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Physical Solubility and Diffusivity of N_2O and CO_2 into Aqueous Solutions of (2-Amino-2-methyl-1-propanol + Monoethanolamine) and (*N*-Methyldiethanolamine + Monoethanolamine)

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In this work, the physical solubility of N_2O in (monoethanolamine + water), (*N*-methyldiethanolamine + monoethanolamine + water), and (2-amino-2-methyl-1-propanol + monoethanolamine + water) and the diffusivity of N_2O in (*N*-methyldiethanolamine + monoethanolamine + water) and (2-amino-2-methyl-1-propanol + monoethanolamine + water) have been measured at (293, 298, 303, 308, and 313) K. For the binary mixture, the amine concentration ranges studied are (2.0, 2.5, and 3.0) $\text{kmol}\cdot\text{m}^{-3}$. For the ternary mixtures, the total amine strength in the solution was kept at 30 mass %, in view of the recent interest in using concentrated amine solutions in gas treatment. A solubility apparatus was used to measure the solubility of N_2O in amine solutions. The diffusivity was measured with a wetted wall column absorber. The accuracy of the measurement is estimated to be ±2%. A semiempirical model of the excess Henry's constant was used to correlate the solubility of N_2O in amine solutions. The parameters of the correlation were determined from the measured solubility data. The experimental N_2O solubility and diffusivity data have been correlated. Comparison with the experimental results indicates that the models for solubility and diffusivity will be useful in estimating the solubility of N_2O in (MEA + H_2O), (MDEA + MEA + H_2O), and (AMP + MEA + H_2O) as well as the diffusivity of N_2O in (MDEA + MEA + H_2O) and (AMP + MEA + H_2O) solvents. The "N_2O analogy" is then used to estimate the solubility and diffusivity of CO_2 in the aqueous alkanolamine solutions.

Introduction

The removal of acid gas impurities, such as CO_2 and H_2S, from natural, refinery, and synthesis gas streams is a significant operation in gas treatment. A wide variety of alkanolamines that have proven to be of commercial interest for acid gas removal by chemical absorption are monoethanolamine (MEA), diethanolamine (DEA), *N*-methyldiethanolamine (MDEA), and di-2-propanolamine (DIPA). A different class of chemical absorbents, the sterically hindered amine (e.g., 2-amino-2-methyl-1-propanol (AMP)), has been suggested recently as an attractive solvent for the removal of acid gases from sour gas streams.

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. Blends of primary and tertiary amines, such as aqueous solutions of MEA and MDEA, have been suggested for CO_2 removal.[1,2] Sterically hindered amine-based blended amine solvents, for example, (AMP + MEA + H_2O), have also been suggested recently as potential attractive new solvents for acid gas treatment processes.[2,3]

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. However, because CO_2 reacts in amine solutions, its physical solubility and diffusivity cannot be measured directly. As a result, one must use a nonreacting gas such as N_2O as a surrogate for CO_2. The N_2O analogy has been frequently used to estimate the solubility and diffusivity of CO_2 in amine solutions.[4−9] Numerous solubility and diffusivity data of N_2O in aqueous amine solutions were reported in the literature for binary systems such as (MEA + H_2O),[4,10−13] (AMP + H_2O),[8−10,14−15] and (MDEA + H_2O),[5−7,15] but there is limited literature on the solubility and the diffusivity of N_2O and CO_2 in ternary systems (MDEA + MEA + H_2O) and (AMP + MEA + H_2O). Li and Lai[13] reported the solubility and diffusivity of N_2O and CO_2 in (MDEA + MEA + H_2O) and (AMP + MEA + H_2O) in the temperature range from (303 to 313) K. The solubility and diffusivity of N_2O in 30 mass % and 40 mass % blends of MEA and MDEA aqueous solutions in the temperature range from (313 to 323) K have been reported by Hagewiesche et al.[16] In this work, the physical solubility of N_2O in (MEA + H_2O), (MDEA + MEA + H_2O), and (AMP + MEA + H_2O) and the diffusivity of N_2O in (MDEA + MEA + H_2O) and (AMP + MEA + H_2O) have been measured at (293, 298, 303, 308, and 313) K. For the binary mixture, the amine concentrations were (2.0, 2.5, and 3.0) $\text{kmol}\cdot\text{m}^{-3}$. For the ternary mixtures, the total amine concentration was 30 mass %.

