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Abstract: Carbon dioxide was absorbed into aqueous polyethylene oxide (PEO) solution containing monoethanolamine (MEA) in a flat-stirred vessel to investigate the effect of non-Newtonian rheological behavior of PEO on the rate of chemical absorption of CO_2, where the reaction between CO_2 and MEA was assumed to be a first-order reaction with respect to the molar concentration of CO_2 and MEA, respectively. The liquid-side mass transfer coefficient (k_L), which was obtained from the dimensionless empirical equation containing the properties of viscoelasticity of the non-Newtonian liquid, was used to estimate the enhancement factor due to chemical reaction. PEO with elastic property of non-Newtonian liquid made the rate of chemical absorption of CO_2 accelerate compared with Newtonian liquid based on the same viscosity of the solution.

Keywords: Chemical absorption, carbon dioxide, PEO, monoethanolamine, 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 a

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fermentation broth, slurry, and fluidized bed, et al. (1–3). Variation of the volumetric liquid-phase mass transfer coefficient (k_La) 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.

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, 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 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 the dimensionless term such as $(1 + n_1De^{n_2})^{n_3}$, which are listed in Table 1. As shown in Table 1, values of in the dimensionless group are different one another.

There is little information about the effect of elastic properties on chemical absorption of gas in non-Newtonian liquid. Park et al.(6) presented the effect of elasticity of polyisobutylene (PIB) in the benzene solution of polybutene (PB) and PIB on chemical absorption of CO_2 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 CO_2. 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 CO_2 and reactant occurs in the aqueous phase.

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

Investigator	n_1	n_2	n_3	Polymer	Contactor
Yagi and Yoshida (1)	2	0.5	−0.67	CMC, PA	Agitated vessel
Ranade and Ulbrecht (2)	100	1	−0.67	CMC, PAA	Stirred tank
Nakanoh and Yoshida (3)	0.13	0.55	−1	CMC, PA	Bubble column
Park et al. (4)	100	1	−0.42	PB, PIB	Agitated vessel
Park et al. (5)	2461.3	1	−0.274	PB, PIB	Agitated vessel

In this study, the chemical absorption mechanism of CO_2 into an aqueous PEO solution with MEA is presented to observe the effect of rheological property on the chemical absorption, and the measured absorption rates of CO_2 are compared with those obtained from the model based on the film 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 (CO_2) dissolves into the liquid phase and then reacts irreversibly with species B according to

$$\mathrm{A} + \nu \mathrm{B} \rightarrow \mathrm{P} \tag{1}$$

The stoichiometric coefficients (ν) in Eq. 1 for MEA was obtained from the reference (7) and its value was 2.

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

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

$$r_A = k_2 C_A C_B \tag{2}$$

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

$$D_A \frac{\partial^2 C_A}{\partial z^2} = k_2 C_A C_B \tag{3}$$

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

Boundary and conditions to be imposed are

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

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

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

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

$$\frac{\partial^2 b}{\partial x^2} = \nu \mathrm{rqab} \tag{8}$$

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

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

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

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

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

β is estimated by a solution of Eq. 7 and 8 using a numerical analysis of the finite element method and used to predict the absorption rate(R_A) of CO_2 with chemical reaction as follows:

$$R_A = \beta R_{Ao} = \beta k_L S C_{Ai} \tag{12}$$

where R_{Ao} is the physical absorption rate multiplied the molar flux by the interfacial area between gas and liquid (S).

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%. The polymers used in this study were PEO with the mean molecular weight of 20000 (by Aldrich chemical company, U.S.A.). MEA (Aldrich, U.S.A.) was used as reagent grade without purification.

Rate of Absorption

An agitated vessel used for measurement of absorption rate of CO_2 was constructed of glass of 0.102 m inside diameter and of 0.151 m in height with four equally spaced vertical baffles. 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 200 rev/min. The absorption rate of CO_2 was measured in the aqueous solution of PEO of $0 \sim 30\,kg/m^3$ and MEA of $0 \sim 2\,kmol/m^3$ under the experimental conditions such as an impeller speed of 50-400 rev/min along the procedure similar to those reported elsewhere(4) at 1 atm and 25°C.

