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Abstract: The chemical absorption rate (R_A) of CO_2 was measured into the aqueous nanometer sized colloidal silica solution of 0–31 wt% and diethanoleamine of 0–2 kmol/m^3 in the flat-stirred vessel with the impeller size of 0.034 m and its agitation speed of 50 rev/min at 25°C and 0.101 MPa, and compared with the values estimated from the model based on the film theory accompanied by chemical reaction. The value of the volumetric liquid-side mass transfer coefficient (k_La) of CO_2, which was used to estimate the value of R_A, was obtained by the empirical correlation formula presenting the relationship between k_La and the rheological behavior of the aqueous colloidal silica solution. The value of R_A in the aqueous colloidal silica solution was decreased by the reduction of k_La due to the elasticity of the solution.

Keywords: Chemical absorption, carbon dioxide, colloidal silica, viscoelastic liquid, diethanolamine

INTRODUCTION

In multiphase systems appearing in agitated reactors, gas-liquid mass transfer may be rate-determining step for the overall process. Therefore, the knowledge of gas-liquid mass transfer rates characterized by the volumetric

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liquid-phase mass transfer coefficient (k_La) is needed for a reliable design of such reactors and many researchers (1) have studied how to enhance the mass transfer rate.

The techniques for the enhancement of heat and mass transfer performance are generally categorized into several methods; the mechanical treatment, the chemical treatment, the acoustic (2) and electromagnetic (3) treatments, and the application of nano technology. The selection of heat transfer mode such as falling film type and bubble type is a kind of mechanical treatment. The representative chemical treatment is the addition of a surfactant into the working fluids. The addition of the surfactant causes interfacial turbulence, which leads to a higher heat and mass transfer performance (4). In the recent decade, the nanofluid becomes one of the most attractive heat transfer media due to the development of nano technology. Nanofluid is the solid/liquid mixture in which nano-sized particles($d_p < 100$ nm) are suspended evenly in the base liquid. It is well known (5) that the nanofluid can enhance the effective thermal conductivity and affects the heat transfer characteristic of the fluid. One suggested four potential mechanisms (5); Brownian motion of the particles, liquid layering at liquid/particle interface, nature of heat transport in nano particles, and effects of nano particle clustering. To the best of our knowledge, the studies on the effect of nanofluid on the mass transfer performance have not been conducted although some researchers (5) have actively studied the heat transfer enhancement by nanofluid. Kim et al. (6) measured the absorption rate of NH_3 in nanofluid of Cu, CuO, and Al_2O_3 of 50 nm in water using a bubble absorber, and they showed that the absorption rate increased with increasing concentration of nano particles.

However, in the slurry or colloidal systems, the effects of milli or micro particles on the absorption have been studied by many researchers (7–15). Absorption of gas into slurries constituted by fine particles is fairly common as a means of intensifying gas absorption rates and even for improving selectivity in the case of multiple gaseous solutes. Improvement of the mass transfer rate by fine particles was explained by the mechanism of the grazing effect, first discovered by Kars and Best (7). Zhou et al. (15) reviewed the effect of fine particles on multiphase mass transfer and concluded that the finer the particles in the slurries were, the stronger the influence was, and they reported that the mass transfer coefficient (k_L) might be increased or reduced via changing film thickness with turbulence or a lowering of the diffusion coefficient of gas. The reduction of the solubility and the diffusivity of dissolved gas in slurries with increasing volume fraction of slurries may be due to the decrease (9) in the portion of continuous phase in the slurries. As increasing of volume fraction of slurries, the covered gas-liquid interface by the fine particles can hinder diffusion of gas and hence reduce k_L. If the nanofluid was treated as the pseudohomogeneous phase (13), in which the diffusion of the solutes as well as the gas-liquid interfacial area are assumed to be unaltered, the hydrodynamics of the nanofluid might be used to predict the gas absorption rate in the nanofluid/gas system.

The dependence of shear stress on shear rate of a fluid in hydrodynamic system is different according to the type of the fluid, i.e., Newtonian or non-Newtonian fluid, and the mass transfer coefficient (k_L) of a solute in one phase is in inverse proportion to the viscosity of its phase due to the inverse proportion of viscosity to diffusivity. Deborah number (De), which is one of the rheological properties and defined as the ratio of the characteristic material time to the characteristic process time, is taken into account to correlate the mass transfer coefficient in the non-Newtonian fluid with that in the Newtonian one.

