

Effect of elasticity of aqueous colloidal silica solution on chemical absorption of carbon dioxide with 2-amino-2-methyl-1-propanol

Sang-Wook Park*, Byoung-Sik Choi and Jae-Wook Lee¹

Division of Chemical Engineering, Pusan National University, Busan 609-735, Korea

¹Department of Chemical Engineering, Sogang University, Seoul 121-742, Korea

(Received, July 6, 2006; final revision received August 26, 2006)

Abstract

Carbon dioxide was absorbed into the aqueous nanometer sized colloidal silica solution of 0-31 wt% and 2-amino-2-methyl-1-propanol of 0-2 kmol/m³ in a flat-stirred vessel with the impeller of various sizes and speeds at 25 °C and 0.101 MPa to measure the absorption rate of CO₂. The volumetric liquid-side mass transfer coefficient ($k_L a$) of CO₂ was used to obtain the empirical correlation formula containing the rheological behavior of the aqueous colloidal silica solution. Reduction of the measured $k_L a$ was explained by the viscoelastic properties of the aqueous colloidal silica solution. The theoretical value of the absorption rate of CO₂ was estimated from the model based on the film theory accompanied by chemical reaction and compared with the measured value.

Keywords : absorption, 2-amino-2-methyl-1-propanol, carbon dioxide, silica, viscoelastic liquid

1. Introduction

In multiphase systems appearing in agitated reactors, the 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 liquid-phase mass transfer coefficient ($k_L a$) is needed for reliable design of such reactors and many researchers (Astarita *et al.*, 1983) 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 (Fan and Cui, 2005) and electromagnetic (Xu *et al.*, 2005) 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 the 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 (Hozawa *et al.*, 1991). In recent decade, the 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 ($d_p < 100$ nm) are suspended evenly in the base liquid. It is well known (Kebblinski *et al.*, 2002) that the

nanofluid can enhance the effective thermal conductivity and affects the heat transfer characteristic of fluid. One suggested four potential mechanisms (Kebblinski *et al.*, 2002); 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 (Kebblinski *et al.*, 2002) have actively carried out the study of heat transfer enhancement by nanofluid. Kim *et al.* (2006) measured the absorption rate of NH₃ in nanofluid of Cu, CuO and Al₂O₃ 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 (Kars and Best, 1979; Hikita *et al.*, 1985; Zhou *et al.*, 2003). 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 (1979). Zhou *et al.* (2003) 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

*Corresponding author: swpark@pusan.ac.kr
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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 (Hikita *et al.*, 1985) 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 (Mehra, 1995), 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.

Only use of the apparent viscosity of non-Newtonian fluids is not sufficient to obtain a unified correlation for $k_L a$. 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 $k_L a$ is due to the viscoelasticity of the aqueous solution (Astarita, 1969; Nakanoh and Yoshida, 1980), 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 $k_L a$ with the properties of non-Newtonian liquids. Unified correlations have been proposed for $k_L a$ in Newtonian as well as non-Newtonian solutions by introducing a dimensionless term, such as $(1 + n_1 De^{n_2})^{n_3}$; they are listed in Table 1. As shown in Table 1, values of numbers in the dimensionless group are different one another, and the polymers in Table 1 act as a reduction or increment agent in the absorption rate of gas.

Park *et al.* presented the effect of elasticity of polyisobutylene (PIB) (Park *et al.*, 2003) in the benzene solution of polybutene (PB) and PIB on the absorption rate of CO_2 , and that (Park *et al.*, 2003) 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 (Park *et al.*, 2005a)

and PEO (Park *et al.*, 2005b) 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 that elastic properties have on the absorption of gas accompanied by a chemical reaction in non-Newtonian liquid. Park *et al.* (2003; 2004) 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 . Park *et al.* (2005a; 2005b) also measured the absorption rate of CO_2 in aqueous PAA solution with NaOH and in aqueous PEO solution with monoethanolamine, respectively. They showed that the viscoelastic polymers, such as PIB, PAA, and PEO influenced the absorption rate of CO_2 .