The N_2O analogy for the solubility and diffusivity of CO_2 in amine solutions is given by eqs 1 and 2, respectively,

$$(\mathrm{H}_{CO_2})_{\text{amine}} = (H_{N_2O})_{\text{amine}}\left(\frac{\mathrm{H}_{CO_2}}{H_{N_2O}}\right)_{\text{water}} \tag{1}$$

$$(\mathrm{D}_{CO_2})_{\text{amine}} = (D_{N_2O})_{\text{amine}}\left(\frac{\mathrm{D}_{CO_2}}{D_{N_2O}}\right)_{\text{water}} \tag{2}$$

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where $(H_{N_2O})_{amine}$ and $(D_{N_2O})_{amine}$ are the solubility and diffusivity of N_2O in the amine solution, respectively. Versteeg and van Swaaij[6] proposed, on the basis of the available solubility and diffusivity data for N_2O and CO_2 in water, the following correlations for the solubility and diffusivity of N_2O and CO_2 in water.

$$H_{N_2O}/\text{kPa·m}^3\text{·kmol}^{-1} = 8.7470 \times 10^{-6} \exp\left(-\frac{2284}{T/\text{K}}\right) \quad (3)$$

$$H_{CO_2}/\text{kPa·m}^3\text{·kmol}^{-1} = 2.8249 \times 10^{6} \exp\left(-\frac{2044}{T/\text{K}}\right) \quad (4)$$

$$D_{N_2O}/\text{m}^2\text{·s}^{-1} = 5.07 \times 10^{-6} \exp\left(-\frac{2371}{T/\text{K}}\right) \quad (5)$$

$$D_{CO_2}/\text{m}^2\text{·s}^{-1} = 2.35 \times 10^{-6} \exp\left(-\frac{2119}{T/\text{K}}\right) \quad (6)$$

Experimental Section

Reagent-grade MEA, AMP, and MDEA of 98 mol % purity were obtained from E. Merck. Double-distilled water degassed by boiling was used to make the amine solutions. The total amine contents of the solutions were determined by titration with standard HCl using methyl orange indicator. A pure CO_2 cylinder, obtained from Hydrogas, India, had 99.995 mol % certified purity. A zero-grade N_2O cylinder was obtained from Hydrogas, India.

Physical Solubility. The experimental setup with a Corning glass equilibrium cell was similar to the one used by Kundu et al.[17] A magnetic stirrer was used for the liquid phase, and two four-bladed impellers mounted on a shaft, passing through a mercury seal and driven by a dc motor, were used for the gas phase. The temperature of the equilibrium cell was controlled to within ±0.2 K of the desired level with a circulator temperature controller (FP 55, Julabo, Germany) operated in external control mode. Precalibrated platinum sensors with temperature indicators (Pt-100, TD 300, Julabo, FRG) were used for the measurement of temperature in the equilibrium cell, in the thermostated bath, and in the eudiometer tube. The uncertainty in the measurement of temperature was ±0.1 K. All solubility measurements were done at atmospheric pressure. A precise manomatric device was employed to maintain atmospheric pressure in the cell throughout the equilibrium experiment. The total pressure was measured for each run within an accuracy of ±0.2 kPa.

For each run, the equilibrium cell was allowed to reach thermal equilibrium with respect to the desired temperature for solubility measurement. The cell was then purged with the desired gas. The gas streams at the outlet of the respective flow meters were passed through water vapor saturators, maintained at the measurement temperature, before being introduced into the cell. The gas-phase stirrer was kept on at 70 rpm during purging to ensure a uniform gas-phase concentration throughout the cell. After purging was complete, 10 mL of a freshly prepared amine solution of the desired concentration was quickly transferred to the cell, and the cell was fully sealed. The liquid-phase and gas-phase stirrers were turned on to commence absorption. The attainment of equilibrium was indicated when there was no absorption for at least 1 h while the temperature was maintained constant at the desired level. It took about (4 to 5) h to reach equilibrium for each run. The measured volume change is equal to the volume of gas absorbed. The partial pressure of N_2O in the experiments was corrected for the vapor pressure of the solution.

Table 1. Measured Solubility and Diffusivity of CO_2 and N_2O in Water

	H/kPa·m^3·kmol^{-1}		10^9D/m^2·s^{-1}	
T/K	CO_2 in water	N_2O in water	CO_2 in water	N_2O in water
293	2647	3581	1.71	1.54
298	3096	4091	1.90	1.80
303	3314	4512	2.13	1.98
308	3765	5023	2.47	2.29
313	4098	5715	2.86	2.58

The temperature of the system was controlled to within ±0.2 K. The reproducibility between the various experiments was within 2%. The experimental uncertainty in the measured solubility was estimated is about ±2%.

Diffusivity. The diffusivity of CO_2 in water and N_2O in water and aqueous amine solutions was measured in a cylindrical stainless steel wetted wall column absorber of outside diameter 2.81×10^{-2} m and height 5 cm. The apparatus and the experimental procedure are the same as those described by Saha et al.[9] The gas−liquid contact time could be varied from (0.3 to 0.8) s by varying the absorption length but keeping the liquid flow rate constant. The gas absorption rate was measured by the volume uptake method using a soap film meter. The liquid flow rate was measured with a rotameter that was calibrated at various experimental temperatures and concentrations. The temperature of absorption was controlled within ±0.2 K. The pressure in the absorption chamber was about 100 kPa. The experimental uncertainty was estimated to be ±2%. The reproducibility between the various experiments was always within ±2%.

