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Mass transfer of carbon dioxide in aqueous colloidal silica solution containing *N*-methyldiethanolamine

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Abstract−The absorption rate (R_A) of carbon dioxide was measured into an aqueous nanometer sized colloidal silica solution of 0-31 wt% and *N*-methyldiethanolamine of 0-2 kmol/m^3 in a flat-stirred vessel for the various sizes and speeds of at 25 °C and 0.101 MPa to obtain the volumetric liquid-side mass transfer coefficient (k_La) of CO_2. The film theory accompanied by chemical reaction between CO_2 and *N*-methyldiethanolamine was used to estimate the theoretical value of absorption rate of CO_2. An empirical correlation formula containing the relationship between k_La and rheological property of the aqueous colloidal silica solution was presented. The value of R_A in the aqueous colloidal silica solution was decreased by the reduction of k_La due to elasticity of the solution.

Key words: Absorption, *N*-methyldiethanolamine, Carbon Dioxide, Silica, Viscoelastic Liquid

INTRODUCTION

In multiphase systems appearing in agitated reactors, the gas-liquid mass transfer may be rate-determining step for the overall process. Therefore, a knowledge of gas-liquid mass transfer rates characterized by the volumetric liquid-phase mass transfer coefficient (k_La) is needed for 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: mechanical treatment, chemical treatment, acoustic [2] and electromagnetic [3] treatments, and 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 surfactant into the working fluids. The addition of surfactant causes interfacial turbulence, which leads to a higher heat and mass transfer performance [4]. In recent decades, nanofluids have become among the most attractive heat transfer media due to the development of nano technology. A nanofluid is a 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 characteristics of fluids. One researcher has 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, 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 bubble absorber, and they showed that the absorption rate increased with increasing concentration of nano particles.

However, in a slurry or colloidal systems, the effects of milli or micro particles on the absorption have been studied by many researchers [7-9]. 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 the fine particles was explained by the mechanism of the grazing effect, first discovered by Kars and Best [7]. Zhou et al. [9] reviewed the effect of fine particles on multiphase mass transfer and concluded that the finer the particles in the slurries, the stronger the influence. They reported that the mass transfer coefficient (k_L) could be increased or reduced through changing film thickness with turbulence or lowering diffusion coefficient of the gas. The reduction of solubility and diffusivity of dissolved gases in slurries upon increasing volume fraction of slurries may be due to the decrease [8] in the portion of continuous phase in the slurries. When the volume fraction of slurries is increased, the gas-liquid interface covered by the fine particles can hinder diffusion of gas and, hence, reduce k_L. If the nanofluid is treated as the pseudohomogeneous phase [10], in which diffusion of the solutes as well as the gas-liquid interfacial area are assumed to be unaltered, the hydrodynamics of the nanofluid can be used to predict the gas absorption rate in the nanofluid/gas system.

The apparent viscosity of non-Newtonian fluids is not sufficient to obtain a unified correlation for k_La_L in the case of gas absorption into non-Newtonian fluids. Because of the complexities of gas absorption in non-Newtonian media, the correlations obtained by these studies have been 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_L is due to the viscoelasticity of the aqueous solution [11,12], 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. A dimensionless number,

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

Investigator	n_1	n_2	n_3	Substance	Contactor
Yagi and Yoshida [27]	2	0.5	−0.67	CMC, PA	Agitated vessel
Ranade and Ulbrecht [28]	100	1	−0.67	CMC, PAA	Stirred tank
Nakanoh and Yoshida [12]	0.13	0.55	−1	CMC, PAA	Bubble column
Park et al. [13]	100	1	−0.42	PB, PIB	Agitated vessel
Park et al. [15]	2461.3	1	−0.274	PB, PIB	Agitated vessel
Park et al. [16]	54.7	1	−0.45	PAA	Agitated vessel
Park et al. [17]	8.33	1.31	1	PEO	Agitated vessel
Park et al. [18]	39.4	1	−0.43	Silica	Agitated vessel

such as Deborah number (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 a dimensionless term, such as $(1+n_1De^{n_2})^{n_3}$; they are listed in Table 1. As shown in Table 1, values of numbers in the dimensionless group are different from one another, and the polymers in Table 1 act as a reduction or increment agent in the absorption rate of gas.

