

Mass Transfer of Carbon Dioxide in Aqueous Polyacrylamide Solution with Methyldiethanolamine

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Abstract—Carbon dioxide was absorbed into aqueous polyacrylamide (PAA) solution containing methyl-diethanolamine (MDEA) in a flat-stirred vessel to investigate the effect of non-Newtonian rheological behavior of PAA on the rate of chemical absorption of CO₂, where the reaction between CO₂ and MDEA was assumed to be a first-order reaction with respect to the molar concentration of CO₂ and MDEA, respectively. The liquid-side mass transfer coefficient (k_L), which was obtained from the dimensionless empirical equation containing the viscoelasticity properties of a non-Newtonian liquid, was used to estimate the enhancement factor due to chemical reaction. PAA with elastic property of non-Newtonian liquid made the rate of chemical absorption of CO₂ accelerate compared with a Newtonian liquid.

Key words: Chemical Absorption, Carbon Dioxide, PAA, MDEA, Non-Newtonian Liquid

INTRODUCTION

Gas-liquid mass transfer in non-Newtonian liquid is an important example of gas absorption in pseudoplastic flow of industrial processes such as fermentation broth, slurry, and fluidized bed. The volumetric liquid-phase mass transfer coefficient ($k_L a$) in gas-dispersed systems consists of the mass transfer coefficient (k_L) and the specific gas-liquid interfacial area (a). The former could be correlated with Reynolds and Schmidt numbers including liquid viscosity. It is likely that the latter varies not only with Newtonian liquid properties such as surface tension but also with some non-Newtonian and/or viscoelastic fluid properties.

The use of only the apparent viscosity of non-Newtonian fluids was not sufficient to obtain a unified correlation for $k_L a$ 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 $k_L a$ is due to the viscoelasticity of the aqueous solution, 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. One of the dimensionless numbers, which relate the elastic properties to the process parameters, is the Deborah number (De) defined as the ratio of the material's characteristic relaxation time to the characteristic flow time. Unified correlations have been proposed for $k_L a$ in Newtonian as well as non-Newtonian solutions by introducing a dimensionless term such as $(1 + n_1 De^{n_2})^{n_3}$, which are listed in Table 1.

There is little information about the effect of elastic properties on chemical absorption of gas in a non-Newtonian liquid. Park et al. [2004] presented the effect of elasticity of polyisobutylene (PIB)

in the benzene solution of polybutene (PB) and PIB on chemical absorption of CO₂ in w/o emulsion composed of aqueous alkaline 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 CO₂. It is worthwhile to investigate the effect of non-Newtonian rheological behavior on the rate of chemical absorption of a gas, where a reaction between CO₂ and reactant occurs in the aqueous phase.

In this study, the chemical absorption mechanism of CO₂ into an aqueous PAA solution with MDEA is presented, and the measured absorption rates of CO₂ are compared with those obtained from the model based on the penetration theory with chemical reaction. The volumetric mass transfer coefficient obtained from the empirical formula is used to estimate the enhancement factor due to chemical reaction.

THEORY

The problem to be considered is that a gaseous species A (CO₂) dissolves into the liquid phase and then reacts irreversibly with species B according to



The stoichiometric coefficient (ν) in Eq. (1) for MDEA was obtained from the reference [Ko and Li, 2000] and its value was 1.

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:

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

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Table 1. Coefficients of dimensionless Deborah number, $(1+n_1De^{n_2})^{n_3}$ for gas-liquid mass transfer correlation

Investigator	n_1	n_2	n_3	Polymer	Contactors
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
Nakanoh and Yoshida, 1980	0.13	0.55	-1	CMC, PA	Bubble column
Park et al., 2003a	100	1	-0.42	PB, PIB	Agitated vessel
Park et al., 2003b	2461.3	1	-0.274	PB, PIB	Agitated vessel

Under the assumptions mentioned above, the conservation equations of species A and B are given as