To investigate the effect that the behavior of non-Newtonian liquid has on the gas absorption in series, an aqueous nano-sized colloidal silica solution was used as a viscoelastic material in this study. Deborah number, which was obtained from a power-law model of the relationship between the measured shear stress and first normal stress difference against the shear rate of the aqueous nano-sized colloidal silica solution, was used to obtain a unified correlation between the values of $k_L a$ in water and that in aqueous nano-sized colloidal silica solutions. The absorption rates of CO_2 were measured in aqueous nano-sized colloidal silica solution with 2-amino-2-methyl-1-propanol (AMP) and were compared with those estimated by the mass transfer mechanism with chemical reaction based on the film theory.

2. Theory

The overall reaction between $CO_2(A)$ and AMP(B) in aqueous solution is



The stoichiometric coefficients (ν) in Eq. (1) for AMP was obtained from the reference (Messouadi, and Sada, 1996) and its value was 1.

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

Investigator	n_1	n_2	n_3	Substance	Contactors
Nakanoh and Yoshida (1980)	0.13	0.55	-1	CMC, PAA	Bubble column
Yagi and Yoshida (1975)	2	0.5	-0.67	CMC, PA	Agitated vessel
Ranade and Ulbrecht (1978)	100	1	-0.67	CMC, PAA	Stirred tank
Park <i>et al.</i> (2003)	100	1	-0.42	PB, PIB	Agitated vessel
Park <i>et al.</i> (2004)	2461.3	1	-0.274	PB, PIB	Agitated vessel
Park <i>et al.</i> (2005a)	54.7	1	-0.45	PAA	Agitated vessel
Park <i>et al.</i> (2005b)	8.33	1.31	1	PEO	Agitated vessel

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 (Messouadi, and Sada, 1996) as follows:

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

where C_i is concentration of species, i .

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

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

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

$$z=0, C_A = C_{Ai}, \frac{dC_B}{dz} = 0 \quad (5)$$

$$z=z_L, C_A = 0, C_B = C_{Bo} \quad (6)$$

where z is diffusion coordinate of CO_2 and z_L , film thickness.

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

$$\frac{\partial^2 a}{\partial x^2} = M a b \quad (7)$$

$$\frac{\partial^2 b}{\partial x^2} = \nu r q a b \quad (8)$$

$$x=0; a=1, \frac{db}{dx} = 0 \quad (9)$$

$$x=1; a=0, b=1 \quad (10)$$

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

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

$$\beta = \left. \frac{da}{dx} \right|_{x=0} \quad (11)$$

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

$$R_A = \beta R_{A0} = \beta k_L a C_{Ai} V_L \quad (12)$$

where R_{A0} is the physical absorption rate, obtain by mul-

tiplying the molar flux by the specific contact area between gas and liquid (a) and the liquid volume (V_L).

3. Experimental

3.1. 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 2200 kg/m^3) was used. The average particle radius, the specific surface area, and PH at 298 K are 12 nm, $220 \text{ m}^2/\text{g}$, 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 .

3.2. 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 $8.047 \times 10^{-3} \text{ m}^2$. 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. Straight impellers that were with 0.034, 0.05, and 0.075 m in length, respectively, 0.017 m in width, and 0.005 m in thickness were used as the liquid phase agitator, and were located at the middle position of the liquid phase of 0.833 dm^3 . The absorption rate of CO_2 was obtained from the difference between the flow rates of CO_2 at inlet and outlet of the absorber in the aqueous colloidal silica solution (silica of 0~31 wt%) and AMP (0~2 kmol/m^3) with the impeller speed of 50-400 rev/min 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 (Park *et al.*, 2005b).

4. 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 (Mehra, 1995), were obtained as follows:

4.1. 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 using the pressure measuring method, which measured the pressure difference of CO₂ before and after equilibrium in the gas and liquid phases, similar to the procedure reported elsewhere (Kennard and Meisen, 1984). The experimental procedure was the same as that as reported in detail previously (Park *et al.*, 2005b).

