

## Effect of PEO viscoelasticity on carbon dioxide absorption in aqueous PEO solution of AMP

Sang-Wook Park\*, Byoung-Sik Choi and Jae-Wook Lee<sup>1</sup>

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

*<sup>1</sup>Department of Chemical Engineering, Sogang University, Seoul 121-742, Korea*

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### Abstract

Carbon dioxide was absorbed into aqueous polyethylene oxide (PEO) solution containing AMP in a flat-stirred vessel to investigate the effect of non-Newtonian rheological behavior of PEO on the rate of chemical absorption of CO<sub>2</sub>, where the reaction between CO<sub>2</sub> and AMP was assumed to be a first-order reaction with respect to the molar concentration of CO<sub>2</sub> and AMP respectively. The liquid-side mass transfer coefficient, 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<sub>2</sub> accelerate compared with Newtonian liquid based on the same viscosity of the solution.

**Keywords** : chemical absorption, carbon dioxide, PEO, AMP, non-Newtonian liquid

### 1. 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 fermentation broth, slurry, and fluidized bed, *et al.* Variation of 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.

Only use of 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. The dimensionless

number such as Deborah number ( $De$ ), which relates the elastic properties to the process parameters, is used to correlate  $k_L a$  with properties of non-Newtonian liquids. Unified correlations have been proposed for  $k_L a$  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 in the dimensionless group are different one another.

There is little information about the effect of elastic properties on the absorption of gas accompanied by chemical reaction in non-Newtonian liquid. Park *et al.* (2004a) presented the effect of elasticity of polyisobutylene (PIB) in the benzene solution of polybutene (PB) and PIB on chemical absorption of CO<sub>2</sub> 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<sub>2</sub>. 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<sub>2</sub> and reactant occurs in the aqueous phase.

In this study, the chemical absorption mechanism of CO<sub>2</sub> into an aqueous PEO solution with 2-amino-2-methyl-1-propanol (AMP) is presented to observe the effect of rheological properties on the absorption of CO<sub>2</sub>, and the measured rates of CO<sub>2</sub> absorption are compared with those obtained from the model based on the film theory with chemical reaction. The  $k_L a$  obtained from the empirical formula is used to estimate the enhancement factor due to chemical reaction.

\*Corresponding author: swpark@pusan.ac.kr  
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**Table 1.** Coefficients of dimensionless group of the gas-liquid mass transfer

Investigator	$n_1$	$n_2$	$n_3$	polymer	contactor
Yagi and Yoshida (1975)	2	0.5	-0.67	CMC, PA	agitated vessel
Ranade and Ulbrecht (1978)	100	1	-0.67	CMC	stirred tank
Nakanoh and Yoshida (1980)	0.13	0.55	-1	PAA	bubble column
Park <i>et al.</i> , 2003a	100	1	-0.42	CMC, PA	agitated vessel
Park <i>et al.</i> , 2003b	2461.3	1	-0.274	PB, PIB	agitated vessel

## 2. Theory

The problem to be considered is that a gaseous species A (CO<sub>2</sub>) dissolves into the liquid phase and then reacts irreversibly with species B (AMP) according to



The stoichiometric coefficients ( $\nu$ ) in Eq. (1) for AMP was obtained from the reference (Kennard and Meisen, 1984) 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:

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

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

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

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

Boundary and initial conditions to be imposed are

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

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

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

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

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

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

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

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

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

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

The  $\beta$  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<sub>2</sub> with chemical reaction as follows:

$$R_A = \beta R_{Ao} = \beta k_L S C_{Ai} \quad (12)$$

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

## 3. Experimental

### 3.1. Chemicals

All chemicals in this study were reagent grade, and used without further purification. Purity of both CO<sub>2</sub> and N<sub>2</sub> was more than 99.9%. The polymers used in this study were polyethylene oxide (PEO) with the mean molecular weight of 200,000 (by Aldrich chemical company, U.S.A.) and AMP (Aldrich, U.S.A.) was used as reagent grade without purification.

