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

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In this work the physical solubility of N_2O in (diethanolamine + water), (2-amino-2-methyl-1-propanol + water), (*N*-methyldiethanolamine + water), (*N*-methyldiethanolamine + diethanolamine + water), and (2-amino-2-methyl-1-propanol + diethanolamine + water) as well as the diffusivity of N_2O in (*N*-methyldiethanolamine + diethanolamine + water) and (2-amino-2-methyl-1-propanol + diethanolamine + water) have been measured at (293, 298, 303, 308, and 313) K. For the binary mixtures the amine concentration ranges studied are (2.0, 2.5, and 3.0) kmol·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 treating. 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 uncertainty of the measurement is estimated to be ±2%. A semiempirical model of the excess Henry's constant proposed by Wang et al. (*Chem. Eng. J.* **1992**, *48*, 31−40) 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 (DEA + H_2O), (AMP + H_2O), (MDEA + H_2O), (MDEA + DEA + H_2O), and (AMP + DEA + H_2O) as well as diffusivity of N_2O in (MDEA + DEA + H_2O) and (AMP + DEA + 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

Aqueous solutions of alkanolamines are widely used for the removal of acid gases such as CO_2 and H_2S from natural, refinery, and synthesis gas streams. A wide variety of alkanolamines that have proved to be of commercial interest for acid gas removal by chemical absorption are monoethanolamine (MEA), diethanolamine (DEA), *N*-methyldiethanolamine (MDEA), and diisopropanolamine (DIPA). A different class of chemical absorbents, the sterically hindered amines, for example, 2-amino-2-methyl-1-propanol (AMP), has been suggested recently as an attractive solvent for removal of acid gases from sour gas streams.

The use of a blended amine, 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.[2,3] Sterically hindered amine based blended amine solvents, for example, (AMP + MEA + H_2O), have also been suggested recently as potential attractive new solvents for the acid gas treating processes.[3,4] Compared to MEA, DEA has a similar thermodynamic capacity to absorb CO_2 and favorable reaction kinetics for CO_2, but with a better thermal stability and less amine degradation. Thus, MEA may be substituted by DEA in blends of alkanolamine aqueous solutions. Consequently, (MDEA + DEA + H_2O) as well as (AMP + DEA + H_2O) may be a potential solvent for acid gas removal.

For the analysis of the experimental absorption rate and for the rational design of the gas absorption units, two of the most important parameters required are physical solubility and diffusivity of the solute gases in the solvents. However, since 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 to CO_2. The N_2O analogy has been frequently used to estimate the solubility and diffusivity of CO_2 in amine solutions.[5−10] Numerous solubility and diffusivity data of N_2O in aqueous amine solutions were reported in the literature for the binary systems, such as (DEA + H_2O),[7,11−14] (AMP + H_2O),[9,10,15,16] and (MDEA + H_2O).[6−8] But there are limited literature results on the solubility and the diffusivity of N_2O and CO_2 in the ternary systems (MDEA + DEA + H_2O) and (AMP + DEA + H_2O). Li and Lee[14] reported the solubility and the diffusivity of N_2O and CO_2 in (MDEA + DEA + H_2O) and (AMP + DEA + H_2O) in the temperature range (303 to 313) K. The diffusivities of N_2O in 50 mass % blends of DEA and MDEA aqueous solutions in the temperature range (313 to 353) K have been reported by Rinker et al.[17] In this work the physical solubility of N_2O in (DEA + H_2O), (AMP + H_2O), (MDEA + H_2O), (MDEA + DEA + H_2O), and (AMP + DEA + H_2O) and the diffusivity of N_2O in (MDEA + DEA + H_2O) and (AMP + DEA + H_2O) have been measured at (293, 298, 303, 308, and 313) K. For the binary mixtures the amine concentrations were (2.0, 2.5, and 3.0) kmol·m^{-3}. For the ternary mixtures the total amine concentration was 30 mass %.

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

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

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

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[7] proposed, on the basis of the available data of solubility and diffusivity of N_2O and CO_2 in water, the following correlations for the solubility and diffusivity of N_2O and CO_2 in water.

