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Diffusion Coefficients Significant in Modeling the Absorption Rate of Carbon Dioxide into Aqueous Blends of *N*-Methyldiethanolamine and Diethanolamine and of Hydrogen Sulfide into Aqueous *N*-Methyldiethanolamine

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Absorption rates of gaseous CO_2 into aqueous blends of *N*-methyldiethanolamine (MDEA) and diethanolamine (DEA) and of gaseous H_2S into aqueous MDEA were measured in a quiescent, inverted-tube diffusiometer by monitoring the rate of pressure drop. A numerical model for absorption, diffusion, and reaction of CO_2 and H_2S in blends of MDEA, DEA, and water was developed. The model was used to regress diffusion coefficients of bicarbonate, carbamate, and $MDEAH_2CO_3$ for the case of CO_2 absorption and of bisulfide ion for the case of H_2S absorption from measured absorption rates. CO_2 absorption rates and diffusion coefficients of bicarbonate, carbamate, and $MDEAH_2CO_3$ were obtained at 298.2 K and 318.2 K in aqueous solutions containing 50 mass % total amine at DEA:MDEA mole ratios of 1:20, 1:4, 1:3, and 2:3. H_2S absorption rates and diffusion coefficients of bisulfide ion were obtained at 298.2 K and 318.2 K in aqueous solutions containing 20, 35, and 50 mass % MDEA.

Introduction

The acid gases CO_2 and H_2S are commonly removed from natural gas streams by treatment with aqueous alkanolamines. Addition of the amine to the water increases absorption and removal of the gases owing to reactions between the gases and the amine. Aqueous mixtures of *N*-methyldiethanolamine (MDEA) and diethanolamine (DEA) are often used to selectively remove H_2S over CO_2. To design efficient absorption systems, accurate physical property data are necessary. In addition to quality solubility and equilibrium data, values of the mutual diffusion coefficients of the important species present in the mixture are also required to model gas uptake rates. Recently, Rowley et al. (1997, 1998) reported the results of absorption measurements and diffusion coefficients of species present in aqueous MDEA systems as well as aqueous DEA systems. In this paper, we report the results of similar measurements made for CO_2 in aqueous blends of MDEA and DEA and for H_2S in aqueous MDEA. These data are important in the design of absorbers for which it is desirable to tune the selectivity of H_2S to CO_2 absorption. Because the tertiary amine MDEA can only react with CO_2 through a slow step involving the formation of the carbamate ion, it exhibits a preference for H_2S absorption kinetically, although at equilibrium more CO_2 than H_2S would be absorbed.

As discussed in the previous works cited above, very few measurements of diffusion coefficients in reacting mixtures have been attempted because of the complexities involved when both reactions and diffusion are involved in the absorption process. Generally, experimentalists have tried to obtain diffusion coefficients independent of the reaction by blocking the reaction so that only diffusion occurs or by measuring diffusion in an analogous system that does not react. For example, the amine solution can be acidified to block the reaction, or the absorption and diffusion of N_2O as a nonreacting analogue for CO_2 can be studied. Both of these methods change the nature of the species or solution from that actually desired and can eliminate important information about the absorption process. This was found to be the case in the MDEA and DEA studies. It was found that the rate of absorption of CO_2 is controlled by the rate of diffusion of the ionic reaction products, the protonated amine and bicarbonate ions, rather than diffusion of the CO_2 gas itself as previously assumed. Very little CO_2 gas is present in the system because the reactions involved are rapid relative to the rate of diffusion. There-

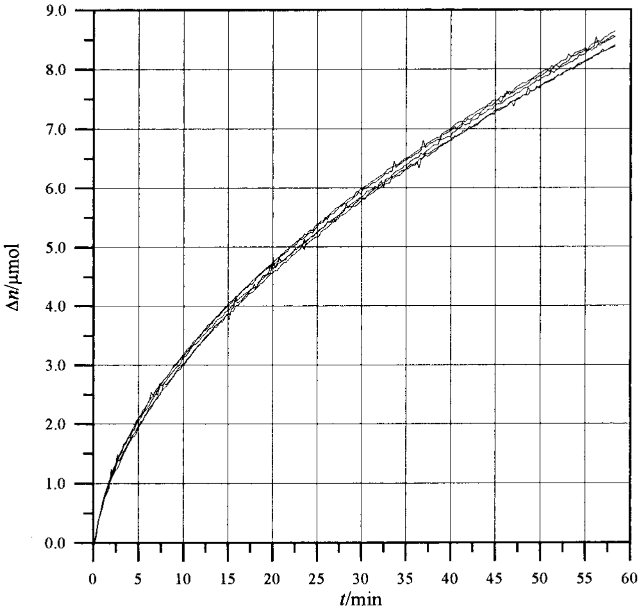

Figure 1. Molar uptake data for H_2S in 20 mass % MDEA at 25 °C. Five replicate runs are shown.