### 3.2. Rate of absorption

An agitated vessel used for measurement of absorption rate of CO<sub>2</sub> was constructed of glass of 0.75 m inside diameter and of 0.131 m in height with four equally spaced vertical baffles. A straight impeller with 0.034, 0.04, and 0.05 m in length and 0.017 m in width was used as the liquid phase agitator, and located at the middle position of the liquid phase. The absorption rate of CO<sub>2</sub> was measured in the aqueous solution of PEO of 10~30 kg/m<sup>3</sup>, AMP of 0~2 kmol/m<sup>3</sup>, and the impeller speed of 50 – 200 rpm at 101.3 N/m<sup>2</sup> and 25°C following the procedure reported elsewhere (Park *et al.*, 2004a).

## 4. Physicochemical and rheological properties

### 4.1. Solubility of CO<sub>2</sub> in aqueous PEO solution

The solubility ( $C_{Api}$ ) of CO<sub>2</sub> in the aqueous PEO solutions was obtained by the pressure measuring method, which measured the pressure difference of CO<sub>2</sub> between before and after equilibrium in gas and liquid phase similar to the procedure reported elsewhere (Kennard and Meison, 1984). The experimental procedure was duplicated that as reported in the published research (Park *et al.*, 2004a) in detail.

The solubility ( $C_{Ai}$ ) of CO<sub>2</sub> in aqueous AMP solution was estimated as follows (Totiwachwuthikul *et al.*, 1991):

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

### 4.2. Density and apparent viscosity of PEO solution

The density of the aqueous solution of PEO were measured at 25°C within 0.1 kg/m<sup>3</sup> 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 with Brookfield viscometer (Brookfield Eng. Lab. Inc, USA).

### 4.3. Reaction Rate Constant

The reaction rate constant ( $k_2$ ) in the reaction of CO<sub>2</sub> with AMP was estimated as follows (Messouadi and Sada, 1996).

$$\log k_2 = 10.986 - \frac{2551.2}{T} \quad (14)$$

### 4.4. Diffusivities of CO<sub>2</sub> and AMP

The diffusivity ( $D_{Aaq}$ ) of CO<sub>2</sub> in aqueous AMP solution was estimated as follows (Danckwerts and Sharma, 1966):

$$D_{Aaq} = 2.1625 \times 10^{-9} - 6.85 \times 10^{-10} C_{Bo} + 7 \times 10^{-11} C_{Bo}^2 \quad (15)$$

Diffusivity ( $D_{Baq}$ ) of AMP in aqueous AMP solution was obtained from assumption that the ratio of  $D_{Baq}$  to  $D_{Aaq}$  was equal to the ratio in water (Nijsing *et al.*, 1959). The diffusivity of CO<sub>2</sub> and AMP in water at 25°C were taken as  $1.97 \times 10^{-9}$  m<sup>2</sup>/s (Hikita *et al.*, 1976) and  $7.24 \times 10^{-10}$  m<sup>2</sup>/s

(Danckwerts and Sharma, 1966), respectively.

The diffusivity of a solute of a small size such as CO<sub>2</sub>, O<sub>2</sub> or CH<sub>4</sub> in a polymer solution depends on the viscosity of the solution and the molecular weight of the polymer. The diffusivity ( $D_A$ ) of CO<sub>2</sub> and that ( $D_B$ ) of AMP in the aqueous PEO 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.

$$D_A/D_{Aaq} = (\mu_w/\mu)^{3.7} \sqrt{M_w/M_p} \quad (16)$$

$$D_B/D_{Baq} = (\mu_w/\mu)^{3.7} \sqrt{M_w/M_p} \quad (17)$$

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

The values of solubility, diffusivity of CO<sub>2</sub>, density, and apparent viscosity of aqueous PEO solution are given in Table 2.