PHYSICOCHEMICAL AND RHEOLOGICAL PROPERTIES

Solubility of CO_2 in Aqueous PEO Solution

The pressure measuring method in this study 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 (8) to get the solubility (C_{APi}) of CO_2 in the aqueous solutions of PEO at 25°C and 0.101 MPa. The experimental procedure was duplicated that as reported in the published research (4) in detail. The solubility (C_{Ai}) of CO_2 in aqueous MEA (7) solution was estimated as follows:

$$\mathrm{Log}(C_{Ai}/C_{APi}) = 0.3C_{Bo}/(1 + 0.963C_{Bo})$$

Density and Apparent Viscosity of Aqueous PEO Solution

The density of the aqueous solution of PEO 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 aqueous solution of PEO was measured at 25°C with Brookfield viscometer. (Brookfield Eng. Lab. Inc, USA)

Reaction Rate Constant

In the reaction of CO_2 with MEA (7), the reaction rate constant (k_2) was estimated as follows.

$$\log k_2 = 10.99 - \frac{2152}{T}$$

Diffusivities of CO_2 and MEA

The diffusivity (D_{AMEA}) of CO_2 in aqueous MEA(9) solution was estimated as follows:

$$D_{AMEA} = (1.9686 - 0.184C_{Bo} - 0.0429C_{Bo}^2) \times 10^{-9}$$

Diffusivity (D_{BMEA}) of MEA in aqueous MEA solution was obtained from assumption that the ratio of D_{BMEA} to D_{AMEA} was equal to the ratio in water(10). The diffusivity of CO_2 and MEA in water at 25°C were taken as $1.97 \times 10^{-9}\ m^2/s$ (11) and $3.24 \times 10^{-9}\ m^2/s$ (9), respectively.

The diffusivity of a solute of a small size such as CO_2, O_2 or CH_4 in a polymer solution depends on the viscosity of the solution and the molecular weight of the polymer. The diffusivity (D_A) of CO_2 and that (D_B) of MEA in the aqueous PEO solution were obtained from the following equations suggested by Lohse et al. (12), which were modified from the Stoke-Einstein equation, and correlated with the molecular weight of the polymer in the solution, respectively.

$$D_A/D_{AMEA} = (\mu_W/\mu)^{3.7\sqrt{M_W/M_P}}$$

$$D_B/D_{BMEA} = (\mu_W/\mu)^{3.7\sqrt{M_W/M_P}}$$

where M_w and M_p are molecular weight of water and polymer, respectively.

The values of solubility, diffusivity of CO_2, density, and apparent viscosity of aqueous PEO solution are given in Table 2.

Rheological Properties of Aqueous PAA Solution

We assume that a power-law model, which has been widely used for shear-dependent viscosity, can be represented the non-Newtonian flow behavior of aqueous PEO solutions.

$$\tau = K\gamma^n \tag{13}$$

$$\mu = K\gamma^{n-1} \tag{14}$$

$$N_1 = A\gamma^b \tag{15}$$

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 in the aqueous solution of various concentration of PEO are given in Table 2. As shown in Table 2, the values of A increased with increasing of the concentration of PEO, which means that PEO has an elastic behavior (2).

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

$$\lambda = \frac{N_1}{\mu\gamma^2} \tag{16}$$

Table 2. The physicochemical and rheological properties of CO_2 and aqueous PEO solution

					Rheological propertie			
C (kg/m^3)	Viscosity $(Ns/m^2) \times 10^3$	Diffusivity $(m^2/s) \times 10^9$	Solubility $(kmol/m^3)$	Density (kg/m^3)	n (–)	K $(Ns^n \cdot m^2)$	b (–)	A $(Ns^n \cdot m^2)$
0	1	1.950	0.039	1000	1.0	0.001	–	–
10	3.05	1.875	0.034	1001.4	0.529	0.019	0.084	0.134
20	8.17	1.811	0.029	1003.2	0.485	0.416	0.099	1.255
30	14.9	1.774	0.028	1004.8	0.453	0.895	0.103	2.766

Using Eq. 14 and Eq. 15, λ is rearranged as

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

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:

$$De = \lambda/t = \frac{A}{K}\gamma^{b-n-1}N \tag{18}$$

where shear rate (γ) is obtained in case of agitation of liquid in a cylindrical vessel as follows (13):

$$\gamma = 4\pi N/n \tag{19}$$

RESULTS AND DISCUSSION

Empirical Correlation of Liquid-Side Mass Transfer Coefficient of CO_2

To observe the effect of the concentration of PEO with the impeller speed and size as parameters on the volumetric mass transfer coefficient (k_La), the measured k_La in the aqueous solution of PEO concentration in the range of $10-30\,kg/m^3$ was plotted against PEO concentration in Fig. 1. As shown in Fig. 1, k_La increases with the increase of the agitation speed of the impeller and decreases with the increase of PEO concentration.

