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Diffusion Coefficients of N₂O in Aqueous Piperazine Solutions Using the Taylor Dispersion Technique from (293 to 333) K and (0.3 to 1.4) mol·dm⁻³

Espen S. Hamborg,^{*,†,||} Peter W. J. Derks,[‡] Sascha R. A. Kersten,[§] John P. M. Niederer,[‡] and Geert F. Versteeg[‡]

University of Twente, Faculty of Science and Technology, P.O. Box 217, 7500 AE Enschede, The Netherlands, and Procede Group BV, P.O. Box 328, 7500 AH Enschede, The Netherlands

The diffusion coefficients of N₂O in aqueous piperazine (PZ) solutions have been determined using the Taylor dispersion technique over a temperature range from (293 to 333) K and a concentration range from (0.3 to 1.4) mol·dm⁻³ PZ. The experimental results have been compared to literature values. Diffusion coefficients of N₂O in aqueous PZ solutions can be estimated by using a modified Stokes–Einstein relation with sufficient accuracy for engineering purposes. The diffusion coefficient of CO₂ in an aqueous PZ solution can be estimated by means of the “N₂O analogy”. The use of the Taylor dispersion technique for determination of gas diffusivities in liquids is given in detail.

Introduction

Aqueous solutions of amines are frequently used for the removal of acid gases, such as CO₂ and H₂S, from a variety of gas streams. In particular, aqueous solutions of alkanolamines and blends of alkanolamines are widely applied in gas treating.¹ Recently, blends of a primary or secondary (alkanol)amine with an aqueous solution of a tertiary (alkanol)amine have found application in the removal and absorption of CO₂ due to higher reaction rates and lower heats of regeneration in the desorber section. There is a growing interest in the use of piperazine (PZ) blended with aqueous solutions of *N*-methyl-diethanolamine (MDEA) as a tertiary alkanolamine for gas treating processes. This type of solution is also called the activated MDEA solvent. PZ is known as the activator or promotor and enhances the reactivity toward CO₂, whereas MDEA contributes to the lower heats of regeneration. In these solvents, the concentration of MDEA is usually kept at about (3 to 4) mol·dm⁻³, and typically a maximum PZ concentration of about 1.0 mol·dm⁻³ is applied.^{2–6}

Knowledge of diffusion coefficients is needed for the design of absorbers and desorbers in a commercial CO₂ capture plant, as they are related to the mass transfer coefficients. They are also essential for a correct and accurate interpretation of many (laboratory scale) absorption rate experiments, e.g., the experiments aimed at the determination of the intrinsic kinetics in a gas–liquid process. The diffusion coefficients of N₂O in aqueous PZ solutions are presented in this work, and these values can be used to estimate the diffusion of CO₂ in aqueous PZ solutions by means of the “N₂O analogy”.

Measurements of gas diffusivities in liquids have previously, in large part, been carried out with the use of experimental techniques like laminar liquid jet, diaphragm cell, and wetted wall absorber. The Taylor dispersion technique has primarily been used to determine liquid diffusivities in liquid systems. However, some

literature sources have reported gas diffusivities in liquids using the Taylor dispersion technique.^{7,8} These literature sources determined the diffusivity of gases in single component liquids, e.g., the N₂O + H₂O system. In the present work, diffusivities of a gas in binary liquid mixtures are determined, e.g., the N₂O + (PZ + H₂O) system. These types of chemical systems contain three compounds, e.g., N₂O, PZ, and H₂O, and there are experimental challenges with these types of systems when determining diffusivities using the Taylor dispersion technique. The advantages of using the Taylor dispersion technique over the other experimental techniques mentioned are the fast measuring time, readily automation of the setup, possibility of measurements at elevated temperatures and/or pressures, and that the setup consists of standard HPLC components.