### 4.5. Rheological properties of aqueous PEO 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.

$$\tau = K\gamma^n \quad (18)$$

$$\mu = K\gamma^{n-1} \quad (19)$$

$$N_1 = A\gamma^b \quad (20)$$

where  $n$ ,  $K$ ,  $b$  and  $A$  are material parameters depending on temperature. These parameters were obtained from the measurement of  $\tau$  and  $N_1$  for the change of  $\gamma$  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 (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 ( $\lambda$ ) of the liquid defined as

**Table 2.** The physicochemical and rheological properties of CO<sub>2</sub> and PEO aqueous solution

C (kg/m <sup>3</sup> )	Viscosity (Ns/m <sup>2</sup> ) × 10 <sup>3</sup>	Diffusivity (m <sup>2</sup> /s) × 10 <sup>9</sup>	Solubility (kmol/m <sup>3</sup> )	Density (kg/m <sup>3</sup> )	Rheological propertie			
					N (-)	K (Ns <sup>n</sup> ·m <sup>2</sup> )	B (-)	A (Ns <sup>n</sup> ·m <sup>2</sup> )
0	1	1.95	0.039	1000.0	1.0	0.001	-	-
10	3.05	1.88	0.034	1001.4	0.53	0.02	0.10	0.13
20	8.17	1.81	0.029	1003.2	0.49	0.42	0.10	1.26
30	14.9	1.77	0.028	1004.8	0.45	0.90	0.08	2.77

$$\lambda = \frac{N_1}{\mu\gamma^2} \quad (21)$$

Using Eqs. (19) and Eq. (20),  $\lambda$  is rearranged as

$$\lambda = \frac{A}{K}\gamma^{b-n-1} \quad (22)$$

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:

$$De = \lambda/t = \frac{A}{K}\gamma^{b-n-1}N \quad (23)$$

where shear rate ( $\gamma$ ) is obtained in case of agitation of liquid in a cylindrical vessel as follows (Metzner and Otter, 1978):

$$\gamma = 4\pi N/n \quad (24)$$

## 5. Results and discussion

### 5.1. Empirical correlation of liquid-side mass transfer coefficient of CO<sub>2</sub>

To observe the effect of the concentration of PEO on the volumetric mass transfer coefficient ( $k_La$ ), the  $k_La$  measured in the aqueous PEO solution in the range of 10~30 kg/m<sup>3</sup> with the impeller speed as a parameter at  $d = 0.034$  m were typically plotted against PEO concentration in Fig. 1. As shown in Fig. 1,  $k_La$  increased with increasing the impeller speed and decreased with increasing PEO con-

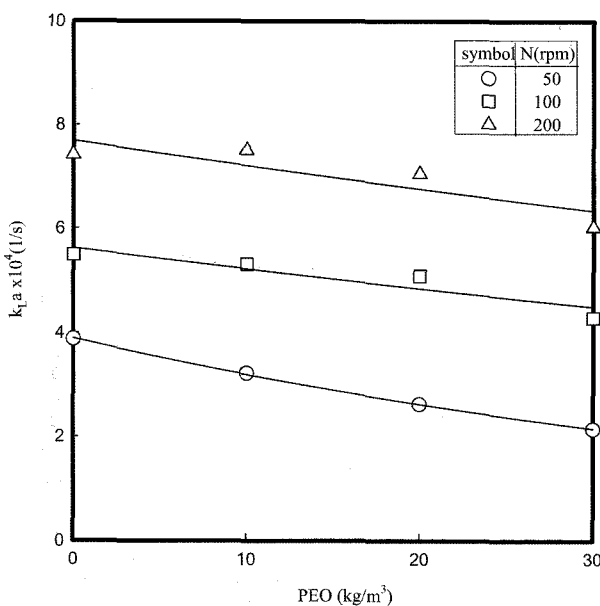


Fig. 1. Effect of PEO concentration on  $k_La$  of CO<sub>2</sub> at  $d = 0.034$  m.

centration.

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 aqueous PEO solution.

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<sub>2</sub> 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 aqueous PEO solution to that of water was used to correlate with  $k_La$  (Sandall and Patel, 1970), because the viscosity in the agitated vessel depends on the speed of impeller and the rheological properties as shown in Eqs. (19)-(24).

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

$$Sh = k_Lad^2/D_A \quad (25)$$

$$Re = d^2N\rho/\mu \quad (26)$$

Fig. 2 shows logarithmic plots of  $Sh$  against  $Re$  in the absorption of CO<sub>2</sub> in water.