There is little information about the effect that elastic properties have on the absorption of gas accompanied by a chemical reaction in non-Newtonian liquids. Park et al. [13-15] presented the effect of elasticity of PIB in the benzene solution and in the w/o (water in oil) emulsion, which is composed of an aqueous alkaline solution as the dispersed phase and a benzene solution as the continuous phase, on the absorption rate of CO_2. The absorption rates of CO_2 were also measured in aqueous PAA solution with MDEA [16], PEO solution with 2-amino-2-methyl-1-propanol (AMP) [17], silica solution with AMP [18] and PE solution with triethanolamine [19], respectively. They showed that the viscoelastic polymers, such as PIB, PAA, and PEO and colloidal silica influenced the absorption rate of CO_2.

To investigate the effect that a non-Newtonian liquid has on the gas absorption in series as mentioned above, *N*-methyldiethanolamine (MDEA) was used as a reactant with CO_2 in an aqueous nano-sized colloidal silica solution in this study. The absorption rates of CO_2 were measured in aqueous nano-sized colloidal silica solution with MDEA, and they were compared with those estimated by the mass transfer, with chemical reaction, based on the film theory. The volumetric mass transfer coefficient (k_La_L) of CO_2 in the aqueous nano-sized colloidal silica solution was obtained by using a unified correlation between the values of k_La_L and Deborah number, which was obtained in the previous paper [18].

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 (MDEA) according to

$$CO_2+\nu R_2NH \rightarrow R_2NCO_2^- + R_2NH_2^+ \tag{1}$$

where R represents the functional groups on the secondary amine.

The stoichiometric coefficient (ν) in Eq. (1) for MDEA was obtained from the reference [20] and its value was 1.

Species B, which is a nonvolatile solute, was dissolved into the liquid phase prior to its introduction into the gas absorber. It is assumed that gas phase resistance to absorption is negligible when using pure species A; 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:

$$r_A=k_2C_AC_B \tag{2}$$

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

$$D_A\frac{d^2C_A}{dz^2}=k_2C_AC_B \tag{3}$$

$$D_B\frac{d^2C_B}{dz^2}=\nu k_2C_AC_B \tag{4}$$

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

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

Eqs. (3)-(6) are put into the dimensionless form as follows:

$$\frac{d^2a}{dx^2}=Mab \tag{7}$$

$$\frac{d^2b}{dx^2}=rqMab \tag{8}$$

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

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

where $M=D_Ak_2C_{Bo}/k_L^2$, $a=C_A/C_{Ai}$, $b=C_B/C_{Bo}$, $x=z/z_L$, $q=\nu C_{Ai}/C_{Bo}$, $r=D_A/D_B$.

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:

$$R_A=\beta R_{Ao}=\beta k_La_L C_{Ai}V_L \tag{12}$$

where R_{Ao} is the physical absorption rate, obtained by multiplying the molar flux by the specific contact area between gas and liquid (a_L) and the liquid volume (V_L).

EXPERIMENTAL

1. 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 2,200 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. According to the Aldrich materials science catalog 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.

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 8.047×10^{-3} m^2. The liquid phase was agitated by 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 of 0.034 m in length, 0.017 m in width, and 0.005 m in thickness was used as the liquid phase agitator, and was located at the middle position of the liquid phase of 0.833 dm^3. An absorption rate of CO_2 was obtained from the difference between the flow rates of CO_2 at inlet and outlet of the absorber by using a mass flow meter (Brook Instrument, U.S.A) in the aqueous colloidal silica solution (silica of 0-31 wt%) and MDEA (0-2 kmol/m^3) with the agitation speed of 50 rpm at 101.3 N/m^2 and 25 °C. The experimental procedure used to obtain the absorption rate was the same as that reported in detail previously [18].

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 [10], were obtained as follows:

1. Solubility of CO_2 in the Aqueous Colloidal Silica Solution

The solubility (C_{Ai}) of CO_2 in the aqueous colloidal silica solutions was obtained by using the pressure measuring method, which measured the pressure difference of CO_2 before and after equilibrium in the gas and liquid phases, similar to a procedure reported elsewhere [21] at 25 °C and 0.101 MPa. The experimental procedure was the same as that as reported in detail previously [18].

2. 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 kg/m^3 by weighing with a pycnometer (Fisher Scientific Co., USA); it was identical within experimental accuracy to the density of water. The apparent viscosity of the aqueous silica colloid solution was measured at 25 °C with Brookfield viscometer (Brookfield Eng. Lab. Inc, USA).

3. Diffusivities of CO_2 in the Aqueous Colloidal Silica Solution

Diffusivity (D_{AB}) of CO_2 in aqueous MDEA [20] solution was estimated as follows:

$$D_{AB}=2.35\times10^{-6}\ \mathrm{Exp}(-2119/T) \tag{13}$$

Diffusivity (D_A) of CO_2 in the aqueous colloidal silica solution with MDEA was estimated from the following equation [22] corrected with viscosity of the aqueous colloidal silica solution.