Only use of the apparent viscosity of the non-Newtonian fluids was not sufficient to obtain a unified correlation for k_La values. Due to the complexities of gas absorption in the non-Newtonian media, the correlations obtained by these studies were limited to just a few kinds of non-Newtonian fluids such as Carbopol, carboxymethylcellulose (CMC), polyacrylate (PA), polyethylene oxide (PEO), polyacrylamide (PAA), and polyisobutylene (PIB) solutions. If a considerable reduction of k_La is due to the viscoelasticity of the aqueous solution (16, 17), then the extent to which data for the viscoelastic solution such as PAA deviate from those for the inelastic solution such as CMC should correlate with some measure of the solution's elasticity. The dimensionless number such as De, which relates the elastic properties with the process parameters, is used to correlate k_La with properties of non-Newtonian liquids. Unified correlations have been proposed for k_La in Newtonian as well as non-Newtonian solutions by introducing the dimensionless term such as $(1 + n_1 De^{n_2})^{n_3}$, which are listed in Table 1. As shown in Table 1, values of in the dimensionless group are different one another.

Park et al. presented the effect of elasticity of polyisobutylene (PIB) (20) in the benzene solution of polybutene (PB) and PIB on the absorption rate of

Table 1. Coefficients of dimensionless group for gas-liquid mass transfer correlation

Investigator	n_1	n_2	n_3	Substance	Contactor
Nakanoh and Yoshida (17)	0.13	0.55	−1	CMC, PAA	Bubble column
Yagi and Yoshida (18)	2	0.5	−0.67	CMC, PA	Agitated vessel
Ranade and Ulbrecht (19)	100	1	−0.67	CMC, PAA	Stirred tank
Park et al. (20)	100	1	−0.42	PB, PIB	Agitated vessel
Park et al. (21)	2461.3	1	−0.274	PB, PIB	Agitated vessel
Park et al. (22)	54.7	1	−0.45	PAA	Agitated vessel
Park et al. (23)	8.33	1.31	1	PEO	Agitated vessel
Park et al. (24)	39.4	1	0.43	Silica	Agitated vessel

CO_2, and that (21) in w/o emulsion composed of aqueous solution as dispersed phase and benzene solution of PB and PIB as continuous phase in an agitation vessel. They showed that PIB accelerated the absorption rate of CO_2. Also, the effect of PAA (22) and PEO (23) in an aqueous solution on the absorption rate of CO_2 were investigated. The polymers used in their papers act as accelerators of the absorption rate of CO_2 in the non-Newtonian viscoelastic liquid based on the same viscosity of the solution.

There is little information about the effect of rheological properties of nano-sized particles in aqueous phase on physical and chemical absorption of gas in non-Newtonian liquid, then, it is worthwhile to investigate the rheological behavior of nano-sized particles in the absorption of CO_2.

In this study, the absorption rate of CO_2 was measured into aqueous nano-sized colloidal silica solution with diethanoleamine(DEA) to observe the effect of elasticity of the solution on the chemical absorption rate of CO_2, and compared with the theoretical value, which was estimated from the model based on the film theory accompanied by chemical reaction using the k_La, which was obtained by the empirical equation (24) for k_La of CO_2 in aqueous nano-sized colloidal silica solution.

THEORY

The problem to be considered is that a gaseous species A (CO_2) dissolves into the liquid phase, and then, reacts irreversibly with species B(DEA) according to

$$CO_2 + \nu R_2NH \longrightarrow R_2NCO_2^- + R_2NH_2^+ \qquad (1)$$

where R represents the functional groups on the secondary amine.

The stoichiometric coefficients (ν)) in Eq. (1) for DEA was obtained from the reference (25) and its value was 2.

Species B is a nonvolatile solute, which has been dissolved into the liquid phase prior to its introduction into the gas absorber. It is assumed that gas phase resistance to absorption is negligible by using pure species A, and thus the concentration of species A at the gas-liquid corresponds to equilibrium with the partial pressure of species A in the bulk gas phase.