Theory and Experimental Procedures

The diffusivities were determined using the Taylor dispersion technique. A square pulse of a solute solution was injected into a solvent solution showing laminar flow through a capillary tube. The solute solution contained the same amounts of liquid components as the solvent, but with additional N₂O gas absorbed. As an example, for measurements of N₂O diffusivity in an aqueous PZ solution of 1.0 mol·dm⁻³, the solute and solvent solution would contain 1.0 mol·dm⁻³ PZ. The solute solution would additionally contain N₂O gas absorbed, and a square pulse of this solution would be injected into the flowing solvent solution. The combined action of axial convection and radial and axial molecular diffusion will eventually change the shape of the solute pulse into a Gaussian shaped curve. The theory and mathematical description of such a measurement have been described in detail by Taylor^{9,10} and Aris.¹¹

The experimental setup used is shown schematically in Figure 1. Two vessels containing the solute solution and the solvent solution were kept under a constant 5·10² kPa pressure of saturated helium to create a constant flow of the solute and solvent solution. Saturated helium was used to prevent any concentration changes in the solute and solvent solution as the liquid level in the two closed vessels containing the solutions decreased during a measurement due to the solution outflow. Introduction of a solute square pulse

* Corresponding author. E-mail: espen.hamborg@procede.nl. Tel.: +31 53 489 4636. Fax: +31 53 489 5399.

† University of Twente.

‡ Procede Group BV.

§ University of Twente, Faculty of Science and Technology, Thermo-Chemical Conversion of Biomass.

|| Current address: Procede Group BV, P.O. Box 328, 7500 AH Enschede, The Netherlands.

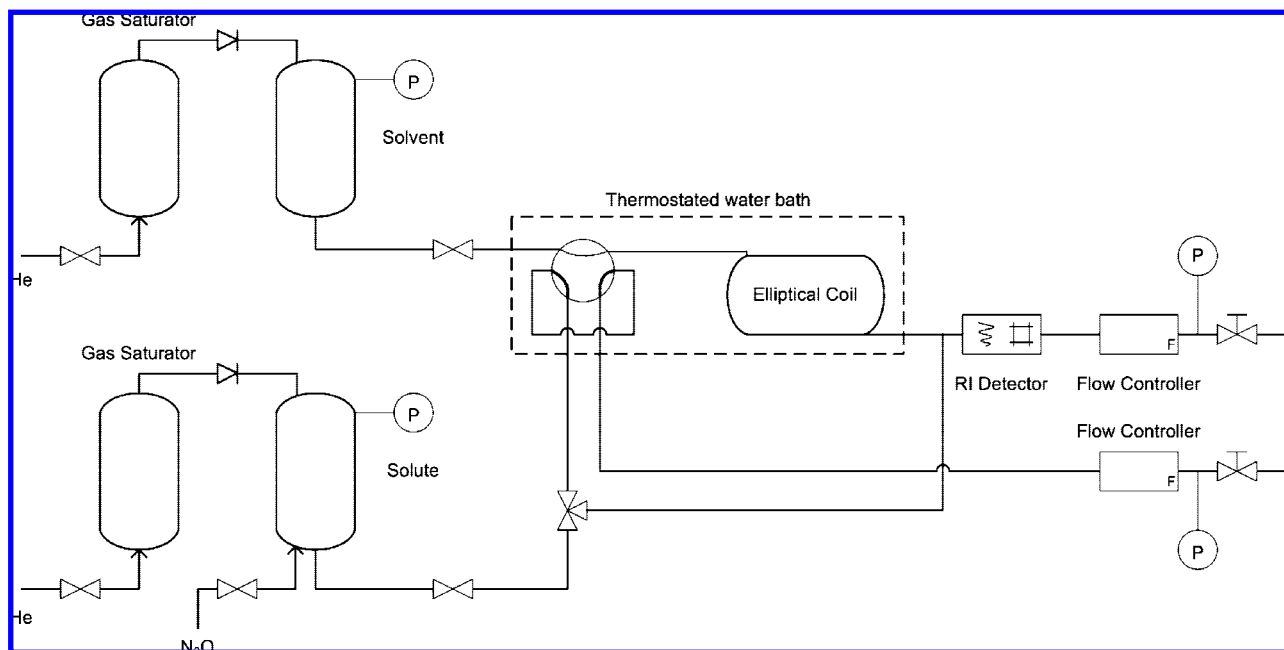


Figure 1. Schematic representation of the Taylor dispersion setup.

