Physicochemical Properties of Aqueous Solutions of 2-Amino-2-hydroxymethyl-1,3-propanediol†

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The density and viscosity of aqueous solutions of 2-amino-2-hydroxymethyl-1,3-propanediol (AHPD) as well as the Henry's law constants and diffusivity of N_2O in these solutions were measured over the temperature range from (298 to 323) K at atmospheric pressure. The mass fractions of AHPD used were (2.17 to 21.7) %. The "N_2O analogy" was used to estimate the Henry's law constants and diffusivity of CO_2 in aqueous AHPD solutions. The experimental density, viscosity, Henry's law constants, and diffusivity data are correlated as a function of temperature and concentration of amine and compared with the available data in the literature.

Introduction

The removal of acid gases is an important process in many presently favored chemical and power producing industries. Particularly, the removal of CO_2 is undoubtedly the technology for mitigating greenhouse gas emissions from existing fossil fuel-fired electric power plants. In the extensive field of gas treating, alkanolamines have become one of the most important classes of chemicals for the removal of CO_2. Sterically hindered amines have been proposed as commercially attractive new solvents for acid gas treating over commercial amines.[1−8] The advantages of these sterically hindered amines are mainly due to their particular structure which results in increased cyclic and thermodynamic capacity for CO_2 absorption. It is reported that due to the instability the hindered amine carbamates readily undergo hydrolysis forming bicarbonate and releasing free amine that again reacts with CO_2 leading to a stoichiometric loading capacity of 1 mol of CO_2 per mole of amine with appreciable rate of absorption.[1,4] Knowledge of the thermophysical properties of the alkanolamines, for example, density, viscosity, and Henry's law constant, and diffusivity of CO_2 in these solutions is essential for the process design of gas treating units and the design of the gas treating equipments. This is also important for the mass transfer rate modeling of absorption and regeneration because these properties influence the values of the liquid side mass transfer coefficients, k_L. However, despite the immense importance of these data, few results are available in the open literature.[9,10] Park et al.[9] studied the densities and viscosities of aqueous solutions of AHPD over the temperature range from (303.15 to 343.15) K and the AHPD mass fractions of (5, 10, 15, 20, and 25) %. Tourneux et al.[10] measured the densities and viscosities of aqueous AHPD solutions and the Henry's law constant of N_2O in these solutions over the temperature range from (303.15 to 343.15) K and the AHPD mass fractions of 0.0015, 0.005, 0.01, and 0.025. They estimated the Henry's law constant of CO_2 in aqueous AHPD solutions using N_2O analogy. However, to the best of our knowledge, the diffusivity of N_2O and CO_2 in aqueous AHPD solutions has not yet been published in the open literature so far, and also for the other physicochemical properties no correlation has been reported.

In this work, the density and viscosity of aqueous solutions of AHPD as well as the Henry's law constants and diffusivity of N_2O in these solutions were measured over the temperature range from (298 to 323) K at atmospheric pressure. The mass fractions of AHPD used were (2.17 to 21.7) %. N_2O analogy is applied to estimate the Henry's law constants and diffusivity of CO_2 in (AHPD + H_2O) solutions. All the experimental data are correlated as a function of temperature and concentration of amine.

Because CO_2 undergoes chemical reaction with these solvents, the Henry's law constant and diffusivity of CO_2 in amine solutions cannot be measured directly. As a result, one must use a nonreacting gas such as N_2O as a surrogate to CO_2 in estimating the Henry's law constant and diffusivity of CO_2 in these solvents. The "N_2O analogy"[11] for estimating the Henry's law constant and diffusivity of CO_2 in amine solutions is given by eqs 1 and 2, respectively, as follows.

$$k_{H,CO_2-Am} = k_{H,N_2O-Am}\left(\frac{k_{H,CO_2-water}}{k_{H,N_2O-water}}\right) \quad (1)$$

$$D_{CO_2-Am} = D_{N_2O-Am}\left(\frac{D_{CO_2-water}}{D_{N_2O-water}}\right) \quad (2)$$

where k_{H,N_2O-Am} and D_{N_2O-Am} are the Henry's law constant and diffusivity of N_2O in amine solutions, respectively.