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

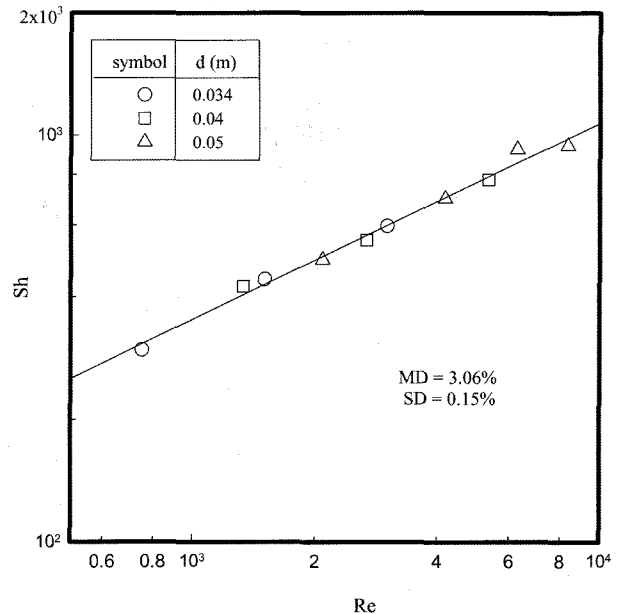


Fig. 2.  $Sh$  vs.  $Re$  at various impeller size in water.

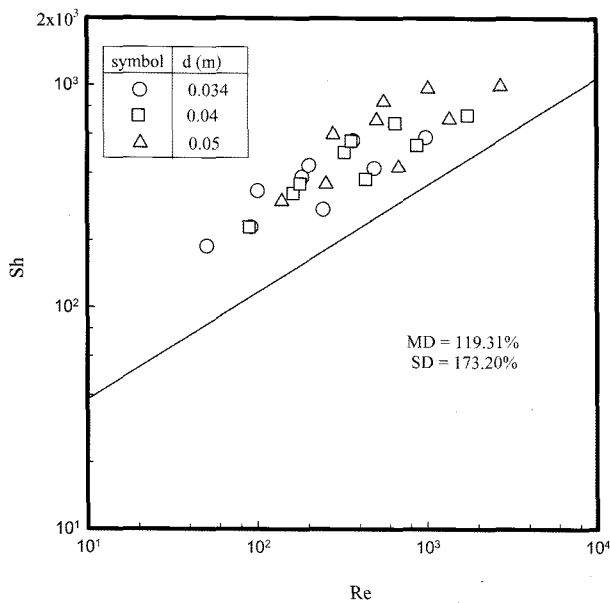


Fig. 3.  $Sh$  vs.  $Re$  at various impeller size PEO solution.

empirical equation between  $Sh$  and  $Re$  as follows:

$$Sh = 12.56 Re^{0.48} \quad (27)$$

The calculated values of  $Sh$  from Eq. (27) approached to the measured  $Sh$  very well with a standard deviation (SD) of 3.06% and a mean deviation (MD) of 0.15%.

Fig. 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. (27). As shown in Fig. 3, the plots were scattered from the straight line with SD of 119.31% and MD of 173.2%. This may be due to non-Newtonian behavior of the aqueous PEO solution.

The new term corrected with  $De$  was used to lessen the deviation of the plots for the aqueous PEO 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  $(1 + c_1 De^{c_2})$  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:

$$k_L a d^2 / D_A = 12.56 (d^2 N \rho / \mu)^{0.48} (1 + 8.33 De^{1.31}) \quad (28)$$

From the work of Yagi and Yoshida (1975), Ranade and Ulbrecht (1978), and 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. (28) for the overall effect of viscosity on the  $k_L a$ , it comes out as  $k_L a \propto \mu^{-0.48}$ , which compares well with the findings of Yagi and Yoshida of  $\mu^{-0.4}$  and Ranade and Ulbrecht of  $\mu^{-0.41}$ , and Moo-Young and Kawase of  $\mu^{-0.38}$ . As a result in Fig. 4, the deviation of plots

