Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Park, Sang-Wook , Lee, Joon-Wook , Choi, Byoung-Sik and Lee, Jae-Wook(2006) 'Absorption of Carbon Dioxide into Aqueous Colloidal Silica Solution', Separation Science and Technology, 41: 8, 1661 — 1677
To link to this Article: DOI: 10.1080/01496390600674760
URL: http://dx.doi.org/10.1080/01496390600674760

Separation Science and Technology, 41: 1661–1677, 2006
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ISSN 0149-6395 print/1520-5754 online
DOI: 10.1080/01496390600674760

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Abstract: On the basis of experimental data for carbon dioxide absorption into aqueous nanometer sized colloidal silica solution as a non-Newtonian fluid, a dimensionless correlation for volumetric liquid-side mass transfer coefficient (k_La) of CO_2 in the flat-stirred vessel was proposed. In addition to ordinary liquid properties and operating parameters such as impeller size and speed in the vessel, Deborah number, which is defined as the product of the characteristic material times of the liquid and agitation speed in the flat-stirred vessel and represents the viscoelastic behavior of non-Newtonian fluid, was used to present unified expressions for k_La in Newtonian as well as non-Newtonian liquid. The values of k_La in the aqueous colloidal silica solution were reduced due to elasticity of the solution.

Keywords: Absorption, carbon dioxide, colloidal silica, non-Newtonian liquid

INTRODUCTION

In multiphase systems appearing in agitated reactors, gas-liquid mass transfer may be the rate-determining step for the overall process. Therefore, the knowledge of gas-liquid mass transfer rates characterized by the volumetric liquid-phase mass transfer coefficient (k_La) is needed for a reliable design

Received 17 January 2006, Accepted 28 February 2006

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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 surfactant into the working fluids. The addition of surfactant causes the interfacial turbulence, which leads to a higher heat and mass transfer performance (4). In the recent decade, nanofluid has become one of the most attractive heat transfer media due to the development of nano technology. The definition of nanofluid is the solid/liquid mixture in which nano-sized particles($d_p < 100\,\text{nm}$) 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 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 carried out the study of 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 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 the 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 diffusion coefficient of gas. The reduction of solubility and 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 a 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 non-Newtonian fluid with that in the Newtonian one.

Only use of the apparent viscosity of non-Newtonian fluids was not sufficient to obtain a unified correlation for k_La values. Due to the complexities of gas absorption in 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 numbers 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 CO_2, and that (21) in w/o emulsion composed of aqueous solution as

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

Investigator	n_1	n_2	n_3	Polymer	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

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 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, rheological properties of colloidal silica from Ludox products of Aldrich Chemicals are measured using a rheometer and the absorption rate of CO_2 is measured into nano-sized colloidal silica using a flat-stirred vessel to observe the effect of rheological property of colloidal silica, whose average size is 24 nm, on the volumetric mass transfer coefficient of carbon dioxide, and to propose a unified correlation for k_La in Newtonian as well as non-Newtonian solutions by introducing the dimensionless number such as Deborah number.

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 t% w/w solid content (silica density 2.2 g/cm^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 absorption of CO_2.

Absorption Rate of CO_2

An agitated vessel, which was used to measure the absorption rate of CO_2 in the aqueous colloidal silica solution, was constructed of glass of 0.102 m inside diameter and of 0.151 m in height with four equally spaced vertical baffles. Three kinds of straight impeller of 0.034, 0.05, and 0.075 m in length and 0.011 m in width were used as the liquid phase agitator, and located at the middle position of the liquid phase. The absorption rate (R_A) of CO_2 was measured in the colloidal silica of $0 \sim 40$ wt% according to the change of the impeller speed of 50–400 rev/min along the procedure similar to those reported elsewhere (23) at 0.101 MPa and 25°C. The k_La of CO_2 was obtained using the measured R_A and solubility of CO_2.

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 (24) 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 as reported in the published research (23) in detail.

Density and Apparent Viscosity of the Aqueous Colloidal Silica Solution

The density of the aqueous silica colloid solution were measured at 25°C within $0.1\,kg/m^3$ by weighing with a pycnometer (Fisher Scientific Co., USA) and were 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 Brookfield viscometer (Brookfield Eng. Lab. Inc, USA).