The solubility (C_{Ai}) of CO₂ in aqueous AMP solution was estimated as follows (Totiwachwuthikul *et al.*, 1991):

$$C_{Ai} = C_{Aw} (1 - 0.1079 C_{Bo} + 0.0115 C_{Bo}^2) \quad (13)$$

4.2. Density and apparent viscosity of the aqueous colloidal silica solution

The density (ρ) of the aqueous silica colloid solution was measured within 0.1 kg/m³ by weighing with a pycnometer (Fisher Scientific Co., USA); it was identical within experimental accuracy to the density of water. The apparent viscosity (μ) of aqueous solution of PEO was measured with Brookfield viscometer (Brookfield Eng. Lab. Inc, USA).

4.3. Diffusivities of CO₂ in the aqueous colloidal silica solution

The diffusivity (D_{Aaq}) of CO₂ in aqueous AMP solution was estimated as follows (Danckwerts and Sharma, 1966):

$$D_{AB} = 2.1625 \times 10^{-9} - 6.85 \times 10^{-10} C_{Bo} + 7 \times 10^{-11} C_{Bo}^2 \quad (14)$$

Diffusivity (D_A) of CO₂ in the aqueous silica colloid solution was estimated from the following equation (Cussler, 1984) corrected with viscosity of the aqueous colloidal silica solution.

$$D_A = D_{AB} (\mu_w/\mu)^{2/3} \quad (15)$$

where μ_w is the viscosity of water.

Diffusivity (D_B) of AMP in aqueous amine solution was obtained from assumption that the ratio of D_B to D_A was equal to the ratio in water (Nijsing *et al.*, 1959). The diffusivity of CO₂ and AMP in water at 25 °C were taken as 1.97×10^{-9} m²/s (Hikita *et al.*, 1976) and 7.24×10^{-10} m²/s (Danckwerts and Sharma, 1966), respectively.

The obtained values of solubility, diffusivity of CO₂, density, and apparent viscosity of the aqueous silica colloid

solution are given in Table 2.

4.4. Reaction rate constant of CO₂ with AMP

The reaction rate constant (k_2) between the reaction of CO₂ and AMP was obtained from the following equation (Messaoudi and Sada, 1996).

$$\log k_2 = 10.986 - \frac{2551.2}{T} \quad (16)$$

4.5. 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 aqueous PEO solutions.

$$\tau = K\dot{\gamma}^n \quad (17)$$

$$\mu = K\dot{\gamma}^{n-1} \quad (18)$$

$$N_1 = A\dot{\gamma}^b \quad (19)$$

where τ , $\dot{\gamma}$, and N_1 are shear stress, shear rate and first normal stress difference, respectively. K , n , A , and b are material parameters that depend on the temperature. These parameters were obtained from the measurement of τ and N_1 for the change of $\dot{\gamma}$ by using the parallel disk type rheometer (Ares, Rheometrics, U.S.A.) having a diameter of 0.05 m and a gap of 0.001 m. The values of torque, axial trust, and shear rate, which were measured according to the change of the angular speed of the disk at a given concentration of silica, were used to obtain τ and N_1 .

The K , n , A , and b at various concentrations of silica are give in Table 2. As shown in Table 2, the values of A increased with increasing the silica concentration, which means that silica shows an elastic behavior (Ranade and Ulbrecht, 1978).

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

$$\lambda = \frac{N_1}{\mu\dot{\gamma}^2} \quad (20)$$

Using Eqs. (18) and (19), λ is rearranged as

Table 2. Physicochemical and rheological properties of CO₂ 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 propertie			
					n	K × 10 ³ (Ns ⁿ /m ²)	b	K × 10 ³ (Ns ⁿ /m ²)
0	1	1.97	0.039	1000	1.0	1.0	-	-
5.8	1.08	1.88	0.032	1039	0.97	1.115	0.097	0.03
11.91	1.27	1.82	0.031	1082	0.94	1.321	0.160	0.057
18.35	1.91	1.77	0.029	1134	0.92	1.525	0.190	0.082
23.06	2.35	1.72	0.028	1160	0.9	1.67	0.217	0.1
30.85	2.74	1.68	0.027	1180	0.85	1.95	0.232	0.129

