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Physical Solubility and Diffusivity of N_2O and CO_2 in Aqueous Sodium Glycinate Solutions

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Physical solubility and diffusivity of N_2O and CO_2 in pure water and of N_2O in aqueous sodium glycinate solutions of mass fraction (0.1 to 0.5) were measured at T = (303.15 to 323.15) K. Since it was not possible to measure these properties for CO_2 due to the chemical reaction between CO_2 and the amine group of sodium glycinate, these were estimated using the well-known N_2O analogy. While the physical solubility increases with a decrease in temperature and with a decrease in mass fraction, diffusivity decreases with a decrease in temperature as well as an increase in mass fraction except for concentrated solution (mass fraction 0.4 and 0.5) where it increases with a decrease in temperature.

Introduction

The increasing anthropogenic CO_2 emission and global warming have challenged the world scientists to find new and better ways to meet the world's increasing needs for energy while mitigating the global warming effect by curtailing the increase in concentration of the major greenhouse gas CO_2 in the atmosphere, mainly due to its emission from combustion of fossil fuels.[1,2] Another goal of CO_2 separation and capture is to isolate CO_2 from its many sources and its further utilization in many technological applications.[3,4] The most likely options for CO_2 separation and capture include chemical absorption, physical and chemical adsorption, gas-separation membranes, mineralization/biomineralization, and vegetation.[4−10]

Aqueous alkanolamines such as monoethanolamine (MEA), methyldiethanolamine (MDEA), and 2-amino-2-methyl-1-propanol (AMP) have been widely used chemical absorbents for removal of acid gases (CO_2, H_2S).[11−13] Especially, aqueous MEA solution has been used as an industrially important absorbent because of its rapid reaction rate, the low cost of the solvent, the thermal stability and low solubility of hydrocarbons as well as high alkalinity. However, the aqueous MEA solution has a disadvantage due to its degradation through oxidation of the amine, high enthalpy of reaction, and corrosion problems for removal acid gases.[14,15] Tertiary alkanolamines as aqueous MDEA solutions are better than primary and secondary alkanolamines such as aqueous MEA and DEA solutions as they have the properties of high loading capacity, less regeneration energy, and high resistance to thermal degradation, but the reaction rates of tertiary alkanolamines are low.[14,16,17] Sterically hindered amines such AMP and 2-piperidineethanol (PE) are definitely superior to the others because of CO_2 loading capacity, absorption rate, selectivity, and regeneration energy, but sterically hindered amines have an estimated lower reaction rate than the aqueous MEA solution.[18−20] To avoid such problems, various absorbents have been widely investigated. In the past, sodium glycinate in glycerol was used in an immobilized liquid membrane in a closed loop life support system, such as in spacecraft or space suits, for removal of carbon dioxide from the atmosphere.[21−23] This prompted us to study the aqueous sodium glycinate solution as an absorbent for CO_2. In a previous paper, we reported the physical properties of sodium glycinate solution.[24] In continuation of this, we have measured/predicted the physical solubility and diffusivity of N_2O and CO_2 in water and aqueous sodium glycinate solutions, which are necessary for the design, operation, and optimization of acid gas treatment equipment and also for measuring other chemical properties such as kinetic reaction, regeneration energy, enthalpy of reaction, and CO_2 loading.[24−27]

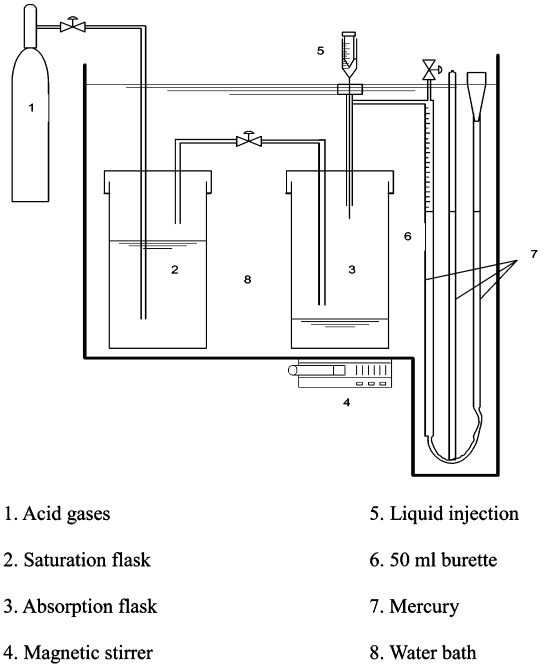

Figure 1. Schematic diagram of physical solubility apparatus.

