Measurement of the Absorption Rate of Carbon Dioxide into Aqueous Diethanolamine

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Absorption rates of gaseous CO_2 into aqueous diethanolamine (DEA) solutions were measured in a quiescent, inverted-tube diffusiometer by monitoring the rate of pressure drop. The absorption rate was found to be insensitive to the diffusion coefficient of CO_2 in solution but very sensitive to the diffusion rate of bicarbonate, protonated DEA, and carbamate ions. Evidence also suggested that chemical reaction equilibrium is rapid relative to diffusion. The diffusion coefficient of DEA in water was also measured using a Taylor dispersion apparatus. A numerical model was developed and used to regress diffusion coefficients of bicarbonate, carbamate, and protonated amine from measured absorption rates. CO_2 absorption rates and diffusion coefficients of bicarbonate, carbamate, and protonated DEA were obtained at 298.2 K and 318.2 K in solutions containing 20, 35, and 50 mass % DEA in water.

Introduction

Aqueous alkanolamine solutions are commonly used in natural gas sweetening processes to remove the acid gases CO_2 and H_2S. The reactions between the amines and acid gases enhance absorption and removal. Mixtures of amines such as *N*-methyldiethanolamine (MDEA) and diethanolamine (DEA) are often used to selectively absorb H_2S over CO_2. Economical and efficient process design for selective absorption of these acid gases requires accurate fundamental property values including gas solubility, diffusion coefficients, kinetic rate data, and heats of absorption. As part of a project to supply some of the required design data, we have been measuring absorption rates of acid gases in amine solutions to obtain mutual diffusion coefficients of species in solution important to the overall modeling of the absorption process. Recently, Rowley et al. (1997) (hereafter referred to as paper 1) reported absorption rates of CO_2 into aqueous MDEA solutions and diffusion coefficients of the aqueous species produced by the reactions. In this paper, we report the results of similar measurements made on the absorption of CO_2 into aqueous DEA solutions.

As discussed in paper 1, 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. Two common methods of circumventing the complexities of these systems are (1) to block the reactions so that only diffusion occurs and (2) to measure diffusion coefficients on an analogous system that does not react. For example, one could acidify the amine solutions to block the reactions or one could use N_2O as a nonreacting analogue to CO_2. In so doing, one not only changes the nature of the species or solution from that actually desired, but one may also eliminate important information about the absorption process. Such was found to be the case in paper 1. It had previously been assumed that an accurate measurement of the diffusion coefficient of CO_2 in aqueous MDEA was necessary to model the rate of CO_2 absorption in aqueous MDEA solutions. However, paper 1 showed that the reactions involved are rapid relative to the diffusion process and so there is very little aqueous CO_2 in solution. Rather, CO_2 is rapidly converted to HCO_3^-, and the rate-controlling step in the absorption of CO_2 is then the diffusion of HCO_3^- away from the gas/liquid interface. We therefore consider it important in developing an overall model for CO_2 absorption in amine solutions to determine insofar as is possible the diffusion coefficients of the species involved in the reacting−diffusing mixture. In this work, we report absorption rate data for the dissolution of CO_2 into aqueous DEA solutions. These data are important in analysis of the diffusion coefficients of important species in solution, and we report here the diffusion coefficients that we obtain from these data using a model similar to that previously reported in paper 1.

Chemicals

DEA was obtained from Aldrich Chemical Co. with a stated purity of 99%. No additional purification was attempted. To avoid contamination with CO_2 in the air, the DEA was initially transferred to smaller bottles in an Ar-filled glovebox. The smaller bottles were only opened and used once. Water was distilled, deionized, and degassed. The water was passed through a Shodex filter/degasser and then further degassed by boiling under vacuum until approximately 20% of the water had been removed. Carbon dioxide was obtained from Air Liquide and had a stated purity of 99.89%. All DEA + water mixtures were made gravimetrically and then stored under their own vapor pressure before introduction into an ISCO high-pressure micropump.