$$\lambda = \frac{A}{K} \dot{\gamma}^{b-n-1} \quad (21)$$

One of 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:

$$De = \lambda/t = \frac{A}{K} \dot{\gamma}^{b-n-1} N \quad (22)$$

where $\dot{\gamma}$ is obtained in case of agitation of liquid in a cylindrical vessel as follows (Metzner and Otter, 1957):

$$\dot{\gamma} = 4\pi N/n \quad (23)$$

5. Results and discussion

5.1. Empirical correlation of volumetric liquid-side mass transfer coefficient of CO₂

To observe the effect of silica in the aqueous colloidal silica solution on R_{A0} and volumetric mass transfer coefficient ($k_L a$), R_A and $k_L a$, which were measured for the aqueous colloidal silica solutions having concentration in the range 0-31 wt% with the impeller speed as a parameter, were plotted against the silica concentration using the typical size (d) of the impeller at 0.05 m in Fig. 1 and 2, respectively. As shown in these figures, R_{A0} and $k_L a$ increases with increasing the agitation speed of the impeller and decreases with increasing the silica concentration. Same results were obtained at other impeller sizes of 0.034 and 0.075 m.

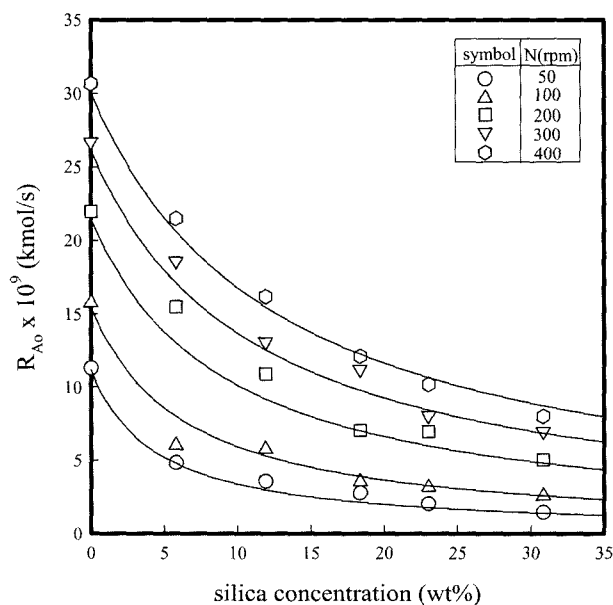


Fig. 1. Effect of silica concentration on R_{A0} at $d = 0.05$ m.

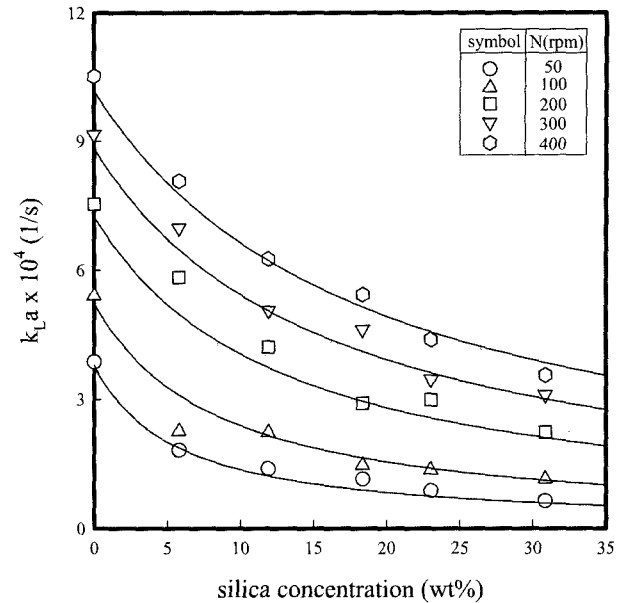


Fig. 2. Effect of silica concentration on $k_L a$ at $d = 0.05$ m.