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fore, it is important to develop a model for CO_2 absorption that accounts for the simultaneous reaction and diffusion of the species present to determine insofar as is possible the diffusion coefficients of the species involved. In this work, we report the results of diffusion and modeling studies performed on the absorption of CO_2 into aqueous blends of DEA and MDEA and of H_2S in aqueous MDEA.

In the previous studies involving aqueous solutions of DEA and MDEA alone, the solubility model of Kent and Eisenberg (1976) and Chakma and Meisen (1987) was used. However, the Chakma−Meisen equilibrium constant for the MDEA protonation reaction was based on compositions, not activities, and was therfore composition-dependent. The composition dependence was developed specifically for the aqueous MDEA system and therefore cannot be used for systems containing both DEA and MDEA. For this reason, the more general solubility model of Deshmukh and Mather (1981) was used in this work. The model includes the reversible reactions

$$H^+ + MDEA_{(aq)} \rightleftarrows MDEAH^+ \tag{1}$$

$$H^+ + DEA_{(aq)} \rightleftarrows DEAH^+ \tag{2}$$

$$DEA_{(aq)} + HCO_3^- \rightleftarrows (HOCH_2CH_2)_2NCOO^- + H_2O \tag{3}$$

$$CO_{2(aq)} + H_2O \rightleftarrows H^+ + HCO_3^- \tag{4}$$

$$HCO_3^- \rightleftarrows H^+ + CO_3^{2-} \tag{5}$$

$$MDEAH^+ + HCO_3^- \rightleftarrows MDEAH_2CO_{3(aq)} \tag{6}$$

$$H_2S_{(aq)} \rightleftarrows HS^- + H^+ \tag{7}$$

$$HS^- \rightleftarrows S^{2-} + H^+ \tag{8}$$

$$MDEAH^+ + HS^- \rightleftarrows MDEAH_2S_{(aq)} \tag{9}$$

$$H_2O \rightleftarrows H^+ + OH^- \tag{10}$$

This model was used to regress species diffusion coefficients from experimental absorption-rate data. The model suggests that diffusion coefficients for MDEA, DEA, $MDEAH^+$, $DEAH^+$, $(HOCH_2CH_2)_2NCOO^-$ (carbamate ion), HCO_3^-, (bicarbonate ion), H^+, OH^-, $MDEAH_2CO_3$, and CO_3^{2-} in aqueous solutions might be required in addition to the diffusion coefficient of CO_2 in order to accurately model the absorption of CO_2 into aqueous mixtures of DEA and MDEA. Similarly, the diffusion coefficients for MDEA, $MDEAH^+$, HS^- (bisulfide ion), S^{2-} (sulfide ion), H^+, OH^-, and $MDEAH_2S$ might be necessary in addition to the diffusivity of H_2S to accurately model the absorption of H_2S into aqueous MDEA.

Chemicals

N-Methyldiethanolamine was obtained from Aldrich Chemical Company with a stated purity of 99%. Diethanolamine was also obtained from Aldrich with a purity of 99%. The DEA originally came in large bottles but was transferred into smaller bottles in an argon-filled glovebox to minimize exposure of DEA to air. Distilled and deionized water was degassed by boiling under vacuum until approximately 20% of the water had been removed. Carbon dioxide was obtained from Air Liquide at a purity of 99.89%. Hydrogen sulfide was obtained from Matheson Gas Products at a purity of 99.95%. No further purification was attempted with any of the chemicals used. All amine + water mixtures were made gravimetrically and then stored under their own vapor pressure before introduction into an ISCO high-pressure micropump.