was done by switching an air-actuated six-way valve back and forth within a few seconds. The capillary tube was elliptical coiled and placed in a water bath for temperature control. The flow velocity was controlled with mass flow controllers (Rosemount Flowmega 5881), located behind the refractive index (RI) detector (Varian 350 RI) and the six-way pulse valve to obtain a constant pulsation-free solute and solvent solution flow throughout the measurement. To avoid bubble formation from the absorbed gas and a disturbance of the laminar fluid flow profile, especially at higher temperatures, pressure reducers were located behind the RI detector and the six-way pulse valve. These reducers pressurized the fluids inside the tubing to $4 \cdot 10^2$ kPa. The influence of the pressure on the diffusion coefficients can be neglected for the pressure applied.^{12,13} Prior to each experiment, both the solute and the solvent solution were degassed by applying a vacuum for a while, and N_2O was further absorbed into the solute solution by bubbling the gas up to $1.5 \cdot 10^2$ kPa into the closed vessel containing the solution. The vessels were kept at room temperature (298.15 K) at all times, and the concentration of N_2O in the solute solution was determined based on the solubility of N_2O in aqueous PZ solutions¹⁴ and the ideal gas law. A computer was connected to the setup for control and data acquisition. The output signal from the RI detector was recorded as a function of time and used to determine the diffusion coefficients. The RI detector showed a linear response to concentration changes of absorbed N_2O in the solutions investigated. Depending on the residence time and the dispersion rate of the injected N_2O pulse, the average concentration of the detected N_2O was lower than the concentration of the N_2O absorbed into the solute solution. The determined diffusivities are characterized to be at infinite dilution, since the average concentration of N_2O in the injected pulse was in the order of 10^{-3} mol·dm⁻³. The dimensions of the experimental setup and the flow conditions are given in Table 1.

A disturbance of the laminar fluid flow profile can occur due to the helical coiling of the tube. The varying path lengths traversed by the fluid at different radial positions in the tube and the secondary flows present in the flow can contribute

Table 1. Dimensions of the Experimental Setup and Flow Conditions

length of the capillary tube	L	14.92 m
inner radius of the capillary tube	R	$5.14 \cdot 10^{-4}$ m
radius of the helical coil	R_C	0.1 m
injection volume	V_{inj}	$(2.5 \text{ to } 4.1) \cdot 10^{-8}$ m ³
liquid flow velocity	u	$(2 \text{ to } 6) \cdot 10^{-3}$ m·s ⁻¹

additionally to the dispersion process. This topic has been extensively discussed by Alizadeh et al.¹⁵ and Snijder et al.¹⁶ To avoid this disturbance, the critical $(De)^2Sc$ was determined for each system. The dimensionless $(De)^2Sc$ number is defined as

$$De = Re \left(\frac{R_C}{R} \right)^{-1/2} \quad (1)$$

$$Sc = \frac{\mu}{\rho D} \quad (2)$$

where Re is the well-known Reynolds number and μ and ρ are the solvent viscosity and density. The other parameters are defined in Table 1. The measurements had to be carried out at a value of $(De)^2Sc$ lower than the critical one.

Chemicals. PZ [110-85-0] (Sigma-Aldrich), N_2O [10024-97-2], and helium [7440-59-7] (Linde Gas) were used as supplied. Water was demineralized and further purified by vacuum distillation. The actual PZ concentration in the prepared solutions was determined by volumetric titration.