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

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

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

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

Experimental Section

Reagent grade DEA, AMP, and MDEA of 98 mol % purity were obtained from E. Merck. Double distilled water degassed by boiling was used for making 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.[18] 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 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 temperatures 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 manometric 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 uniform gas-phase concentration throughout the cell. After completion of purging, 10 mL of freshly prepared amine solution of desired concentration was quickly transferred into 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.

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

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

	H/kPa·m^3·kmol^{-1}		$D_{CO_2}/10^9$ 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

Diffusivity. The diffusivities of CO_2 in water and N_2O in water and aqueous amine solutions were measured in a cylindrical stainless steel wetted wall absorber of outside diameter 2.81 × 10^{-2} m. The apparatus and the experimental procedure are the same as those described by Saha et al.[10] 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 amine 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 ±4%. 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[8,14] and those obtained in this study for the solubility of CO_2 in water. The comparison between the literature results[8,14] 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 and CO_2 in water as a function of temperature, as shown in Figures 2 and 1, respectively. While the measured N_2O solubility in water of this study and that reported by Li and Lee[14] agreed well with the solubility calculated from eq 3, the solubility values reported by Al-Ghawas et al.[8] are smaller than the calculated values using eq 3.

The measured solubility of N_2O and the estimated solubility of CO_2 in (DEA + H_2O), (AMP + H_2O), (MDEA + H_2O), (MDEA + DEA + H_2O), and (AMP + DEA + H_2O) for the temperatures (293, 298, 303, 308, and 313) K are presented in Tables 2 and 3. The solubility measurements

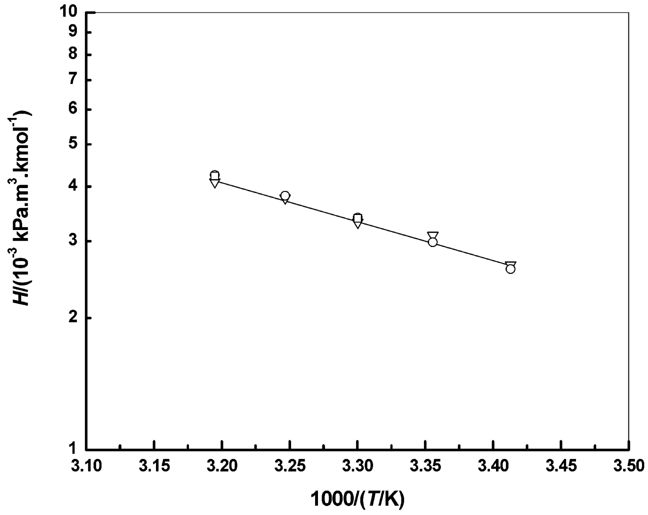

Figure 1. Henry's constant of CO_2 in water as a function of temperature: ▽, this study; ○, Al-Ghawas et al.;[8] □, Li and Lee;[14] —, calculated using eq 4.

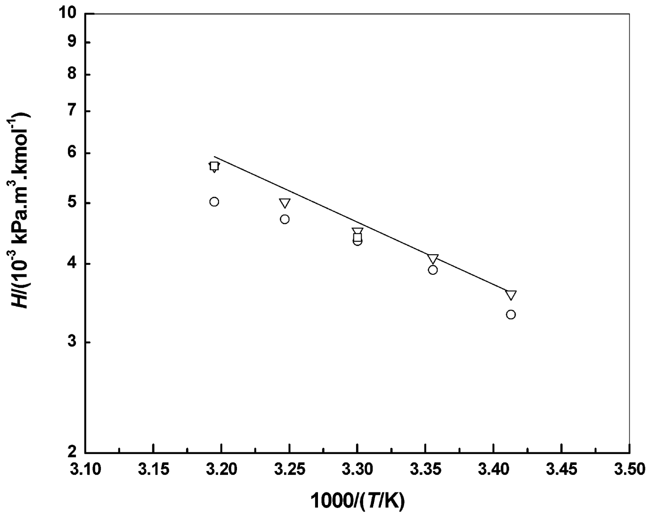

Figure 2. Henry's constant of N_2O in water as a function of temperature: ▽, this study; ○, Al-Ghawas et al.;[8] □, Li and Lee;[14] —, calculated using eq 3.

are in good agreement with the literature results. While the average absolute deviation (AAD%) values of the measured solubilities in the binary systems are in the range 1.5 to 15.6% when compared with the literature results,[8,10,19−22] for the ternary systems of this study the deviations with the literature results[14] are in the range 4.7 to 11.7%. As shown in Table 2, the solubilities of N_2O and CO_2 in the binary mixtures have been found to decrease with increasing temperature. At constant temperature the solubilities decrease with increase in the concentration of DEA, AMP, and MDEA. As shown in Figure 5, for the ternary system (AMP + DEA + H_2O), the N_2O solubility has been found to decrease with the increase in DEA concentration at constant temperature. The solubility also decreases with increase in temperature at constant DEA concentration.