Diffusivities of CO_2 in the Aqueous Colloidal Silica Solution

The diffusivity (D_{Aw}) of CO_2 in water at 25°C was taken as $1.97 \times 10^{-9}\,m^2/s$ (25). The diffusivity (D_A) of CO_2 in the aqueous silica colloid solution was estimated from the following equation (26) corrected with viscosity of the colloidal silica.

$$D_A = D_{Aw}(\mu/\mu_w)^{(-2/3)} \tag{1}$$

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

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

					Rheological properties			
Silica (wt%)	Viscosity $(Ns/m^2) \times 10^3$	Diffusivity $(m^2/s) \times 10^9$	Solubility $(kmol/m^3)$	Density (kg/m^3)	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

Rheological Properties of the Aqueous Colloidal Silica Solution

We assume that a power-law model, which has been widely used for shear-dependent viscosity, can represent the non-Newtonian flow behavior of the aqueous silica colloid solution.

$$\tau = K\gamma^{n} \tag{2}$$

$$\mu = K\gamma^{n-1} \tag{3}$$

$$N_1 = A\gamma^{b} \tag{4}$$

where n, K, b, and A are material parameters depending on temperature. These parameters 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. As shown in Table 2, the values of A increased with increasing of the concentration of silica, which means that silica has an elastic behavior (19).

One of the parameters used frequently to represent the characteristics of viscoelasticity is known as the material's characteristic relaxation time (λ) of the liquid defined as

$$\lambda = \frac{N_1}{\mu\gamma^2} \tag{5}$$

Using Eqs. 2 and 3, λ is rearranged as

$$\lambda = \frac{A}{K}\gamma^{b-n-1} \tag{6}$$

One of the dimensionless numbers, which relate the elastic properties with the process parameters, is Deborah number (De) defined as ratio of the material's characteristic relaxation time to the characteristic flow time. The characteristic flow time is measured against a characteristic process time (t), which is related to the reciprocal of the impeller speed (N) in case of stirred tanks, and De is derived as follows:

$$De = \lambda/t = \frac{A}{K}\gamma^{b-n-1}N \tag{7}$$

where shear rate (γ) is obtained in case of agitation of liquid in a cylindrical vessel as follows (27):

$$\gamma = 4\pi N/n \tag{8}$$

RESULTS AND DISCUSSION

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

To observe the effect of silica in the aqueous colloidal silica solution on the k_La, absorption experiments were performed to get the R_A and k_La of CO_2, in which silica concentration, impeller speed and size were varied. R_A and k_La were typically plotted against silica concentration at the impeller size of 50 mm in Fig. 1 and Fig. 2, respectively. As shown in these figures, R_A and k_La increases with the increase of the agitation speed of the impeller and decreases with the increase of silica concentration. Same results were obtained at other impeller sizes of 34 and 75 mm.

In analyzing the relationship between k_La and the experimental variables such as the silica concentration, and the speed and size of the impeller, the following influences may be considered: diffusivity, viscosity, and rheological properties of the liquid phase.

It is customary to express the influence of viscosity upon the mass transfer coefficient in terms of Schmidt number defined as $\mu/\rho D_A$, in which the