Results and Discussion

Solubility. To validate the solubility apparatus and the experimental procedure of the measurement, the solubilities of N_2O in water and CO_2 in water at (293, 298, 303, 308, and 313) K were measured. These are presented in Table 1. Figure 1 compares the literature results[7,13] and those obtained in this study for the solubility of CO_2 in water. The comparison between the literature results[7,13] and results obtained in this study for the solubility of N_2O in water is shown in Figure 2. The results of this study for the solubilities of CO_2 and N_2O in water are in excellent agreement with the literature results. Equations 3 and 4 appear to be accurate correlations for the solubility of N_2O

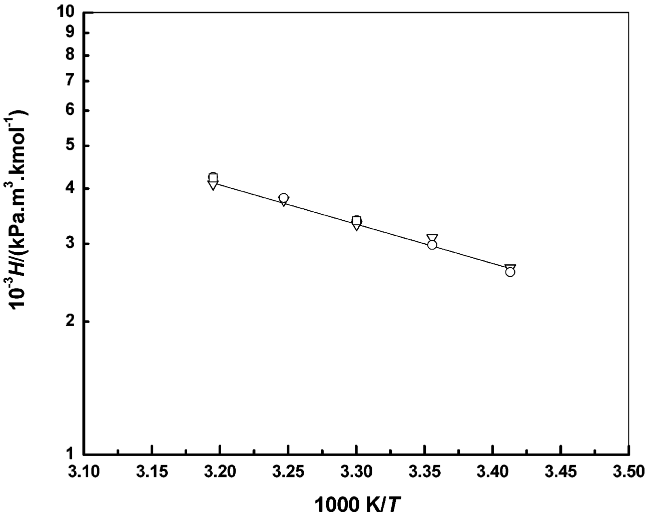

Figure 1. Henry's constant of CO_2 in water as function of temperature: ▽, this study; ○, Al-Ghawas et al.;[7] □, Li and Lai;[13] −, calculated using eq 4.

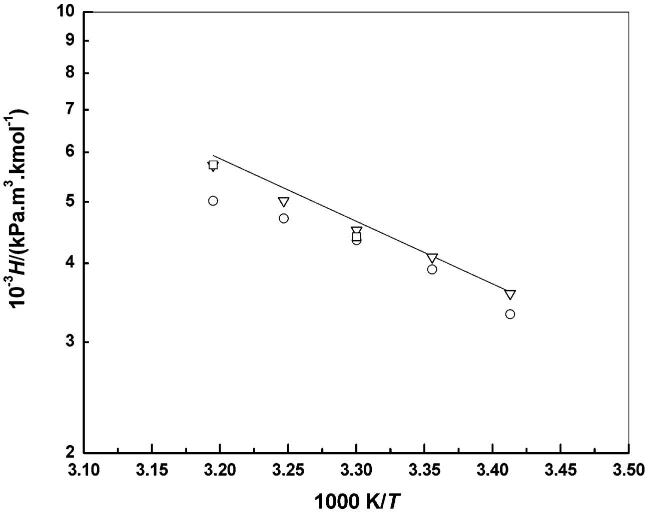

Figure 2. Henry's constant of N_2O in water as function of temperature: ▽, this study; ○, Al-Ghawas et al.;[7] □, Li and Lai;[13] −, calculated using eq 3.

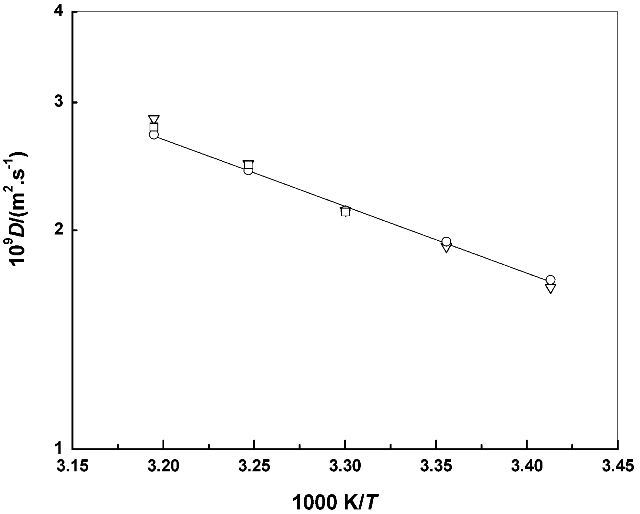

Figure 3. Diffusivity of CO_2 in water as a function of temperature: ▽, this study; ○, Al-Ghawas et al.;[7] □, Li and Lai;[13] −, calculated using eq 6.

and CO_2 in water as a function of temperature, as shown in Figures 2 and 1, respectively. Although the measured N_2O solubility in water from this study and that reported by Li and Lai[13] agreed well with the solubility calculated from eq 3, the solubility values reported by Al-Ghawas et al.[7] are smaller than the calculated values using eq 3.

