

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

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Abstract

Absorption rate of carbon dioxide was measured in the aqueous xanthan gum (XG) solution in the range of 0-0.15 wt% containing 2-amino-2-methyl-1-propanol (AMP) of 0-2 kmol/m³ in a flat-stirred vessel with an impeller of 0.05 m and agitation speed of 50 rpm at 25°C and 0.101 MPa. The volumetric liquid-side mass transfer coefficient ($k_{L,a}$) of CO₂, which was correlated with the viscosity and the elastic behavior of XG solution containing Deborah number as an empirical formula, was used to estimate the chemical absorption rate of CO₂ (R_A). R_A , which was estimated by mass transfer mechanism based on the film theory using the physicochemical properties and the kinetics of reaction between CO₂ and AMP, was compared with the measured rate. The aqueous XG solution with elastic property of non-Newtonian liquid made R_A increased compared with Newtonian liquid based on the same viscosity of the solution.

Keywords : absorption, 2-amino-2-methyl-1-propanol, carbon dioxide, xanthan gum, viscoelastic 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* (Yagi and Yoshida, 1975; Ranade and Ulbrecht, 1978; Nakanoh and Yoshida, 1980). 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.

The apparent viscosity of non-Newtonian fluids is not sufficient to obtain a unified correlation for $k_{L,a}$ values in the case of gas absorption into non-Newtonian fluids. Because of the complexities of gas absorption in non-Newtonian media, the correlations obtained by these studies have been 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 an

viscoelastic solution, such as PAA, deviate from those for the inelastic solution, such as CMC, should correlate with some measure of the elasticity of solution. A 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 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.

There is some information about the effect that elastic properties have on the absorption of gas accompanied by a chemical reaction in non-Newtonian liquid. As shown in Table 1, Park and coworkers (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 CO₂. Park and coworkers (2005, 2006, 2007) also measured the absorption rate of CO₂ in aqueous PEO solution with AMP, nanometer sized silica colloid solution with AMP, and PAA solution with monoethanolamine, respectively. They showed that the viscoelastic materials, such as PIB, PAA, PEO, and nanometer sized silica influenced the absorption rate of CO₂ using $k_{L,a}$. The $k_{L,a}$ increased in PEO and decreased in w/o emulsion, PAA, and silica compared

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

Investigator	n_1	n_2	n_3	Substance	Contactor
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, PAA	Bubble column
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> (2005)	8.33	1.31	1	PEO	Agitated vessel
Park <i>et al.</i> (2006)	39.4	1	-0.43	silica	Agitated vessel
Park <i>et al.</i> (2007)	54.7	1	-0.45	PAA	Agitated vessel

with Newtonian liquid based on the same viscosity of the solution in their works.

Xanthan gum (XG) is an extracellular heteropolysaccharide produced by microorganisms of the genus *Xanthomonas*. Since its first commercial production in 1964, XG has been used in numerous applications in the food and chemical industries as a thickening agent, stabilizer and emulsifier (Sandford and Baird, 1983). Even solutions having a low concentration of XG show high viscosity and insensitivity to changes in temperature, PH and sanitary. XG production usually has been performed in batch fermentation using the bacterium *Xanthomonas campestris*. The conditions and performance of such fermentations in stirred tank reactors (Herbst *et al.*, 1992), bubble columns (Pons *et al.*, 1989; Suh *et al.*, 1992) and airlifts (Kessler *et al.*, 1993) have been studied by some researchers. XG fermentation is probably the most complex fermentation process in terms of rheological property variation and associated mixing problems. The changes in viscosity during culture exceed 4 orders of magnitude, which is greater even than that found in high-viscosity polymerization processes in the chemical industry. The change in rheological properties of broths during the course of a batch fermentation leads also to increasing pseudoplasticity, viscoelasticity, and yield stress values. This last characteristic, the yield stress, makes it especially difficult to achieve good mixing because, beyond certain distance from the impeller, the fluid is stagnant. In these stagnant regions where only diffusional mass and heat transfer can occur, the productivity is practically reduced to zero, and mass transfer in stirred tank reactors for XG was related to the geometry of the tank and viscosity of liquid based on the power-law liquid (Galindo and Nienow, 1992). As was reviewed in Song *et al.* (2006), there exists a relatively large amount of literature published on the rheological properties of both diluted and semi-dilute (or moderately concentrated) XG solutions prepared from aqueous media. However, only a little attention has been given to the effect of rheological properties of XG on mass transfer of a solute in XG media (Vashitz *et al.*, 1988, 1989; Garcia-Ochoa and Gomez, 1998). Terasaka and Shibata (2003) presented that $k_L a$ of oxygen in XG fermentation of a bubble column was a

function of gas hold up, yield stress and diffusivity of oxygen.