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

Diffusivity (D_B) of MDEA in the aqueous colloidal silica solution with MDEA was obtained from the assumption [23] that the ratio of D_B to D_A was equal to the ratio (D_{Bw}/D_{Aw}) in water. The diffusivity of CO_2 and MDEA in water at 25 °C was taken as 1.97×10^{-9} m^2/s [24] and 8.1×10^{-10} m^2/s [20], respectively.

The obtained values of solubility, diffusivity of CO_2, density, and apparent viscosity of the aqueous silica colloid solution are given in Table 2.

4. Reaction Rate Constant of CO_2 with MDEA

For the reaction of CO_2 with MDEA [20], the reaction rate constant (k_2) was estimated as follows:

$$k_2=4.01\times10^{8}\mathrm{Exp}(-5400/T) \tag{15}$$

5. Volumetric Mass Transfer Coefficient of CO_2 (k_La_L)

The k_La_L was obtained, which presents the relationship (the standard deviation of 3.16% and mean deviation of 15.48%) between k_La_L and rheological behavior of the aqueous colloidal silica solution [18] under the conditions of the agitation speed of 50-400 rpm with the impeller size of 0.034, 0.05, and 0.075 m, as follows:

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

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

Silica (wt%)	Viscosity (Ns/m^2)×10^3	Diffusivity (m^2/s)×10^9	Solubility (kmol/m^3)	Density (kg/m^3)	Rheological properties			
					n	K×10^3 (Ns^n/m^2)	b	A (Ns^n/m^2)
0	1	1.97	0.039	1,000	1.0	1.0	-	-
5.8	1.075	1.88	0.032	1,039	0.97	1.115	0.097	0.03
11.91	1.127	1.82	0.031	1,082	0.94	1.321	0.160	0.057
18.35	1.191	1.77	0.029	1,134	0.92	1.525	0.190	0.082
23.06	1.235	1.72	0.028	1,160	0.9	1.67	0.217	0.1
30.85	1.274	1.68	0.027	1,180	0.85	1.95	0.232	0.129

where De is defined as the ratio of the characteristic material time (λ) to the characteristic process time (t) as follows:

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

where K and n are the index of $\tau=\mathrm{K}\gamma^n$, A and b, the index of $\mathrm{N}_1=\mathrm{A}\gamma^b$, N, the agitation speed of impeller and the shear rate is obtained in case of agitation of liquid in a cylindrical vessel as follows [25]:

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

RESULTS AND DISCUSSION

1. Effect of Rheological Properties on the k_La_L of CO_2

To observe the effect of rheological properties of the aqueous colloidal silica solution on k_La_L, the absorption rate of CO_2 was measured in the colloidal silica of 0-31 wt% to get k_La_L. The circles in Fig. 1 show the typical plots of the k_La_L of CO_2 against the silica concentration of weight and volume fraction under the experimental conditions of the agitation speed of 50 rev/min with the impeller size of 0.034 m. The solid line in Fig. 1 represents the calculated values of k_La_L using Eq. (16). As shown in Fig. 1, the value of k_La_L decreases with increasing silica concentration.

In absorption of gas into aqueous slurries, which have an adsorption capacity of the dissolved gas, k_La_L increases with increasing the concentration of fine particles, and this behavior was explained by the grazing effect [7]. In contrast, the value of k_La_L in this study decreased with increasing silica concentration, as shown in Fig. 1, because the aqueous colloidal silica solution made by diluting Ludox HS-40 with distilled water does not have an adsorption capacity. On the other hand, the behavior of decrease of the k_La_L with increasing the silica concentration might be explained due to the inverse proportion of viscosity to diffusivity, because the diffusivity of CO_2 decreases with increasing the apparent viscosity of the aqueous colloidal silica solution as shown in Table 2. Hikita et al. [8] have shown that the diffusivity of CO_2 decreases with increasing the volume fraction of fine particle in aqueous slurries.