Experimental Section

The chemical absorbent (sodium glycinate) used in this study was obtained from Sigma-Aldrich with a mass purity of > 99 %, and its aqueous solution was prepared from doubly distilled water. All solutions were prepared by mass with a balance

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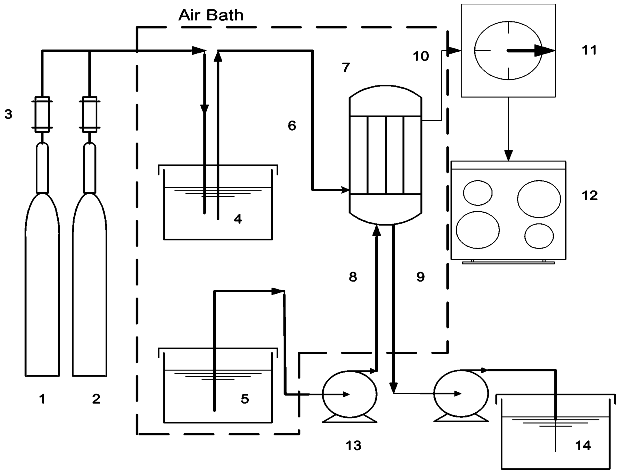

1. N_2O gas
2. CO_2 gas
3. Mass flow controller
4. Saturation flask
5. Sodium glycinate solution
6. Gas input
7. Wet-wall column
8. Absorbent input
9. Absorbent output
10. Gas output
11. Wet gas meter
12. GC-TCD
13. Pump
14. Reservoir

Figure 2. Schematic diagram of diffusivity apparatus.

precision of ± 1 × 10^{-4} g. Nitrous oxide and carbon dioxide of high purity (> 99.8 %) were used during the experiment.

Physical Solubility. Physical solubility of gas in liquids (A) can be calculated from Henry's law, which is defined as

$$H_A/(\mathrm{kPa \cdot m^3 \cdot kmol^{-1}}) = P_A(\mathrm{kPa})/C^*_A(\mathrm{kmol \cdot m^{-3}}) \qquad (1)$$

where H_A is Henry's law constant, P_A is the partial pressure in gas phase, and C^*_A is the equilibrium concentration of gas (A) absorbed by liquid. A higher Henry's constant value corresponds to a lower solubility.

The physical solubility of N_2O was measured by using the apparatus, shown schematically in Figure 1, in the manner described by Al-Ghwas et al.[27] Physical solubility was measured in a glass flask with a volume of 500 mL at atmospheric pressure. About 30 mL of solution, which was weighted with a balance precision of ± 1 × 10^{-4} g, was injected into the flask. The apparatus was kept in a water thermostat, and the temperature of bath was controlled to within ± 0.05 K. The partial pressures of N_2O in the experiments were obtained from the measured total pressure corrected for water vapor pressure using the following equation:[27]

$$P^{\mathrm{vap}}_{H_2O}/(\mathrm{kPa}) = 1.35337 \times 10^6 \exp(-5243.04/(T/\mathrm{K})) \qquad (2)$$

The experimental uncertainty in the solubility measurements was estimated to be less than ± 2 %.

Diffusivity. A schematic diagram of experimental set up was shown in Figure 2. The diffusion coefficients were measured using the wetted-wall column absorber consists of outer diameter 2.54 cm and length 10.05 cm made of 316 stainless steel in an air bath. The temperature of air thermostat was kept constant within ± 0.1 K, and the temperature of the air bath was recorded by a digital thermometer (Hanyang AT3) with an accuracy of 0.1 K. The sodium glycinate solution was made to flow using gear pumps (Cole-Parmer, model 7553-70), which were calibrated with experimental liquids of different concentrations and at different temperatures. The liquid sample distributed uniformly as a thin film on the outside of the cylinder. To prevent the ripple on the liquid surface, 0.04 % (v/v) of a surface active agent, Tween 80, was added. The effect of the surface active agent Tween 80 was negligible.[28] The absorption rate was measured by gas uptake method using a soap-film meter. The input flow rate of input gas was controlled with an accuracy of ± 0.03 % by mass flow controller (Tylon model FC-280S). The composition of exhaust gases was determined by a gas chromatograph (Hewlett-Packard, 5890 series II) having a thermal conductivity detector (TCD) and column packed with Porapak Q. The analysis conditions of the gas chromatograph were a detector temperature of 140 °C, a column temperature of 30 °C, and a carrier gas total flow rate of 75 mL·min^{-1}.