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

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

Boundary and initial conditions to be imposed are

$$z=0, t>0; C_A=C_{Ai}, \frac{\partial C_B}{\partial z}=0 \quad (5)$$

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

$$z=\infty, t>0; C_A=0, C_B=C_{Bo} \quad (7)$$

Eqs. (3)–(7) are put into the dimensionless form as follows:

$$\frac{\partial^2 a}{\partial x^2} = \frac{\partial a}{\partial \theta} + ab \quad (8)$$

$$\frac{\partial^2 b}{\partial x^2} = r \frac{\partial b}{\partial \theta} + \nu r q ab \quad (9)$$

$$x=0, \theta>0; a=1, \frac{\partial b}{\partial x}=0 \quad (10)$$

$$x>0, \theta=0; a=0, b=1 \quad (11)$$

$$x=\infty, \theta>0; a=0, b=1 \quad (12)$$

where $a=C_A/C_{Ai}$, $b=C_B/C_{Bo}$, $x=z\sqrt{k_2 C_{Bo}/D_A}$, $\theta=k_2 C_{Bo} t$, $r=D_A/D_B$, $q=C_{Ai}/C_{Bo}$

The molar flux of CO₂ with chemical reaction at any contact time, t is defined as

$$N_A = -D_A \left. \frac{\partial C_A}{\partial z} \right|_{z=0} \quad (13)$$

The mean molar flux of CO₂ during contact time, t is written as

$$\bar{N}_A = \frac{1}{t} \int_0^t N_A dt \quad (14)$$

The mean molar flux without chemical reaction based on the penetration model during contact time has been derived as follows [Higbie, 1935]:

$$\bar{N}_A^o = 2C_{Ai} \sqrt{\frac{D_A}{\pi t}} \quad (15)$$

From comparison of the penetration model with the film model, the relation between t and k_L is derived as follows [Higbie, 1935]:

$$k_L = 2 \sqrt{\frac{D_A}{\pi t}} \quad (16)$$

The enhancement factor (β) here defined as the ratio of molar flux with chemical reaction to that without chemical reaction, \bar{N}_A/\bar{N}_A^o , is described by using Eq. (14) and Eq. (15) as follows:

$$\beta = -\frac{\pi}{4H_A} \int_0^\theta \left. \frac{\partial a}{\partial x} \right|_{x=0} d\theta \quad (17)$$

where $H_A = \sqrt{D_A k_2 C_{Bo}}/k_L$

The absorption rate of CO₂ (R_A) is expressed from β defined in Eq. (17) as follows:

$$R_A = \beta R_{Ao} = \beta k_L a C_{Ai} \quad (18)$$

where R_{Ao} is the absorption rate multiplied the mean molar flux (\bar{N}_A^o) by the contact area of gas and liquid. The value of β in Eq. (18) is estimated from Eq. (17) by using a numerical solution with FEM-LAB at the contact time of $4D_A/\pi k_L^2$ calculated from Eq. (16).

EXPERIMENTAL

1. Chemicals

All chemicals in this study were reagent grade, and used without further purification. Purity of both CO₂ and N₂ was more than 99.9%. The polymers used in this study were polyacrylamide (PAA) with the mean molecular weight of 10000 (by Aldrich chemical company, U.S.A.). MDEA (Aldrich, U.S.A.) was used as reagent grade without purification.

2. Rate of Absorption

Absorption experiments were carried out in an agitated vessel constructed of glass 0.102 m inside diameter and 0.157 m in height. Four equally spaced vertical baffles, each one-tenth the vessel diameter in width, were attached to the internal wall of the vessel. The liquid phase was agitated with an agitator driven by a 1/4 Hp variable speed motor without agitation in gas phase because of pure CO₂ gas. A straight impeller with 0.034, 0.05, and 0.07 m in length and 0.011 m in width was used as the liquid phase agitator, and located at the middle position of the liquid phase. The gas and liquid in the vessel were agitated in the range of 50 to 400 rpm. The absorption rate of CO₂ was measured in the aqueous solution of PAA of 0–100 kg/m³ and MDEA of 0–2 kmol/m³ under the experimental conditions such as an impeller speed of 50–400 rpm at 101.3×10^3 N/m² and 25 °C following the procedure reported elsewhere [Park et al., 2003a].