Diffusivity of N_2O in H_2O . Figure 2 shows a typical experimental result from a pulse recorded using the Taylor dispersion technique. The figure shows a dimensionless result from an $N_2O + H_2O$ system. The recorded pulse is the response from absorbed N_2O in the solution flowing through the RI detector. The experimental result of the diffusion coefficient from this typical experiment is derived from fitting eqs 3 and 4⁹⁻¹¹ to the experimental data where $N_{inj}(N_2O)$, u , and $D_{N_2O}(\text{in } H_2O)$ are the independent variables.

$$c_m = \frac{N_{inj}(N_2O)}{2\pi R^2 \sqrt{\pi K(N_2O)t}} \exp\left(-\frac{(L-ut)^2}{4K(N_2O)t}\right) \quad (3)$$

$$K(N_2O) = \frac{u^2 R^2}{48D_{N_2O}(\text{in H}_2\text{O})} + D_{N_2O}(\text{in H}_2\text{O}) \quad (4)$$

where c_m is the measured concentration profile; t is the time; $N_{inj}(N_2O)$ is the number of moles of N_2O injected; and $D_{N_2O}(\text{in H}_2\text{O})$ is the diffusion coefficient of N_2O . The other parameters are defined in Table 1.

Diffusivity of N_2O in (PZ + H_2O). Figure 3 shows a dimensionless recorded experimental result from a N_2O + (PZ + H_2O) system. It is shown that a combination of two pulses appears. The degassed solvent and solute solutions used were prepared containing a specific and equal concentration of PZ. N_2O was further absorbed into the solute solution as aforementioned. As N_2O is absorbed into the solution, a small liquid volume increase takes place because of the volume occupied by the N_2O molecules. Consequently, the concentration of PZ is decreased in the solute solution, as compared to the solvent solution. The “negative” peak in Figure 3 must be attributed to this phenomena. The “positive” peak is the effect of the N_2O present in the solution.

A RI detector shows a higher sensitivity toward concentration changes of liquid compounds than concentration changes of absorbed gas compounds in the fluid under investigation. This leads to an amplification of the recorded RI signal from concentration changes of liquid compounds relative to concentration changes of absorbed gas compounds. The concentration peak responsible for the “negative” pulse in Figure 3 was found to be in the order of $10^{-6} \text{ mol}\cdot\text{dm}^{-3}$ after calibration of a PZ + H_2O system, whereas the “positive” peak was found to be in the order of $10^{-3} \text{ mol}\cdot\text{dm}^{-3}$. The concentration differences between the positive and negative peak are thus in the order of $10^3 \text{ mol}\cdot\text{dm}^{-3}$, and the negative peak in Figure 3 appears to be larger than the actual in relation to the positive peak. As the dispersions of the positive and negative pulse are unequal, apparent molecular diffusion coefficients can be determined from the recorded experimental data by the principle of superposition. The total response of the RI detector is the sum of two separate Gaussian curves. The experimental measured result was fitted to eqs 5 to 7

$$c_m = \frac{N_{inj}(N_2O)}{2\pi R^2 \sqrt{\pi K(N_2O)t}} \exp\left(-\frac{(L-ut)^2}{4K(N_2O)t}\right) + \frac{N_{inj}(PZ)}{2\pi R^2 \sqrt{\pi K(PZ)t}} \exp\left(-\frac{(L-ut)^2}{4K(PZ)t}\right) \quad (5)$$

$$K(N_2O) = \frac{u^2 R^2}{48D_{N_2O}(\text{in PZ sol.})} + D_{N_2O}(\text{in PZ sol.}) \quad (6)$$

$$K(PZ) = \frac{u^2 R^2}{48D_{PZ}(\text{in H}_2\text{O})} + D_{PZ}(\text{in H}_2\text{O}) \quad (7)$$

where c_m is the measured concentration profile, t is the time and $N_{inj}(N_2O)$ and $N_{inj}(PZ)$ are the respective number of moles of N_2O and excess number of moles of PZ injected. The fluid flow, u , is identical for both injected N_2O and PZ. $D_{N_2O}(\text{in PZ sol.})$ and $D_{PZ}(\text{in H}_2\text{O})$ are the respective binary diffusion coefficients of N_2O and PZ. The parameters $N_{inj}(N_2O)$, $N_{inj}(PZ)$, u , and $D_{N_2O}(\text{in PZ sol.})$ in eqs 5 to 7 are the independent variables used to fit the equations to the experimental data. $D_{PZ}(\text{in H}_2\text{O})$ is taken from Derks,⁶ and the influence of N_2O present in the current work on this value is neglected. The value of $D_{PZ}(\text{in H}_2\text{O})$ represents the dispersion of the negative pulse in Figure 3 through eq 7. The other parameters are defined in Table 1.