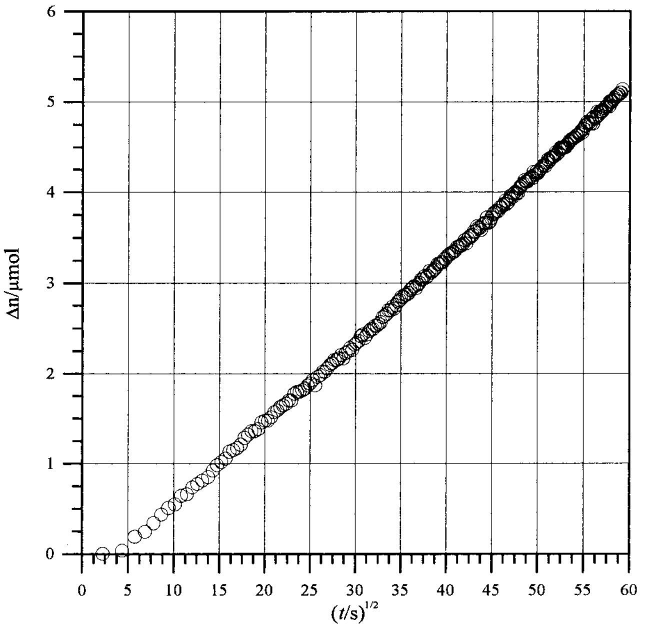

Figure 2. Typical raw absorption data from experiment exhibiting diffusion-controlled linearity when plotted versus square root of time.

Measurement of CO_2 and H_2S Absorption

The absorption of CO_2 into aqueous solutions of MDEA and DEA and of H_2S into aqueous MDEA was measured in an inverted tube (liquid on top, held up by capillary action) diffusion apparatus by monitoring the decrease in pressure as a function of time after contact of the gas (saturated with water vapor) with the liquid solution. The inverted tube method was used to avoid free convection due to the increase in density of the solution upon acid gas loading (Al-Ghawas et al., 1989). The intent was to fit absorption rate data to the model by adjusting one or more diffusion coefficients. Details of the apparatus and experimental procedure were given previously in the MDEA study (Rowley et al., 1997). The consistency of raw data obtained for replicate measurements of H_2S absorption into 20 mass % MDEA under similar conditions is illustrated in Figure 1. As indicated in the previous study (Rowley et al., 1997), there is some ambiguity in the initial time because of the injection process of the liquid into the capillary diffusion tube. Hence, there is an uncertainty in the absolute times of about 5 s. Relative uncertainties in time after the initial start up are less than 1 ms. Uncertainties in the reported temperatures are less than 5.0 mK. Uncertainties in compositions of the prepared solutions are less than 0.1 mass %.

Analysis of Absorption Data

An equilibrium model was used to analyze the absorption rate data in the previous MDEA and DEA studies because it was shown that the reactions occur rapidly relative to the rate of diffusion. Evidence of this rapid equilibrium was the linearity of the absorption data when plotted with respect to $t^{1/2}$, which is indicative of a diffusion-controlled process (Rowley et al., 1997, 1998). The same observation was true for this study indicating that reactions are rapid enough to be in steady state relative to the immediate local

Table 1. Coefficients for Use in Eqs 11−13, 16

eq	C_1	C_2	C_3	C_4	C_5	C_6	C_7	ref
1	1974.40	−7.5329	56.008					Oscarson et al. (1989)
2	3741.82	−4.2726	32.241					Oscarson et al. (1989)
3	8071.5	16.4262	−117.012					Oscarson et al. (1995)
4	−12092.1	−36.7816	235.482					Renon (1980)
5	−12431.7	−35.4819	220.067					Renon (1980)
6	−1404.6	−14.3427	85.455					Oscarson et al. (1995)
7	−12995.4	−33.5471	218.599					Renon (1980)
8	−2049.0	15.65	−114.45					Renon (1980)
9	−2227.6	−19.4842	11.857					Oscarson et al. (1995)
10	−4.098	−3245.2	2.2363×10^5	-3.984×10^7	13.957	−1262.3	8.5641×10^5	Marshall and Franck (1981)
11(CO_2)	22.2819	−13830.6	6.9135×10^6	-1.5586×10^9	1.2004×10^{11}			Chakma and Meisen (1987)
11 (H_2S)	104.518	−136808	7.3774×10^7	-1.7472×10^{10}	1.5216×10^{12}			Kent and Eisenberg (1976)
16	−61.44534	2.864468	183.5379	−0.6820223	7.8757×10^{-4}	58.9579		Chen et al. (1982)

compositions in the diffusing mixture. Figure 2 shows typical raw data as a function of $t^{1/2}$. We thus apply a similar model to that used previously, namely, a diffusion-controlled system in which local compositions are consistent with the reaction equilibrium constants.