Experimental

Material. Reagent grade AHPD (> 99.8 % pure) was obtained from Merck and used without further purification. Distilled water degassed by boiling was used for making the amine solutions. The amine contents of the solutions were determined by acidimetric titration with standard HCl using an autotitrator (DL 50, Mettler Toledo). The nitrous oxide (> 99.9 % pure) and carbon dioxide (> 99.9 % pure) used for the Henry's law constant and diffusivity studies were obtained from Assam Air Products, India.

Density and Viscosity. The densities of the amine solutions were measured using a $26.76 \cdot 10^{-6}$ m^3 Gay-Lussac pycnometer. The pycnometer containing the amine solution was immersed

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in a constant-temperature bath. The bath temperature was controlled within ± 0.3 K of the temperature range using a circulator temperature controller (RW 2025G, Jeio Tech). Once the solution reached the desired temperature, it was weighed to within ± 0.0002 g with an analytical balance (AND GR-200). Each reported density data was the average of three measurements. The experimental uncertainty in the measured density was estimated to be ± $3.5 \cdot 10^{-5}$ g·cm^{-3}.

The viscosity was measured using an Ostwald viscometer. The viscometer was immersed in a thermostatted bath. The bath temperature was controlled within ± 0.3 K of the desired level using a circulator temperature controller (RW 2025G, Jeio Tech). Each reported value was the average of three measurements. The experimental uncertainty was estimated to be ± 1.0 %. The validity of the procedure and apparatus for density and viscosity measurements was reported in our previous publications.[12−14]

Henry's Law Constant. The method for measuring Henry's law constants of N_2O is to bring a known volume of liquid into contact with gas in a closed system at constant temperature and pressure. A $6.5 \cdot 10^{-4}$ m^3 glass flask was used as the equilibrium absorption cell. The temperature of the equilibrium cell was controlled within ± 0.3 K of the desired level with a circulating temperature controller operated on external control mode (RW 2025G, Jeio Tech). All measurements were done at atmospheric pressure. A precise manomatric device was employed to maintain atmospheric pressure in the cell throughout each experimental run. The total pressure was measured for each run within an accuracy of ± 0.2 kPa. The procedures of the Henry's law constant measurements are the same as those described by Mandal et al.[15,16] The estimated uncertainties in the measured Henry's law constant were estimated to be ± 1.5 %. The reproducibility between the various experiments was within ± 1 %.

Diffusivity. The diffusivity of N_2O in aqueous AHPD solutions was measured using a wetted-wall column absorber similar to the one used by Mandal et al.[15,16] with the outside diameter of $2.81 \cdot 10^{-2}$ m. The length of the absorption surface was maintained at $8 \cdot 10^{-2}$ m. The temperature of absorption was controlled within ± 0.2 K of the desired level with a circulating temperature controller (Jeio Tech, RW-2025G). A jacketed glass soap film storage was used to store the saturated and thermostatted gas. The liquid film and the gas phase temperature were controlled by circulating thermostatted water. The gas absorption rate was measured by the volume uptake method using a soap film meter. The pressure in the absorption chamber was about 100 kPa. The experimental uncertainty in the measured diffusivity was estimated to be ± 2.0 %. Reproducibility between the various experiments was within ± 1 %.

Results and Discussion

Density and Viscosity. The measured densities and viscosities of solutions of (AHPD + H_2O) are presented in Figures S1 and S2 (Supporting Information). w_1 is the mass fraction of individual amine present in the solution. The densities and viscosities of the solutions increase with mass fraction of AHPD in the solutions. The density and viscosity measurements of this study are in good agreement with the literature results. For the aqueous solution of mass fraction of 10.3 % AHPD over the temperatures of (298 to 313) K, the experimental density and viscosity data of this study show 0.18 % and 2.65 % deviations, respectively from the experimental data of Tourneux et al.[10] For the aqueous solution of mass fractions of (5.10 to 15.8) % AHPD over the temperatures of (303 to 323) K, the experimental density and viscosity results of this study show 0.32 % and 2.96 % deviations from the experimental data of Park et al.[9]

Table 1. Parameters and AAD for Density Correlation for AHPD (1) + H_2O (2)

	A_i	B_i	C_i
0	24.376	7.0798	−0.0123
1	19.715	8.3926	−0.0102
2	12.377	4.5610	−0.1178
100 AAD[a]		0.04	

[a] 100 AAD = $(1/N)\sum_{i=1}^{N} (|\rho_{exptl,i} - \rho_{calcd,i}|)/(\rho_{exptl,i})$.