Measurement of the DEA + Water Mutual Diffusion Coefficient

The mutual diffusion coefficient of DEA in water was measured as a function of composition and temperature

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S0021-9568(97)00213-6 CCC: $15.00

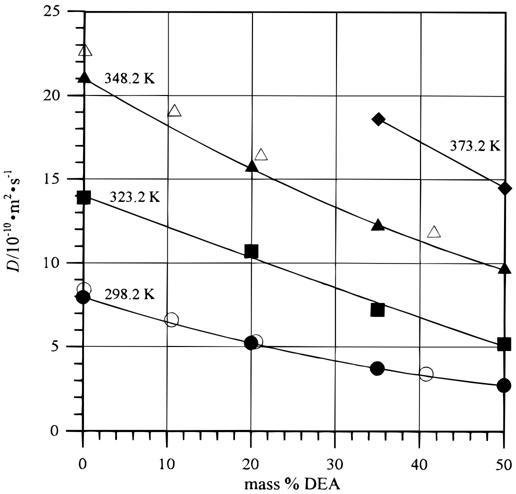

Figure 1. Mutual diffusion coefficients for DEA in water at 298.2 K (●), 323.2 K (■), 348.2 K (▲), and 373.2 K (◆). The corresponding open symbols are values reported by Snijder et al. (1993).

Table 1. Measured Diffusivities of DEA in Water

	$D/10^{-9}\cdot m^2\cdot s^{-1}$			
T/K	0 mass % DEA	20 mass % DEA	35 mass % DEA	50 mass % DEA
298.2	0.795 ± 0.008	0.523 ± 0.006	0.373 ± 0.002	0.274 ± 0.009
323.2	1.39 ± 0.02	1.07 ± 0.03	0.724 ± 0.002	0.544 ± 0.042
348.2	2.10 ± 0.02	1.57 ± 0.01	1.22 ± 0.02	0.963 ± 0.008
373.2			1.86 ± 0.03	1.45 ± 0.01

using the Taylor dispersion method. The specifications of the instrument, its use, and its accuracy have been previously reported (Rowley et al. 1988). The method used for measurement and analysis is the same as in paper 1. Operation and accuracy of the diffusiometer were checked by measuring infinite dilution diffusion coefficients of methanol in toluene, benzene, and water at 40 °C. The results of these measurements in comparison to published literature values indicate an uncertainty in the instrument of about 1%, which is consistent with previous calibrations and measurements made with it.

The mutual diffusion coefficients of DEA in water were measured at 0, 20, 35, and 50 mass % DEA and at 25, 50, and 75 °C. The higher concentrations of DEA (35 and 50 mass %) lowered the vapor pressure sufficiently so that measurements could also be made at 100 °C for these compositions. Generally, four replicate measurements were made at each temperature and composition, and the results were averaged. Table 1 contains the results of these measurements. The standard deviations from the replicates were used to obtain the 95%-probability confidence intervals shown in Table 1. From these data, we estimate the relative uncertainty of the measured diffusion coefficients to be less than 2%; from experience with the apparatus, we estimate the absolute uncertainty to be less than 3.5%. Mutual diffusion coefficients of DEA in water have also been measured by Snijder et al. (1993), and Figure 1 shows a comparison between those values and values measured in this work. The smoothing curves in the figure were obtained by fitting

$$D_{12}/(10^{-10}\ \mathrm{m^2\cdot s^{-1}}) = d_0 + d_1 w_{\mathrm{DEA}} + d_2 w_{\mathrm{DEA}}^2 \qquad (1)$$

to our experimental data. In this equation, w is mass fraction and values of the parameters d_i are given in Table 2.