PHYSICOCHEMICAL AND RHEOLOGICAL PROPERTIES

Table 2. The physicochemical and rheological properties of CO₂ and PAA aqueous solution

PAA (kg/m ³)	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	b	A×10 ³
0	1	1.970	0.039	1000	1.0	1.0	-	-
0.1	1.002	1.9694	0.039	1000	0.99	1.07	0.43	27.3
0.5	1.004	1.9688	0.039	1003	0.94	1.25	0.38	28.5
1	1.011	1.9666	0.038	1005	0.92	1.34	0.25	30.9
5	0.172	1.9486	0.038	1008	0.89	1.71	0.18	46.5
10	2.155	1.7463	0.036	1010	0.86	3.71	0.13	57.6
50	2.811	1.6750	0.036	1016	0.78	6.16	0.11	188.2
100	3.451	1.6219	0.035	1032	0.76	7.15	0.1	294.8

1. Solubility of CO₂ in Aqueous PAA Solution

The pressure measuring method in this study involved measuring the pressure difference of CO₂ between before and after equilibrium between gas and liquid phase, similar to the procedure reported elsewhere [Kennard and Meiser, 1984] to get the solubility (C_{Ai}) of CO₂ in the aqueous solutions of PAA at 25 °C and 0.101 MPa. The experimental procedure duplicated that as reported in published research [Park et al., 2003a] in detail. The solubility (C_{Ai}) of CO₂ in aqueous MDEA [Ko and Li, 2000] solution was estimated as follows:

$$C_{Ai} = 1/[2824900 \exp(-2119/T)/101.3]$$

2. Density and Apparent Viscosity of Aqueous PAA Solution

The density of the aqueous solution of PAA was measured at 25 °C within 0.1 kg/m³ 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 aqueous solution of PAA was measured at 25 °C with Brookfield viscometer. (Brookfield Eng. Lab. Inc, USA).

3. Reaction Rate Constant

In the reaction of CO₂ with MDEA [Ko and Li, 2000], the reaction rate constant (k_2) was estimated as follows:

$$k_2 = 4.01 \times 10^8 \exp(-5400/T)$$

4. Diffusivities of CO₂ and MDEA

The diffusivity (D_{AMDEA}) of CO₂ in aqueous MDEA [Ko and Li, 2000] solution was estimated as follows:

$$D_{AMDEA} = 2.35 \times 10^{-6} \exp(-2119/T)$$

The diffusivity (D_{BMEA}) of MDEA in aqueous MDEA solution was obtained from the assumption that the ratio of D_{BMEA} to D_{AMDEA} was equal to the ratio in water [Nijing et al., 1959]. The diffusivity of CO₂ and MDEA in water at 25 °C were taken as 1.97×10^{-9} m²/s [Danckwerts and Sharma, 1966] and 8.1×10^{-10} m²/s [Ko and Li, 2000], respectively.

The diffusivity of a solute of a small size such as CO₂, O₂ or CH₄ in a polymer solution depends on the viscosity of the solution and the molecular weight of the polymer. The diffusivity (D_A) of CO₂ and that (D_B) of MDEA in the aqueous PAA solution were obtained from the following equations suggested by Lohse et al. [1981], which were modified from the Stoke-Einstein equation, and correlated with the molecular weight of the polymer in the solution, respectively.

$$D_A/D_{AMDEA} = (\mu_w/\mu)^{3.7\sqrt{M_w/M_p}}$$

$$D_B/D_{BMDEA} = (\mu_w/\mu)^{3.7\sqrt{M_w/M_p}}$$

The values of solubility and diffusivity of CO₂, density and apparent viscosity of aqueous PAA solution are given in Table 2.