Table 2. Parameters and AAD for Viscosity Correlation for AHPD (1) + H_2O (2)

a	2.2234
b	0.5467
c	0.2032
h	−1792.3
100 AAD[a]	2.06

[a] 100 AAD = $(1/N)\sum_{i=1}^{N} (|\eta_{exptl,i} - \eta_{calcd,i}|)/(\eta_{exptl,i})$.

The experimental density and viscosity data are correlated as a function of amine concentration and temperature using eqs 3 and 4, respectively.

$$\rho \cdot 10^3/\mathrm{g \cdot cm^{-3}} = \sum_{i=0}^{2} [A_i x^i + B_i x^i (T/\mathrm{K}) + C_i x^i (T/\mathrm{K})^2] \quad (3)$$

$$\eta \cdot 10^3/\mathrm{mPa \cdot s} = (a + bM + cM^2)\exp(-h/(T/\mathrm{K})) \quad (4)$$

where x is the mole fraction and M is the molarity of amine in the solutions. For convenience, we have considered AHPD as the first component and H_2O as the second component in the mixtures. The determined parameters for density and viscosity are obtained by regression analysis of the experimental data of this work and are presented in Tables 1 and 2, respectively. The calculated densities and viscosities from the correlation (eqs 3 and 4) are in excellent agreement with the experimental data, the average absolute deviation between the correlated and the experimental density and viscosity data for the (AHPD + H_2O) systems being about 0.04 % and 2.06 %, respectively, for 20 data points. The measured and calculated densities and viscosities from the correlation (eqs 3 and 4) are also compared in Figures S1 and S2 (Supporting Information).

Henry's Law Constant. To confirm the validity of the experimental method and the apparatus used for the Henry's law constant measurements, the Henry's law constants of CO_2 and N_2O in water were measured at (298, 303, 313, and 323) K. The results are compared with the literature data[16−19] in Figures S3 and S4 (Supporting Information). There is a good agreement between the literature results and the results of the present study, thus supporting the applicability of the present experimental method and confirming the authenticity of the experimental data. The results of the Henry's law constant of CO_2 and N_2O in water have been correlated as a function of temperature by eqs 5 and 6 as follows.

$$k_{H,CO_2-water}/\mathrm{kPa \cdot m^3 \cdot kmol^{-1}} = 3.00 \cdot 10^6 \cdot \exp(-2060/(T/\mathrm{K})) \quad (5)$$

$$k_{H,N_2O}/\mathrm{kPa \cdot m^3 \cdot kmol^{-1}} = 1.04 \cdot 10^7 \exp(-2350/(T/\mathrm{K})) \quad (6)$$

While the measured Henry's law constant of N_2O in water reported by Mandal et al.,[16] Versteeg and van Swaaij,[18] and

Table 3. Estimated Solubility of CO_2, H_{CO_2}, in AHPD (1) + H_2O (2) from (298 to 323) K Using the N_2O Analogy

	H_{N_2O}/(kPa·m³·kmol⁻¹)				H_{CO_2}/(kPa·m³·kmol⁻¹)			
100 w_1	T = 298 K	T = 303 K	T = 313 K	T = 323 K	T = 298 K	T = 303 K	T = 313 K	T = 323 K
2.17	4183	4672	6126	7784	3170	3535	4417	5527
5.10	4225	4730	6230	7950	3202	3579	4492	5644
10.3	4324	4851	6440	8123	3277	3670	4643	5767
15.8	4473	5048	6672	8301	3390	3820	4810	5894
21.7	4647	5263	6791	8489	3522	3982	4896	6027

Table 4. Parameters and AAD for N_2O Solubility Correlation for AHPD (1) + H_2O (2)

a	$1.4838 \cdot 10^7$
b	$1.0143 \cdot 10^6$
c	−24868
h	2443.1
100 AAD[a]	0.82

[a] 100 AAD = $(1/N)\sum_{i=1}^{N} (|H_{exptl,i} - H_{calcd,i}|)/(H_{exptl,i})$.