In analyzing the relationship between k_La and the experimental variables such as the PEO 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 PEO 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 PEO solution to that of water (14) 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. 13 through Eq. 19.

To correlate k_La with the experimental variables such as the PEO concentrations and the speed and size of the impeller, the dimensionless groups such

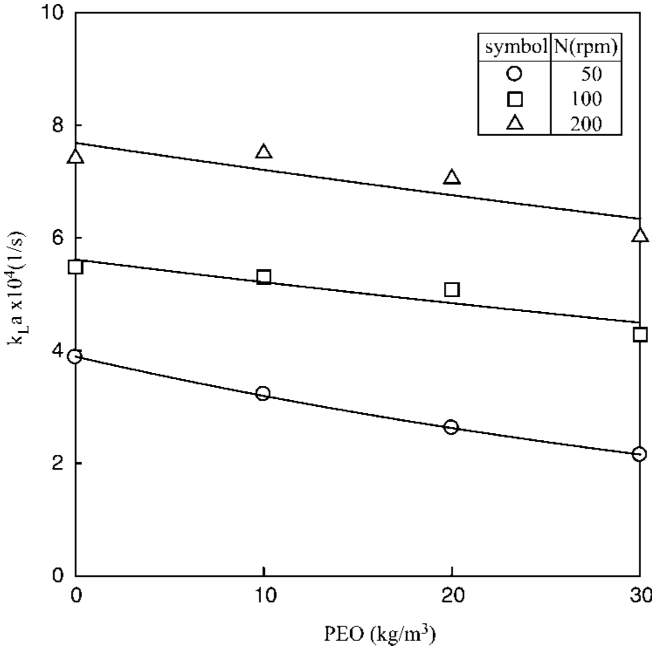

Figure 1. Effect of PEO concentration on k_La of CO_2 at $d = 0.034$ m.

as Sherwood number (Sh) and Reynolds number (Re) are used, and they are defined as follows, respectively:

$$Sh = k_Lad^2/D_A \tag{20}$$

$$Re = d^2N\rho/\mu \tag{21}$$

Figure 2 shows logarithmic plots of Sh against Re in case of water as absorbent of CO_2. As shown in Fig. 2, 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:

$$Sh = 12.56Re^{0.48} \tag{22}$$

The calculated values of Sh from Eq. 22 approached to the measured Sh very well with a standard deviation (SD) of 0.15% and a mean deviation (MD) of 3.06%.

Figure 3 shows logarithmic plots of Sh against Re for the aqueous PEO solutions, and the solid line in Fig. 3 presents the Sh calculated from Eq. 22. As shown in Fig. 3, the plots were scattered from the straight line with SD of 173.2% and MD of 119.31%. This may be due to non-Newtonian behavior of the aqueous PEO solution.

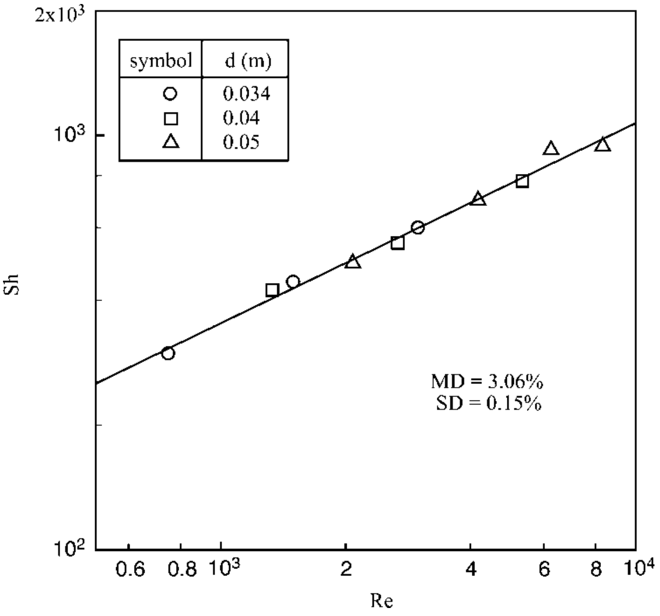

Figure 2. Sh vs. Re in water at various impeller size.