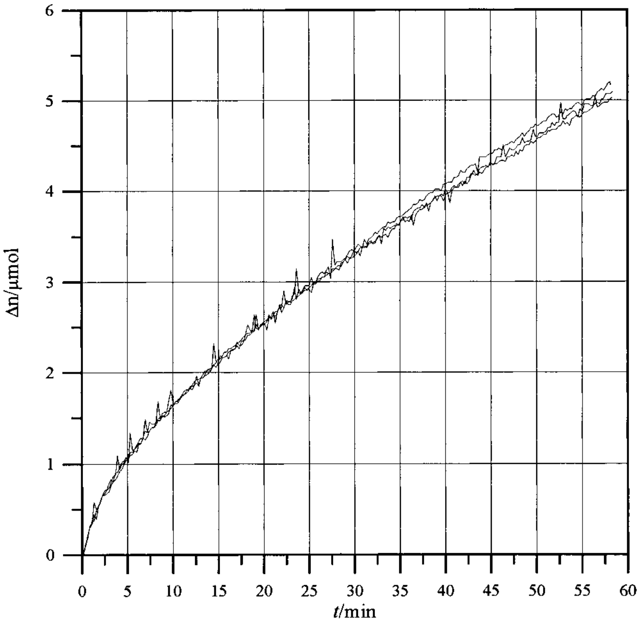

Figure 2. Reproducibility of CO_2 absorption runs for 20 mass % DEA and 80 mass % water at 298.2 K.

Table 2. Values of d_i for Use in Eq 1

T/K	d_0	d_1	d_2
298.2	7.951	−15.8	10.72
323.2	14.004	−19.3	3.66
348.2	21.021	−29.8	13.90
373.2	28.167	−27.3	0

Measurement of CO_2 Absorption

The absorption of CO_2 into aqueous DEA solutions was measured in an inverted tube (liquid on top, held up by capillary action) diffusion apparatus. The intent was to fit absorption rate data to the model by adjusting one or more diffusion coefficients. Absorption was measured by monitoring the decrease in pressure as a function of time after contact of the CO_2 (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 with CO_2 loading (Al-Ghawas et al., 1989). Details of the apparatus and experimental procedure were given previously in paper 1. Generally, four replicate experiments were performed at each temperature and composition. Figure 2 illustrates the typical consistency of the replicate measurements. Pressures were measured at approximately 20-s intervals for a period of 1 h. To conserve space, we report in Table 3 only the average, obtained from the replicates, number of micromoles CO_2 absorbed at 100-s intervals. But, because the moles of absorbed CO_2 is linear with the square root of time, as can be seen from Figure 3, and because replicate runs are quite consistent, as seen in Figure 2, little if any information is lost by using this abbreviated table of the raw data.

Analysis of Absorption Data

Analysis of the absorption data must be done in terms of a coupled transport and reaction model, including appropriate thermodynamic information about the mixtures. This is a difficult task, and many models have been proposed. Littel et al. (1991) have reviewed many of the models and have tested three numerical models of their own. These authors divide the models into two catego-

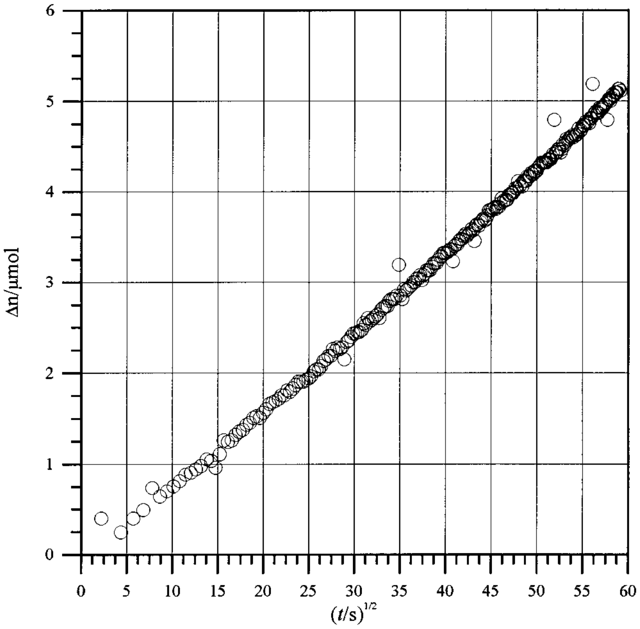

Figure 3. Typical raw absorption data from experiment exhibiting diffusion-controlled linearity when plotted versus the square root of time for 20 mass % DEA in water at 298.2 K.

ries: analytical models that are approximate because of the necessary simplifying assumptions and numerical models that may be more accurate but consume more computational time. Versteeg et al. (1989) indicate that because of the approximations in analytical models, their predictions should be checked with numerical models. Models by their very nature are not exact representations of the process, but a successful model must incorporate the important physics of the problem. Thus, the wide range of models that have been used to represent this complex physicochemical system have varied in complexity and in focus. For example, the models used by Glasscock and Rochelle (1989) focused heavily on the transport mechanisms, while the models studied by Littel et al. (1991) focused more on the ionic diffusivities of the species.