The chemical reaction of Eq. (1) is assumed to be second-order as follows:

$$r_A = k_2 C_A C_B \qquad (2)$$

Under the assumptions mentioned above, the mass balances of species A and B from the film theory with chemical reaction are given as

$$D_A \frac{\partial^2 C_A}{\partial z^2} = k_2 C_A C_B \qquad (3)$$

$$D_B \frac{\partial^2 C_B}{\partial z^2} = \nu k_2 C_A C_B \tag{4}$$

Boundary and conditions to be imposed are

$$z = 0, C_A = C_{Ai}, \frac{dC_B}{dz} = 0 \tag{5}$$

$$z = z_L, C_A = 0, C_B = C_{Bo} \tag{6}$$

Eq. (3)–(6) are put into the dimensionless form as follows:

$$\frac{\partial^2 a}{\partial x^2} = \text{Mab} \tag{7}$$

$$\frac{\partial^2 b}{\partial x^2} = \nu \text{rq ab} \tag{8}$$

$$x = 0;\ \ a = 1, \frac{db}{dx} = 0 \tag{9}$$

$$x = 1;\ \ 1a = 0, b = 1 \tag{10}$$

where $M = D_A k_2 C_{Bo}/k_L^2$, $a = C_A/C_{Ai}$, $b = C_B/C_{Bo}$, $x = z/z_L$, $q = \nu C_{Ai}/C_{Bo}$, $r = D_A/D_B$.

The enhancement factor (β) here defined as the ratio of molar flux with chemical reaction to that without chemical reaction is described as follows:

$$\beta = -\left.\frac{da}{dx}\right|_{x=0} \tag{11}$$

The absorption rate(R_A) of CO_2 with chemical reaction can be predicted as follows:

$$R_A = \beta\, R_{Ao} = \beta\, k_L a C_{Ai} V_L \tag{12}$$

where R_{Ao} is the absorption rate of CO_2 without chemical reaction.

EXPERIMENTAL

Chemicals

All chemicals in this study were reagent grade, and used without further purification. Purity of both CO_2 and N_2 was more than 99.9%. Ludox HS-40 suspension(Aldrich chemical company, U.S.A.) having a 40% w/w solid content(silica density 2200 kg/m^3) was used. The average particle radius, the specific surface area, and PH at 298 K are 12 nm, 220 m^2/g, 9.8, respectively, as indicated by the manufacturer. It was reported from Aldrich materials science catalog that the Ludox products are not used as an adsorbent. An aqueous solution of colloidal silica was made by diluting Ludox HS-40 with distilled water and neutralized with aqueous diluted HCl to remove the chemical reaction with CO_2.

Absorption Rate of CO_2

The gas-liquid contactor used was a stirred tank made of glass (10.2 cm inside diameter, 15.1 cm in height) having a planar unbroken gas-liquid interface; it was operated continuously with respect to the gas and batch-wise with respect to the liquid phase. Four equally spaced vertical baffles, each one-tenth of the vessel diameter in width, were attached to the internal wall of the vessel. The contact area between the gas and the liquid was measured as $8.047 \times 10^{-3}\ m^2$. The liquid phase was agitated using an agitator driven by a 1/4 Hp variable speed motor without agitation in gas phase, because it was pure CO_2 gas. A straight impeller (3.4 cm length, 1.7 cm width, and 0.5 cm thickness) was used as the liquid phase agitator; it was located at the middle position of the liquid phase of $0.833\ dm^3$. The absorption rate of CO_2 was measured in the aqueous colloidal silica solution along the procedure similar to that reported elsewhere (24) at 0.101 MPa and 25°C, in which silica concentration of $0 \sim 31$ wt% and concentration of DEA of $0-2\ kmol/m^3$ were varied at agitation speed of impeller of 50 rev/min.

PHYSICOCHEMICAL AND RHEOLOGICAL PROPERTIES

The physicochemical and rheological properties of the aqueous colloidal silica solution, which is assumed to be nanofluid of the pseudohomogeneous phase (13), were obtained as follows:

Solubility of CO_2 in the Aqueous Colloidal Silica Solution

The pressure measuring method was used by measuring the pressure difference of CO_2 between before and after equilibrium between gas and liquid phase similar to the procedure reported elsewhere (26) to get the solubility of CO_2 in the aqueous colloidal silica solution at 25°C and 0.101 MPa. The experimental procedure was duplicated that as reported in the published research (24) in detail.The solubility (C_{Ai}) of CO_2 in aqueous DEA solution was estimated as follows (25):

$$\mathrm{Log}(C_{Ai}/C_{Aw}) = -(1.0406 \times 10^{-4} + 6.8433 \times 10^{-6} C_{Bo} + 1.33633 \times 10^{-8} C_{Bo}^2 - 1.1549 \times 10^{-12} C_{Bo}^3)$$

Density and Apparent Viscosity of the Aqueous Colloidal Silica Solution

The density of the aqueous silica colloid solution was measured at 25°C within $0.1\ \mathrm{kg/m^3}$ by weighing with a pycnometer (Fisher Scientific Co., USA) and was found to be identical within experimental accuracy to the density of water. The apparent viscosity of the aqueous silica colloid solution was measured at 25°C with a Brookfield viscometer (Brookfield Eng. Lab. Inc, USA).