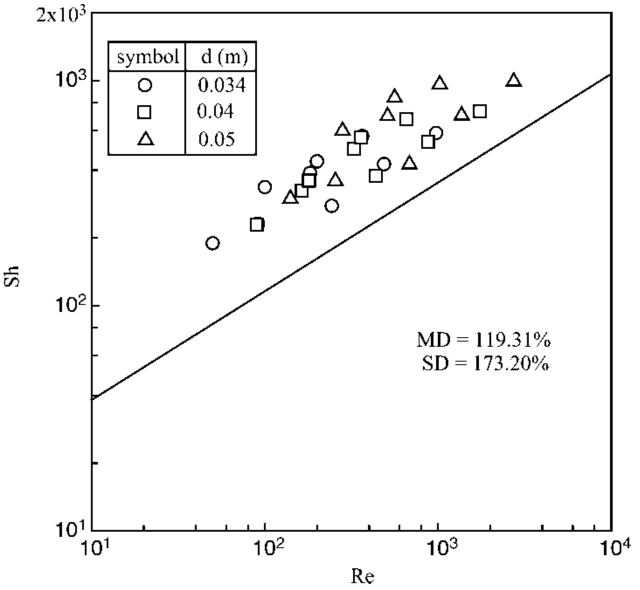

Figure 3. Dimensionless correlation of volumetric mass transfer coefficient of CO_2 in PEO aqueous solution at various impeller size.

The new term corrected with De was used to lessen the deviation of the plots for the aqueous PEO solution from those for water as shown in Fig. 2. A simple multiple regression exercise was used for the plots of Sh combined with $(1 + c_1\ De^{c_2})$ against Re, which gave the values of c_1 and c_2 are 8.33 and 1.31, respectively, with SD of 4.45% and MD of 14.41% as shown in Fig. 4.

Using a total of 36 data points, a multiple regression analysis came up with a correlation as follows:

$$k_L a d^2/D_A = 12.56\left(d^2\, N\rho/\mu\right)^{0.48}\left(1 + 8.33De^{1.31}\right) \qquad (23)$$

As shown in Eq. 23, k_L is affected by both viscosity and elasticity, and increased with increasing PEO concentration.

Effect of Rheological Properties on the Rate of Chemical Absorption

To observe the effect of rheological properties of aqueous PEO solution on the rate of chemical absorption, the absorption rate of CO_2 into aqueous PEO solution with MEA was measured according to change of MEA concentration

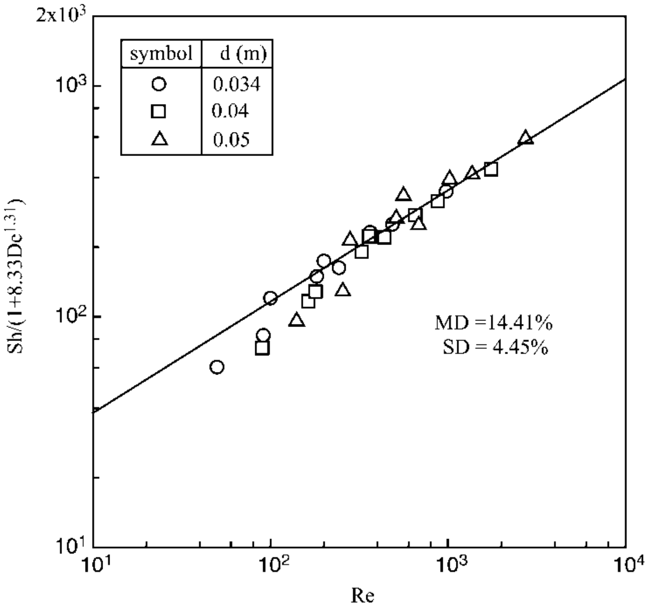

Figure 4. Dimensionless correlation of volumetric mass transfer coefficient of CO_2 in PEO aqueous solution with corrected specific viscosity and Deborah number at various impeller size.

in the range of $0 \sim 2\,\mathrm{kmol/m^3}$, and Deborah numbers in the aqueous PEO solution were calculated at impeller size with 0.034 m and agitation speed with 50 rev/min using Eq. 18. Figures 5 and 6 show the typical plots of the absorption rate of CO_2 against the concentration of MEA at PEO concentration of 10 and $30\,\mathrm{kg/m^3}$, respectively, under the experimental conditions of the agitation speed of 50 rev/min with the impeller size of 0.034 m. The triangle and circle in Figs. 5 and 6 represent the measured values of absorption rate of CO_2 in water with MEA and aqueous PEO solution with MEA, respectively. As shown in these figures, R_A increases with increasing MEA concentration. And R_A in water is larger than that in aqueous PEO solution, because k_L in water is larger than that in the solution. The solid lines in Figs. 5 and 6 are R_A estimated by Eq. 12, with k_L in water with $\mu = \mu_w$ and $De = 0$, k_L in aqueous PEO solution with $De > 0$, and k_L in aqueous PEO solution with $De = 0$ under assumption that the solution would act as a Newtonian liquid, respectively, and the estimated R_A in aqueous PEO solution with $De > 0$ is larger than that with $De = 0$. This means that the effect of the elasticity of the aqueous PEO solution on R_A is stronger than the effect of viscosity on R_A, in other words, the elasticity of PEO accelerates the rate of absorption of CO_2 due to the increase of the volumetric mass transfer coefficient as shown in Eq. 23.