Results and Discussion

Diffusivity of N_2O in H_2O . Measurements were conducted on the system N_2O + H_2O from (293 to 368) K to evaluate the accuracy of the equipment used. The critical $(De)^2Sc$ number was found to be 130 for this system. Figure 4 shows the experimental results of N_2O diffusion in H_2O , and the results are tabulated in Table 2. The experimental uncertainty is estimated to be 10 %. The values are the average value of at least three measurements, and the reproducibility of the measurements is within 2 %. The concentration of N_2O in the injected square pulse was calculated¹⁴ to be $0.036 \text{ mol}\cdot\text{dm}^{-3}$. The results of the present study are in agreement with the results reported by previous authors using different experimental techniques.^{17–24}

Diffusivity of N_2O in Aqueous PZ Solutions. The experimental determined diffusivities of N_2O + (PZ + H_2O) were measured and averaged over at least three measurements from (293 to 333) K and $(0.3 \text{ to } 1.4) \text{ mol}\cdot\text{dm}^{-3}$. These results are presented in Table 3. The experimental uncertainty is estimated to be 10 %, and the reproducibility is within 2 %. The critical $(De)^2Sc$ number

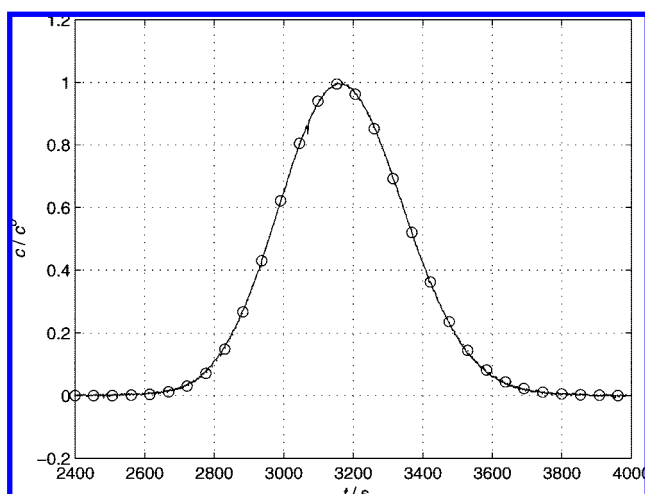


Figure 2. Typical result from an N_2O + H_2O system: —, exptl result; \circ , fit.

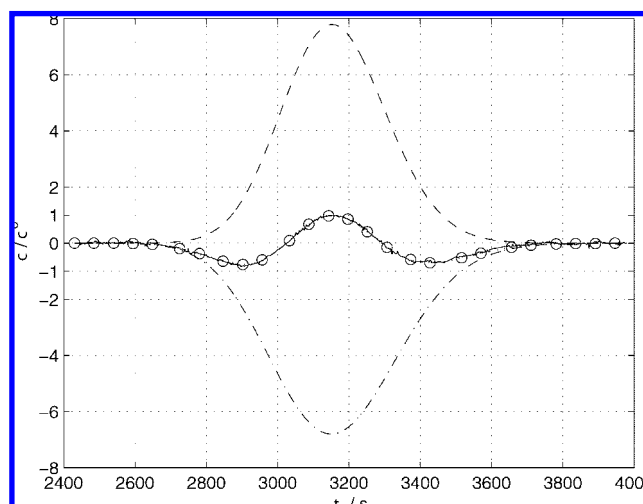


Figure 3. Typical result from an N_2O + (PZ + H_2O) system: —, exptl result; \circ , fit; --, N_2O response; - · - ·, PZ response.