To analyze the absorption data using the equilibrium reaction model of eqs 1−10, a numerical algorithm was devised based on the assumptions that (a) the concentrations of all species satisfy the equilibrium constants for eqs 1−10 at all times and positions in the cell, (b) there is no resistance to mass transfer at the interface, (c) the Henry's law constants given by Chakma and Meisen (1987) for CO_2 and by Kent and Eisenberg (1976) for H_2S apply, (d) the diffusion coefficients are independent of composition, (e) binary mutual diffusion coefficients can be used in place of the diffusion coefficients in the actual mixture, and (f) electroneutrality exists locally. Henry's law constants were used as obtained from Chakma and Meisen (1987) and Kent and Eisenberg (1976) in the form

$$H = \frac{1}{7.500\ 61} \exp\left(\sum_{i=1}^{5} C_i T^{1-i}\right) \qquad (11)$$

with the constants, C_i, given in Table 1, H in $Pa \cdot m^3 \cdot mol^{-1}$, and T in K. The equilibrium constants for the reactions in eqs 1−9 are calculated using an equation in the form

$$\ln K_j = (C_1/T) + C_2 \ln T + C_3 \qquad j = 1, ..., 9 \qquad (12)$$

where T is in K and j refers to the reaction number in Table 1. The equilibrium constant for the ionization of water in eq 10 is in the form (Marshall and Franck, 1981)

$$\log K_{10} = \sum_{i=1}^{4} C_i T^{i-1} + \left(\sum_{i=1}^{3} C_{i+4} T^{i-1}\right) \log \rho_w \qquad (13)$$

where T is in K and ρ_w is the density of water in g/cm^3.

The activity coefficients of the species in solution as well as the activity of water were calculated using a modified form of the Pitzer equation given by Edwards et al. (1978). For activity coefficients based on molality, the equation has the form

$$\ln \gamma_i = -z_i^2 A_\phi \left[\frac{I^{1/2}}{1 + 1.2I^{1/2}} + \frac{2}{1.2} \ln(1 + 1.2I^{1/2})\right] + 2\sum_{j \neq w} m_j \left\{B_{ij}^{(0)} + \frac{B_{ij}^{(1)}}{2I}[1 - (1 + 2I^{1/2}) \exp(-2I^{1/2})]\right\} - z_i^2 \sum_{j \neq w} \sum_{k \neq w} m_j m_k \frac{B_{jk}^{(1)}}{4I^2}[1 - (1 + 2I^{1/2} + 2I) \exp(-2I^{1/2})] \qquad (14)$$

and the activity of water is given by

$$\ln a_w = \frac{M_w}{1000}\left[\frac{2A_\phi I^{1.5}}{I + 1.2I^{0.5}} - \sum_{i \neq w} \sum_{j \neq w} m_i m_j [B_{ij}^{(0)} + B_{ij}^{(1)} \exp(-2I^{0.5})] - \sum_{j \neq w} m_i\right] \qquad (15)$$

where m_j is the molality of species j, I is the ionic strength on a mole fraction basis, M_w is the molecular weight of water, A_ϕ is the Debye−Hückel limiting slope, and $B_{ij}^{(k)}$ are interaction parameters listed in Table 2. The Debye−Hückel limiting slope was calculated using the expression of Chen et al. (1982)

$$A_\phi = C_1 \exp\left(\frac{T - T_0}{T_0}\right) + C_2\left[\exp\left(\frac{T - T_0}{T_0}\right)\right]^2 + C_3 \ln\left(\frac{T}{T_0}\right) + C_4(T - T_0) + C_5(T^2 - T_0^2) + C_6\left(\frac{T_0}{T}\right) \qquad (16)$$

where T is in K, T_0 equals 273.15 K, and the constants are listed in Table 1. The numerical method used to solve the equations has been described previously (Rowley et al., 1997, 1998). The total amount of CO_2 or H_2S absorbed as a function of time, α, is taken as the sum of the amount of CO_2 or H_2S in solution in its various forms and is given by