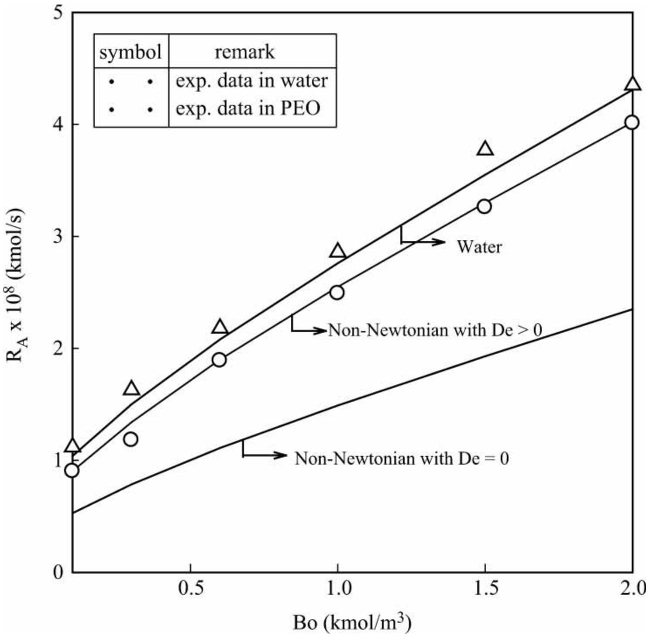

Figure 5. Effect of C_{Bo} on R_A at aqueous solution of PEO of $10\,\mathrm{kg/m^3}$. ($d = 0.034\,\mathrm{m}$, $N = 50$ rev/min).

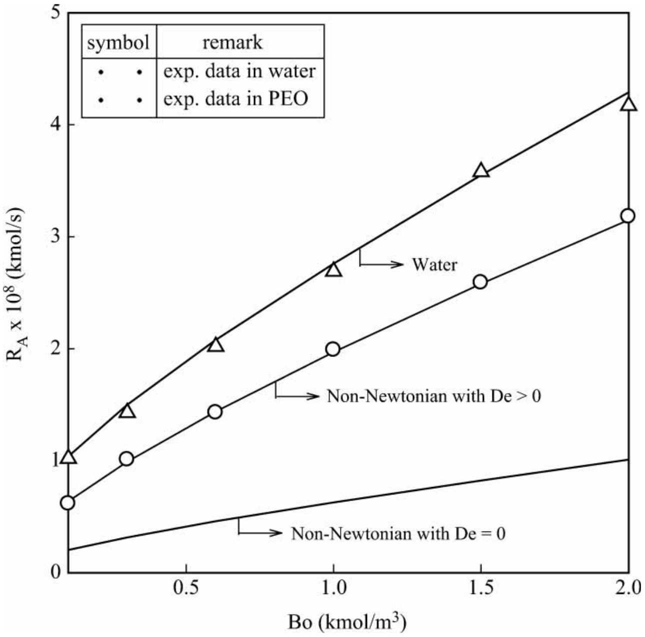

Figure 6. Effect of C_{Bo} on R_A at aqueous solution of PEO of 30 kg/m^3. (d=0.034 m, N = 50 rev/min).

CONCLUSIONS

Rates of the chemical absorption of CO_2 in the aqueous solution of PEO at $10 \sim 30$ kg/m^3 with MEA at $0 \sim 2$ kmol/m^3 were measured in a flat-stirred vessel to get the influence of the rheological properties of PEO 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 $50 \sim 200$ rev/min at 25°C and 0.101 MPa. The elastic property such as Deborah number of the aqueous PEO solution was considered to get 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. The elasticity of PEO accelerates the rate of absorption of CO_2 from the comparison of effect of the elasticity of the aqueous PEO solution on R_A with effect of viscosity using Deborah number.

NOMENCLATURE

C_A	concentration of CO_2 (kmol/m^3)
C_B	concentration of MEA (kmol/m^3)
d	diameter of impeller (m)

N	speed of impeller (1/s)
N_1	primary normal stress difference (N/m^2)
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	MEA
i	gas-liquid interface
o	bulk body

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