The measured solubility of N_2O in (MEA + H_2O), (MDEA + MEA + H_2O), and (AMP + MEA + H_2O) for temperatures of (293, 298, 303, 308, and 313) K are presented in Tables 2 and 3. The solubility measurements are in good agreement with the literature results as shown in Tables 4 and 5. As shown in Table 2, the solubilities of N_2O and CO_2 in the binary mixture decrease with increasing temperature. At constant temperature, the solubilities decrease with increasing concentration of MEA. As shown in Figure 5, for the ternary system (AMP + MEA + H_2O) the N_2O solubility has been found to increase with increasing MEA

Table 2. Experimental Solubility of N_2O, H_{N_2O}, and Estimated Solubility of CO_2, H_{CO_2}, for (MEA + H_2O), (AMP + H_2O), and (MDEA + H_2O) from 293 K to 313 K

C	*T*/K					*T*/K				
$\mathrm{kmol{\cdot}m^{-3}}$	293	298	303	308	313	293	298	303	308	313
	$H_{N_2O}/\mathrm{kPa{\cdot}m^3{\cdot}kmol^{-1}}$					$H_{CO_2}/\mathrm{kPa{\cdot}m^3{\cdot}kmol^{-1}}$				
MEA										
2.0	3821	4205	4605	4972	5475	2824	3182	3382	3727	3926
2.5	3840	4218	4643	5003	5493	2838	3192	3410	3750	3939
3.0	3848	4244	4675	5042	5524	2844	3212	3434	3779	3961
AMP										
2.0	4271	4805	5236	5877	6318	3157	3636	3846	4405	4530
2.5	4385	4917	5325	5984	6441	3241	3721	3911	4485	4619
3.0	4492	5045	5451	6072	6545	3320	3818	4004	4551	4693
MDEA										
2.0	4276	4617	4903	5343	5811	3161	3494	3601	4005	4167
2.5	4445	4890	5215	5475	5968	3286	3701	3830	4104	4279
3.0	4697	5075	5345	5602	6095	3472	3841	4115	4199	4371

Table 3. Experimental Solubility of N_2O, H_{N_2O}, and Estimated Solubility of CO_2, H_{CO_2}, for (AMP + MEA + H_2O) and (MDEA + MEA + H_2O) from 293 K to 313 K

mass %	*T*/K					*T*/K				
mass %	293	298	303	308	313	293	298	303	308	313
	$H_{N_2O}/\mathrm{kPa{\cdot}m^3{\cdot}\ kmol^{-1}}$					$H_{CO_2}/\mathrm{kPa{\cdot}m^3{\cdot}\ kmol^{-1}}$				
MDEA/MEA										
30/0	4423	4879	5205	5498	5979	3269	3692	3823	4121	4287
28.5/1.5	4394	4845	5171	5447	5945	3248	3667	3798	4083	4263
27/3	4502	4803	5145	5402	5892	3222	3635	3779	4049	4225
25.5/4.5	4472	4769	5096	5375	5854	3194	3609	3743	4027	4198
24/6	4435	4715	5045	5346	5837	3167	3568	3706	4007	4186
22.5/7.5	4392	4682	4991	5315	5809	3144	3543	3666	3984	4165
21/9	4357	4635	4943	5290	5769	3118	3508	3631	3965	4137
0/30	4305	4311	4789	5146	5613	2895	3262	3517	3857	4025
AMP/MEA	4274									
30/0	4502	5060	5475	6095	6582	3328	3829	4021	4569	4720
28.5/1.5	4472	4995	5405	5997	6445	3306	3780	3970	4495	4622
27/3	4435	4930	5326	5915	6382	3278	3731	3912	4434	4576
25.5/4.5	4392	4885	5250	5825	6305	3247	3697	3856	4366	4521
24/6	4357	4846	5181	5746	6219	3221	3667	3805	4307	4459
22.5/7.5	4305	4775	5105	5613	6143	3182	3614	3750	4207	4405
21/9	4275	4704	5045	5525	6065	3159	3560	3706	4141	4349

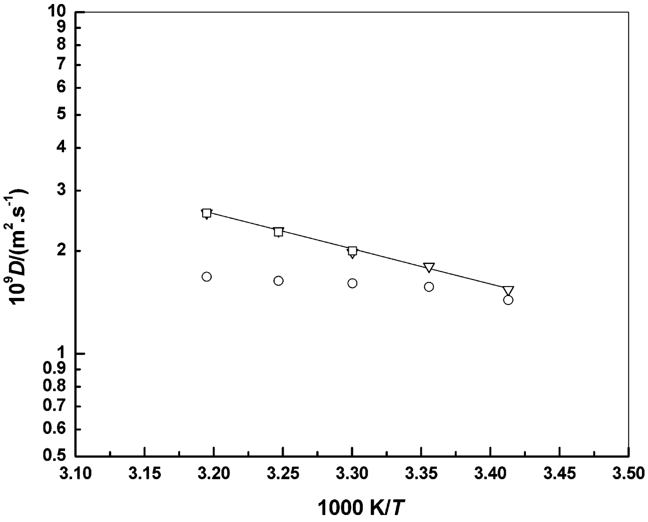

Figure 4. Diffusivity of N_2O in water as a function of temperature: ▽, this study; ○, Al-Ghawas et al.;[7] □, Li and Lai;[13] −, calculated using eq 5.