The diffusivity, viscosity, and rheological properties of the aqueous colloidal silica solution may be considered when analyzing the relationship between $k_L a$ and the experimental variables such as the silica concentration, and the speed and size of the impeller.

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 viscosity is related to the diffusion coefficient. As shown in Table 2, the diffusivity of CO₂ 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 (Sandall and Patel, 1970) was used to correlate with $k_L a$, because the viscosity in the agitated vessel depends on the speed of impeller and the rheological properties as shown in Eq. (17) through Eq. (22).

To correlate $k_L a$ 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:

$$Sh = k_L a d^2 / D_A \quad (24)$$

$$Re = d^2 N \rho / \mu \quad (25)$$

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

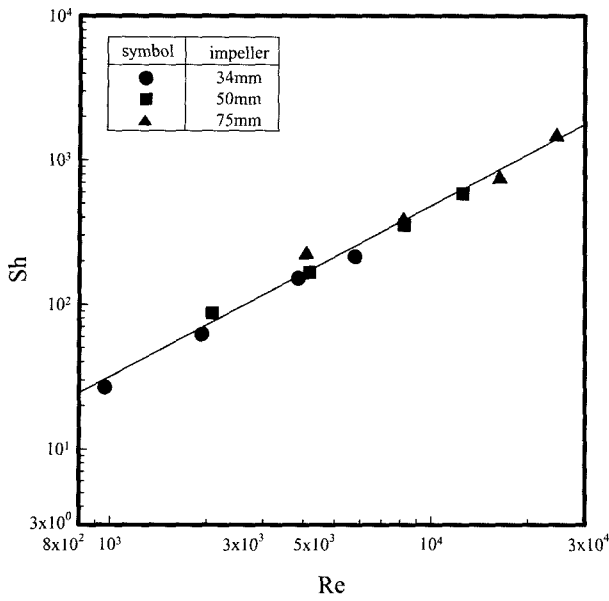


Fig. 3. Sh vs. Re in water for various impeller sizes.

$$Sh = 12.56 Re^{0.48} \quad (26)$$

The calculated values of Sh from Eq. (26) approached to the measured Sh very well with a standard deviation (SD) of 0.15 % and a mean deviation (MD) of 3.06 %. Eq. (26) 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. (24) and (25) 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. (26). 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 were 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 $(\mu/\mu_w)^{c_1}(1+c_2De)^{c_3}$ against Re, 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:

$$k_L ad^2/D_A = 12.56(d^2 N \rho/\mu)^{0.48} (\mu/\mu_w)^{0.11} (1+39.4De)^{-0.43} \quad (27)$$

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 $k_L a$. From the work of Perez and Sandall (1974), Yagi and Yoshida (1975), and

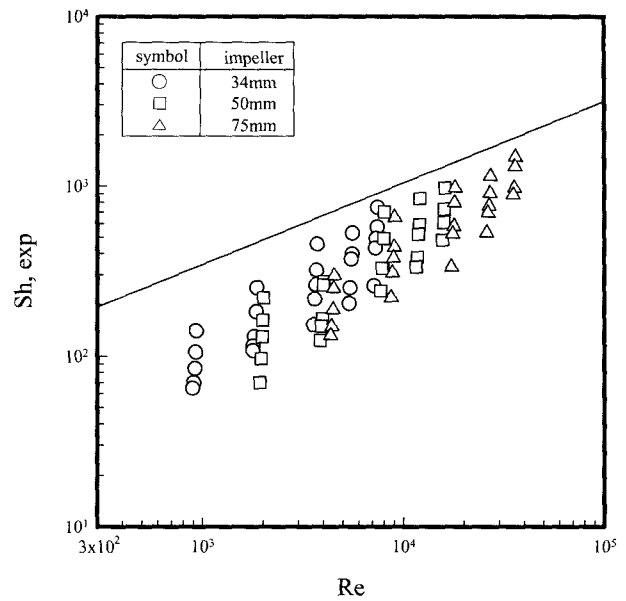


Fig. 4. Sh vs. Re in aqueous colloidal silica solution for various impeller sizes.