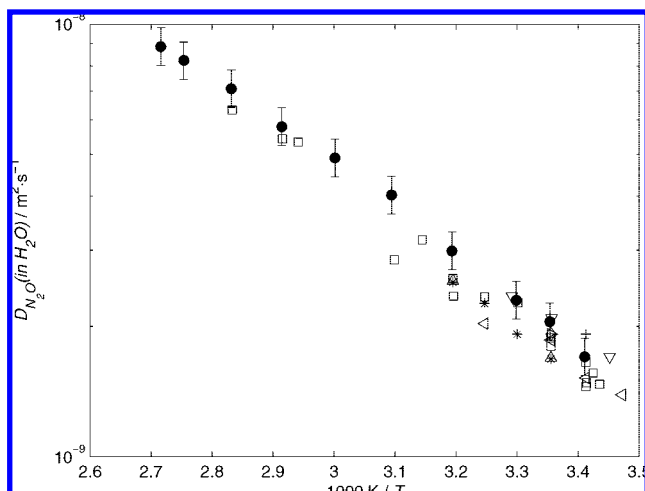


Figure 4. Experimental results of D_{N_2O} (in H_2O): ●, exptl results in the present work with error bars representing the uncertainty of 10 %; ▽, Davidson & Cullen;¹⁷ △, Duda & Vrentas;¹⁸ open arrow pointing left, Haimour & Sandall;¹⁹ open arrow pointing right, Sada et al.;²⁰ ◇, Joosten & Danckwerts;²¹ □, Versteeg & Van Swaaij;²² +, Thomas & Adams;²³ *, Samanta et al.²⁴

Table 2. Diffusivity of N_2O in H_2O , D_{N_2O} (in H_2O), at Various Temperatures

T/K	D_{N_2O} (in H_2O) $10^{-9} \cdot m^2 \cdot s^{-1}$	refs	method ^a
288.00	1.39	Haimour & Sandall	19 LLJ
289.70	1.70	Davidson & Cullen	17 WWA
291.10	1.47	Versteeg & Van Swaaij	22 DC
292.00	1.56	Versteeg & Van Swaaij	22 DC
292.90	1.48	Versteeg & Van Swaaij	22 DC
293.00	1.52	Haimour & Sandall	19 LLJ
293.00	1.92	Thomas & Adams	23 LLJ
293.00	1.45	Versteeg & Van Swaaij	22 DC
293.00	1.65	Versteeg & Van Swaaij	22 DC
293.15	1.70	this work	TDT
297.90	2.09	Davidson & Cullen	17 WWA
298.00	1.86	Haimour & Sandall	19 LLJ
298.00	1.69	Duda & Vrentas	18 LLJ
298.00	1.92	Joosten & Danckwerts	21 LLJ
298.00	1.78	Sada et al.	20 LLJ
298.00	1.88	Versteeg & Van Swaaij	22 DC
298.00	1.80	Versteeg & Van Swaaij	22 DC
298.00	1.68	Samanta et al.	24 WWA
298.15	2.05	this work	TDT
302.90	2.27	Versteeg & Van Swaaij	22 DC
303.00	1.92	Samanta et al.	24 WWA
303.15	2.30	this work	TDT
303.80	2.35	Davidson & Cullen	17 WWA
308.00	2.03	Haimour & Sandall	19 LLJ
308.00	2.34	Versteeg & Van Swaaij	22 DC
308.00	2.26	Samanta et al.	24 WWA
312.90	2.35	Versteeg & Van Swaaij	22 DC
313.00	2.55	Duda & Vrentas	18 LLJ
313.00	2.58	Versteeg & Van Swaaij	22 DC
313.00	2.53	Samanta et al.	24 WWA
313.15	2.99	this work	TDT
318.00	3.17	Versteeg & Van Swaaij	22 DC
322.70	2.85	Versteeg & Van Swaaij	22 DC
323.15	4.02	this work	TDT
333.15	4.90	this work	TDT
340.00	5.33	Versteeg & Van Swaaij	22 DC
343.00	5.43	Versteeg & Van Swaaij	22 DC
343.15	5.79	this work	TDT
353.00	6.32	Versteeg & Van Swaaij	22 DC
353.15	7.09	this work	TDT
363.15	8.23	this work	TDT
368.15	8.87	this work	TDT