A semiempirical model proposed by Wang et al.[1] 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 Henry's constant of N_2O in the mixed solvent, $H_{1,i}$ is 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

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

where v_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 $H_{1,i}$.

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

Table 2. Estimated Solubility of CO_2, H_{CO_2}, for DEA (1) + H_2O (3) and AMP (2) + H_2O (3) and MDEA (2) + H_2O (3) from 293 K to 313 K Using the N_2O Analogy

amine	H_{N_2O}/kPa·m^3·kmol^{-1} at the following T/K					H_{CO_2}/kPa·m^3·kmol^{-1} at the following T/K				
kmol·m^{-3}	293	298	303	308	313	293	298	303	308	313
DEA										
2.0	4525	5004	5471	6104	7247	3345	3787	4018	4575	5197
2.5	4676	5101	5592	6235	7423	3456	3860	4107	4674	5323
3.0	4795	5245	5690	6394	7621	3544	3969	4179	4793	5465
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. Estimated Solubility of CO_2, H_{CO_2}, for AMP (2) + DEA (1) + H_2O (3) and MDEA (2) + DEA (1) + H_2O (3) from 293 K to 313 K Using the N_2O Analogy

mass %/ mass %	H_{N_2O}/kPa·m^3·kmol^{-1} at the following T/K					H_{CO_2}/kPa·m^3·kmol^{-1} at the following T/K				
	293	298	303	308	313	293	298	303	308	313
MDEA/DEA										
30/0	4423	4879	5205	5498	5979	3269	3692	3823	4121	4287
28.5/1.5	4462	4901	5247	5545	6061	3298	3709	3854	4156	4346
27/3	4505	4945	5297	5607	6145	3330	3742	3891	4203	4406
25.5/4.5	4547	4986	5346	5662	6297	3361	3773	3927	4244	4515
24/6	4596	5015	5399	5742	6401	3397	3795	3965	4304	4590
22.5/7.5	4643	5045	5445	5886	6545	3432	3818	3999	4412	4693
21/9	4675	5097	5502	6015	6723	3456	3857	4041	4509	4821
0/30	4802	5234	5701	6405	7625	3550	3961	4187	4801	5468
AMP/DEA										
30/0	4502	5060	5475	6095	6582	3328	3829	4021	4569	4720
28.5/1.5	4532	5103	5470	6105	6819	3350	3862	4018	4576	4890
27/3	4574	5105	5479	6125	7125	3381	3863	4024	4591	5109
25.5/4.5	4601	5135	5481	6142	7284	3401	3886	4026	4604	5223
24/6	4607	5140	5492	6167	7345	3405	3890	4034	4623	5267
22.5/7.5	4645	5175	5521	6179	7385	3434	3916	4055	4632	5296
21/9	4663	5192	5542	6198	7402	3447	3929	4071	4646	5308

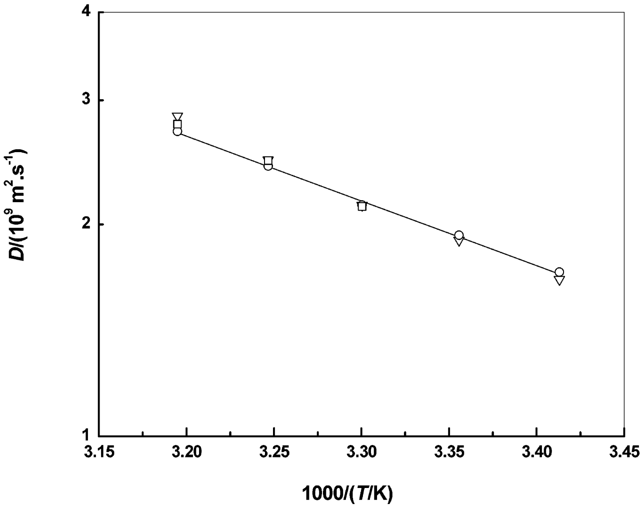

Figure 3. Diffusivity of CO_2 in water as a function of temperature: ▽, this study; ○, Al-Ghawas et al.;[8] □, Li and Lee;[14] —, calculated using eq 6.