Li and Lai[19] agreed well with the Henry's law constant calculated from eq 6, the Henry's law constant values reported by Al-Ghawas et al.[17] are smaller than the calculated values using eq 6.

The data for the Henry's law constant of N_2O and the estimated Henry's law constant of CO_2 using the N_2O analogy in aqueous solutions of AHPD for the temperatures (298, 303, 313, and 323) K are presented in Table 3. Henry's constant of N_2O in aqueous AHPD increases with an increase in temperature and increases with the mass percent of AHPD in the mixture. The Henry's law constant measurements of this study are in good agreement with the literature results. For the aqueous solution of mass fraction of 10.3 % AHPD over the temperatures of (298 and 313) K, the experimental Henry's law constant data of this study show 2.3 % deviation from the experimental data of Tourneux et al.[10]

The experimental Henry's law constant data of N_2O in (AHPD + H_2O) are correlated as a function of amine concentration and temperature using a similar equation as eq 4.

$$k_{H,N_2O}/\text{kPa}\cdot\text{m}^3\cdot\text{kmol}^{-1} = (a + bM + cM^2)\exp(-h/(T/\text{K})) \quad (7)$$

The calculated parameters and average absolute deviation (AAD) are listed in Table 4 for the (AHPD + H_2O) systems. The calculated Henry's law constants from eq 7 are in good agreement with the experimental results of this study. The average absolute deviation between the correlated and experimental data for (AHPD + H_2O) is 0.82 % for 20 data points. The measured and calculated Henry's law constants from the correlation (eq 7) are compared in Figure S5 (Supporting Information).

Diffusivity. To confirm the validity of the experimental method and the apparatus used for the diffusivity measurements, the diffusivites of CO_2 and N_2O in water were measured at (298, 303, 313, and 323) K. The results are compared with the literature data[16−19] in Figures S6 and S7 (Supporting Information). There is a good agreement between the literature results and the results of the present study. The results of the diffusivity of CO_2 and N_2O in water have been correlated as a function of temperature by eqs 8 and 9 as follows.

$$D_{CO_2-water}/\text{m}^2\cdot\text{s}^{-1} = 10.75 \cdot 10^{-6} \exp(-2579/(T/\text{K})) \quad (8)$$

$$D_{N_2O-water}/\text{m}^2\cdot\text{s}^{-1} = 7.12 \cdot 10^{-6} \exp(-2485/(T/\text{K})) \quad (9)$$

While the measured N_2O diffusivities in water reported by Mandal et al.[16] and Versteeg and van Swaaij[18] agreed well with the diffusivity calculated from eq 9, the diffusivity values reported by Al-Ghawas et al.[17] are smaller than the calculated values using eq 9.

The data for the diffusivity of N_2O and the estimated diffusivity of CO_2 using the N_2O analogy in aqueous solutions of AHPD for the temperatures (298, 303, 313, and 323) K are presented in Table 5. Diffusivity of N_2O in aqueous AHPD increases with an increase in temperature and decrease in the mass percent of AHPD in the mixture. The experimental diffusivity data are correlated as a function of amine concentration and temperature using the following polynomial equation.

$$\ln D_{N_2O} = \sum_{i=0}^{2} [A_i x^i + B_i x^i (T/\text{K}) + C_i x^i (T/\text{K})^2] \quad (10)$$

The calculated parameters and average absolute deviation are listed in Table 6 for the (AHPD + H_2O) systems. The calculated diffusivities from eq 10 are in good agreement with the experimental results of this study. The average absolute deviation between the correlated and experimental data for (AHPD + H_2O) is 1.85 % for 20 data points. The measured and calculated diffusivities from the correlation (eq 10) are compared in Figure S8 (Supporting Information).

The Stokes−Einstein relation ($D_{N_2O}\eta/T$ = constant) has often been used to correlate the diffusivity of N_2O in aqueous solutions of alkanolamines.[20,21] The viscosity, η, of the AHPD solutions, required for calculation of diffusivity using the Stokes−Einstein equation, was measured in the present study. It is observed from Table 7 that the experimental diffusivities of N_2O in (AHPD + H_2O) do not follow the Stokes−Einstein relation strictly.