^a LLJ = laminar liquid jet; DC = diaphragm cell; WWA = wetted wall absorber; TDT = Taylor dispersion technique.

was found to be 80 for this system. The concentration of N_2O in the injected square pulse was calculated¹⁴ to be (0.036, 0.036, 0.035, and 0.034) $mol \cdot dm^{-3}$ at (0.295, 0.629, 1.067, and 1.368) $mol \cdot dm^{-3}$ PZ, respectively. Diffusivities were, in addition, esti-

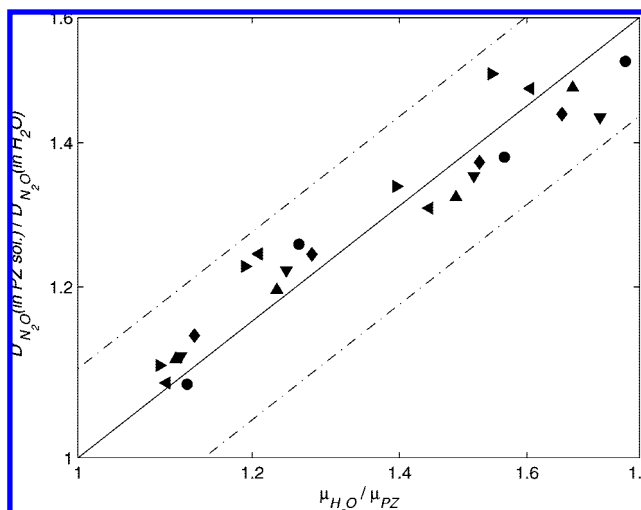


Figure 5. Logarithmic Stokes–Einstein plot for D_{N_2O} (in PZ sol.): ●, $T = 293.15$ K; ▼, $T = 298.15$ K; ▲, $T = 303.15$ K; solid triangle pointing left, $T = 313.15$ K; solid triangle pointing right, $T = 323.15$ K; ◆, $T = 333.15$ K. The solid line represents eq 8, and the dashed lines represent 10 %.

Table 3. Diffusivity of N_2O in Aqueous PZ Solutions, D_{N_2O} (in PZ sol.), at Various Temperatures, T , and PZ Concentrations, c_{PZ}

c_{PZ} $mol \cdot dm^{-3}$	T/K					
	293.15	298.15	303.15	313.15	323.15	333.15
	D_{N_2O} in PZ sol./ $10^{-9} \cdot m^2 \cdot s^{-1}$					
0.295	1.44	1.60	1.83	2.41	2.99	3.61
0.629	1.24	1.46	1.70	2.10	2.69	3.31
1.067	1.13	1.32	1.54	2.00	2.47	3.00
1.368	1.02	1.24	1.37	1.76	2.19	2.85

ated at different temperatures and concentrations with the modified Stokes–Einstein relation²²

$$\frac{D'_{N_2O}(\text{in PZ sol.})}{D'_{N_2O}(\text{in } H_2O)} = \left(\frac{\mu_{H_2O}}{\mu_{PZ}}\right)^{0.8} = \text{constant} \quad (8)$$

where the viscosities μ_{H_2O} and μ_{PZ} were taken from Lide²⁵ and Derks et al.,¹⁴ respectively. The viscosities from Derks et al.¹⁴ were extrapolated to 333.15 K. The diffusivity of N_2O in H_2O was calculated by the equation presented by Versteeg & Van Swaaij²²

$$D'_{N_2O}(\text{in } H_2O)/10^{-9} \cdot m^2 \cdot s^{-1} = 5.07 \cdot 10^{-6} \exp\left(\frac{-2371}{T/K}\right) \quad (9)$$