5. Rheological Properties of Aqueous PAA 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 PAA solutions.

$$\tau = K\dot{\gamma}^n \quad (19)$$

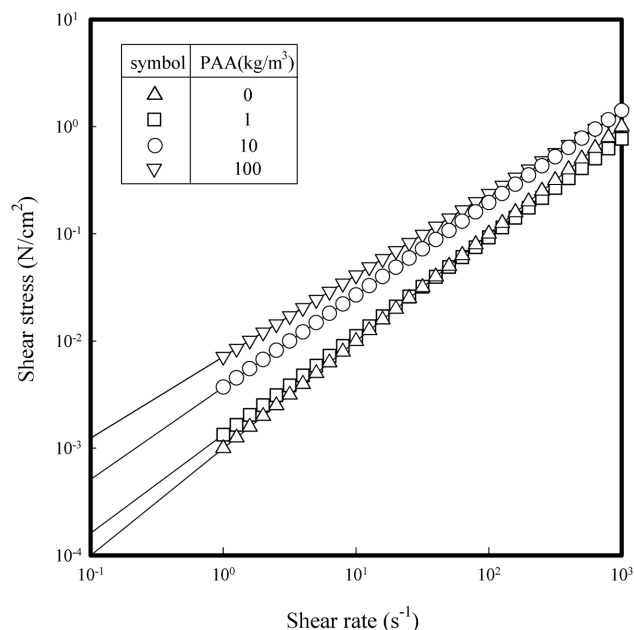
$$\mu = K\dot{\gamma}^{n-1} \quad (20)$$

$$N_1 = A\dot{\gamma}^b \quad (21)$$

where n , K , b , and A are material parameters depending on temperature.

These parameters were obtained from the dependence of τ and N_1 on $\dot{\gamma}$.

To observe the dependence of τ and N_1 on $\dot{\gamma}$, τ and N_1 of the aqueous PAA solution were measured according to the change of $\dot{\gamma}$ by

**Fig. 1. Shear stress of PAA aqueous solution as a function of shear rate.**

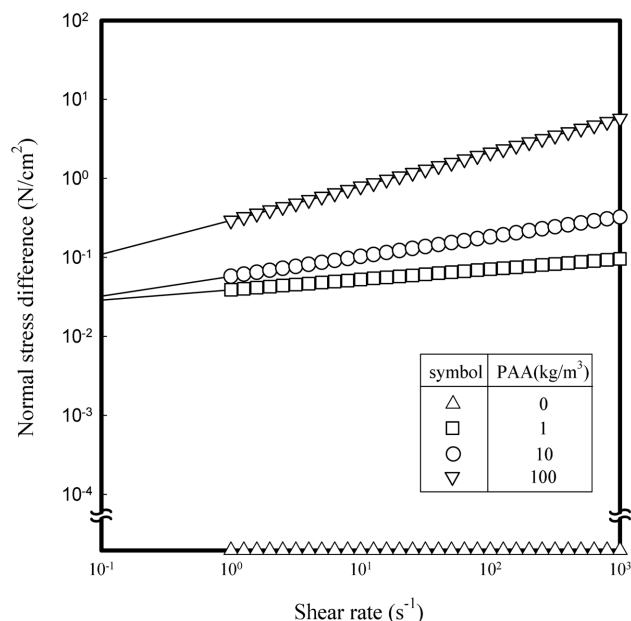


Fig. 2. Normal stress difference of PAA aqueous solution as a function of shear rate.

the parallel disk type rheometer (Ares, Rheometrics, U.S.A.) of the diameter of 0.05 m and the gap of 0.001 m.