Results and Discussion

Due to the chemical reaction between amine and CO_2, it is difficult to experimentally measure the physical solubility and molecular diffusivity of CO_2 in sodium glycinate solution. It was found that the ratio of diffusivities or solubilities of CO_2 and N_2O in water is similar (within 5 %) to that in aqueous

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

T	$H^0_{N_2O}$	$H^0_{CO_2}$		T	D_{N_2O}	$D^0_{CO_2}$	
K	kPa·m^3·kmol^{-1}	kPa·m^3·kmol^{-1}	reference	K	10^9·m^2·s^{-1}	10^9·m^2·s^{-1}	reference
303.15	4450.2	3289.57	this study	298.15	1.75	1.98	this study
	4568.17	3376.78	eqs 9 and 10		1.78	1.93	eqs 11 and 12
	4349.88	3394.38	27		1.57	1.93	27
	4835	3428	34		1.80	1.90	33
	4649.4	3391.7	32		1.68	1.98	35
	4512	3314	33	303.15	2.09	2.15	this study
313.15	5504.92	3965.96	this study		2.03	2.16	eqs 11 and 12
	5810.73	4186.28	eqs 9 and 10		1.61	2.16	27
	5020.65	4249.57	27		2.04	2.17	32
	6108	4306	34	313.15	2.46	2.83	this study
	5600.2	3983.3	32		2.61	2.71	eqs 11 and 12
	5715	4098	33		1.68	2.71	27
323.15	7184.98	5053.03	this study		2.58	2.86	33
	7282.04	5121.29	eqs 9 and 10		2.55	2.80	35
	5369.21	5166.56	27		2.66	2.76	32
	7445	5234	34	323.15	3.19	3.35	this study
					3.30	3.34	eqs 11 and 12
				323	1.89	3.34	27
				322.7	2.85	-	31

Table 2. Physical Solubility and Diffusivity of N_2O and CO_2 in Aqueous Sodium Glycinate (SG) Solution

T	SG	H_{N_2O}	H_{CO_2}	D_{N_2O}	D_{CO_2}
K	$kmol \cdot m^{-3}$	$kPa \cdot m^3 \cdot kmol^{-1}$	$kPa \cdot m^3 \cdot kmol^{-1}$	$10^9 \cdot m^2 \cdot s^{-1}$	$10^9 \cdot m^2 \cdot s^{-1}$
303.15	1.065	4788.19	3539.42	1.513	1.610
	2.226	4977.19	3679.13	1.424	1.515
	3.466	5043.56	3728.18	1.373	1.462
	4.78	5181.73	3830.33	1.289	1.372
	6.217	5327.70	3938.22	1.224	1.269
313.15	1.061	5833.57	4202.74	1.902	1.972
	2.218	6107.02	4399.74	1.711	1.773
	3.455	6797.31	4897.10	1.478	1.531
	4.764	7042.32	5073.57	1.198	1.242
	6.196	7663.61	5521.24	0.886	0.896
323.15	1.056	7695.46	5386.72	2.178	2.202
	2.209	7882.55	5543.61	2.092	2.115
	3.440	8119.02	5709.92	1.817	1.838
	4.057	8457.32	5947.83	1.344	1.393
	6.170	9332.21	6563.13	0.713	0.721