$$\alpha_{CO_2} = [CO_2] + [HCO_3^-] + [CO_3^{2-}] + [(HOCH_2CH_2)_2NCOO^-] + [MDEAH_2CO_3]$$

$$\alpha_{H_2S} = [H_2S] + [HS^-] + [S^{2-}] + [MDEAH_2S] \qquad (17)$$

According to the model, for the case of CO_2 absorption in blends of DEA and MDEA, there are 11 species in solution: CO_2, MDEA, DEA, $MDEAH^+$, $DEAH^+$, R_2NCOO^-, HCO_3^-, H^+, OH^-, $MDEAH_2CO_3$, and CO_3^{2-}. For the case of H_2S absorption in aqueous MDEA there are 8 species: H_2S, MDEA, $MDEAH^+$, HS^-, S^{2-}, H^+, OH^-, and $MDEAH_2S$. The mutual diffusion coefficients for MDEA and DEA in water were measured previously (Rowley et al., 1997, 1998) and were applied in this model as a function of total amine concentration. The diffusion coefficient of CO_2 in amine solutions used in the model was obtained from the N_2O-analogy measurements reported by Rinker et al. (1995). Since no appropriate analogy exists for the diffusivity of H_2S in amine solutions, the modified Stokes−Einstein equation for the diffusivity of H_2S in water, as reported by Tamimi et al. (1994), was used and corrected for the viscosity of the amine solutions. Viscosity data for the amine solutions were obtained from Snijder et al. (1993).

Table 2. Interaction Parameters for Activity Coefficients in Eqs 14 and 15

interaction type	$B_{ij}^{(0)}$	$B_{ij}^{(1)}$	ref
between like-charged ions	0.0	0.0	Oscarson et al. (1995)
cation−anion	0.0067	0.0223	Oscarson et al. (1995)
ion−molecule	0.0017	0.0052	Oscarson et al. (1995)
molecule-molecule (CO_2 case)	$-0.1333 + 40.84/T$	0.0	Grimsrud (1995)
molecule-molecule (H_2S case)	$-0.084 + 25.77/T$	0.0	Oscarson et al. (1995)

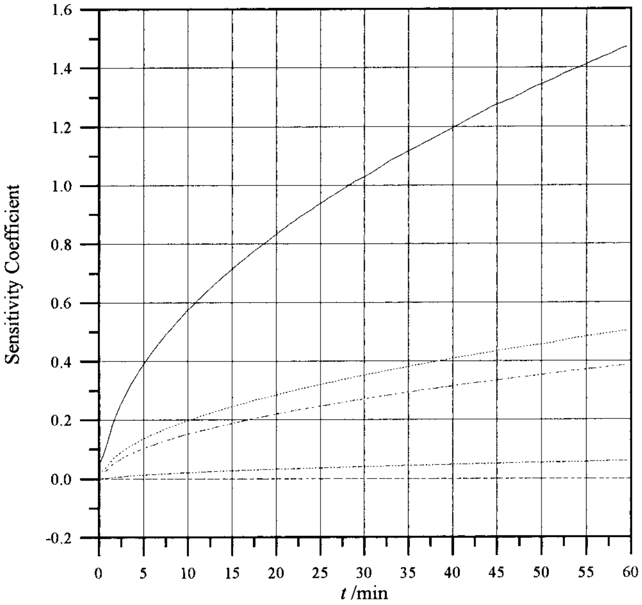

Figure 3. Sensitivity coefficient (derivative of moles absorbed with respect to the diffusivity of a single species) versus time for the significant species in the CO_2/MDEA/DEA/H_2O absorption system. Species shown: HCO_3^- (—); R_2NCOO^- (•••); $MDEAH_2CO_3$ (- — -); CO_3^{2-} (— - - - —); $MDEAH^+$, $DEAH^+$, H^+, and OH^- (all coincident) (- - -).