Fig. 1 shows the typical logarithmic plot of shear stress vs. shear rate for the aqueous PAA solutions in the range of 0–100 kg/m³. The best straight-line fit was determined by the least-squares method with the plots in Fig. 1. From the intercept and slope of the line, the values of K and n were evaluated. Also, Fig. 2 shows the logarithmic plot of primary normal stress difference vs. shear rate for the same solution in Fig. 1. As shown in Fig. 2, the plots are linear, and the values of A and b were evaluated from the intercept and slope of the straight line. The parameters, K , n , A and b in the aqueous solution of various concentration of PAA are given in Table 2. As shown in Table 2, the values of b and A increased with increasing the concentration of PAA, which means that PAA has an elastic behavior.

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

$$\lambda = \frac{N_1}{\mu \dot{\gamma}} \quad (22)$$

Using Eq. (20) and Eq. (21), λ is rearranged as

$$\lambda = \frac{A}{K} \dot{\gamma}^{b-n-1} \quad (23)$$

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, which is related to the reciprocal of the impeller speed in case of stirred tanks, and De is derived as follows:

$$De = \lambda/t = \frac{A}{K} \dot{\gamma}^{b-n-1} N \quad (24)$$

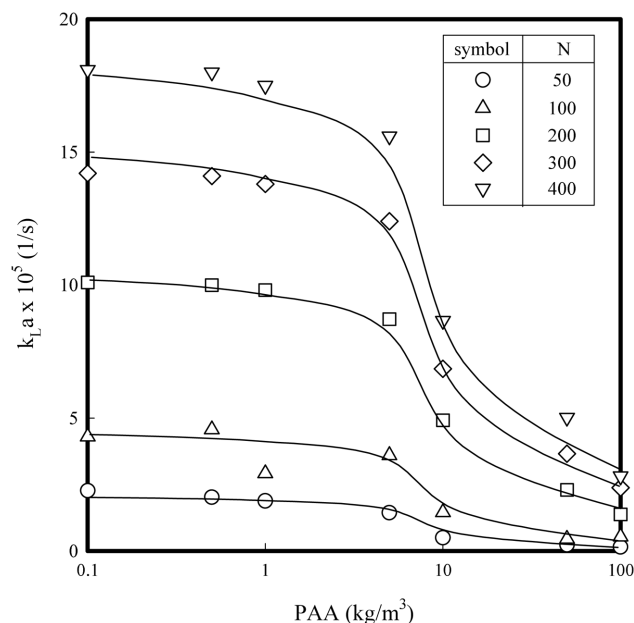


Fig. 3. Effect of PAA concentration on $k_L a$ of CO_2 at $d=0.034$ m.

The shear rate ($\dot{\gamma}$) in Eq. (24) is obtained in case of agitation of liquid in a cylindrical vessel as follows [Metzner and Otter, 1957]:

$$\dot{\gamma} = 4\pi N/n \quad (25)$$

RESULTS AND DISCUSSION

1. Empirical Correlation of Liquid-Side Mass Transfer Coefficient of CO_2

To observe the effect of the concentration of PAA with the impeller speed and size as parameters on the volumetric mass transfer coefficient ($k_L a$), the measured $k_L a$ in the aqueous solution of PAA concentration in the range of 0.1–100 kg/m³ was plotted against PAA concentration in Fig. 3. As shown in Fig. 3, $k_L a$ increases with the increase of the speed of the impeller and decreases with the increase of PAA concentration.

In analyzing the relationship between $k_L a$ and the experimental variables such as the PAA 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 viscosity is related to the diffusion coefficient. As shown in Table 2, the diffusivity of CO_2 in the aqueous PAA 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 PAA 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. (20) through Eq. (25).