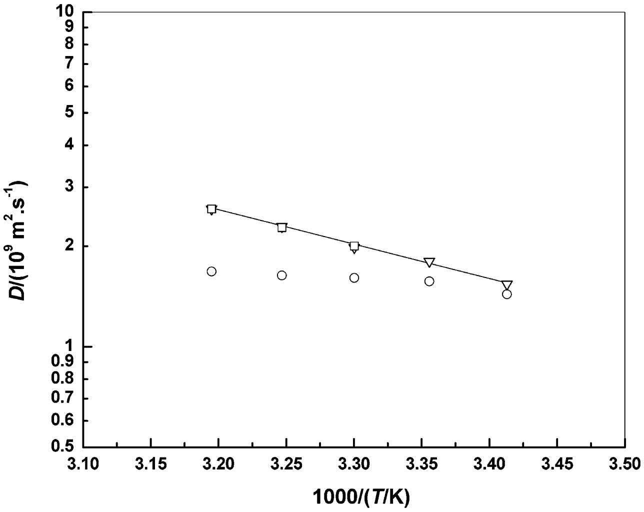

Figure 4. Diffusivity of N_2O in water as a function of temperature: ▽, this study; ○, Al-Ghawas et al.;[8] □, Li and Lee;[14] —, calculated using eq 5.

regression method. The average regression deviation between calculated values and experimental data was calculated from

$$\sigma = \frac{1}{n}\sum_{i=1}^{n}\frac{|H_{\text{m,calc}} - H_{\text{m,exp}}|}{H_{\text{m,exp}}} \tag{11}$$

Wang et al.[1] proposed the solubility of N_2O in pure amine solvent as follows.

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

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.[1] 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.[1] was adopted directly in 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 1 and 2.[7] The densities of amine aqueous solutions, required in the calculation of volume fraction, were estimated on the basis of the correlation of Hsu and Li[23] and Mandal et al.[24]

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 4. It is noted that the temperature in eq 10 is, in this study, in kelvin (K) while degree centigrade (°C) was used in the correlation of Wang et al.[1]

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

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

where subscripts i and j stand for amine and water, respectively; the 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, (MDEA + H_2O), (AMP + H_2O), and (DEA + H_2O). The remaining two-body interaction parameters between MDEA and DEA as well as AMP and DEA and the one three-body interaction parameter were regressed from the (MDEA + DEA + H_2O) and

Table 4. Parameters, k_1, k_2, k_3, and 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_{234} for the Excess Henry's Constant for the Binary and Ternary Solvent Systems

system	k_1	k_2	k_3	k_4	α_{234}	AAD %
(DEA + H_2O)	307.401 05	−2.023 024 3	0.003 341 4	0.413 234 3		0.87
(AMP + H_2O)	−29.760 815	0.236 224 8	−0.000 417 1	−1.577 179 4		0.77
(MDEA + H_2O)	81.968 179	−0.472 605 2	0.000 713 6	−3.567 293 2		0.81
(DEA + AMP)	9224.624 3	−24.041 875	0.038 887 9	−819.861 49		
(DEA + MDEA)	−144.843 78	3.014 200 2	−0.004 884 3	−38.221 012		
(DEA + AMP + H_2O)					−7669.155 0	1.25
(DEA + MDEA + H_2O)					−435.707 31	0.84