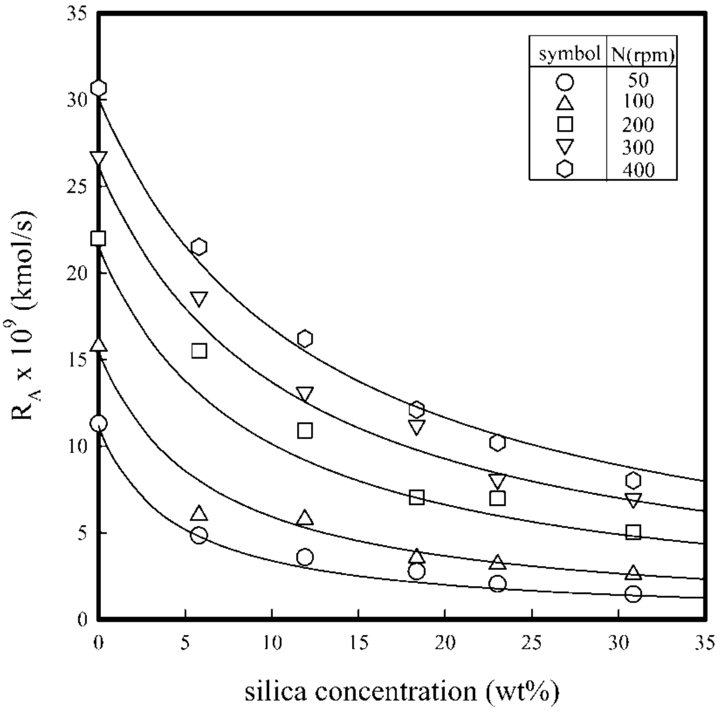

Figure 1. Effect of silica concentration in aqueous colloidal silica solution on R_A at $d = 0.05$ m.

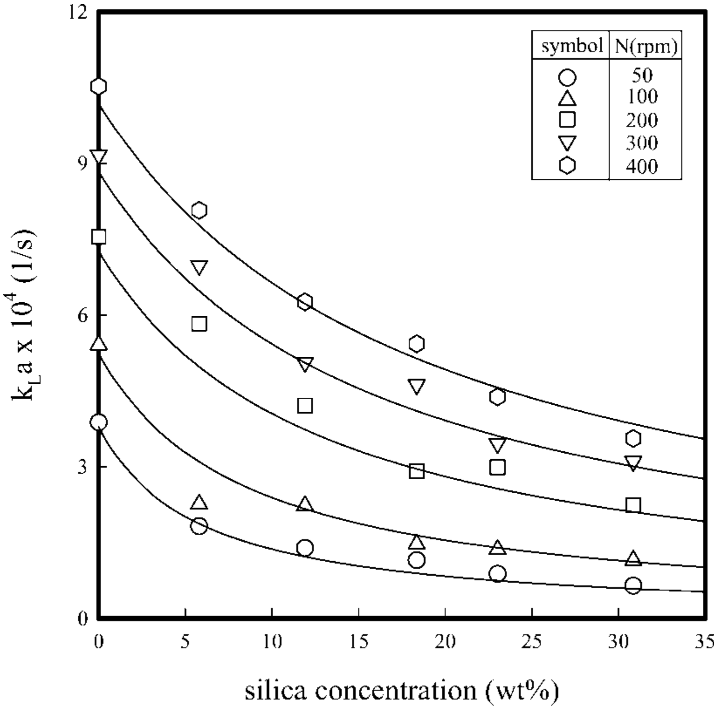

Figure 2. Effect of silica concentration in aqueous colloidal silica solution on k_La at $d = 0.05\,m$.

viscosity is related to the diffusion coefficient. As shown in Table 2, the diffusivity of CO_2 in the aqueous colloidal silica solution does not vary significantly from that in water, and thus the use of a dimensionless Schmidt number is not warranted. Instead, the ratio of viscosity of silica solution to that of water (28) was used to correlate with k_La, because the viscosity in the agitated vessel depends on the speed of impeller and the rheological properties as shown in Eq. (3) through Eq. (8).

To correlate k_La with the experimental variables such as the silica concentrations and the speed and size of the impeller, the dimensionless groups such as Sherwood number (Sh) and Reynolds number (Re) are used, and they are defined as follows, respectively:

$$Sh = k_Lad^2/D_A \tag{9}$$

$$Re = d^2N\rho/\mu \tag{10}$$

Figure 3 shows logarithmic plots of Sh against Re in water, which were obtained according to the change of impeller speed and size. As shown in Fig. 3, the plots are linear, and the slope and intercept from the straight line