Diffusivities of CO_2 in the Aqueous Colloidal Silica Solution

Diffusivity (D_{AB}) of CO_2 in aqueous DEA (27) solution was estimated as follows:

$$D_{AB} = (1.9886 - 0.8103C_{Bo} - 0.1771\ C_{Bo}^2) \times 10^{-9} \qquad (13)$$

Diffusivity (D_A) of CO_2 in the aqueous silica colloid solution was estimated from the following equation (28) corrected with the viscosity of the aqueous colloidal silica solution.

$$D_A = D_{AB}(\mu_w/\mu)^{2/3} \qquad (14)$$

Diffusivity (D_B) of DEA in the aqueous amine solution was obtained from the assumption that the ratio of D_B to D_A was equal to the ratio in water (29). The diffusivity of CO_2 and DEA in water at 25°C were taken as $1.97 \times 10^{-9}\ \mathrm{m^2/s}$ (30) and $6.67 \times 10^{-10}\ \mathrm{m^2/s}$ (27), respectively. The obtained values of the solubility, the diffusivity of CO_2, the density, and the apparent viscosity of the aqueous silica colloid solution are given in Table 2.

Reaction Rate Constant of CO_2 with DEA

The reaction rate constant (k_2) between the reaction of CO_2 and DEA in aqueous solution was obtained from the following equation (27).

$$\log k_2 = 10.4493 - \frac{2274.5}{T} \qquad (15)$$

Rheological Properties of the Aqueous Colloidal Silica Solution

Material parameters of K, n, A and b in a power-law model such as $\tau = K\gamma^n$ and $N_1 = A\gamma^b$ were obtained from the measurement of τ and N_1 for the change of γ by the parallel disk type rheometer (Ares, Rheometrics, U.S.A.) of the diameter of 0.05 m and the gap of 0.001 m. The obtained values of K, n, A and b of various concentration of silica are give in Table 2.

Table 2. Physicochemical and rheological properties of CO_2 and aqueous colloidal silica solution

Silica (wt%)	Viscosity $(Ns/m^2) \times 10^3$	Diffusivity $(m^2/s) \times 10^9$	Solubility $(kmol/m^3)$	Density (kg/m^3)	Rheological properties			
					n	$K \times 10^3\ (Ns^n/m^2)$	b	$A\ (Ns^n/m^2)$
0	1	1.97	0.039	1000	1.0	1.0	–	–
5.8	1.075	1.88	0.032	1039	0.97	1.115	0.097	0.03
11.91	1.127	1.82	0.031	1082	0.94	1.321	0.160	0.057
18.35	1.191	1.77	0.029	1134	0.92	1.525	0.190	0.082
23.06	1.235	1.72	0.028	1160	0.9	1.67	0.217	0.1
30.85	1.274	1.68	0.027	1180	0.85	1.95	0.232	0.129

Empirical Correlation Formula for Volumetric Liquid-Side Mass Transfer Coefficient of CO_2

The value of liquid-side mass transfer coefficient (k_L) was obtained from Eq. (12) presenting the relationship between k_La and rheological behavior of the aqueous colloidal silica solution (24) such as follows:

$$k_La d^2/D_A = 12.56(d^2N\rho/\mu)^{0.48}(\mu/\mu_w)^{0.11}(1+39.4De)^{-0.43} \tag{16}$$

where De is defined as the ratio of the characteristic material time to the characteristic process time as follows:

$$De = \lambda/t = \frac{A}{K}\gamma^{b-n-1}N \tag{17}$$

where the shear rate (γ) is obtained in case of agitation of the liquid in a cylindrical vessel as follows (31):