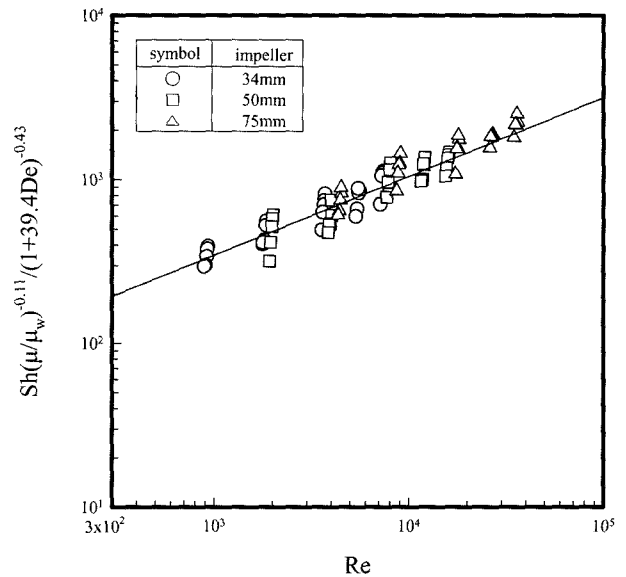


Fig. 5. Sh corrected with specific viscosity and Deborah number vs. Re for various impeller sizes in aqueous colloidal silica solution.

Moo-Young and Kawase (1987), it may be concluded that the exponent m in the relationship $k_L a \propto \mu^{-m}$ for stirred tanks is between 0.35 and 0.6. When our data were analyzed by Eq. (27) for the overall effect of viscosity on the $k_L a$, it comes out as $k_L a \propto \mu^{-0.37}$, which compares well with the findings of Yagi and Yoshida (1975) of $\mu^{-0.4}$ and Rande and Ulbrecht (1978) of $\mu^{-0.41}$, and Moo-Young and Kawase (1987) 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.

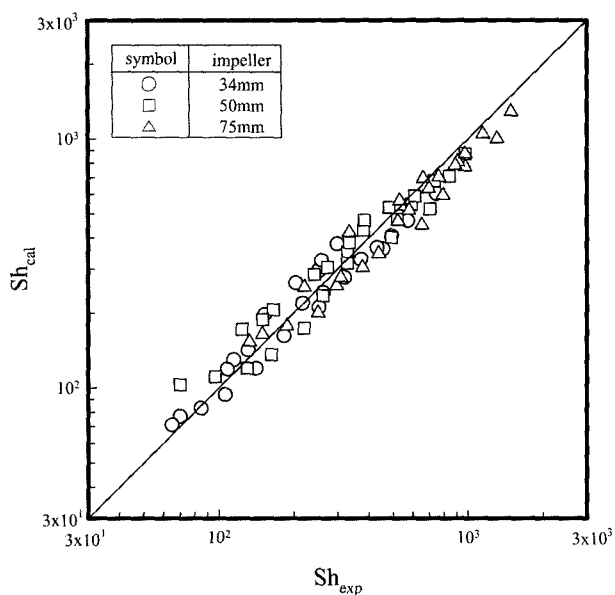


Fig. 6. Comparison of calculated Sh with experimental values.

Fig. 6 compares the experimental Sh with those predicted by Eq. (27). It can be seen that the agreement between the observed Sh and those predicted by Eq. (27) is very good; the mean deviation between the 75 experimental predicted points is 14.88%, and the standard deviation, 2.95 %.

5.2. Effect of rheological properties on the $k_L a$ of CO_2

To observe the effect of rheological properties of the aqueous colloidal silica solution on $k_L a$, the absorption rate of CO_2 was measured in the colloidal silica of 0~31 wt% to get $k_L a$. The circles in Fig. 7 shows the typical plots of the $k_L a$ 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. 7 represents the calculated values of $k_L a$ using Eq. (27). As shown in Fig. 7, the value of $k_L a$ decreases with increasing silica concentration.