A Stokes–Einstein logarithmic plot for the diffusivity of N_2O in aqueous PZ solutions is shown in Figure 5. The modified Stokes–Einstein relation can be used to calculate N_2O diffusivities in aqueous PZ solutions satisfactorily from (293 to 333) K and up to approximately 1.4 $mol \cdot dm^{-3}$ for engineering purposes. The average and maximum absolute relative deviations between the experimental determined values and the calculated values are 3.0 % and 6.4 %, respectively. The diffusivity of CO_2 in an aqueous PZ solution can be estimated by the N_2O analogy.²²

Diffusivities of N_2O in aqueous PZ solutions have been reported by Samanta et al.²⁴ and Sun et al.²⁶ at (303 and 313) K. These data are compared to the results from the present work at (303.15 and 313.15) K in Figure 6 and the calculated values from the modified Stokes–Einstein relation. The values from the present work have a relative absolute average deviation of 2.4 % and 2.8 % from the calculated values at (303.15 and 313.15) K, respectively. The values from Samanta et al.²⁴ deviate with 1.9 % and 5.6 % at (303 and 313) K, and the values of Sun et al.²⁶ deviate 5.2 % and 3.0 %, respectively, from the

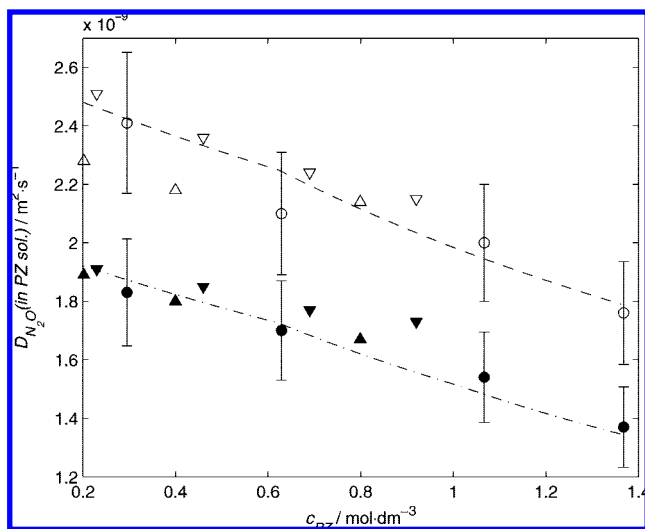


Figure 6. Comparison of D_{N_2O} (in PZ sol.) to literature sources: ●, $T = 303.15$ K, present work with error bars representing the uncertainty of 10 %; ▼, $T = 303$ K, Sun et al.;²⁶ ▲, $T = 303$ K, Samanta et al.;²⁴ - - -, $T = 303.15$ K, modified Stokes–Einstein; ○, $T = 313.15$ K, present work with error bars representing the uncertainty of the results of 10 %; ▽, $T = 313$ K, Sun et al.;²⁶ △, $T = 313$ K, Samanta et al.;²⁴ - - -, $T = 313.15$ K, modified Stokes–Einstein.

calculated values. Samanta et al.²⁴ and Sun et al.²⁶ both used a wetted wall absorber to determine the diffusivities. Figure 6 shows that the method developed and described in the present work can be used to determine gas diffusivities in binary liquid systems with sufficient accuracy.

Conclusion

Gas diffusivities have been measured by the Taylor dispersion technique in a binary liquid system. The diffusivities of N_2O in aqueous PZ solutions have been determined from (293 to 333) K and (0.3 to 1.4) $\text{mol} \cdot \text{dm}^{-3}$. The results have been compared to literature values determined with the use of experimental techniques different from the Taylor dispersion technique. The diffusivity of N_2O in aqueous PZ solutions can be calculated with a modified Stokes–Einstein relation for engineering purposes. The results from the present work have an average and maximum deviation of 3.0 % and 6.4 % from the calculated values, whereas the results from the literature have an average deviation of up to 5.6 % for the results considered. As a result, the Taylor dispersion technique can be used as an experimental technique to determine gas diffusivities in binary liquid systems with sufficient accuracy.

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