$$\gamma = 4\pi N/n \tag{18}$$

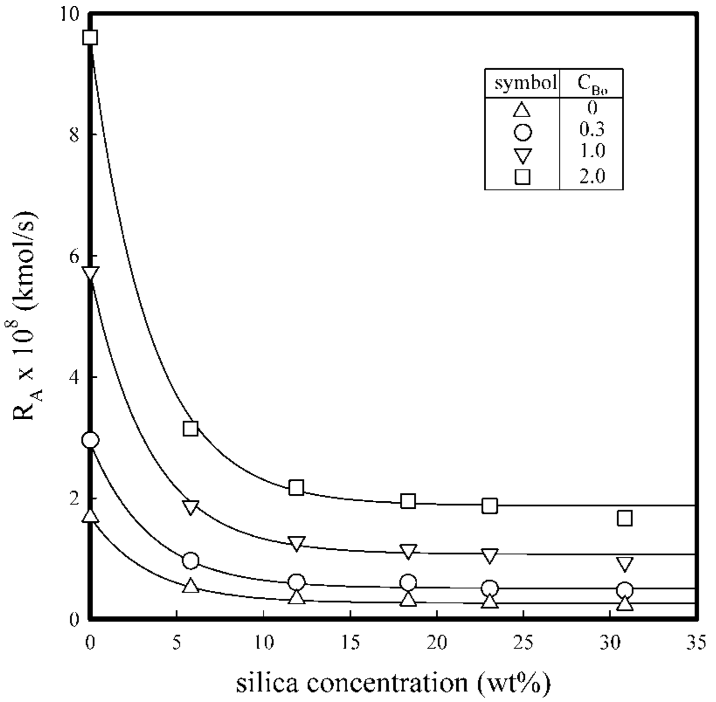

Figure 1. Effect of silica concentration on absorption rate for various DEA concentrations.

RESULTS AND DISCUSSION

To observe the effect of elasticity of the aqueous colloidal silica solution on the chemical reaction of CO_2 with DEA, the absorption rate of CO_2 into the aqueous colloidal silica solution with DEA was measured according to the change of silica concentration in the range of 0–31 wt% at DEA concentration $0 \sim 2\ \mathrm{kmol/m^3}$ under the experimental conditions of the agitation speed of 50 rev/min with the impeller size of 0.034 m.

Figure 1 shows the plots of the absorption rate of CO_2 against the silica concentration for various DEA concentrations. As shown in Fig. 1, R_A decreases with increasing silica concentration, and increases with increasing DEA concentration.

To explain the trend of R_A as shown in Fig. 1, the dependence of variables such as k_La, β, and C_{Ai} on R_A is studied as mentioned below:

The values of k_La were obtained from Eq. (16) using the physicochemical and rheological properties as listed in Table 2 in the range of the silica concentration of 0–31 wt%, and plotted in Fig. 2. As shown in Fig. 2, k_La decreases with increasing the silica concentration. The solid line in Fig. 2 is the value of k_La calculated from Eq. (16), and the symbols with the open circle are the

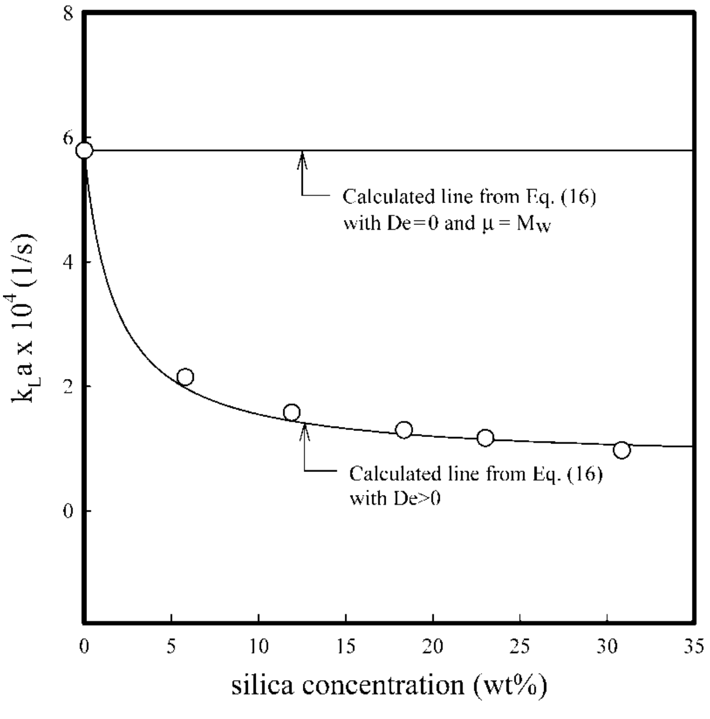

Figure 2. Effect of silica concentration on k_La at d = 0.034 m and N = 50 rpm.

experimental values. As shown in Fig. 2, the experimental values approach the calculated values very well. This result comes from the fact that the viscosity and De of the aqueous colloidal silica solution increase with increasing the concentration of silica as shown in Eq. (16) and Table 2.