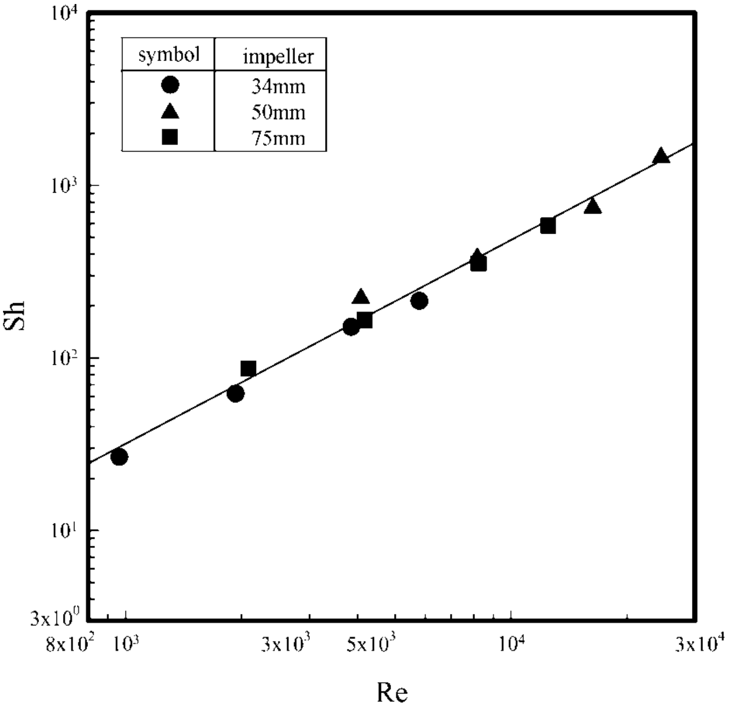

Figure 3. Sh vs. Re in water at various impeller size.

of the plots by a least-squares method were obtained, which were used to get an empirical equation between Sh and Re as follows:

$$\mathrm{Sh} = 12.56\,\mathrm{Re}^{0.48} \tag{11}$$

The calculated values of Sh from Eq. (11) approached to the measured Sh very well with a standard deviation (SD) of 0.15% and a mean deviation (MD) of 3.06%. Eq. (11) would be used as a base for comparison of the behavior of non-Newtonian liquid with that of Newtonian liquid.

The values of Sh and Re were obtained from Eq. (9) and (10) using the R_A measured according to the change of silica concentration, impeller speed, and size in the aqueous colloidal silica solution, and logarithmic plots of Sh against Re are shown in Fig. 4. The solid line in Fig. 4 presents the Sh calculated by Eq. (11). As shown in Fig.4, the plots were scattered from the straight line with SD of 37.38% and MD of 58.41%. This deviation may be due to non-Newtonian behavior of the aqueous colloidal silica solution.

The new terms corrected with viscosity and De was used to lessen the deviation of the plots for the aqueous colloidal silica solution from the plots for water as shown in Fig. 2. A simple multiple regression exercise was used for the plots of Sh combined with $(\mu/\mu_w)^{c_1}$ $(1 + c_2\mathrm{De})^{c_3}$ against Re,