Because k_La_L is affected by both the viscosity and elasticity of the aqueous colloidal silica solution as shown in Eq. (16), the estimated values of k_La_L are obtained according to the kinds of the solutions, i.e., water, the aqueous colloidal silica solution without elasticity, and with elasticity, and presented as symbols of the dotted, dashed, and solid lines in Fig. 1, respectively. In water, μ is μ_w and De is 0, in the aqueous colloidal silica solution with elasticity, μ is that in aqueous colloidal silica solution and De has some value >0. The aqueous colloidal silica solution without elasticity (De=0) is an imaginary solution, which is assumed to be a Newtonian liquid with the same viscosity as that of the actual aqueous colloidal silica solution with elasticity. As shown in Fig. 1, the k_La_L decreases in the dotted, dashed, and solid line order. In general, the mass transfer coefficient in the solution is inversely proportional to the viscosity. The viscosity of water is smaller than that of the aqueous colloidal silica solution as listed in Table 2, and then, k_La_L in water is larger that that in the aqueous colloidal silica solution. If the aqueous colloidal silica solution in this study would have only viscous behavior, the k_La_L should be the k_La_L represented as the dashed line. But, the actual k_La_L of the solid line is smaller than that of the dashed line. This result means that the effect of the elasticity of the aqueous colloidal silica solution on the decrease of k_La_L is stronger than the effect of viscosity on k_La_L; in other words, the elasticity of the aqueous colloidal silica solution reduces k_La_L according to Eq. (16) based on the same viscosity of the solution.

Astarita et al. [11] and Seyer and Metzner [26] have shown that even very dilute solutions of drag reducing polymers show a considerable resistance to stretching under turbulent flow conditions, and this phenomenon is considered as an effect of the viscoelastic-

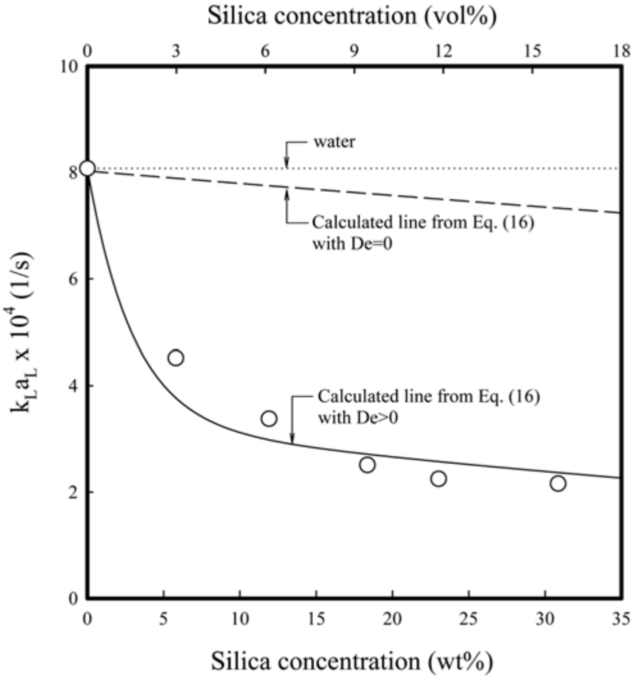

Fig. 1. Effect of De on k_La_L at d=0.034 m and N=50 rpm.

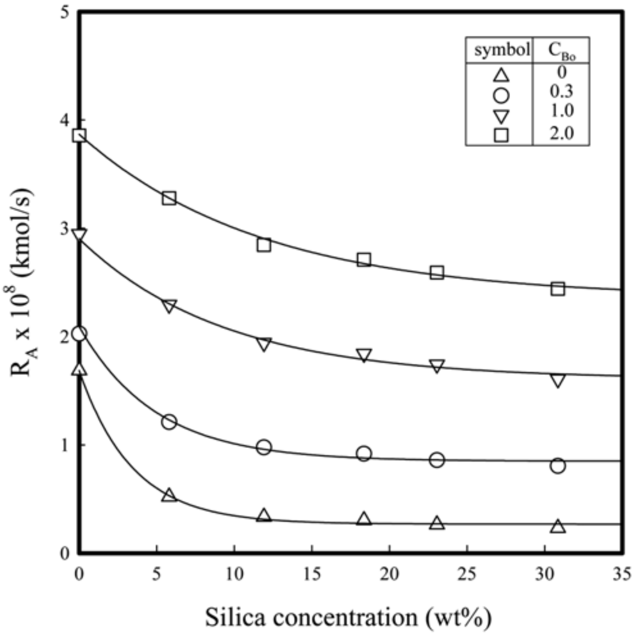

Fig. 2. Effect of silica concentration on absorption rate for various MDEA concentrations.

ity. They have presented that the reduction of the k_La_L in a viscoelastic non-Newtonian liquid such as PAA is attributable to the elasticity of the liquid phase. The reduction of k_La_L from the dashed line to the solid one shown in Fig. 1 might be attributed to the elasticity of the aqueous colloidal silica solution. In other words, the aqueous colloidal silica solution used in this study might play the part of reducing agent of k_La_L.