Table 3. Regression Parameters of Equation 13 and AADs

w_{SG}	A_1	A_2	A_3	AAD %
		N_2O Physical Solubility		
0.1	509109.43	−300627.37	44788.25	$6.2\ e^{-5}$
0.2	425285.83	−248026.64	36563.13	$9.5\ e^{-5}$
0.3	−98276.13	80770.73	−14990.07	$1.8\ e^{-4}$
0.4	−97703.69	81544.47	−15264.51	$2.9\ e^{-4}$
0.5	−180686.45	137410.33	−24560.57	$5.4\ e^{-5}$
		CO_2 Physical Solubility		
0.1	−168963.34	121990.41	−21091.28	$5.3\ e^{-4}$
0.2	−102452.52	77620.93	−13763.03	$6.1\ e^{-4}$
0.3	−101036.67	75960.97	−13399.34	$4.9\ e^{-4}$
0.4	−102452.52	77620.93	−13763.03	$7.7\ e^{-6}$
0.5	−168963.34	121990.41	−21091.28	$2.8\ e^{-4}$
		N_2O Diffusivity		
0.1	−30.4507	23.4911	−4.1837	0.063
0.2	71.2972	−40.2659	5.7853	0.058
0.3	132.11	−79.5507	12.1011	0.054
0.4	119.7125	−73.8723	11.5112	0.059
0.5	64.2016	−42.1081	6.9773	0.021
		CO_2 Diffusivity		
0.1	−42.2787	30.5774	−5.2361	$4.1\ e^{-4}$
0.2	64.0635	−36.0343	5.1757	$5.3\ e^{-4}$
0.3	131.7546	−79.6277	12.1652	$4.0\ e^{-4}$
0.4	140.2143	−86.8367	13.5649	$2.5\ e^{-4}$
0.5	79.1531	−51.6362	8.4959	$5.3\ e^{-4}$

solution of organic compounds having an amine group because of somewhat similar molecular volume and electronic structure of CO_2 and N_2O. This is known as N_2O analogy[29] and used by many workers for estimation of molecular diffusivity and physical solubility of CO_2 in various solutions containing amine.[12,27,28,30−33] Thus

$$H_{CO_2}/H_{N_2O} = H^0_{CO_2}/H^0_{N_2O} \tag{3}$$

and

$$D_{CO_2}/D_{N_2O} = D^0_{CO_2}/D^0_{N_2O} \tag{4}$$

where H and D represent the physical solubility and diffusivity of gases in aqueous solution of same concentration, and H^0 and D^0 are the same in pure water, respectively.

According to the Higbie penetration theory, the absorption rate of sparingly soluble gas at short contact time with a degassed liquid is related to the diffusion coefficient by the following relation:

$$D_{N_2O}/(m^2 \cdot s^{-1}) = \left(\frac{N_{N_2O}H_{N_2O}}{2P_{N_2O}}\right)^2 \pi t_c \tag{5}$$

where t_c is the contact time and can be computed from the wetted wall column hydrodynamics using the relation

$$t_c/(s) = \frac{2h}{3}\left(\frac{\pi d_s}{L}\right)^{2/3}\left(\frac{3\eta}{\rho g}\right)^{1/3} \tag{6}$$

where

$$d_s = d + (3\eta L/\pi\rho d) \tag{7}$$

In these equations, N_{N_2O}, H_{N_2O}, and P_{N_2O} are the absorption rate of gas in liquid per unit area, Henry's law constant, and partial pressure of gas, respectively. The parameters d, η, ρ, and h are the outside diameter, viscosity, density of absorber, and height of the wetted surface, respectively. N_{N_2O} is calculated from the total absorption rate from

$$N_{N_2O}/(kmol \cdot m^{-2} \cdot s^{-1}) = N^T_{N_2O}/\pi d_s h \tag{8}$$

The experimental uncertainty in the diffusivity was ± 2 %. The experimental data for physical solubility and diffusivity of CO_2 and N_2O in water ($H^0_{CO_2}$, $H^0_{N_2O}$, $D^0_{CO_2}$, $D^0_{N_2O}$) were recorded in Table 1 along with the values estimated from eqs 4 to 7 proposed by Versteeg and Swaaij:[31]

$$H^0_{CO_2}/(kPa \cdot m^3 \cdot kmol^{-1}) = 2.8249 \times 10^6 \times \exp(-2044/(T/K)) \tag{9}$$

$$H^0_{N_2O}/(kPa \cdot m^3 \cdot kmol^{-1}) = 8.5470 \times 10^6 \times \exp(-2284/(T/K)) \tag{10}$$

$$D^0_{CO_2}/(m^2 \cdot s^{-1}) = 2.35 \times 10^{-6} \times \exp(-2119/(T/K)) \tag{11}$$

$$D^0_{N_2O}/(m^2 \cdot s^{-1}) = 5.07 \times 10^{-6} \times \exp(-2371/(T/K)) \tag{12}$$

It was found that our experimental values for $H^0_{CO_2}$, $H^0_{N_2O}$, $D^0_{CO_2}$, and $D^0_{N_2O}$ compared well with the values estimated from eqs 9 to 12 and these were also in good agreement with the previously reported values in the literature[27,32−35] (Table 1). This indicates that the experimental apparatus and the procedure used in this work were precise enough for performing such measurements.