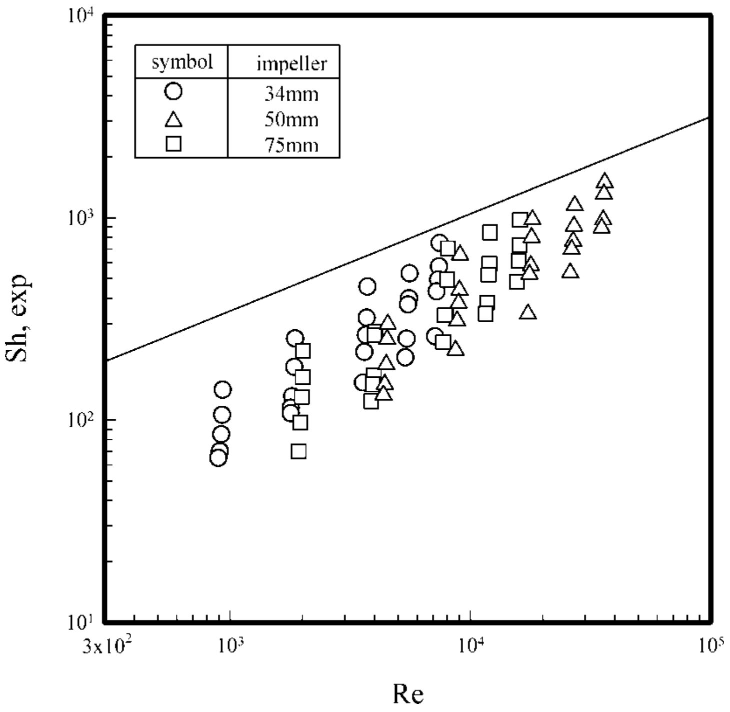

Figure 4. Sh vs. Re in aqueous colloidal silica solution at various impeller size.

which gave the values of c_1, c_2, and c_3 are 0.11, 39.4, and −0.43, respectively, with SD of 3.16% and MD of 15.48% as shown in Fig. 5.

Using a total of 75 data points, a multiple regression analysis came up with a correlation as follows:

$$k_L ad^2/D_A = 12.56(d^2N\rho/\mu)^{0.48}(\mu/\mu_w)^{0.11}(1+39.4De)^{-0.43} \tag{12}$$

There is enough experimental evidence in the literature which shows that an increase in the viscosity of the aqueous phase results in a reduction of the k_La. From the work of Yagi ans Yoshida (18), Perez and Sandall (29), and Moo-Young and Kawase (30), it may be concluded that the exponent m in the relationship $k_La \propto \mu^{-m}$ for stirred tanks is between 0.35 and 0.6. When our data were analyzed by Eq. (12) for the overall effect of viscosity on the k_La, it comes out as $k_La \propto \mu^{-0.37}$, which compares well with the findings of Yagi and Yoshida (18) of $\mu^{-0.4}$ and Rande and Ulbrecht (19) of $\mu^{-0.41}$, and Moo-Young and Kawase (30) of $\mu^{-0.38}$. As a result, in Fig. 5 the deviation of plots of Sh against Re in colloidal silica can be eliminated by incorporating De into the correlation.

Figure 6 compares the experimental Sh with those predicted by Eq. (12). It can be seen that the agreement between the observed Sh and those predicted

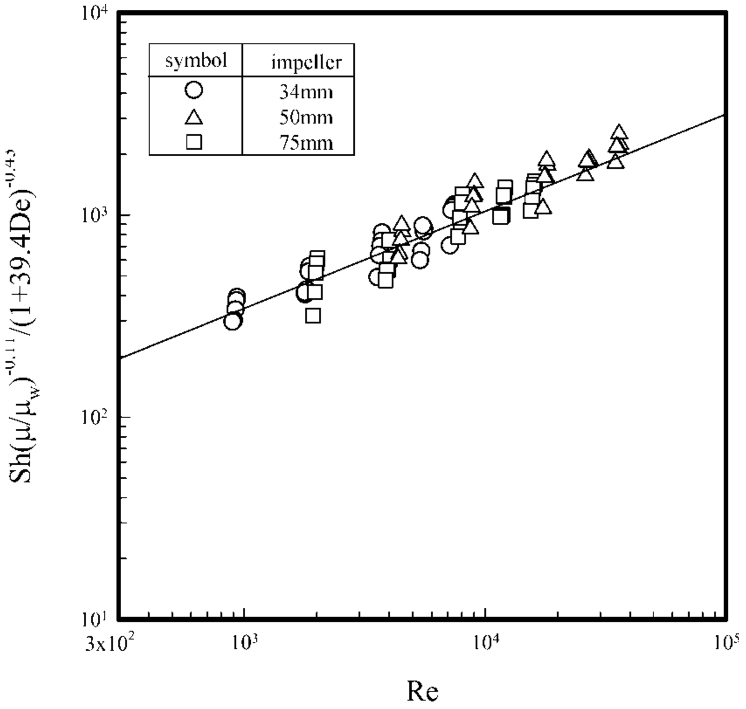

Figure 5. Dimensionless correlation of Sh vs. Re corrected with specific viscosity and Deborah number in aqueous colloidal silica solution at various impeller size.

by Eq. (12) is very good; the mean deviation between the 75 experimental predicted points is 14.88%, and the standard deviation, 2.95%.