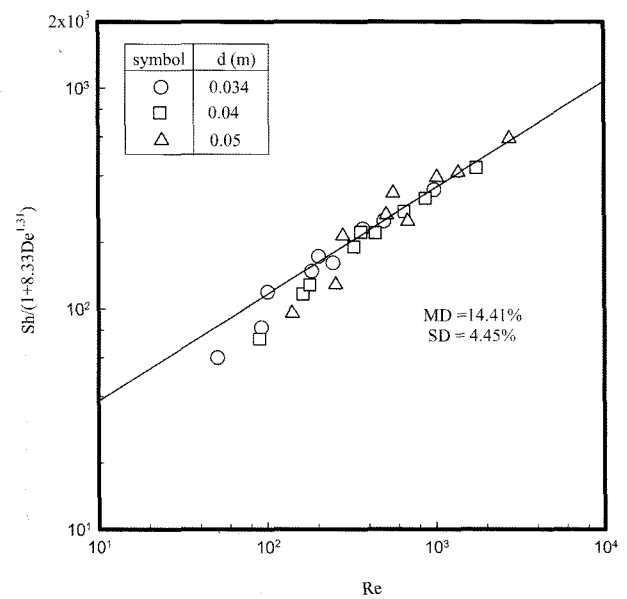


Fig. 4.  $Sh(1 + 8.33De^{1.31})$  vs.  $Re$  for change of PEO concentration at various impeller size in PEO solution.

of  $Sh$  against  $Re$  in aqueous PEO solution can be eliminated by incorporating  $De$  into the correlation.

## 5.2. 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 rates of  $CO_2$  into aqueous PEO solution with AMP were measured according to change of AMP concentration in the range of 0 – 2 kmol/m<sup>3</sup>. Fig. 5 and 6 show the typical plots of the absorption rate of  $CO_2$  against the concentration of AMP at PEO concentration of 10 and 30 kg/m<sup>3</sup>, 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 Fig. 5 and 6 represent the measured values of absorption rate of  $CO_2$  in water with AMP and aqueous PEO solution with AMP, respectively.

As shown in these figures, the measured  $R_A$  increases with increasing AMP concentration, and the  $R_A$  in water is larger than that in aqueous PEO solution. The estimated values of  $R_A$  are calculated using Eq. (12) with various  $k_L$  obtained by Eq. (28) according to the kinds of the solutions, i.e., water, the aqueous PEO solution with elasticity, and without elasticity, and presented as symbols of the dotted, solid and short dash lines in Fig. 5 and 6, respectively. In water,  $\mu$  is  $\mu_w$  and  $De$  is 0, in the aqueous PEO solution with elasticity,  $\mu$  is that in aqueous PEO solution and  $De$  has some value  $> 0$ . The aqueous PEO solution without elasticity is an imaginary solution, which is assumed to be a Newtonian liquid with the same viscosity as that of the aqueous PEO solution with elasticity. As shown in Fig. 5

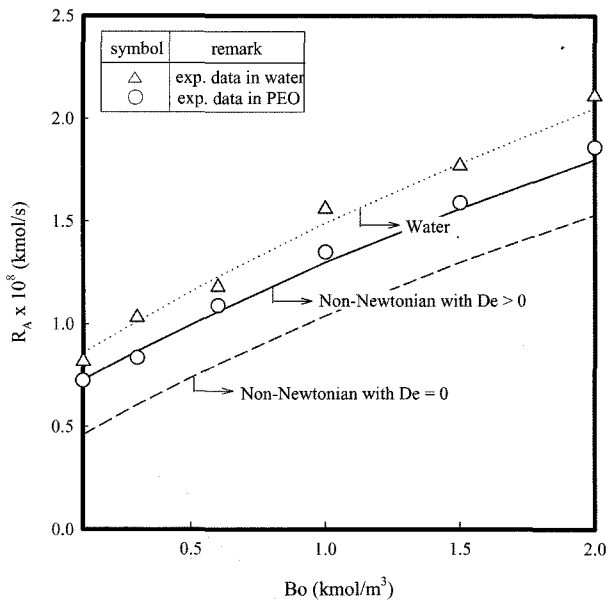


Fig. 5. Comparison of elasticity with viscosity of PEO in aqueous AMP solution (PEO = 10 kg/m<sup>3</sup>,  $d = 30$  mm,  $N = 50$  rpm).