After the experimental measurements of H_{N_2O}, D_{N_2O}, $H^0_{CO_2}$, $H^0_{N_2O}$, $D^0_{CO_2}$, and $D^0_{N_2O}$ in different concentration of sodium glycinate solution or water at T = (303.15 to 323.15) K, physical solubility of CO_2 (H_{CO_2}), and diffusivity of CO_2 (D_{CO_2}) in aqueous sodium glycinate solutions of mass fraction (0.1 to 0.5) at T = (303.15 to 323.15) K was estimated by using the N_2O analogy (eqs 3 and 4) for physical solubility and diffusivity.[31] Such H_{N_2O}, D_{N_2O}, H_{CO_2}, and D_{CO_2}values for different concentrations of sodium glycinate solution at all temperatures are reported in Table 2 and shown graphically in Figures 3 to 6. We are unaware of any previously published data on the physical solubility and diffusivity of N_2O and CO_2 in aqueous sodium glycinate solutions with which to compare our results.

The measured and estimated properties were then regressed using the following equation:

$$Y = A_1 + A_2(T/K) + A_3(T/K)^2 \tag{13}$$

where Y is the physical solubility or diffusivity of N_2O and CO_2

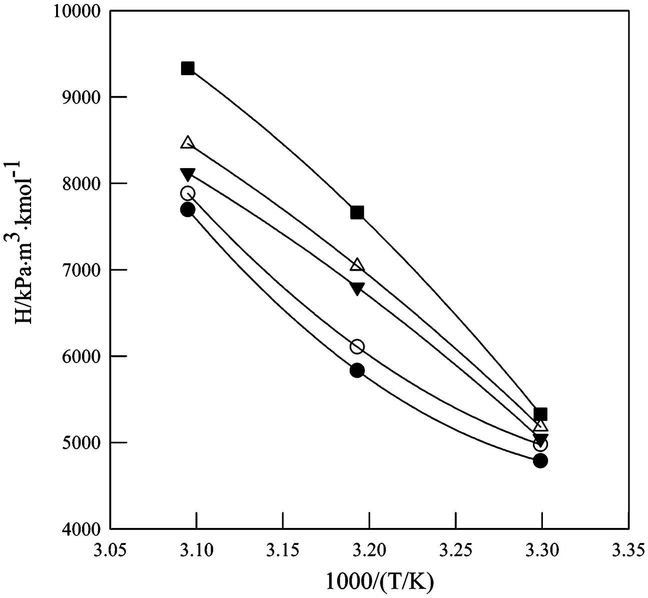

Figure 3. Solubility of N_2O in aqueous sodium glycinate solution of different mass fraction: ●, 0.1; ○, 0.2; ▼, 0.3; □, 0.4; ■ 0.5; —, eq 13.

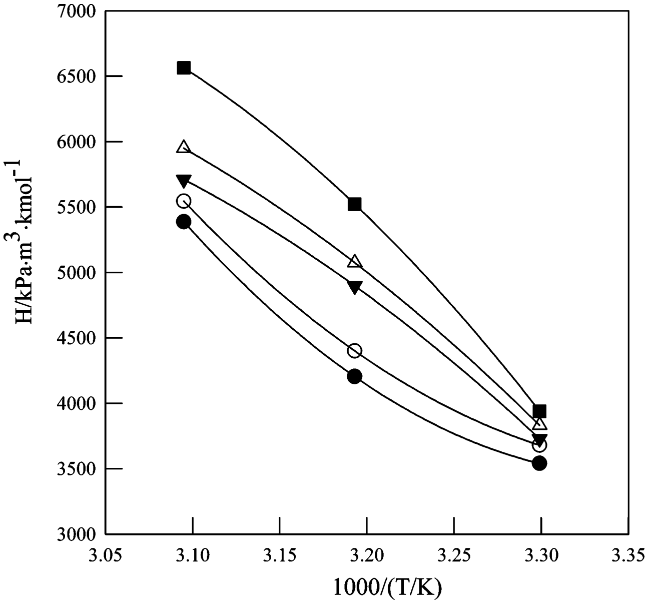

Figure 4. Solubility of CO_2 in aqueous sodium glycinate solution of different mass fraction: ●, 0.1; ○, 0.2; ▼, 0.3; □, 0.4; ■ 0.5; —, eq 13.

in aqueous sodium glycinate solutions, and A_i (i = 1 to 3) is the regression parameters calculated by least-squares method and reported in Table 3 along with average absolute deviations (AADs) between calculated and measured values.