Effect of Rheological Properties on the k_La of CO_2

To observe the effect of rheological properties of the aqueous colloidal silica solution on k_La, the absorption rate of CO_2 was measured in the colloidal silica of $0 \sim 40$ wt% to get k_La. The circles in Fig. 7 show the typical plots of the k_La of CO_2 against the silica concentration of weight and volume fraction under the experimental conditions of the agitation speed of 100 rev/min with the impeller size of 0.034 m. The solid line in Fig. 7 represents the calculated values of k_La using Eq. (12). As shown in Fig. 2, the value of k_La decreases with increasing silica concentration.

In absorption of gas into aqueous slurries, which have an adsorption capacity of the dissolved gas, k_La increases with increasing the concentration of fine particles, and this behavior was explained by the gazing effect (7). But, the k_La in this study does not increase with increasing silica concentration as

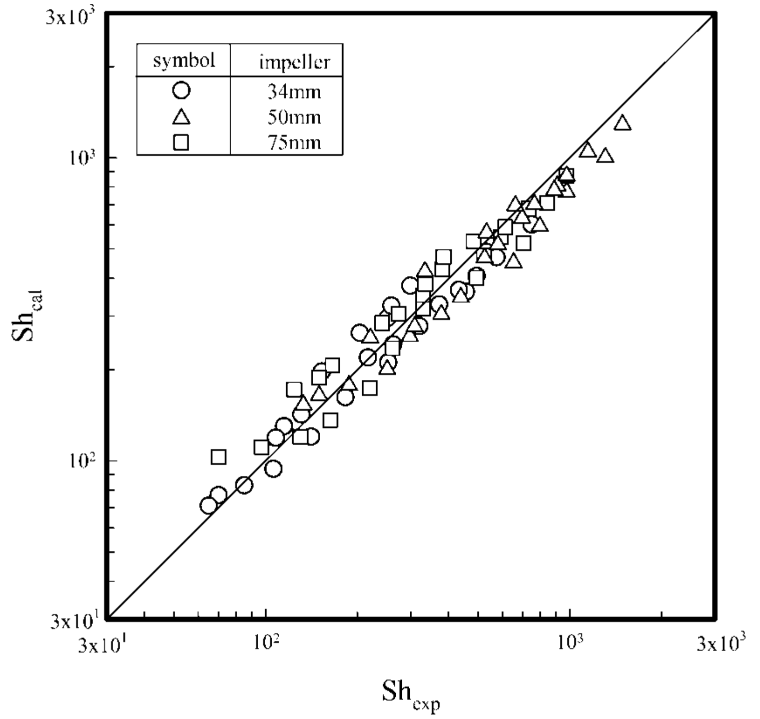

Figure 6. Comparison of calculated Sh with experimental values.

shown in Fig. 7, 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 with increasing silica concentration might be explained due to the inverse proportion of viscosity to diffusivity, because the diffusivity decrease of CO_2 with increasing the apparent viscosity of the aqueous colloidal silica solution as shown in Table 2. Hikita et al. (9) have shown that the diffusivity of CO_2 decreases with increasing the volume fraction of fine particle in aqueous slurries.

Because k_La is affected by both of viscosity and elasticity of the aqueous colloidal silica solution as shown in Eq. (12), the estimated values of k_La 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. 7, 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 aqueous colloidal silica solution with elasticity. As shown in Fig. 7, the k_La decreases in the dotted, dashed, and solid line