Conclusion

The density and viscosity of aqueous AHPD solutions and the Henry's law constant and diffusivity of N_2O in these solutions were measured over the temperature range from (298 to 323) K at atmospheric pressure. The mass fractions of AHPD

Table 5. Estimated Diffusivity of CO_2, D_{CO_2}, in AHPD (1) + H_2O (2) from (298 to 323) K Using the N_2O Analogy

	$D_{N_2O}\cdot 10^9$/(m²·s⁻¹)				$D_{CO_2}\cdot 10^9$/(m²·s⁻¹)			
100 w_1	T = 298 K	T = 303 K	T = 313 K	T = 323 K	T = 298 K	T = 303 K	T = 313 K	T = 323 K
2.17	1.55	1.75	2.22	2.70	1.66	1.88	2.61	3.16
5.10	1.34	1.6	2.12	2.63	1.43	1.72	2.49	3.08
10.3	1.19	1.38	1.89	2.34	1.27	1.48	2.22	2.74
15.8	1.06	1.27	1.76	2.25	1.13	1.36	2.07	2.64
21.7	0.95	1.20	1.69	2.22	1.01	1.29	1.99	2.60

Table 6. Parameters and AAD for N_2O Diffusivity Correlation for AHPD (1) + H_2O (2)

	A_i	B_i	C_i
0	−10.826	−11193	925.72
1	−2484.7	−3241.6	−1444.1
2	$-0.3559 \cdot 10^{-2}$	0.3352	−2.2512
100 AAD[a]	1.85		

[a] $100\text{AAD} = (1/N)\sum_{i=1}^{N} (|D_{\text{exptl},i} - D_{\text{calcd},i}|)/(D_{\text{exptl},i})$.

Table 7. Diffusivity of N_2O in AHPD (1) + H_2O (2) and Verification of the Stokes−Einstein Relation

T/(K)	w_1	$D_{N_2O} \cdot 10^9$ / ($m^2 \cdot s^{-1}$)	η / (mPa·s)	$(D_{N_2O}\eta)/(T) \cdot 10^{15}$ / ($m^2 \cdot Pa \cdot K^{-1}$)
298	2.17	1.55	0.913	4.751
	5.10	1.34	0.995	4.473
	10.3	1.19	1.148	4.584
	15.8	1.06	1.353	4.814
	21.7	0.95	1.605	5.115
303	2.17	1.75	0.874	5.051
	5.10	1.6	0.940	4.963
	10.3	1.38	1.059	4.822
	15.8	1.27	1.223	5.128
	21.7	1.20	1.449	5.737
313	2.17	2.22	0.733	5.197
	5.10	2.12	0.799	5.413
	10.3	1.89	0.884	5.340
	15.8	1.76	1.008	5.669
	21.7	1.69	1.172	6.327
323	2.17	2.70	0.600	5.016
	5.10	2.63	0.650	5.293
	10.3	2.34	0.711	5.149
	15.8	2.25	0.808	5.629
	21.7	2.22	0.930	6.391

used were (2.17 to 21.7) %. N_2O analogy is applied to estimate the Henry's law constant and diffusivity of CO_2 in (AHPD + H_2O) solutions. All the experimental data are correlated as a function of temperature and concentration of amine. The calculated results from the correlations were found in excellent agreement with the experimental data.

Supporting Information Available:

Figure S1 shows the variation of density of (AHPD + H_2O) solutions at various temperatures. Figure S2 shows the variation of viscosity of (AHPD + H_2O) solutions at various temperatures. Figure S3 shows the variation of Henry's law constant of CO_2 in water as a function of temperature. Figure S4 shows the variation of Henry's law constant of N_2O in water as a function of temperature. Figure S5 shows the variation of Henry's law constant of N_2O in (AHPD + H_2O) solutions at various temperatures. Figure S6 shows the variation of diffusivity of CO_2 in water as a function of temperature. Figure S7 shows the variation of diffusivity of N_2O in water as a function of temperature. Figure S8 shows the variation of diffusivity of N_2O in (AHPD + H_2O) solutions at various temperatures. This material is available free of charge via the Internet at http://pubs.acs.org.

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