In absorption of gas into aqueous slurries, which have an adsorption capacity of the dissolved gas, $k_L a$ increases with increasing the concentration of fine particles, and this behavior was explained by the gazing effect (Kars and Best, 1979). But, the $k_L a$ in this study does not increase with increasing silica concentration as 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 $k_L a$ with increasing the silica concentration might be explained due to the inverse proportion of viscosity to diffusivity, because the diffusivity of CO_2 decrease with increasing the apparent viscosity of the aqueous colloidal silica solution as shown in Table 2. Hikita *et al.* (1985) have shown that the diffusivity of CO_2 decreases

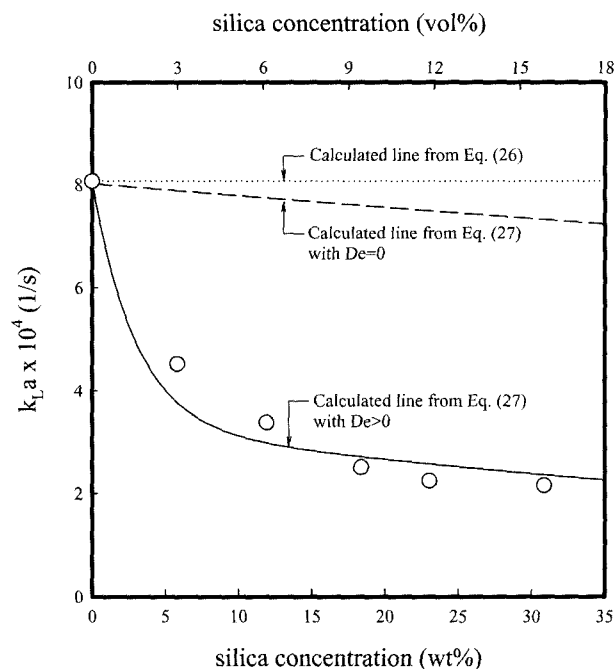


Fig. 7. Effect of De on $k_L a$ at $d = 0.034$ m and $N = 50$ rpm.

with increasing the volume fraction of fine particle in aqueous slurries.

Because $k_L a$ is affected by both of viscosity and elasticity of the aqueous colloidal silica solution as shown in Eq. (27), the estimated values of $k_L a$ 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 actual aqueous colloidal silica solution. As shown in Fig. 7, the $k_L a$ 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, $k_L a$ in water is larger than that in the aqueous colloidal silica solution. If the aqueous colloidal silica solution in this study would have only viscous behavior, the $k_L a$ should be the $k_L a$ represented as the dashed line. But, the actual $k_L a$ 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 $k_L a$ is stronger than the effect of viscosity on $k_L a$, in other words, the elasticity of the aqueous colloidal silica solution reduces $k_L a$ due to the decrease of $k_L a$ as shown in Eq. (27) based on the same viscosity of

the solution.

Astarita *et al.* (1969) and Seyer and Metzner (1969) 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 $k_L a$ in a viscoelastic non-Newtonian liquid such as PAA is attributable to the elasticity of the liquid phase.

The reduction of $k_L a$ 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 $k_L a$.

5.3. Effect of rheological properties on the chemical absorption of CO₂

To observe the effect of elasticity of the aqueous colloidal silica solution on the chemical reaction of CO₂ with AMP, the absorption rate of CO₂ into aqueous colloidal silica solution with AMP was measured according to change of silica concentration in the range of 0-31 wt% at AMP concentration 0~2 kmol/m³ under the experimental conditions of the agitation speed of 50 rev/min with the impeller size of 0.034 m.

Fig. 8 shows the plots of the absorption rate of CO₂ against the silica concentration for various AMP concentrations. As shown in Fig. 8, R_A decreases with increasing silica concentration, and increases with increasing AMP concentration. Increase of R_A with increasing AMP concentration is due to the reactant of AMP in reaction of CO₂.