In paper 1, it was shown that for the case of CO_2 absorption in aqueous MDEA the reactions occurring in solution equilibrate rapidly relative to the rate of absorption. As was the case in paper 1, measured absorption rates of CO_2 in DEA were also linear with respect to $t^{1/2}$ as shown in Figure 3. This is evidence of rapid chemical equilibration and a diffusion-controlled mechanism. A parametric study of coupled diffusion with a first-order irreversible reaction indicated that for a specific rate constant less that about 5×10^{-4} s^{-1} the molar uptake of CO_2 is linear with $t^{1/2}$. Larger rate constants give ever increasing deviations from linearity with $t^{1/2}$. However, all of our measured absorption rates were linear with respect to $t^{1/2}$. This is obviously not an indication that the rate constants are small, because large amounts of absorption occurred, but rather an indication that the reactions are rapidly reversible rather than irreversible. We therefore used the model described in paper 1 to represent the absorption data. The model is fully described in the previous paper, and only details specific to the DEA system will be given here.

The solubility of CO_2 in aqueous DEA has been modeled by Kent and Eisenberg (1976). Experimental solubilities were quite accurately modeled in this study with the reversible reactions

$$\mathrm{DEA} + \mathrm{H}^+ \rightleftharpoons \mathrm{DEAH}^+ \tag{2}$$

$$\mathrm{R_2NH} + \mathrm{HCO_3}^- \rightleftharpoons \mathrm{R_2NCOO}^- + \mathrm{H_2O} \tag{3}$$

$$\mathrm{H_2O} + \mathrm{CO_2} \rightleftharpoons \mathrm{H}^+ + \mathrm{HCO_3}^- \tag{4}$$

$$\mathrm{H_2O} \rightleftharpoons \mathrm{H}^+ + \mathrm{OH}^- \tag{5}$$

$$\mathrm{HCO_3}^- \rightleftharpoons \mathrm{H}^+ + \mathrm{CO_3}^{2-} \tag{6}$$

where R is CH_2CH_2OH.

The equilibrium and Henry's law constants for eqs 2−6 were used directly as obtained from Kent and Eisenberg (1976) and Chakma and Meisen (1987), namely

$$K_j = \exp\left(\sum_{i=1}^{5} A_{ji} T^{1-i}\right) \quad j = 1, ..., 5 \tag{7}$$

$$H = \frac{1}{7.500\ 61} \exp\left(\sum_{i=1}^{5} A_{Hi} T^{1-i}\right) \tag{8}$$

with the constants given in Table 4. The equilibrium constants are K_c, not K_a, values, and they are functions of composition because mixture nonidealities are lumped into the K_c's. The model is not a full thermodynamic model with activity coefficients but was correlated specifically for this system and for the range of compositions studied here. We also checked this against a full thermodynamic model (Grimsrud, 1995) with activity coefficients that fully represents both solubility and heat of solution data over the range of compositions studied in this work. Similar results were obtained with both models.

It is of interest to analyze the absorption data in terms of species diffusion coefficients. To do so, we can only regress a single diffusion coefficient from the experimental absorption-rate data as a sensitivity analysis shows that coupling between the parameters occurs if more than one diffusion coefficient is treated as a parameter. The model indicates that diffusion coefficients of DEA, $DEAH^+$, R_2NCOO^-, HCO_3^-, H^+, OH^-, and CO_3^{2-} in aqueous solutions might be required, in addition to the diffusion coefficient of CO_2, to accurately model the absorption of CO_2 into aqueous DEA.