To correlate $k_L a$ with the experimental variables such as the PAA 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:

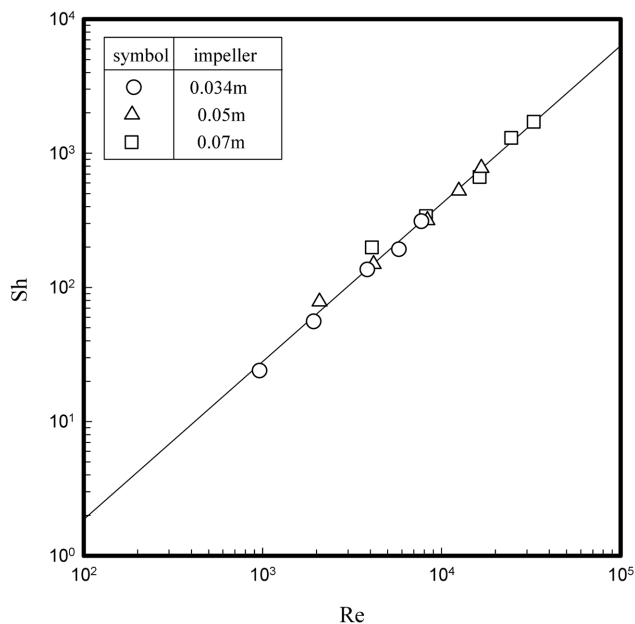


Fig. 4. Sh vs. Re in water at various impeller size.

$$Sh = k_L a d^2 / D_A \quad (26)$$

$$Re = d^2 N \rho / \mu \quad (27)$$

Fig. 4 shows logarithmic plots of Sh against Re in case of water as absorbent of CO₂. As shown in Fig. 4, 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:

$$Sh = 0.082 Re^{1.17} \quad (28)$$

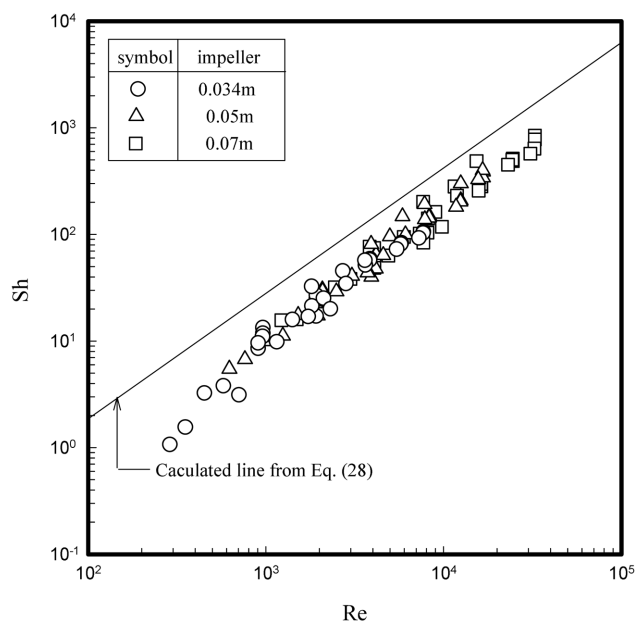


Fig. 5. Dimensionless correlation of volumetric mass transfer coefficient of CO₂ in PAA aqueous solution at various impeller size.

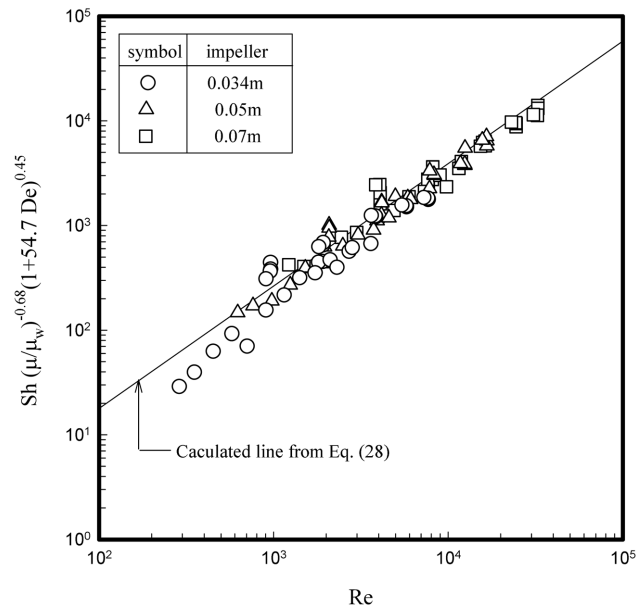


Fig. 6. Dimensionless correlation of volumetric mass transfer coefficient of CO₂ in PAA aqueous solution with corrected specific viscosity and Deborah number at various impeller size.