Table 4. Comparison of the Measured Solubility of N_2O in (MEA + H_2O) with Literature Values

C	T	% deviation from literature	ref
kmol·m^{-3}	K		
MEA			
2−3	303−313	9	Tsai et al.[12]
2−3	298	5	Saha[11]

Table 5. Comparison of the Measured Solubility of N_2O in (MDEA + MEA + H_2O) and (AMP + MEA + H_2O) with Literature Values

mass %	T	% deviation from literature	ref
mass %	K		
MEA/MDEA			
30/0	303−313	9	Li and Lai[13]
1.5/28.5	303−313	5	Hagewiesche et al.[16]
4.5/25.5	303−313	5	Hagewiesche et al.[16]
6/24	303−313	2	Li and Lai[13]
MEA/AMP			
6/24	303−313	11	Li and Lai[13]

concentration at constant temperature. However, the solubility decreases with increasing temperature at constant MEA concentration.

A semiempirical model proposed by Wang et al.[18] was used to correlate the solubility of N_2O in amine solutions. In this method, the excess Henry's coefficient for the mixed solvent system has the following form

$$R = \ln H_{1,\mathrm{m}} - \sum_{i=2}^{n} \varphi_i \ln H_{1,i} \tag{7}$$

where $H_{1,\mathrm{m}}$ is the Henry's constant of N_2O in the mixed solvent, $H_{1,i}$ is the Henry's constant of N_2O in pure solvent i, and φ_i is the volume fraction of solvent i. The volume fraction is calculated as

$$\varphi_i = \frac{x_i v_i}{\sum_{i=1}^{n} x_i v_i} \tag{8}$$

where ν_i is the molar volume of pure solvent i and x_i is the mole fraction of solvent i. From eq 7, the excess Henry's quantity R can be calculated from the measured $H_{1,\mathrm{m}}$ and the estimated $H_{1,i}$.

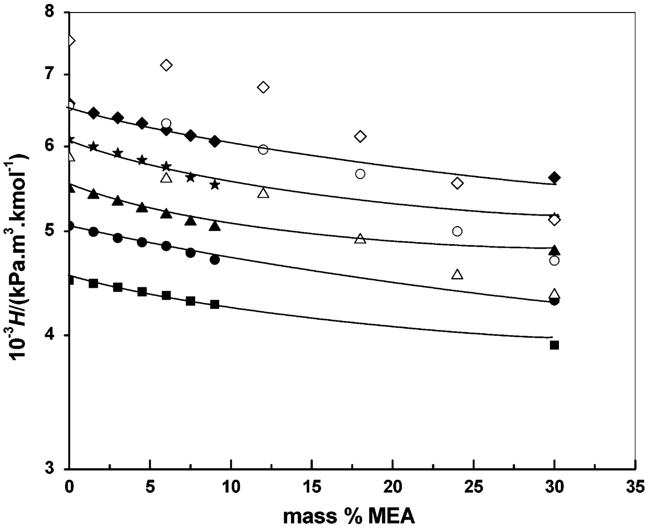

Figure 5. Henry's constant of N_2O in (AMP + MEA + H_2O) with the total amine strength in the solution at 30 mass % as a function MEA concentration at different temperatures: ■, 293 K; ●, 298 K; ▲, 303 K; ★, 308 K; ◆, 313 K; △, 303 K;[13] ○, 308 K;[13] ◇, 313 K;[13] −, calculated using eq 7.

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

$$R_{ij} = \varphi_i \varphi_j \alpha_{ij} \tag{9}$$

where φ_i and φ_j are the volume fractions of amine and water, respectively. To correlate the excess quantity R_{ij} in the binary solvent systems involved in this work, the two-body interaction parameter α_{ij} for the absorption solvent may be estimated as a polynomial function of the absorption temperature and the volume fraction of the solvent j as follows:

$$\alpha_{ij} = k_1 + k_2(T/\mathrm{K}) + k_3(T/\mathrm{K})^2 + k_4\varphi_j \tag{10}$$

R_{ij} is a third-order polynomial of the volume fraction of the solvent and a second-order polynomial of the temperature. The four parameters k_1, k_2, k_3, and k_4 in eq 10 were all obtained from corresponding experimental data by a regression method using MATLAB version 6.0. The average regression deviation between calculated values and experimental data was calculated from

$$\sigma = \frac{1}{n}\sum_{i=1}^{n} \frac{|H_{\mathrm{m,calcd}} - H_{\mathrm{m,exptl}}|}{H_{\mathrm{m,exptl}}} \tag{11}$$

Wang et al.[18] proposed the solubility of N_2O in pure amine solvent as follows:

$$H_{\mathrm{N_2O-pure\ amine}} = b_1 \exp\left(\frac{b_2}{T/\mathrm{K}}\right) \tag{12}$$

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.[18] for temperatures between (293 and 358) K, the average regression deviations between the calculated solubilities of N_2O in pure amines and experimental data are <2.5%, which is satisfactory for estimating the solubilities of N_2O in pure amines. Thus, eq 12 with the parameters b_i of Wang et al.[18] was adopted directly in