According to the model shown in eqs 2−6, there are nine species in solution: H^+, OH^-, H_2O, DEA, $DEAH^+$, CO_2, HCO_3^-, CO_3^{2-}, and R_2NCOO^-. The diffusion coefficient of CO_2 in amine solutions was used instead of the value in pure water. As in paper 1, the concentrations of H^+, OH^-, and CO_3^{2-} are everywhere so small that the CO_2 absorption rate is independent of the diffusion coefficients for these species; therefore, the diffusion coefficients for these ions were set to 1.0×10^{-9} m^2/s. This fact was verified by independently varying each diffusion coefficient over 4 decades, 2 decades on either side of the initial value. This sensitivity analysis indicated that the rate of absorption is essentially controlled by the diffusion rates of DEA, $DEAH^+$, HCO_3^-, and R_2NCOO^-. The mutual diffusion coefficient for DEA in water was measured as part of this work and the correlation given in eq 1 was used in the analysis. The values used were obtained from the N_2O-analogy measurements by Tamimi et al. (1994). This left the diffusion coefficients for $DEAH^+$, HCO_3^-, and R_2NCOO^- undefined. In paper 1 it was shown that to satisfy electroneutrality, the protonated amine and bicarbonate ion must diffuse at the same rate. In this case, there is

Table 3. Measured Absorption Rates of CO_2 into Aqueous DEA Solutions

	$\Delta n/\mu$mol					
	298.15 K			318.15 K		
t/s	20 mass % DEA, 120.66 kPa	35 mass % DEA, 108.38 kPa	50 mass % DEA, 109.44 kPa	20 mass % DEA, 117.60 kPa	35 mass % DEA, 116.11 kPa	50 mass % DEA, 111.75 kPa
91	0.463	0.352	0.644	0.998	1.052	0.507
191	0.803	0.639	1.045	1.394	1.447	0.848
289	1.060	0.884	1.318	1.722	1.765	1.075
387	1.243	1.144	1.554	1.998	2.066	1.213
487	1.437	1.262	1.757	2.244	2.330	1.379
585	1.683	1.436	1.936	2.475	2.573	1.485
670	1.764	1.653	2.082	2.688	2.786	1.637
768	1.877	1.696	2.243	2.886	2.993	1.771
866	2.160	1.837	2.384	3.079	3.185	1.899
965	2.211	1.974	2.549	3.253	3.376	2.023
1063	2.362	2.092	2.662	3.431	3.561	2.132
1148	2.550	2.252	2.752	3.595	3.728	2.235
1246	2.600	2.313	2.889	3.750	3.891	2.341
1346	2.776	2.497	3.030	3.914	4.043	2.436
1444	2.871	2.615	3.143	4.068	4.198	2.532
1542	2.990	2.714	3.261	4.218	4.358	2.617
1627	3.116	2.790	3.360	4.358	4.510	2.716
1725	3.229	2.912	3.469	4.503	4.640	2.818
1824	3.361	3.016	3.573	4.619	4.781	2.907
1922	3.449	3.148	3.686	4.754	4.919	3.002
2021	3.581	3.257	3.804	4.879	5.049	3.091
2105	3.682	3.351	3.856	5.005	5.187	3.158
2203	3.788	3.450	3.926	5.121	5.310	3.257
2303	3.902	3.544	4.068	5.246	5.440	3.342
2401	4.009	3.657	4.172	5.372	5.567	3.427
2500	4.134	3.733	4.247	5.483	5.675	3.508
2598	4.216	3.856	4.431	5.598	5.812	3.603
2682	4.304	3.917	4.487	5.705	5.918	3.663
2781	4.436	4.011	4.511	5.825	6.030	3.734
2879	4.493	4.058	4.591	5.927	6.142	3.833
2978	4.631	4.252	4.737	6.033	6.247	3.900
3076	4.719	4.280	4.775	6.134	6.360	3.954
3161	4.851	4.365	4.841	6.245	6.468	4.038
3260	4.882	4.440	4.935	6.351	6.580	4.095
3358	4.951	4.544	5.020	6.453	6.685	4.166
3457	5.065	4.615	5.101	6.540	6.787	4.243
3498	5.096	4.662	5.124	6.627	6.903	4.236