Table 5. Estimated Diffusivity of CO_2, D_{CO_2}, for AMP (2) + DEA (1) + H_2O (3) and MDEA (2) + DEA (1) + H_2O (3) from 293 K to 313 K Using the N_2O Analogy

mass %/ mass %	$D_{N_2O}/(10^9\ m^2{\cdot}s^{-1})$ at the following T/K					$D_{CO_2}/(10^9\ m^2{\cdot}s^{-1})$ at the following T/K				
	293	298	303	308	313	293	298	303	308	313
MDEA/DEA										
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	0.94	1.04	1.12	1.15	1.24	1.04	1.10	1.21	1.24	1.38
27/3	0.95	1.04	1.13	1.17	1.27	1.06	1.10	1.22	1.26	1.41
25.5/4.5	0.96	1.05	1.15	1.19	1.32	1.07	1.11	1.24	1.28	1.46
24/6	0.96	1.06	1.17	1.22	1.37	1.07	1.12	1.26	1.32	1.52
22.5/7.5	0.97	1.07	1.17	1.25	1.39	1.08	1.13	1.26	1.35	1.54
21/9	0.98	1.08	1.19	1.27	1.44	1.09	1.14	1.28	1.37	1.60
0/30	1.04	1.12	1.25	1.38	1.57	1.16	1.18	1.35	1.49	1.74
AMP/DEA										
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	0.72	0.84	1.02	1.20	1.33	0.80	0.89	1.10	1.29	1.47
27/3	0.74	0.85	1.04	1.23	1.34	0.82	0.90	1.12	1.33	1.49
25.5/4.5	0.77	0.87	1.05	1.24	1.36	0.86	0.92	1.13	1.34	1.51
24/6	0.79	0.88	1.07	1.26	1.38	0.88	0.93	1.15	1.36	1.53
22.5/7.5	0.80	0.91	1.08	1.27	1.39	0.89	0.96	1.16	1.37	1.54
21/9	0.84	0.93	1.12	1.28	1.42	0.93	0.98	1.21	1.38	1.57

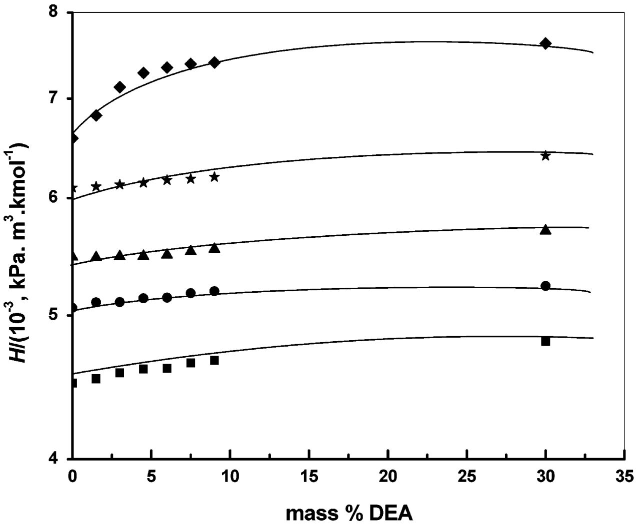

Figure 5. Henry's constant of N_2O in (AMP + DEA + H_2O) as a function of DEA concentration at different temperatures: ■, 293 K; ●, 298 K; ▲, 303 K; ★, 308 K; ◆, 313 K; —, calculated using eq 7.

(AMP + DEA + H_2O) ternary systems' solubility data. 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 comparisons between the literature values[8,14] and the values obtained in this study for the diffusivities of CO_2 and N_2O in water are shown in Figures 3 and 4. As shown in Figures 3 and 4, the diffusivities of CO_2 and N_2O in water obtained in this study are in excellent agreement with the literature values. While the measured N_2O diffusivity in water of this study and that reported by Li and Lee[14] agreed well with the diffusivity calculated from eq 5, the diffusivity values reported by Al-Ghawas et al.[8] are smaller than the calculated values using eq 5. The measured diffusivities of N_2O as well as the estimated diffusivities of CO_2 in (AMP + DEA + H_2O) and (MDEA + DEA + H_2O) are presented in Table 5. For (24 mass % MDEA + 6 mass % DEA) and 30 mass % DEA, over the temperature range (303 to 313) K, the experimental data of this study are within 3.83% deviation when compared with the experimental data of Li and Lee.[14] For (24 mass % AMP + 6 mass % DEA), over the temperature range (303 to 313) K, the deviation of the experimental data of this study is within 2.5% of the experimental data reported by Li and Lee.[14] Recently Ko et al.[25] measured the diffusivities of N_2O in aqueous single alkanolamines, MEA, DEA, DIPA, TEA, and AMP at 30, 35, and 40 °C. Although the present work reports the diffusivities of N_2O in the blended alkanolamines (MDEA + DEA + H_2O) and (AMP + DEA + H_2O), the diffusivity of N_2O in water of this work has been found to agree well with that of Ko et al.[25] Besides, the diffusivities of N_2O in 30 mass % aqueous MDEA and 30 mass % aqueous AMP have been found to agree well with those calculated from the correlation proposed by Ko et al.[25] (eq 13). The diffusivity of N_2O in 30 mass % aqueous DEA of this study is, however, about 30% higher than those calculated from the correlation proposed by Ko et al.[25] As shown in Figure 6, for the ternary system (AMP + DEA + H_2O), the N_2O diffusivity has been found to increase with the increase in concentration of DEA at constant temperature. The diffusivity also increases with increase in temperature at constant DEA concentration.