Sensitivity analysis shows that the concentrations of H^+, OH^-, CO_3^{2-}, S^{2-}, and $MDEAH_2S$ are everywhere so small that the gas absorption rate is independent of their diffusion coefficients. This is illustrated for the case of CO_2 absorption by the sensitivity coefficients plotted in Figure 3. Therefore, the diffusion coefficients for these ions were set to 1.0×10^{-9} m^2/s without loss of generality. Likewise, the CO_2 absorption rate is shown in Figure 3 to be insensitive to the diffusion coefficients of the protonated amines, $MDEAH^+$ and $DEAH^+$, and they likewise were given generic values in the model. The H_2S absorption rate was also found to be insensitive to the value of the diffusivity of the protonated amine, $MDEAH^+$. Only the mutual diffusion coefficients for R_2NCOO^-, HCO_3^-, and $MDEAH_2CO_3$ remain to be defined for the case of CO_2 absorption, and only the diffusion coefficient for HS^- was found to be significant for H_2S mixtures.

Of the three undefined species in CO_2 absorption, the absorption rate is seen to be far more sensitive to the diffusion rate of HCO_3^- than the other two species. Sensitivity analysis of the experiment indicates that only one parameter can be regressed from the absorption rate data; the remaining diffusion coefficients are tightly coupled. Local electroneutrality is imposed, but this does not completely decouple the parameters. This does not present a problem for the analysis of the H_2S absorption data, where only one parameter, the diffusivity of the HS^- ion, needs to be regressed.

Table 3. Diffusion Coefficients for HCO_3^- in 50 Mass % Total Amine Aqueous DEA/MDEA Mixtures along with 95% Confidence Intervals

DEA:MDEA mole ratio	$D_{HCO_3^-}$/ 10^{-10} $m^2 \cdot s^{-1}$	DEA:MDEA mole ratio	$D_{HCO_3^-}$/ 10^{-10} $m^2 \cdot s^{-1}$
$T = 298.2$ K			
1:20	0.65 ± 0.17	1:3	0.88 ± 0.11
1:4	0.95 ± 0.09	2:3	0.92 ± 0.20
$T = 318.2$ K			
1:20	1.38 ± 0.25	1:3	1.40 ± 0.04
1:4	1.64 ± 0.15	2:3	1.36 ± 0.17

For the case of CO_2 absorption in aqueous blends of MDEA and DEA, decoupling the remaining diffusion coefficients would require performing multiple experiments, each with different composition gradients for the species. This does not appear experimentally possible. We could also regress just a single *effective* diffusion coefficient from the data. However, use of this effective coefficient to predict the absorption rate at various blend compositions would be questionable. A more appropriate procedure seemed to be to make an additional assumed constraint between the bicarbonate and carbamate ions. Reid et al. (1987) relates the diffusion coefficient of a molecule to the solute's molar volume raised to a power of approximately −0.42. The molar volumes of ionic species are not readily available, so the additive contribution method of Le Bas (1915) was used to estimate ionic molar volumes. The validity of this assumption was tested by estimating molar volumes for 25 anions of sizes similar to bicarbonate and carbamate using the Le Bas method and plotting known diffusivities of these species in water at 298.2 K (*CRC Handbook*, 1992) against the estimated molar volumes. A linear regression on a log−log plot resulted in a slope of −0.42, consistent with the Le Bas model. The molar volumes of the bicarbonate and carbamate ions were estimated using the Le Bas method as 40.7 cm^3/mol and 161.8 cm^3/mol, respectively. The diffusivity of carbamate was then estimated using the relationship

$$D_{R_2NCOO^-} \simeq D_{HCO_3^-}\left(\frac{V_{R_2NCOO^-}}{V_{HCO_3^-}}\right)^{-0.42} = 0.560 \cdot D_{HCO_3^-} \qquad (18)$$

where V is the estimated molar volume of the ion. Similarly, the diffusion coefficient of $MDEAH_2CO_3$ was estimated using the Le Bas method and a molar volume of 185.9 cm^3/mol to be proportional to the diffusivity of the bicarbonate ion by a factor of 0.528. These relations allow the reaction−diffusion model to be entirely written in terms of a single unknown diffusion coefficient: HCO_3^- for CO_2 mixtures and HS^- for H_2S mixtures. The absorption data were fitted to the model by regressing this diffusion coefficient.