The numerical solutions of the simultaneous differential equations (7) and (8) were obtained by the finite element method at given concentrations of silica and DEA using the physicochemical properties listed in Table 2 and k_L from Eq. (16), from which the value of β was estimated from Eq. (11). Figure 3 shows the plots of β against the silica concentration for various DEA concentrations. As shown in Fig. 3, the values of β hold to be constant with increasing silica concentration and increase with increasing the DEA concentration.

The value of C_{Ai} decreases with increasing silica concentration as shown in Table 2.

From Eq. (12) and the results in Figs. 2, 3, and Table 2, it might be said that decrease of R_A with increasing silica concentration was caused mainly by k_La rather than elasticity of the aqueous colloidal silica solution.

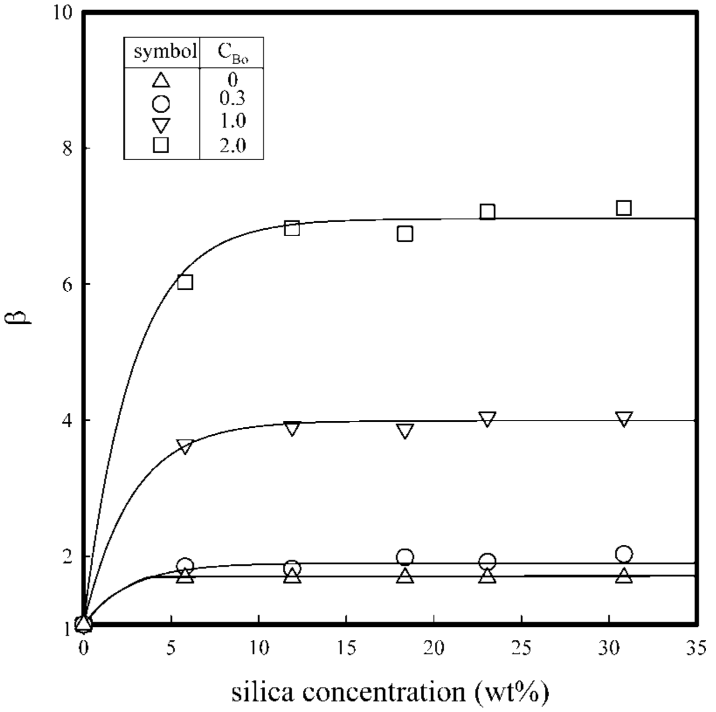

Figure 3. Effect of silica concentration on enhancement factor for various DEA concentrations.

CONCLUSIONS

The measured rate of chemical absorption of carbon dioxide into the aqueous colloidal silica solution of 0–31 wt% with DEA of 0–2 kmol/m^3 in a flat-stirred vessel with the impeller size of 0.034 m and its agitation speed of 50 rev/min at 25°C and 0.101 MPa was compared with that estimated from the model based on the film theory accompanied by chemical reaction using the value of k_La. The chemical absorption rate was decreased due to the reduction effect of k_La by elasticity of the aqueous colloidal silica solution The value of k_La, which was used to estimate the enhancement factor, was obtained from the empirical equation as follow:

$$k_L a d^2/D_A = 12.56(d^2 N\rho/\mu)^{0.48}(\mu/\mu_w)^{0.11}(1 + 39.4De)^{-0.43}$$

NOMENCLATURE

a	specific gas-liquid area (1/m)
C_i	concentration of species, i (kmol/m^3)
d	diameter of impeller (m)
De	Deborah number defined as λ/t
D_i	diffusivity of species, i (m^2/s)
k_2	reaction rate constant in reaction (1) (m^3/kmol s)
k_L	liquid-side mass transfer coefficient of CO_2 in absorbent (m/s)
N_1	primary normal stress difference (kg/m · s^2)
V_L	volume of the liquid phase (m^3)
k_L	liquid-side mass transfer coefficient of CO_2 in absorbent (m/s)
r_A	reaction rate in Eq. (2) (kmol/m^3 · s)
R_A	chemical absorption rate of CO_2 (kmol/s)
R_{Ao}	physical absorption rate of CO_2 (kmol/s)
t	the characteristic process time (sec)
T	temperature (°K)
z	diffusion coordinate of CO_2 (m)
z_L	film thickness (m)

Greek Letters

γ	shear rate (1/s)
λ	the materials' characteristic relaxation time (sec)
μ	viscosity of liquid (Ns/m$^{2)}$
μ_w	viscosity of water (Ns/m^2)
ρ	density of liquid (kg/m^3)
τ	shear stress (N/m^2)

Subscripts

A	CO_2
B	DEA
i	gas-liquid interface

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