To investigate the effect of the rheological behavior of non-Newtonian liquid on the gas absorption as one of series works, XG was used as a viscoelastic material in this study. The absorption rates of CO₂ were measured in aqueous XG solution containing 2-amino-2-methyl-1-propanol (AMP) and they were compared with those estimated by the mass transfer with chemical reaction based on the film theory. Deborah number, which was obtained from a power-law model of the relationship between the measured shear stress and primary normal stress difference against the shear rate of XG solution, was used to obtain a unified correlation between the values of $k_L a$ in water and that in XG solutions.

2. Theory

The overall reaction between CO₂ (A) and AMP (B) in aqueous solution is



The stoichiometric coefficients (ν) in Eq. (1) for AMP was obtained from the reference (Messaoudi and Sada, 1996) 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 (Messaoudi and Sada, 1996) as follows:

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

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

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

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

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

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

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

$$\frac{d^2 a}{dx^2} = Mab \quad (7)$$

$$\frac{d^2 a}{dx^2} = \nu r q Mab \quad (8)$$

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

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

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

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

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

The value of β is used to predict the absorption rate (R_A) of CO_2 with chemical reaction 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, which is obtained by multiplying the molar flux by the specific contact area between gas and liquid (a) and the liquid volume (V_L).

3. Experimental

3.1. Chemicals

The xanthan gum sample used in this study is a commercially available product supplied from the Sigma-Aldrich Corporation (St. Louis, MO, USA). An accurate determination of the molecular weight of XG is extremely difficult for several reasons including its relatively high value, the stiffness of a molecule and the presence of aggregates (Song *et al.*, 2006). The weight average molecular weight of our XG is approximately estimated to be 2×10^6 kg/kmol (Song *et al.*, 2006). XG is soluble in both cold and hot water. Like most other hydrocolloids, XG needs an intensive agitation upon introduction into an aqueous medium in order to avoid a formation of lumps. In this work, XG solutions with concentrations of 0.05, 0.1, and 0.15 wt% were prepared by slowly adding the required amount of XG powder weighed using an electric balance into a known volume of gently agitated medium filled in a glass container, which was maintained at room temperature with constant stirring using a magnetic bar for 24 hr. The preparation procedure was identical to that described else-

where (Song *et al.*, 2006).

3.2. Absorption rate of CO_2

The agitated absorber used for measurement of the absorption rate of CO_2 was constructed of glass (0.102 m inside diameter; 0.151 m in height) with four equally spaced vertical baffles. Straight impeller with 0.05 m in length, 0.017 m in width, and 0.005 m in thickness was used as the liquid phase agitator, and was located at the middle position of the liquid phase of 3×10^{-4} m³. The contact area between the gas and the liquid was measured as 4.32×10^{-3} m². The absorption rates of CO_2 were measured along the procedure similar to those reported elsewhere (Yu *et al.*, 1985) using a semi-batch absorber in the aqueous XG solution of 0-0.015 wt% and AMP of 0~2 kmol/m³ with the impeller speed of 50 rpm at 25°C and 0.101 MPa.

4. Physicochemical and rheological properties

The physicochemical and rheological properties in the CO_2 -XG-AMP system, which are used to calculate the value of b , are obtained as follows:

4.1. Physicochemical properties of carbon dioxide and AMP in aqueous XG solution

The physicochemical properties of carbon dioxide and AMP in aqueous XG solution, such as solubility of CO_2 , density, apparent viscosity of aqueous XG solution, diffusivity of CO_2 and AMP, and overall reaction rate constant (k_2) in the reaction of CO_2 with AMP, were obtained in the paper reported elsewhere (Park *et al.*, 2006).