Table 6. Parameters k_1, k_2, k_3, k_4 of the Equation $\alpha_{ij} = k_1 + k_2(T/\mathrm{K}) + k_3(T/\mathrm{K})^2 + k_4\varphi_j$ and α_{ijk} for the Excess Henry's Constant for the Binary and Ternary Solvent Systems

system	k_1	k_2	k_3	k_4	α_{ijk}	AAD %
(MEA + H_2O)	23.9632	−0.10320	0.00011	−2.33110		1.5
(AMP + H_2O)	−29.7608	0.23622	−0.00042	−1.57718		0.8
(MDEA + H_2O)	81.9682	−0.47261	0.00071	−3.56729		0.8
(MEA + AMP)	3796.13	−6.82751	0.01068	−212.645		
(MEA + MDEA)	3561.00	1.98104	−0.00383	92.9584		
(MEA + AMP + H_2O)					−3859.47	0.9
(MEA + MDEA + H_2O)					−5422.52	1.1

Table 7. Experimental Diffusivity of N_2O, D_{N_2O}, and Estimated Diffusivity of CO_2, D_{CO_2}, for (AMP + MEA + H_2O) and (MDEA + MEA + H_2O) from 293 K to 313 K

mass%	T/K					T/K				
mass%	293	298	303	308	313	293	298	303	308	313
	$10^9 D_{N_2O}/\mathrm{m^2 \cdot s^{-1}}$					$10^9 D_{CO_2}/\mathrm{m^2 \cdot s^{-1}}$				
MDEA/MEA										
30/0	0.84	0.93	1.10	1.16	1.21	0.93	0.98	1.18	1.25	1.34
28.5/1.5	0.87	1.01	1.15	1.22	1.28	0.97	1.07	1.24	1.32	1.42
27/3	0.92	1.09	1.21	1.30	1.39	1.02	1.15	1.30	1.40	1.54
25.5/4.5	1.00	1.15	1.32	1.39	1.47	1.11	1.21	1.42	1.50	1.63
24/6	1.12	1.22	1.37	1.45	1.52	1.24	1.29	1.47	1.56	1.69
22.5/7.5	1.19	1.27	1.40	1.47	1.55	1.32	1.34	1.51	1.59	1.72
21/9	1.24	1.35	1.44	1.52	1.60	1.38	1.43	1.55	1.64	1.77
0/30	1.45	1.52	1.62	1.78	1.93	1.61	1.60	1.74	1.92	2.14
AMP/MEA										
30/0	0.71	0.83	1.01	1.19	1.32	0.79	0.88	1.09	1.28	1.46
28.5/1.5	0.75	0.89	1.04	1.25	1.37	0.83	0.94	1.12	1.35	1.52
27/3	0.77	0.93	1.09	1.29	1.42	0.86	0.98	1.17	1.39	1.57
25.5/4.5	0.82	0.97	1.15	1.35	1.51	0.91	1.02	1.24	1.46	1.67
24/6	0.87	1.01	1.20	1.39	1.55	0.97	1.07	1.29	1.50	1.72
22.5/7.5	0.91	1.08	1.24	1.43	1.58	1.01	1.14	1.33	1.54	1.75
21/9	0.97	1.13	1.29	1.47	1.62	1.08	1.19	1.39	1.59	1.80

Table 8. Comparison of the Measured Diffusivity of N_2O in (MDEA + MEA + H_2O) and (AMP + MEA + H_2O) with Literature Values

mass %	T	% deviation	
mass %	K	from literature	ref
MEA/MDEA			
0/30	303−313	4	Li and Lai[13]
		3	Hagewiesche et al.[16]
1.5/28.5	303−313	3	Hagewiesche et al.[16]
3/27	303−313	3	Hagewiesche et al.[16]
4.5/25.5	303−313	3	Hagewiesche et al.[16]
6/24	303−313	4	Li and Lai[13]
30/0	303−313	4	Li and Lai[13]
MEA/AMP			
0/30	303−313	3	Li and Lai[13]
6/24	303−313	3	Li and Lai[13]

this study. Also, the solubilities of N_2O and CO_2 in pure water, H_{N_2O} and H_{CO_2}, are calculated using eqs 3 and 4.[6] The densities of amine aqueous solutions, required in the calculation of the volume fraction, were estimated on the basis of the correlation of Hsu and Li[19] and Mandal et al.[20]

Using the solubility data in this work, the parameters k_1, k_2, k_3, and k_4 in eq 10 are determined for each binary system, and the results are presented in Table 6. We note that the temperature in eq 10 is, in this study, in Kelvin (K), whereas degrees centigrade (°C) was used in the correlation of Wang et al.[18]

For the ternary solvent system, the excess Henry's constant has the form

$$R_{ijk} = \varphi_i\varphi_j\alpha_{ij} + \varphi_i\varphi_k\alpha_{ik} + \varphi_j\varphi_k\alpha_{jk} + \varphi_i\varphi_j\varphi_k\alpha_{ijk} \quad (13)$$

where subscripts i and j stand for amine and water, respectively; subscript k stands for another amine solvent. In this approach, two-body and three-body interactions are considered to describe the excess Henry's constant for the ternary system. Two of the two-body interaction parameters in eq 13 were taken from the excess quantity of the binary systems, (MEA + H_2O) studied in this work and (MDEA + H_2O) and (AMP + H_2O) studied in our previous work.[15] The remaining two-body interactions between MDEA and MEA and between AMP and MEA and one three-body interaction parameter were regressed using (MDEA + MEA + H_2O) and (AMP + MEA + H_2O) ternary systems' N_2O solubility data studied in this work. The α_{ijk} parameter in eq 13 is set to be a constant. When any of the volume fractions in the above equation is zero, eq 13 will be identical to that of the binary system.