Table 4. Constants from Kent and Eisenberg (1976) and Chakma and Meisen (1987) Used in Eqs 7 and 8

	j				
coeff.	1	2	3	4	5
A_{1j}	−2.5510	−5 652.2	0	0	0
A_{2j}	4.8255	−1 884.8	0	0	0
A_{3j}	−241.818	298 253.0	$-1.485\,28 \times 10^{8}$	$3.326\,48 \times 10^{10}$	$-2.823\,94 \times 10^{12}$
A_{4j}	39.554	−98 790.0	$5.688\,28 \times 10^{7}$	$-1.464\,51 \times 10^{10}$	$1.361\,46 \times 10^{12}$
A_{5j}	−294.74	364 385.0	$-1.841\,58 \times 10^{8}$	$4.157\,93 \times 10^{10}$	$-3.542\,91 \times 10^{12}$
A_{Hj}	22.2819	−13 830.6	$6.913\,46 \times 10^{6}$	$-1.558\,95 \times 10^{9}$	$1.200\,37 \times 10^{11}$

one cation and two anions. It is not unreasonable to again make the assumption that the diffusion of the major anions is coupled with that of the predominant cation. We therefore make the approximation that the effective diffusion coefficients for $DEAH^+$, HCO_3^-, and R_2NCOO^- are equal; i.e., there is one coupled diffusivity for the ion pairs. This is not an exact electroneutrality relation as it was in paper 1, but it is a reasonable assumption based on electroneutrality arguments. For example, one of the models studied by Littel et al. (1991) assumed equal diffusivities of the ions present in solution to achieve electroneutrality. Certainly other models are possible and have been used in the literature.

The model thus incorporates the following assumptions: (a) the concentrations of all species satisfy the equilibrium constants for eqs 2−7 at all times and positions in the cell; (b) there is no resistance to mass transfer at the interface; (c) the Henry's law constant given by Chakma and Meisen (1987) applies; (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; (f) the absorption is insensitive to the values of the diffusion coefficients for H^+, OH^-, and CO_3^{2-}; (g) the diffusion coefficients for $DEAH^+$, HCO_3^-, and R_2NCOO^- are equal.

The model was applied to the analysis of the absorption experiments by discretizing the diffusion cell into equally spaced nodes beginning at the vapor−liquid interface and proceeding in the z-direction (direction of diffusion). Each node, j, had a volume of $V_j = \pi r^2 \Delta z$ associated with it except the first node, which had a volume of half that associated with it to ensure equal nodal spacing at the interface. The initial compositions of H^+, OH^-, $DEAH^+$, and DEA were set to their equilibrium values on the basis of nominal DEA composition with no CO_2 present, calculated using eqs 2−7. The initial concentrations for all other species were set to 0.0 mol·L^{-1}. The saturation concentration of CO_2 was

Table 5. Diffusion Coefficients for $DEAH^+-HCO_3^-$/ R_2NCOO^- Complexes Obtained from CO_2 Absorption Rate Data and the Model Developed in This Work

	$D/10^{-10}\cdot m^2\cdot s^{-1}$	
mass % DEA	$T = 298$ K	$T = 318$ K
20	1.56 ± 0.06	4.50 ± 0.05
35	0.41 ± 0.05	1.97 ± 0.04
50	0.39 ± 0.05	0.39 ± 0.04

obtained using the Henry's law expression in eq 8 and the experimental CO_2 pressure. An accumulator was initialized to the amount of CO_2 absorbed. Computations then proceeded in the following order for each time step:

1. Mass was transferred away from the interface to the next node on the basis of the current concentrations at a node, *j*, using the forward finite difference representation of the flux equation for that component; i.e.

$$\Delta C_{j+1} = -\frac{DA(C_{j+1} - C_j)\Delta t}{V_j \Delta z} \tag{9}$$

where *t* is time, *A* is the cross-sectional area of the diffusion cell, *D* is the binary mutual diffusion coefficient for the species in question, and C_j is the molarity of that species at node *j*.

2. Chemical equilibrium was then established at these two nodes by solving eqs 2–7 using a series-reactor technique. In this way, chemical equilibrium was represented as occurring instantaneously with diffusion as the rate-controlling step.