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

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

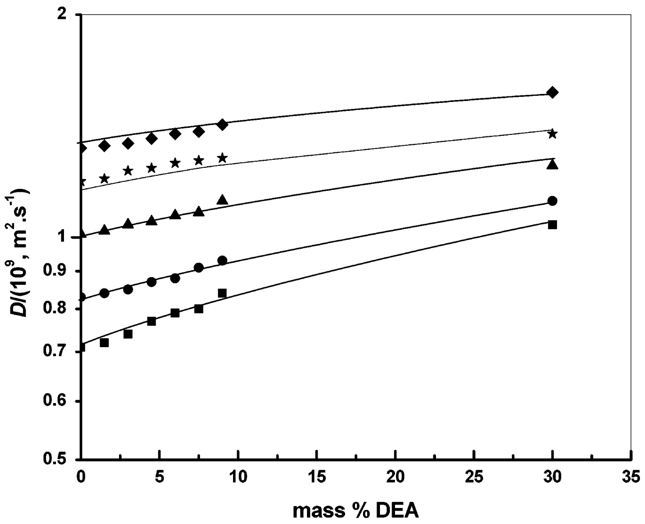

Figure 6. Diffusivity of N_2O in ($AMP + DEA + H_2O$) as a function of the DEA concentration at different of temperatures: ■, 293 K; ●, 298 K; ▲, 303 K; ★, 308 K; ◆, 313 K; —, calculated using eq 14.

Table 6. 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 ($MDEA + DEA + H_2O$)

	ternary systems	
parameter	($AMP + DEA + H_2O$)	($MDEA + DEA + H_2O$)
b_0	$4.463\,882 \times 10^{-7}$	$-5.593\,534 \times 10^{-8}$
b_{11}	$-2.713\,784 \times 10^{-7}$	$5.126\,698 \times 10^{-8}$
b_{12}	$4.124\,287 \times 10^{-8}$	$-1.056\,576 \times 10^{-8}$
b_{21}	$-2.742\,821 \times 10^{-7}$	$5.267\,597 \times 10^{-8}$
b_{22}	$4.211\,549 \times 10^{-8}$	$-1.182\,803 \times 10^{-8}$
c_{12}	$8.341\,194 \times 10^{-8}$	$-2.224\,859 \times 10^{-8}$
c	$4.149\,272 \times 10^{-3}$	$-2.939\,735 \times 10^{-3}$
AAD %	1.8	1.9

where M_1 is the molarity of DEA and M_2 is the molarity of MDEA or AMP. The parameters in eq 14 were determined using the diffusivity data of Table 5. The overall average absolute deviation (AAD%) values for the calculations of the diffusivity of N_2O in ($AMP + DEA + H_2O$) and ($MDEA + DEA + H_2O$) are 1.8 and 1.9%, respectively. The parameters are presented in Table 6.

The Stokes–Einstein relation ($D\eta/T$ = a constant, η is viscosity) has often been used to correlate the diffusivity of N_2O in aqueous single amine solutions.[6,10] The viscosity, η, values of the blended amine solutions required to calculate the Stokes–Einstein relation, that is, $D\eta/T$, were taken from Mandal et al.[24] It was observed from Tables 7 and 8 that the experimental diffusivities of N_2O in ($AMP + DEA + H_2O$) and ($MDEA + DEA + H_2O$) do not follow the Stokes–Einstein relation strictly.