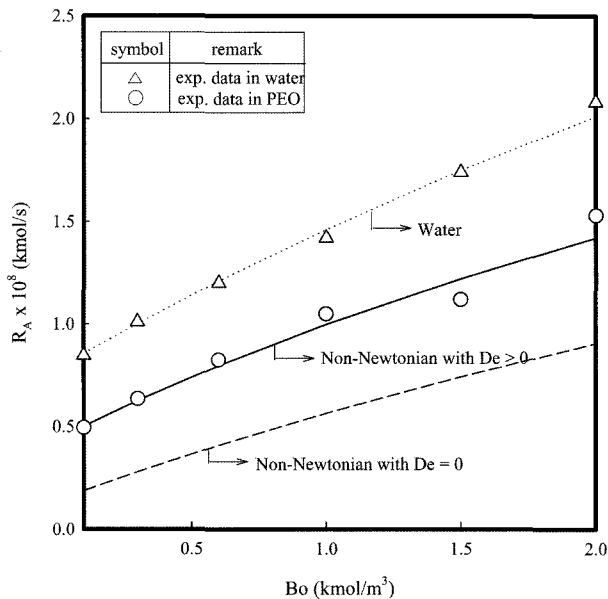


Fig. 6. Comparison of elasticity with viscosity of PEO in aqueous AMP solution (PEO = 30 kg/m<sup>3</sup>,  $d = 30$  mm,  $N = 50$  rpm).

and 6, the  $R_A$  increases in the dotted, solid and short dash line order. In general, the mass transfer coefficient in the solution is inversely proportional to the viscosity. The viscosity of water is larger than that of the aqueous PEO solution as listed in Table 2, and then,  $R_A$  in water is larger than that in the aqueous PEO solution. If the aqueous PEO solution in this study would have only viscous behavior, the  $R_A$  should be the  $R_A$  represented as the short dash line. But, the  $R_A$  in this study represented as the solid line is larger than

that represented as the short dash line. This means that the effect of the elasticity of the aqueous PEO solution on increase of  $R_A$  is stronger than the effect of viscosity on  $R_A$ , in other words, the elasticity of PEO accelerates  $R_A$  due to the increase of  $k_L a$  as shown in Eq. (28) based on the same viscosity of the solution. The behavior that the elasticity of PEO accelerates  $R_A$  was mentioned in other polymer such as polyisobutylene (Park *et al.*, 2004b). The increase of  $R_A$  in the aqueous PEO solution might be explained by the fluctuation of the liquid surface occurred by the elasticity of PEO, which makes  $k_L$  increase.

## 6. Conclusions

Rates of the chemical absorption of CO<sub>2</sub> in the aqueous solution of PEO at 10–30 kg/m<sup>3</sup> with AMP at 0–2 kmol/m<sup>3</sup> 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 50–200 rev/min at 25°C and 101.3 kPa. 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<sub>2</sub> by the comparison of effect of the elasticity of the aqueous PEO solution on  $R_A$  with effect of viscosity.

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## Nomenclature

$C_A$	: concentration of CO <sub>2</sub> (kmol/m <sup>3</sup> )
$C_B$	: concentration of AMP (kmol/m <sup>3</sup> )
$d$	: diameter of impeller (m)
$N$	: speed of impeller (1/s)
$N_1$	: primary normal stress difference (N/m <sup>2</sup> )
$t$	: Time (s)
$T$	: Temperature (K)
$z$	: coordinate in film thickness direction in benzene phase (m)

## Greek letters

$\gamma$	: shear rate (1/s)
$\mu$	: viscosity of liquid (Ns/m <sup>2</sup> )
$\mu_w$	: viscosity of water (Ns/m <sup>2</sup> )
$\rho$	: density of liquid (kg/m <sup>3</sup> )
$\tau$	: shear stress (N/m <sup>2</sup> )

## Subscripts

- A* : CO<sub>2</sub>  
*B* : AMP  
*i* : gas-liquid interface  
*o* : bulk body

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