3. Steps 1 and 2 were repeated for each two nodes until there was no significant gradient between neighboring nodes.

4. The concentration of CO_2 at the interfacial node was again set to the saturation value and eqs 2–8 were again solved for the concentrations at the interfacial node. The amount of CO_2 absorbed during that time step was taken as the difference between α at this time step and its previous value, where α, the total amount of CO_2 in solution in its various forms, is given by

$$\alpha = [CO_2] + [HCO_3^-] + [CO_3^{2-}] + [R_2NCOO^-] \tag{10}$$

The amount of CO_2 absorbed during each time step was then added to the accumulator. Additional details about the stability of the method and testing are given in paper 1.

Discussion of Results

The values of the diffusion coefficient for $DEAH^+$, HCO_3^-, and R_2NCOO^- obtained from the absorption measurements are given in Table 5 along with their uncertainties. Because the diffusion coefficients are inferred from a model, it is difficult to assess their absolute accuracy, so the uncertainties reported were calculated by a nonlinear, weighted least-squares program using the variance–covariance matrix and the pairwise partial correlation coefficients. The model represents the absorption data quite well in every case, as illustrated in Figure 4. As one would expect, the values at 318.2 K are larger than those at 298.2 K. Figure 5 shows that the diffusion coefficient decreases with increasing amine concentration which is consistent with the corresponding increase in solution viscosity. A comparison of Tables 1 and 5 reveals that the diffusion coefficient for the $DEAH^+ - HCO_3^-/R_2NCOO^-$ complex is smaller than that for DEA in water. This is explicable in terms of the much larger size and mass of the complex (than the DEA molecule itself) that must diffuse together to maintain local electroneutrality.

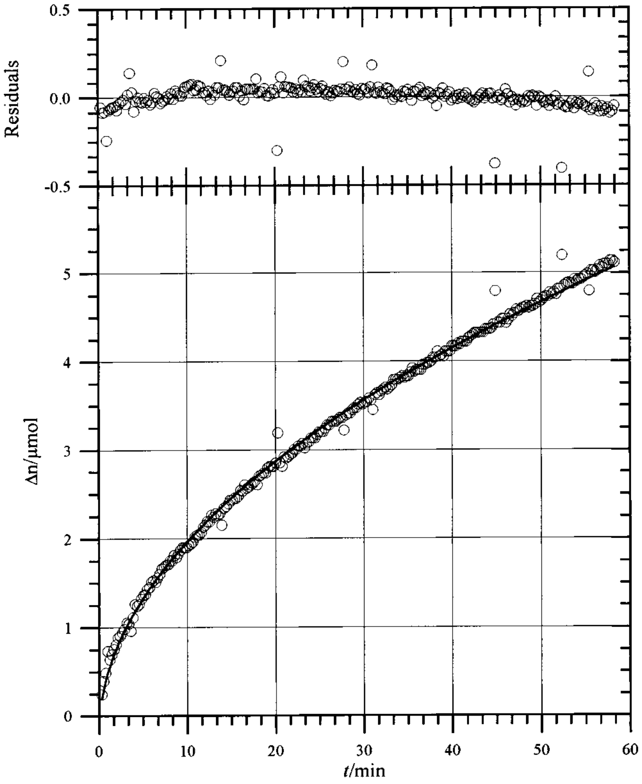

Figure 4. Absorption of CO_2 in 20 mass % DEA in water at 298.2 K showing the measured values (○), values calculated from the numerical model (—), and the residual (top graph) error of the calculated value from the experimental value.