Results and Discussion

The data were analyzed using the simultaneous equilibrium and diffusion model detailed previously to regress values of the diffusion coefficients for HS^- and HCO_3^-. The diffusivities obtained are listed in Tables 3 and 4. Because these values are inferred from a model, it is difficult to assess the absolute accuracy of the data, but sensitivity analysis and the reproducibility of the measurements

Table 4. Diffusion Coefficients for HS^- in Aqueous MDEA Solutions along with 95% Confidence Intervals

mass % MDEA	$D/10^{-10}$ $m^2 \cdot s^{-1}$	mass % MDEA	$D/10^{-10}$ $m^2 \cdot s^{-1}$
	$T = 298.2$ K		
20	4.23 ± 0.13	50	1.03 ± 0.04
35	3.07 ± 0.27		
	$T = 318.2$ K		
20	5.09 ± 0.27	50	1.49 ± 0.21
35	3.91 ± 0.40		

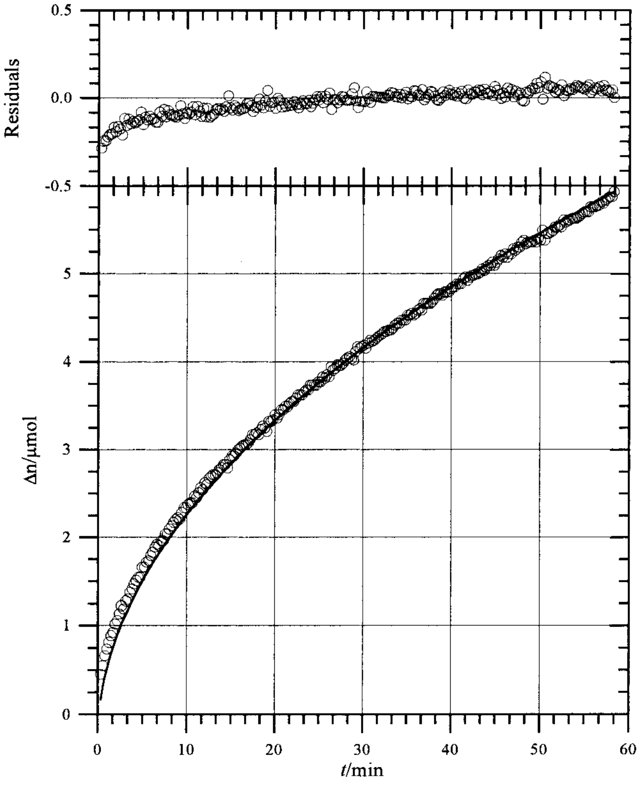

Figure 4. Absorption of CO_2 in 50 mass % total amine in water, 2:3 mole ratio DEA:MDEA at 298.2 K showing the measured values (○), values calculated from the numerical model (—), and the residual (top graph) error of the calculated from the experimental value.

suggest a relative uncertainty of approximately 12% for the reported diffusion coefficients.

The model fitted the data quite well as shown for a typical run in Figure 4. The values for the diffusion coefficients also follow the trends predicted by the Stokes–Einstein equation. For example, the diffusion coefficient of HCO_3^- increases as the temperature increases for a given composition as shown in Figure 5. The bicarbonate diffusion coefficient is reasonably constant with respect to the blend ratio. The value for the 1:4 DEA:MDEA blend is larger than the value for any of the other blends. This effect is reproduced at both temperatures studied in this work. Often diffusion coefficients can be correlated with the inverse viscosity of the mixture. This is true for the HS^- diffusivities in Figure 6 where viscosity is seen to increase with increasing mass fraction of MDEA, but the mixture diffusion coefficients for HCO_3^- are seen to be nearly constant with DEA/MDEA mole ratio (constant total amine) while the viscosity increases in Figure 5. Although the amine blends containing a greater proportion of DEA are more viscous, the change in viscosity is not very pronounced for the low concentrations of DEA studied here. The effect of increased viscosity due to additional DEA on calculated diffusivities is offset to some extent by the smaller size of the DEA molecules in the solvent and of the diffusing $DEAH^+$ ions.

