$  at 25 °C of Sada et al. (1978) are higher than those values of Littel et al. (1992) and values obtained in this study. The AAD % values of the calculation of the solubility of  $N_2O$  in TEA +  $H_2O$  are 2.3% and 1.8% for the data of Littel et al. (1992) and values obtained in this study, respectively. As shown in Figure 10, the calculated Henry's constants for  $N_2O$  in TEA +  $H_2O$  are reasonable to represent the data from (20 to 60) °C and for concentrations of TEA up to 3.0 kmol·m<sup>-3</sup>.

For the AMP +  $H_2O$  system, the results of the  $N_2O$  solubility calculation are presented in Table 8 and also shown in Figures 11 and 12. Similar results were obtained for both methods, the correlation of Wang et al. (AAD % =



**Figure 11.** Solubility of  $N_2O$  in AMP +  $H_2O$  as a function of AMP concentration for temperatures up to 30 °C.



**Figure 12.** Solubility of  $N_2O$  in AMP +  $H_2O$  as a function of AMP concentration for temperatures from 35 to 75 °C.

2.8%), and eq 4 of the present study (AAD % = 2.7%). At 25 °C, the values of Saha et al. (1993) are higher than those of Bosch et al. (1990). Equation 4 fits the data of Xu et al. (1991) quite well; the AAD % is 1.5%, as shown in Table 8. As shown in Figures 11 and 12, the calculation of Henry's constants for N<sub>2</sub>O in AMP + H<sub>2</sub>O from eq 4 is satisfactory (the overall AAD % = 2.7%) to represent the N<sub>2</sub>O solubility data for a wide temperature range from (10 to 75) °C and for concentrations of AMP up to 3.25 kmol·m<sup>-3</sup>.

### Conclusion

The solubility of nitrous oxide ( $N_2O$ ) in alkanolamine aqueous solutions has been measured for (30, 35, and 40) °C. The alkanolamines studied are MEA, DEA, DIPA, TEA, and AMP aqueous solutions. The concentration of amine for MEA ranges from (1 to 6) kmol·m<sup>-3</sup> and for other amines from (0.5 to 3) kmol·m<sup>-3</sup>. A semiempirical model of the excess Henry's constant proposed by Wang et al. (1992) was used to correlate the solubility of  $N_2O$  in amine solutions. The parameters of the correlation were determined from both the solubility of  $N_2O$  obtained in this study and the literature values. For a wide temperature range from (15 to 75) °C, the obtained correlation has been shown to represent reasonably the solubility of N<sub>2</sub>O in six amine aqueous solutions: MEA, DEA, DIPA, TEA, MDEA, and AMP. For the purpose of process design, the obtained correlations are, in general, satisfactory for estimating the solubility of N<sub>2</sub>O in amine solutions, which in turn can be used to estimate the correct free-gas solubility of CO2 in amines.

### **Literature Cited**

- 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.
- Blauwhoff, P. M. M.; Versteeg, G. F.; van Swaaij, W. P. M. A Study of the Reaction Between CO<sub>2</sub> and Alkanolamines in Aqueous Solutions. Chem. Eng. Sci. 1984, 39, 207-225.
- Bosch, H.; Versteeg, G. F.; van Swaaij, W. P. M. Kinetics of the Reaction of CO<sub>2</sub> with the Sterically Hindered Amine 2-Amino-2-methyl-1-propanol at 298 K. Chem. Eng. Sci. 1990, 45, 1167-1173.
- Clarke, J. K. A. Kinetics of Absorption of Carbon Dioxide in Monoethanolamine Solutions at Short Contact Times. Ind. Eng. Chem. Fundams. 1964, 3, 239-245.
- Duda, J. F.; Vrentas, J. C. Laminar Liquid Jet Diffusion Studies. AIChE J. 1968, 14, 286-294.
- Haimour, N. M. Solubility of N<sub>2</sub>O in Aqueous Solutions of Diethanol-amine at Different Temperatures. J. Chem. Eng. Data 1990, 35, 177 - 178
- Haimour, N.; Sandall, O. C. Absorption of Carbon Dioxide into Aqueous Methyldiethanolamine. *Chem. Eng. Sci.* **1984**, *39*, 1791–1796. Hsu, C. H.; Li, M. H. Densities of Aqueous Blended Amines. *J. Chem.*
- Eng. Data 1997, 42, 502-507.
  Ko, J. J.; Li, M. H. Kinetics of Absorption of Carbon Dioxide into Solutions of N-Methyldiethanolamine + Water. Chem. Eng. Sci. 1999 (submitted)
- Kohl, A. L.; Riesenfeld, F. C. Gas Purification, 4th ed.; Gulf: Houston, 1985

- Li, M. H.; Lai, M. D. Solubility and diffusivity of N<sub>2</sub>O and CO<sub>2</sub> in (Monoethanolamine + N-Methyldiethanolamine + Water) and in (Monoethanolamine + 2-Amino-2-methyl-1-propanol + Water). J. Chem. Eng. Data 1995, 40, 486-492.
- Li, M. H.; Lee, W. C. Solubility and Diffusivity of N<sub>2</sub>O and CO<sub>2</sub> in (Diethanolamine + *N*-Methyldiethanolamine + Water) and in (Diethanolamine + 2-Amino-2-methyl-l-propanol + Water). *J. Chem.*
- (Diethanolamine + 2-Amino 2-methyr r proparation 1.1007) *Eng. Data* **1996**, *41*, 551–556. Littel, R. J.; Versteeg, G. F.; van Swaaij, W. P. M. Solubility and Diffusivity Data for the Absorption of COS, CO<sub>2</sub>, and N<sub>2</sub>O in Amine Solutions. *J. Chem. Eng. Data* **1992**, *37*, 49–55.
- Sada, E.; Kito, S. Solubilities of Gases in Aqueous Monoethanolamine Solutions. Kagaku Kogaku 1972, 36, 218–220.
- Sada, E.; Kumazawa, H.; Butt, M. A. Solubility of Gases in Aqueous Solutions of Amine. *J. Chem. Eng. Data* **1977**, *22*, 277–278.
- 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
- Saha, A. K.; Bandyopadhyay, S. S.; Biswas, A. K. Solubility and Diffusivity of N2O and CO2 in Aqueous Solutions of 2-Amino-2-
- methyl-1-propanol. *J. Chem. Eng. Data* **1993**, *38*, 78–82. Versteeg, G. F.; van Swaaij, W. P. M. Solubility and Diffusivity of Acid Gases (CO<sub>2</sub>, N<sub>2</sub>O) in Aqueous Alkanolamine Solutions. J. Chem. Eng. Data **1988**, *33*, 29–34.
- Versteeg, G. F.; Oyevaar, M. H. The Reaction Between CO2 and
- Diethanolamine at 298 K. Chem. Eng. Sci. 1989, 44, 1264–1268. Wang, Y. W.; Xu, S.; Otto, F. D.; Mather, A. E. Solubility of N<sub>2</sub>O in Alkanolamines and in Mixed Solvents. Chem. Eng. J. 1992, 48, 31– 40.
- Xu, S.; Otto, F. D.; Mather, A. E. Physical Properties of Aqueous AMP Solutions. J. Chem. Eng. Data 1991, 36, 71-75.
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