Table 7. Diffusivity of N_2O in ($AMP + DEA + H_2O$) for the Stokes–Einstein Relation

T/K	mass % AMP/ mass % DEA	D_{N_2O}/10^9 $m^2 \cdot s^{-1}$	η/10^3 $N \cdot s \cdot m^{-2}$	$D\eta/T$/10^{15} $N \cdot K^{-1}$
293	30/0	0.71	4.89	11.84
	28.5/1.5	0.72	4.84	11.89
	27/3	0.74	4.82	12.17
	25.5/4.5	0.77	4.78	12.56
	24/6	0.79	4.77	12.86
	22.5/7.5	0.80	4.75	12.96
	21/9	0.84	4.72	13.53
	0/30	1.04	4.22	14.98
303	30/0	1.01	3.21	10.70
	28.5/1.5	1.02	3.18	10.70
	27/3	1.04	3.16	10.84
	25.5/4.5	1.05	3.15	10.92
	24/6	1.07	3.13	11.05
	22.5/7.5	1.08	3.12	11.12
	21/9	1.12	3.10	11.46
	0/30	1.25	2.57	10.60
313	30/0	1.32	2.24	9.446
	28.5/1.5	1.33	2.21	9.391
	27/3	1.34	2.19	9.376
	25.5/4.5	1.36	2.18	9.472
	24/6	1.38	2.16	9.523
	22.5/7.5	1.39	2.15	9.548
	21/9	1.42	2.13	9.663
	0/30	1.57	1.97	9.881

Table 8. Diffusivity of N_2O in ($MDEA + DEA + H_2O$) for the Stokes–Einstein Relation

T/K	mass % MDEA/ mass % DEA	D_{N_2O}/10^9 $m^2 \cdot s^{-1}$	η/10^3 $N \cdot s \cdot m^{-2}$	$D\eta/T$/10^{15} $N \cdot K^{-1}$
293	30/0	0.84	3.46	9.919
	28.5/1.5	0.94	3.61	11.58
	27/3	0.95	3.68	11.93
	25.5/4.5	0.96	3.79	12.42
	24/6	0.96	3.69	12.09
	22.5/7.5	0.97	3.73	11.36
	21/9	0.98	3.85	12.88
	0/30	1.04	4.22	14.98
303	30/0	1.10	2.68	9.729
	28.5/1.5	1.12	2.62	9.684
	27/3	1.13	2.59	9.659
	25.5/4.5	1.15	2.67	10.13
	24/6	1.17	2.58	9.962
	22.5/7.5	1.17	2.61	10.07
	21/9	1.19	2.69	10.56
	0/30	1.25	2.57	10.60
313	30/0	1.21	1.92	7.422
	28.5/1.5	1.24	1.85	7.329
	27/3	1.27	1.86	7.546
	25.5/4.5	1.32	1.94	8.181
	24/6	1.37	1.89	8.273
	22.5/7.5	1.39	1.86	8.260
	21/9	1.44	1.98	9.109
	0/30	1.57	1.97	9.881

Conclusions

The solubility of N_2O in (diethanolamine + water), (2-amino-2-methyl-1-propanol + water), (*N*-methyldiethanolamine + water), (*N*-methyldiethanolamine + diethanolamine + water), and (2-amino-2-methyl-1-propanol + diethanolamine + water) as well as the diffusivity of N_2O in (*N*-methyldiethanolamine + diethanolamine + water) and (2-amino-2-methyl-1-propanol + diethanolamine + 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 (diethanolamine + water), (2-amino-2-methyl-1-propanol + water), (*N*-methyldiethanolamine + water), (*N*-methyldiethanolamine + diethanolamine + water), and (2-amino-2-methyl-1-propanol + diethanolamine + water) as well as the diffusivity of CO_2 in (*N*-methyldiethanolamine + diethanolamine + water) and (2-amino-2-methyl-1-propanol + diethanolamine + 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 1.78% and 2.09%, for (*N*-methyldiethanolamine + diethanolamine + water) and (2-amino-2-methyl-1-propanol + diethanolamine + water) solutions, respectively. In the case of diffusivities of CO_2 and N_2O in (*N*-methyldiethanolamine + diethanolamine + water) and (2-amino-2-methyl-1-propanol + diethanolamine + water) solutions, the literature results have been found to be in good agreement with the results calculated from the correlation.

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