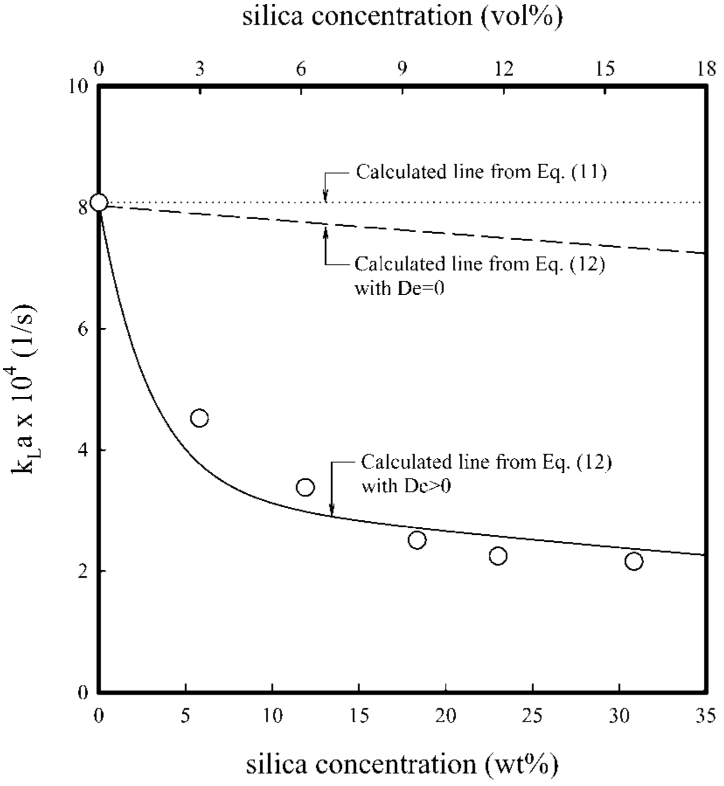

Figure 7. Effect of De on k_La at $d = 0.034\,m$ and $N = 100\,rpm$.

order. In general, the mass transfer coefficient in the solution is inversely proportional to the viscosity. The viscosity of water is smaller compared to that of the aqueous colloidal silica solution as listed in Table 2, and then, k_La in water is larger that in the aqueous colloidal silica solution. If the aqueous colloidal silica solution in this study would have only viscous behavior, the k_La should be the k_La represented as the dashed line. But, the actual k_La of the solid line is smaller than that of the dashed line. This means that the effect of the elasticity of the aqueous colloidal silica solution on decrease of k_La is stronger than the effect of viscosity on k_La, in other words, the elasticity of the aqueous colloidal silica solution reduces k_La due to the decrease of k_La as shown in Eq. (12) based on the same viscosity of the solution.

Astarita et al. (16) and Seyer and Metzner (31) 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 viscoelasticity. They have presented that the reduction of the k_La in a viscoelastic non-Newtonian liquid such as PAA is attributable to the elasticity of the liquid phase. The reduction of k_La from the dashed line to the solid one in shown in Fig.7 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 $k_L a$.

CONCLUSIONS

The absorption rates of carbon dioxide in the aqueous colloidal silica solution were measured using a flat-stirred vessel to observe the influence of the rheological properties of the aqueous colloidal silica solution on the volumetric liquid-side mass transfer coefficient ($k_L a$) under the experimental conditions such as silica concentration of 0–40 wt%, impeller size of 0.034, 0.05, and 0.075 m, agitation speed of $0 \sim 400$ rev/min at 25°C, and 0.101 MPa. The Deborah number presenting the elasticity of the aqueous colloidal silica solution was used to get an empirical correlation containing both viscosity and elasticity of the non-Newtonian viscoelastic liquid. The reduction effect of the elasticity of the aqueous colloidal silica solution on $k_L a$ was stronger than the effect of viscosity.

NOMENCLATURE

a	specific gas-liquid area ($\mathrm{m^2/m^3}$)
d	diameter of impeller (m)
N_1	primary normal stress difference ($\mathrm{kg/m \cdot s^2}$)

Greek Letters

γ	shear rate (1/s)
μ	viscosity of liquid (Ns/m$^{2)}$
μ_w	viscosity of water ($\mathrm{Ns/m^2}$)
ρ	density of liquid ($\mathrm{kg/m^3}$)
τ	shear stress ($\mathrm{N/m^2}$)

Subscripts

A	CO_2
i	gas-liquid interface

ACKNOWLEDGMENTS

This work was supported with the Basic Research Program of the Korea Science and Engineering Foundation (KOSEF) through ARC.

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