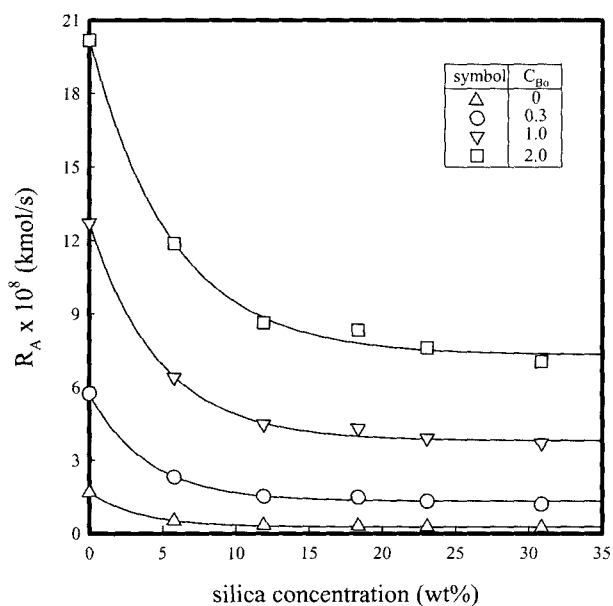


Fig. 8. Effect of silica concentration on absorption rate for various AMP concentrations.

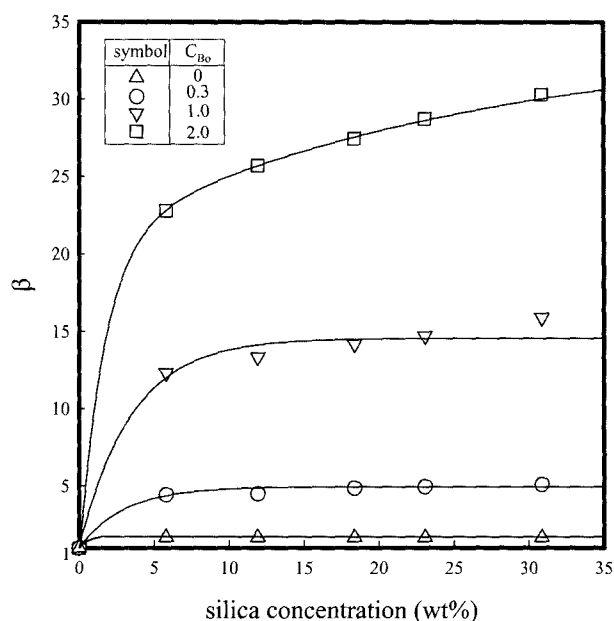


Fig. 9. Effect of silica concentration on enhancement factor for various AMP concentrations.

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

R_A is obtained from Eq. (12) using $k_L a$, β , and C_{Ai} . As shown in Fig. 7, $k_L a$ 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) using the numerical solution of Eq. (7) and (8) by the finite element method at given concentrations of silica and AMP, and plotted against the silica concentration in Fig. 9. As shown in Fig. 9, the values of β hold to be constant almost with increasing silica concentration and increase with increasing AMP concentration, *i.e.*, decrease of $k_L a$ by increase of silica concentration did not affect β , although k_L obtained from Eq. (27) was used to estimate a theoretical β . Therefore, decrease of R_A was affected mainly by decrease of $k_L a$ rather than β and C_{Ai} . Reduction of $k_L a$ by the elastic properties of the aqueous colloidal silica solution causes decrease of chemical absorption rate of CO₂.

6. Conclusions

The measured rate of chemical absorption of carbon dioxide into the aqueous colloidal silica solution of 0-31 wt% with AMP of 0-2 kmol/m³ in a flat-stirred vessel for the various sizes and speeds of impeller at 25 °C and 0.101 MPa were compared with that estimated from the model based on the film theory accompanied by chemical reaction using the value of $k_L a$. The value of $k_L a$, which was used to estimate the enhancement factor, was obtained from the empirical equation as follow:

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

The chemical absorption rate was decreased due to the reduction of $k_L a$ by elasticity of the aqueous colloidal silica solution.

Acknowledgments

This work was supported with the Basic Research Program of the Korea Science and Engineering Foundation (KOSEF) through ARC and Brain Korea 21 Project.

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