Diffusivity. To validate the diffusivity apparatus and the experimental procedure of the measurement, the diffusivities of N_2O and CO_2 in water were measured at (293, 298, 303, 308, and 313) K. The results are presented in Table 1. The comparison between the literature values[7,13] and the values obtained in this study for the diffusivity of CO_2 and N_2O in water is shown in Figures 3 and 4. As shown in Figures 3 and 4, the diffusivity values of CO_2 and N_2O in water obtained in this study are in excellent agreement with the literature values. Although the measured N_2O diffusivity in water from this study and that reported by Li and Lai[13] agreed well with the diffusivity calculated from eq 5, the diffusivity values reported by Al-Ghawas et al.[7] are smaller than the calculated values using eq 5. The measured diffusivities of N_2O as well as estimated diffusivities of CO_2 in (MEA + AMP + H_2O) and (MEA + MDEA + H_2O) are presented in Table 7. The diffusivity measurements are in good agreement with the literature results as shown in Table 8. As shown in Figure 6, for the ternary system (AMP + MEA + H_2O), the N_2O diffusivity has been found to increase with increasing concentration of MEA at constant temperature. The diffusivity also increases with increasing temperature at constant MEA concentration.

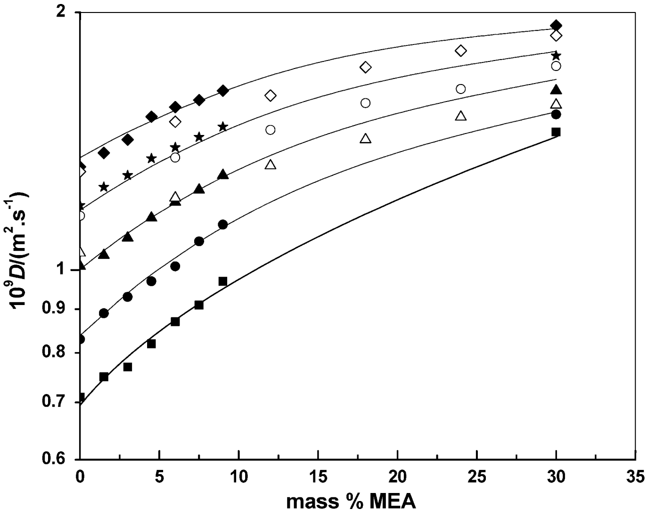

Figure 6. Diffusivity of N_2O in (AMP + MEA + H_2O) with the total amine strength in the solution at 30 mass % as a function MEA concentration at different temperatures: ■, 293 K; ●, 298 K; ▲, 303 K; ★, 308 K; ◆, 313 K; △, 303 K;[13] ○, 308 K;[13] ◇, 313 K;[13] −, calculated using eq 14.

Table 9. Parameters b_0, b_{11}, b_{12}, b_{21}, b_{22}, c_{12}, and c of the Equation $D = (b_0 + b_{11}M_1 + b_{12}M_1^2 + b_{21}M_2 + b_{22}M_2^2 + c_{12}M_1M_2 \exp(-c/T)$ for the Diffusivity of N_2O for (AMP + MEA + H_2O) and (MDEA + MEA + H_2O)

	ternary systems	
parameter	(AMP + MEA + H_2O)	(MDEA + MEA + H_2O)
b_0	2.3850×10^{-7}	-7.4355×10^{-7}
b_{11}	-8.8426×10^{-8}	3.0659×10^{-7}
b_{12}	8.1990×10^{-9}	-3.1521×10^{-8}
b_{21}	-1.4019×10^{-7}	5.8376×10^{-7}
b_{22}	2.0536×10^{-8}	-1.1437×10^{-7}
c_{12}	2.5975×10^{-8}	-1.2000×10^{-7}
c	4.2363×10^{-3}	6.2722×10^{-3}
AAD %	1.4	1.8

The diffusivity of N_2O in the blended amine solutions is calculated as follows

$$D = (b_0 + b_{11}M_1 + b_{12}{M_1}^2 + b_{21}M_2 + b_{22}{M_2}^2 + c_{12}M_1M_2 \exp\left(-\frac{c}{T}\right) \quad (14)$$

where M_1 is the molarity of MEA and M_2 is the molarity of MDEA or AMP. The parameters in eq 14 were determined using the diffusivity data of Table 7. The overall absolute deviations for the calculations of the diffusivity of N_2O in (AMP + MEA + H_2O) and (MDEA + MEA + H_2O) are 1.4 and 1.8%, respectively. The parameters are presented in Table 9.