2. Effect of Rheological Properties on the Chemical Absorption of CO_2

To observe the effect of elasticity of the aqueous colloidal silica solution on the chemical reaction of CO_2 with MDEA, the absorption rate of CO_2 into aqueous colloidal silica solution with MDEA was measured according to change of silica concentration in the range of 0-31 wt% at MDEA concentration 0-2 kmol/m^3 under the experimental conditions of the agitation speed of 50 rpm with the impeller size of 0.034 m.

Fig. 2 shows the plots of the absorption rate of CO_2 against the silica concentration for various MDEA concentrations. As shown in Fig. 2, R_A decreases with increasing silica concentration, and increases with increasing MDEA concentration. Increase of R_A with increasing MDEA concentration is due to the reactant of MDEA in reaction of CO_2.

Decrease of R_A with increasing silica concentration is explained as follows:

R_A is obtained from Eq. (12) using k_La_L, β, and C_{Ai}. As shown in Fig. 1, k_La_L decreased with increasing the silica concentration. As shown in Table 2, C_{Ai} in the aqueous colloidal silica solution did not vary significantly from that in water. The value of β was obtained from Eq. (11) from the numerical solution of Eqs. (7) and (8) by the finite element method using the dimensionless parameters such as r, q, and M, which were obtained from the physicochemical properties for the given concentrations of silica and MDEA, and plotted against the silica concentration in Fig. 3. As shown in Fig. 3, the values of β increased with increasing silica concentration and increased with increasing MDEA concentration, i.e., decrease of k_La_L by increase of silica concentration did not affect β, although k_L obtained from Eq. (16) was used to estimate a theoretical β. Therefore, decrease of R_A was affected mainly by decrease of k_La_L rather than β and C_{Ai}. Reduction of k_La_L by the elastic properties of the aqueous colloidal silica solution causes a decrease of the chemical absorption rate of CO_2.

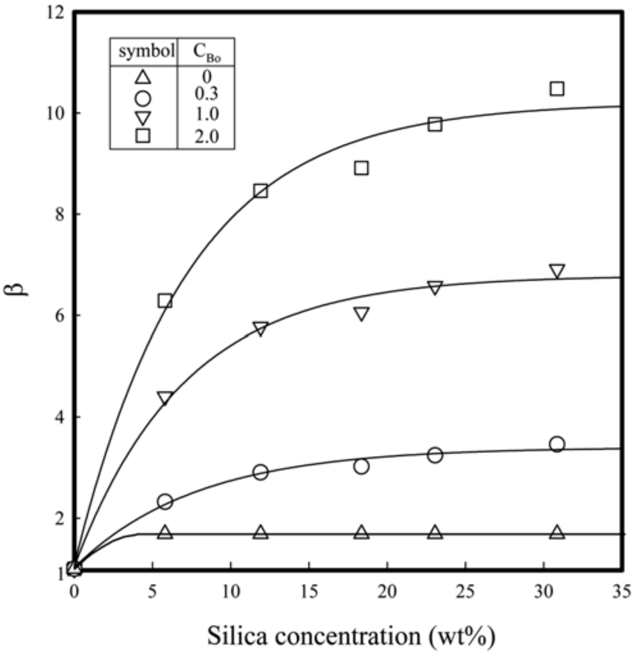

Fig. 3. Effect of silica concentration on enhancement factor for various MDEA concentrations.

CONCLUSIONS

The measured rate of chemical absorption of carbon dioxide into the aqueous colloidal silica solution of 0-31 wt% with MDEA 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 k_La, which was obtained from a unified correlation equation containing Deborah number. The elasticity reduced the absorption rate of CO_2 compared with that of a Newtonian liquid based on the same values of viscosity. The chemical absorption rate was decreased due to the reduction effect of k_La_L by elasticity of the aqueous colloidal silica solution.

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NOMENCLATURE

a_L : specific gas-liquid area [m^2/m^3]
C_i : concentration of species, i [kmol/m^3]
d : diameter of impeller [m]
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]
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 : temperature [°K]
z : diffusion coordinate of CO_2 [m]
z_L : film thickness [m]

Greek Letters

β : enhancement factor of absorption rate by the chemical reaction
γ : shear rate [1/s]
μ : viscosity of liquid [N·s/m^2]
ρ : density of liquid [kg/m^3]
τ : shear stress [N/m^2]

Subscripts

A : CO_2

B : MDEA
i : gas-liquid interface
o : feed
w : water

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