Variation of Henry's law constant of N_2O and CO_2 in aqueous sodium glycinate solutions of various mass fractions (0.1 to 0.5) with temperature are shown in Figures 3 and 4, respectively. A higher Henry's constant value corresponds to a lower solubility. It was found that while physical solubility decreases with increase in sodium glycinate mass fraction, it increases with decrease in temperature. The diffusivity of N_2O and CO_2 in aqueous sodium glycinate solution of various mass fractions (0.1 to 0.5) at different temperatures was plotted in Figures 5 and 6, respectively. It was found that diffusivity decreases with increase in sodium glycinate mass fraction and decrease is more severe at higher temperature. In case of variation with temperature, while diffusivity decreases with a decrease in temperature from mass fraction 0.1 to 0.3, it starts increasing with a decrease in temperature for higher mole fraction. The increase with rise in temperature is prominent at mass fraction 0.5.

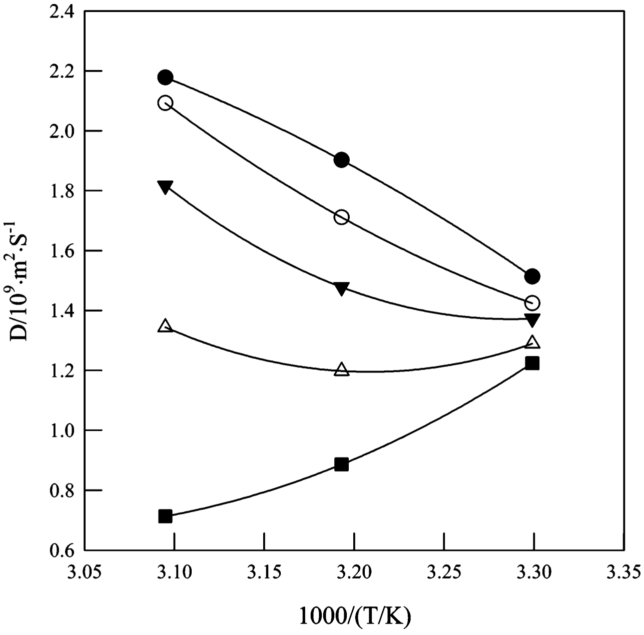

Figure 5. Diffusivity of N_2O in aqueous sodium glycinate solution of different mass fraction: ●, 0.1; ○, 0.2; ▼, 0.3; □, 0.4; ■ 0.5; —, eq 13.

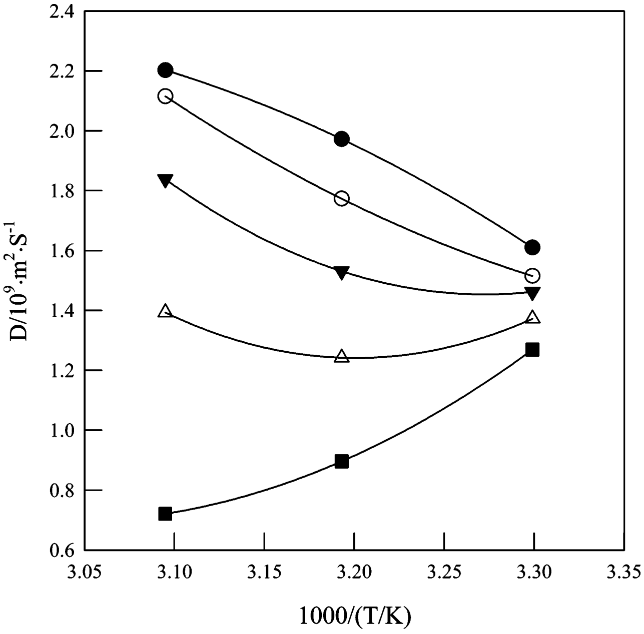

Figure 6. Diffusivity of CO_2 in aqueous sodium glycinate solution of different mass fractions: ●, 0.1; ○, 0.2; ▼, 0.3; □, 0.4; ■ 0.5; —, eq 13.