The Stokes−Einstein relation ($D\eta/T$ = a constant, η is viscosity) has often been used to correlate the diffusivity of N_2O in single amine aqueous solutions.[5,9] The viscosity, η, of the blended amine solutions required to calculate the Stokes−Einstein relation, was taken from Mandal et al.[20] It was observed from Tables 10 and 11 that the experimental diffusivities of N_2O in (AMP + MEA + H_2O) and (MDEA + MEA + H_2O) do not follow the Stokes−Einstein relation strictly.

Table 10. Diffusivity of N_2O in (AMP + MEA + H_2O) for the Stokes−Einstein Relation

T	mass % AMP/	$10^9D_{N_2O}$	$10^3\eta$	$10^{15}(D\eta/T)$
K	mass % MEA	$m^2 \cdot s^{-1}$	$N \cdot s \cdot m^{-2}$	$N \cdot K^{-1}$
293	30/0	0.71	4.89	11.84
	28.5/1.5	0.75	4.78	12.24
	27/3	0.77	4.61	12.12
	25.5/4.5	0.82	4.45	12.45
	24/6	0.87	4.35	12.91
	22.5/7.5	0.91	4.22	13.11
	21/9	0.97	4.09	13.54
	0/30	1.45	2.63	13.02
303	30/0	1.01	3.21	10.70
	28.5/1.5	1.04	3.17	10.88
	27/3	1.09	3.11	11.19
	25.5/4.5	1.15	3.04	11.54
	24/6	1.20	2.98	11.81
	22.5/7.5	1.24	2.91	11.91
	21/9	1.29	2.85	12.13
	0/30	1.62	2.10	11.23
313	30/0	1.32	2.24	9.446
	28.5/1.5	1.37	2.19	9.586
	27/3	1.42	2.16	9.799
	25.5/4.5	1.51	2.13	10.28
	24/6	1.55	2.09	10.34
	22.5/7.5	1.58	2.07	10.45
	21/9	1.62	2.04	10.56
	0/30	1.93	1.60	9.866

Table 11. Diffusivity of N_2O in (MDEA + MEA + H_2O) for the Stokes−Einstein Relation

T	mass % MDEA	$10^9D_{N_2O}$	$10^3\eta$	$10^{15}(D\eta/T)$
K	mass % MEA	$m^2 \cdot s^{-1}$	$N \cdot s \cdot m^{-2}$	$N \cdot K^{-1}$
293	30/0	0.84	3.46	9.919
	28.5/1.5	0.87	3.44	10.21
	27/3	0.92	3.44	10.80
	25.5/4.5	1.00	3.43	11.71
	24/6	1.12	3.42	13.07
	22.5/7.5	1.19	3.38	13.73
	21/9	1.24	3.33	14.09
	0/30	1.45	2.63	13.02
303	30/0	1.10	2.68	9.729
	28.5/1.5	1.15	2.55	9.678
	27/3	1.21	2.53	10.10
	25.5/4.5	1.32	2.51	10.93
	24/6	1.37	2.49	11.26
	22.5/7.5	1.40	2.46	11.37
	21/9	1.44	2.45	11.64
	0/30	1.62	2.10	11.23
313	30/0	1.21	1.92	7.422
	28.5/1.5	1.28	1.88	7.688
	27/3	1.39	1.86	8.260
	25.5/4.5	1.47	1.84	8.642
	24/6	1.52	1.8	8.838
	22.5/7.5	1.55	1.80	8.914
	21/9	1.60	1.78	9.099
	0/30	1.93	1.60	9.866

Conclusions

The solubility of N_2O in (monoethanolamine + water), (*N*-methyldiethanolamine + monoethanolamine + water), and (2-amino-2-methyl-1-propanol + monoethanolamine + water) and the diffusivity of N_2O in (*N*-methyldiethanolamine + monoethanolamine + water) and (2-amino-2-methyl-1-propanol + monoethanolamine + water) have been measured at (293, 298, 303, 308, and 313) K and at atmospheric pressure. The N_2O analogy was used to estimate the solubility of CO_2 in (monoethanolamine + water), (*N*-methyldiethanolamine + monoethanolamine + water), and (2-amino-2-methyl-1-propanol + monoethanolamine + water) as well as diffusivity of CO_2 in (*N*-methyldiethanolamine + monoethanolamine + water) and (2-amino-2-methyl-1-propanol + monoethanolamine + water). The experimental N_2O solubility and diffusivity data have been correlated. For the purpose of process design, the obtained correlations are, in general, satisfactory for estimating the solubility and diffusivity of N_2O in amine solutions. The solubility of N_2O and CO_2 in water as well as in binary and ternary amine solvents has been found to be in good agreement with the literature results and the results calculated from the correlation. The physical solubilities of CO_2 and N_2O in aqueous blended amine solvents have been correlated in this work with standard deviations of 1.5 and 2.3% for (*N*-methyldiethanolamine + monoethanolamine + water) and (2-amino-2-methyl-1-propanol + monoethanolamine + water) solutions, respectively. In the case of the diffusivities of CO_2 and N_2O in (*N*-methyldiethanolamine + monoethanolamine + water) and (2-amino-2-methyl-1-propanol + monoethanolamine + water) systems, the literature results have been found to be in good agreement with the results calculated from the correlation.

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