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

4.2. Volumetric mass transfer coefficient of CO_2 ($k_L a$) in the aqueous XG solution

The $k_L a$, which presents the relationship (correlation coefficient=0.996) between $k_L a$ and rheological behavior of the aqueous XG solution under the conditions of the agitation speed of 50-400 rpm with the impeller size of 0.034, 0.05, and 0.075 m, was obtained with the identical procedure (Park *et al.*, 2007) as follows:

$$k_L a = k_L a \left(\frac{\mu_w}{\mu} \right)^{0.33} (1 + 0.541 De^{0.828}) \quad (13)$$

where De is defined as the ratio of the characteristic material time(λ) to the characteristic process time(t) as follows:

$$De = \lambda / t = \frac{A \cdot b \cdot n - 1}{K} N \quad (14)$$

where K and n are the index of $\tau = K \dot{\gamma}^n$, A and b , the index of $N_1 = A \dot{\gamma}^b$. These parameters were obtained from the measurement of τ and N_1 for the changes of $\dot{\gamma}$ by the par-

Table 2. Physicochemical and rheological properties of CO₂ and aqueous XG solution

XG (wt%)	Viscosity (Ns/m ²)	Diffusivity (m ² /s)×10 ⁹	Solubility (kmol/m ³)	Density (kg/m ³)	Rheological properties			
					n	K (Ns ⁿ /m ²)	b	A (Ns ⁿ /m ²)
0	0.0009	1.95	0.035	1000.0	1.0	0.001	-	-
0.05	0.0331	1.92	0.032	1468.0	0.622	0.0372	0.112	2.81
0.10	0.0777	1.91	0.030	1468.3	0.520	0.1050	0.138	10.20
0.15	0.1571	1.89	0.023	1468.6	0.458	0.2094	0.181	23.13

allel disk type rheometer (ARES, Rheometrics, U.S.A.) with the diameter of 0.05 m and the gap of 0.001 m along the identical procedure in previous paper (Park *et al.*, 2007), and are given in Table 2.

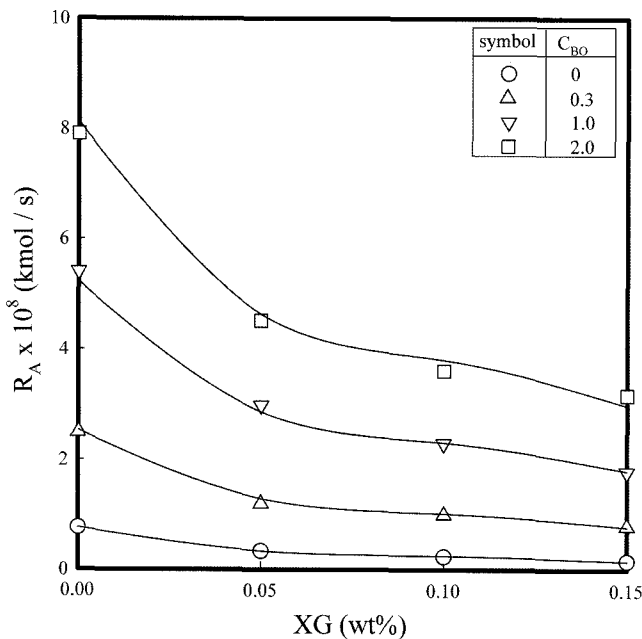
The shear rate is obtained in case of agitation of liquid in a cylindrical vessel as follows (Metzner and Otter, 1957):

$$\dot{\gamma} = 4\pi N/n \quad (15)$$

5. Results and discussion

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

To investigate the effect of elasticity of XG solution on the chemical absorption rate of CO₂, the absorption rate (R_A) of CO₂ into aqueous XG solution containing AMP was measured against XG concentration in the range 0-0.15 wt% at AMP concentration in the range of 0~2 kmol/m³ at the agitation speed of 50 rev/min and the impeller size of 0.05 m. Fig. 1 shows the plots of the absorption rate of CO₂ against XG concentration for various AMP concentrations.