The physical solubility and diffusivity of N_2O in sodium glycinate solution at 303.15 K was also found to be higher (Figures 7 and 8) than those reported[16,20,27] for other absorbents such as 2-amino-2-methyl-1,3-propanediol and MDEA. The similar trend would also be expected for CO_2.

Conclusions

We studied the physical solubility and diffusivity of N_2O and CO_2 with aqueous sodium glycinate solutions of various mass fractions at different temperature. The physical solubility and diffusivity of N_2O and CO_2 in pure water was found to be in agreement with the literature data. Also, physical solubility decreases with an increase in sodium glycinate mass fraction and with an increase in temperature, while diffusivity decreases

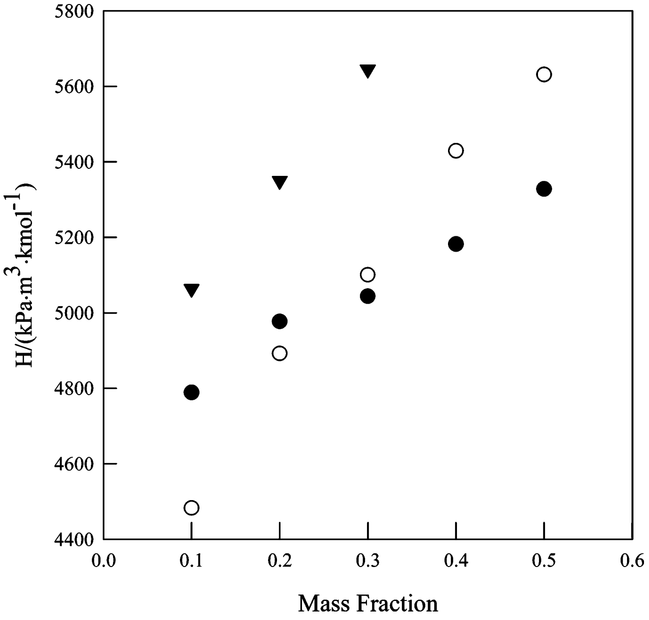

Figure 7. Solubility of N_2O in aqueous absorbent at T = 303.15 K: ●, sodium glycinate; ○, MDEA, Al-Ghawas et al.;[27] ▼, AEPD, Baek et al.[20]

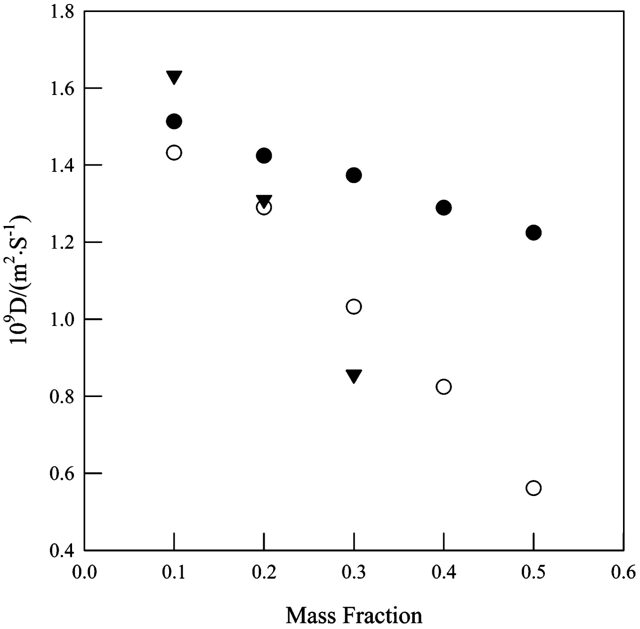

Figure 8. Diffusivity of N_2O in aqueous absorbent at T = 303.15 K: ●, sodium glycinate; ○, MDEA, Al-Ghawas et al.;[27] ▼, MDEA, Rinker et al.[16]

linearly with an increase in sodium glycinate mass fraction and increases with a decrease in temperature. These results could be applied in the basic absorption process such as design of acid gas treatment, reaction rate constant, and kinetics of the absorption reaction for acid gas removal in future work.

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