The calculated values of Sh from Eq. (28) approached to the measured Sh very well with a standard deviation (SD) of 0.81% and a mean deviation (MD) of 6.37%.

Fig. 5 shows logarithmic plots of Sh against Re for the aqueous PAA solutions, and the solid line in Fig. 5 presents the Sh calculated from Eq. (28). As shown in Fig. 5, the plots were scattered from the straight line with SD of 33.75% and MD of 56.93%. This may be due to non-Newtonian behavior of the aqueous PAA solution.

The new terms corrected with viscosity and De was used to lessen the deviation of the plots for the PAA solution from the plots for water as shown in Fig. 4. A simple multiple regression exercise was used for the plots of Sh combined with $(\mu/\mu_w)^{c_1} (1+c_2 De)^{c_3}$ against Re, which gave the values of c_1 , c_2 and c_3 are 0.68, 54.7, and -0.45, respectively, with SD of 8.07% and MD of 22.49% as shown in Fig. 6.

Using a total of 105 data points, a multiple regression analysis came up with a correlation as follows:

$$k_L a d^2 / D_A = 0.082 (d^2 N \rho / \mu)^{1.17} (\mu / \mu_w)^{0.68} (1 + 54.7 De)^{-0.45} \quad (29)$$

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

To observe the effect of rheological properties of aqueous PAA solution on the rate of chemical absorption, the absorption rate of CO₂ into aqueous PAA solution with MDEA was measured according to change of MDEA concentration in the range of 0-2 kmol/m³. Fig. 7 and 8 show the typical plots of the absorption rate of CO₂ against the concentration of MDEA at PAA concentration of 1 and 100 kg/m³, respectively, under the experimental conditions of the agitation speed of 50 rpm with the impeller size of 0.034 m. The triangle and circle in Fig. 7 and 8 represent the measured values of absorption rate of CO₂ in water with MDEA and aqueous PAA solution with MDEA, respectively.

The three lines in both the figures represent the estimated absorp-

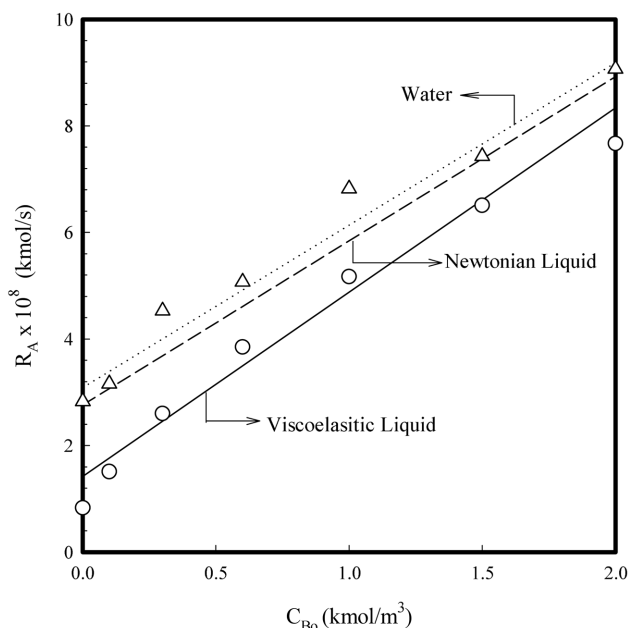


Fig. 7. Effect of C_{B0} on R_A at aqueous solution of PAA of 1 kg/m³ ($d=0.034$ m, $N=50$ rpm).