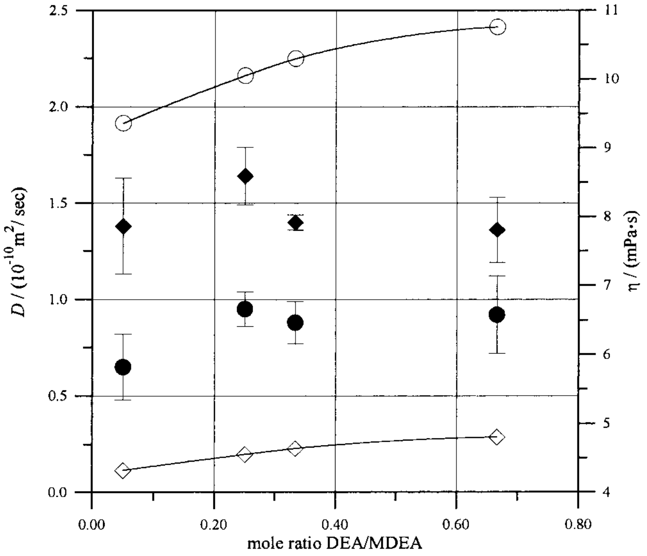

Figure 5. Diffusion coefficients for HCO_3^- (closed symbols) and mixture viscosity (open symbols) in 50 mass % total amine aqueous DEA/MDEA solutions as a function of composition. Viscosities were estimated using the Grunberg–Nissan equation and data from Teng et al. (1994). $T = 298.2$ K (●, ○); 318.2 K (◆, ◇). Lines are only smoothing curves for the viscosities.

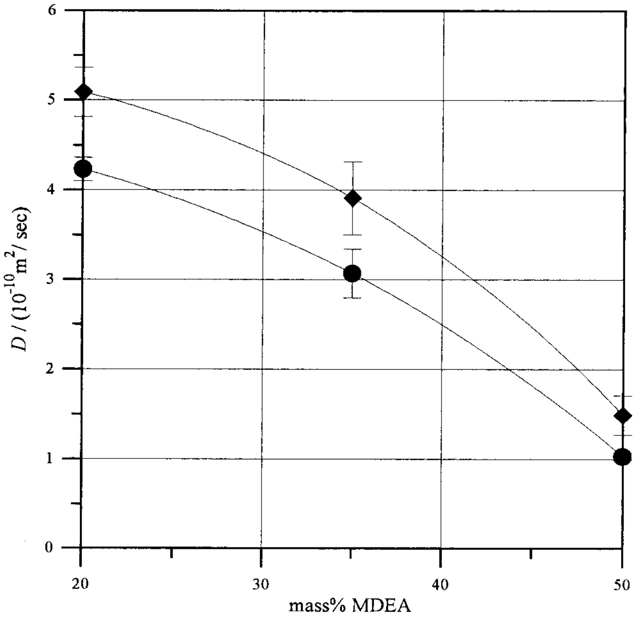

Figure 6. Diffusion coefficients for HS^- in aqueous MDEA solutions as a function of composition. $T = 25$ °C (●); 45 °C (◆).

As was the case with single amine solutions, CO_2 reacts rapidly in the blend, and the rate of absorption is controlled by the rate of diffusion of the CO_2-containing species away from the surface. The same was the case for H_2S absorption in aqueous MDEA.

Conclusions

A quiescent, inverted-tube diffusiometer was used to measure absorption rates of CO_2 in aqueous mixtures of MDEA and DEA and of H_2S in aqueous MDEA. The diffusiometer had been previously used for measurements in aqueous MDEA and aqueous DEA solutions. In this work, the apparatus was used to measure absorption rates of CO_2 into aqueous blends of MDEA and DEA at 298.2

and 318.2 K and 50 mass % total amine as a function of the ratio of amine concentrations, and of H_2S into aqueous MDEA solutions at 298.2 K and 318.2 K at amine concentrations of 20, 35, and 50 mass % MDEA. CO_2 absorption in the blends as well as H_2S absorption in MDEA is diffusion-controlled, but it is diffusion of the CO_2 and H_2S reaction products that limits the absorption rate. Absorption experiments should therefore be done in the actual reacting mixture, and any model developed for prediction of absorption rates must consider the diffusion of these reaction products away from the interface.

Using just such a numerical model, we were able to reduce absorption data of CO_2 into the aqueous alkanolamine blends and of H_2S into aqueous MDEA to obtain the diffusion coefficients of the main species. This required two additional, reasonable, but not rigorous, assumed relationships between the diffusion coefficients of these reaction products for the case of CO_2 absorption. The model developed contains absorption, reaction, and diffusion processes, and it correlated the measured CO_2 and H_2S absorption rates well. Values of the diffusion coefficient regressed from the measurements were of appropriate magnitude, exhibited the correct temperature and concentration dependence, and were consistent with expectations based on molecular size.

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