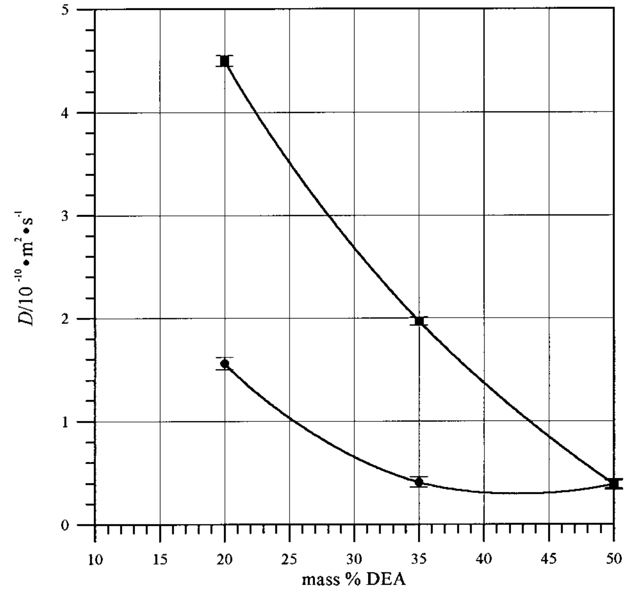

Figure 5. Concentration dependence of the $DEAH^+/HCO_3^-/R_2NCOO^-$ diffusion coefficients at 298.2 K (●) and at 318.2 K (■) with smoothed fits of data.

The significance of this work in modeling the reaction–absorption process is seen in that the CO_2 reacts rapidly and the rate of absorption is actually controlled by the rate of diffusion of the ionic species (bicarbonate and carbamate) away from the surface. Concentration profiles of the

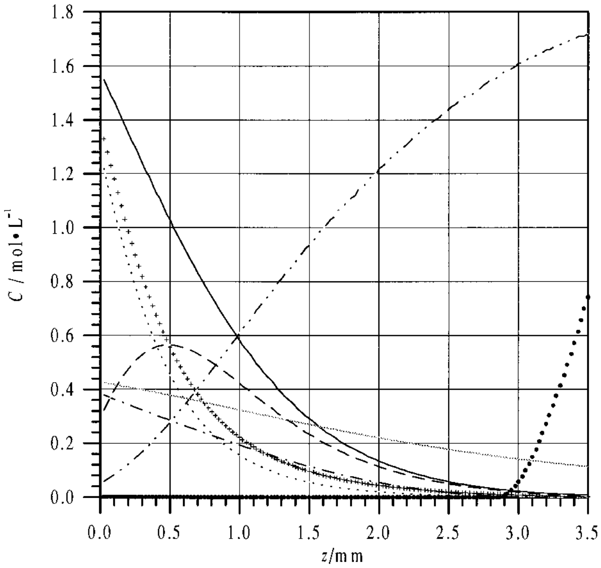

Figure 6. Predicted molar concentrations of species in absorption cell after 1 h (at 298.2 K in 20 mass % DEA solution) for [$DEAH^+$] (—); [HCO_3^-] (---); [DEA] (••—); [R_2NCOO^-] (— —); 1000[OH^-] (•••); 10^8[H^+] (+++); 10[CO_2]$_{aq}$ (•—•); 100[CO_3^{2-}] (gray solid line).

various species, as calculated from the model, are shown as a function of spatial position in Figure 6.

Conclusions

A quiescent, inverted-tube diffusiometer was used to measure absorption rates of CO_2 in aqueous DEA solutions. The diffusiometer had been previously used for measurements in aqueous MDEA solutions. In this work, the apparatus was used to measure absorption rates of CO_2 into aqueous solutions of DEA at 298.2 and 318.2 K as a function of DEA concentration. It was found that CO_2 absorption was diffusion-controlled and that it cannot be modeled with irreversible reactions. This supports a model in which reversible reactions equilibrate rapidly relative to diffusion.

Using a numerical model based on the above findings, we were able to reduce the absorption data in terms of a single diffusion coefficient for the $DEAH^+$, HCO_3^-, and $(HOCH_2CH_2)_2NCOO^-$ ions. The diffusion coefficient of DEA in water was measured with a Taylor dispersion apparatus as a function of temperature and DEA concentration for inclusion in the model. The model for absorption, reaction, and diffusion correlated the measured CO_2 absorption rates well. Values of the diffusion coefficient for $DEAH^+/HCO_3^-/(HOCH_2CH_2)_2NCOO^-$ 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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Received for review September 8, 1997. Accepted February 13, 1998. Financial support for this work by the Gas Research Institute (GRI) and the Gas Processors Association (GPA) under GRI contract 5092-260-2356 is gratefully acknowledged.

JE970213C