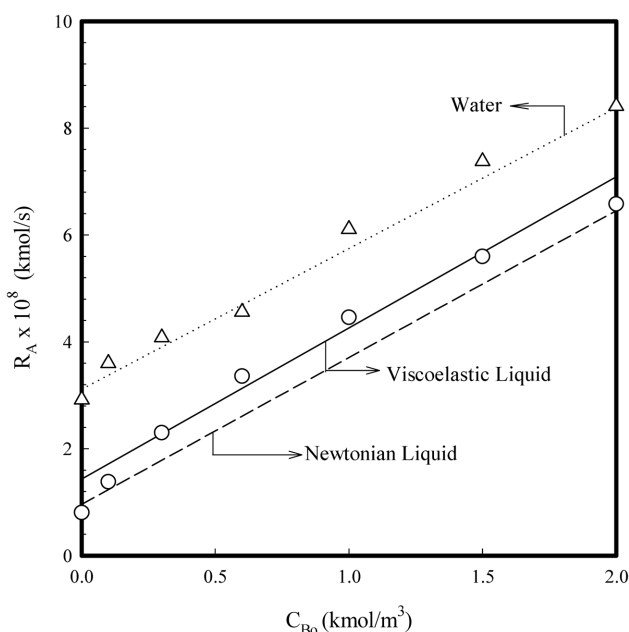


Fig. 8. Effect of C_{B0} on R_A at aqueous solution of PAA of 100 kg/m³ ($d=0.034$ m, $N=50$ rpm).

tion rate of CO₂ according to the kind of the absorbents, i.e., $k_L a$. The value of $k_L a$ used to get the solid line, dashed line, and dotted line comes from Eq. (29), Eq. (29) without De , and Eq. (28), respectively. The value of $k_L a$ in the solid line is affected by both of viscosity and elasticity, that in the dashed line by viscosity of aqueous PAA solution, and that in the dotted line by viscosity of water. As shown in Figs. 7 and 8, R_A increases with increasing MDEA concentration, and R_A in the solid line is smaller than that in the dashed line in Fig. 7; on the other hand, R_A of the solid line is larger than that in the dashed line in Fig. 8. From comparison of R_A of Fig. 7

with that of Fig. 8, the effect of the elasticity of the aqueous solution with PAA of 100 kg/m³ on R_A is stronger than that of 1 kg/m³, on the other hand, the effect of the viscosity of that of 1 kg/m³ on R_A is stronger than that of 100 kg/m³.

CONCLUSIONS

Rates of the chemical absorption of CO₂ in the aqueous solution of PAA at 0.1–100 kg/m³ with MDEA at 0–2 kmol/m³ were measured in a flat-stirred vessel to determine the influence of the rheological properties of PAA on the absorption rate under the experimental conditions such as the impeller size of 0.034, 0.05 and 0.07 m and the agitation speed of 0–400 rpm at 25 °C and 0.101 MPa. The elastic property such as Deborah number of the aqueous PAA solution was considered to obtain an empirical correlation of the volumetric mass transfer coefficient in the non-Newtonian liquid, which is used to estimate the enhancement factor for the chemical absorption. There is a concentration range of PAA which makes the mass transfer coefficient and absorption rate increase due to the comparison of the relative magnitude of viscosity and elasticity of the aqueous PAA solution.

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NOMENCLATURE

C_A	: concentration of CO ₂ [kmol/m ³]
C_B	: concentration of MDEA [kmol/m ³]
d	: diameter of impeller [m]
k	: reaction rate constant [m ³ /kgmol·s]
$k_L a$: volumetric mass transfer coefficient of liquid side [1/s]
N	: speed of impeller [1/s]
N_1	: primary normal stress difference [N/m ²]
t	: time [s]
T	: temperature [K]
z	: coordinate in film thickness direction in benzene phase [m]

Greek Letters

γ	: shear rate [1/s]
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	: MDEA
i	: gas-liquid interface
o	: bulk body

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