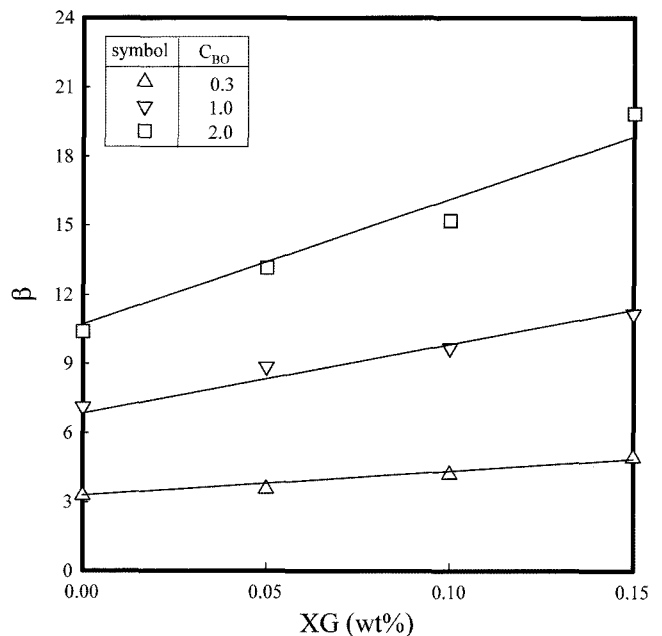
**Fig. 1.** Effect of XG concentration on R_A for various AMP concentrations.

As shown in Fig. 1, R_A increases with increasing AMP concentration and decreases with increasing XG concentration. Increase of R_A with increasing AMP concentration is due to the reactant of AMP in reaction of CO₂. Decrease of R_A with increasing XG concentration is explained from the dependences of $k_L a$, β , and C_{Ai} on XG concentration as follows:

Because $k_L a$ decreases with increasing XG concentration as shown in Eq. (13), R_A decreases with increasing XG concentration.

The values of C_{Ai} in the aqueous XG solution decrease with increasing XG concentration, but, they do not vary significantly from that in water, as shown in Table 2.

The calculated value of β was obtained from Eq. (11) and solutions of Eqs. (7) and (8) with the known parameters, such as k_L , D_A , D_B , k_2 , and C_{Ai} at given concentrations of XG and AMP. Eqs. (7) and (8) were solved by the finite element method using the FEMLAB of a packed program. The measured values of β were obtained as the ratio of R_A to R_{A0} . The measured and calculated values of β were plotted against the XG concentration in Fig. 2 as marks and

**Fig. 2.** Effect of XG concentration on enhancement factor for various AMP concentrations.

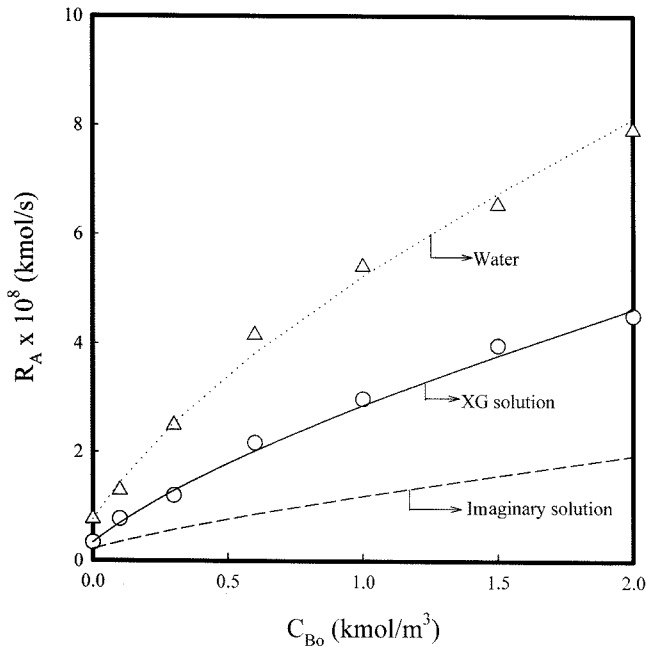


Fig. 3. Effect of C_{B0} on R_A at aqueous XG solution of 0.05 wt%.

solid lines, respectively.

As shown in Fig. 2, β increases with increasing concentrations of AMP and XG. This result coincides with the fact (Danckwerts, 1970) that β increases with increasing M , which is defined as a dimensionless parameter of $D_A k_2 C_{B0} / k_L^2$ in Eqs. (7) and (8). R_A should increase with increasing XG concentration due to the result of Fig. 2, but, R_A decreases actually with increasing XG concentration as shown in Fig. 1. Therefore, decrease of R_A was affected mainly by decrease of $k_L a$ rather than β and C_{A0} , *i.e.*, reduction of $k_L a$ by the elastic properties of the aqueous XG solution causes decrease of chemical absorption rate of CO_2 .

The values of R_A in water, the imaginary and actual solution of aqueous XG solution, which are obtained from Eq. (12) using $k_L a$ in Eq. (13), are plotted as a dotted, solid, and dashed line against AMP concentration at a typical XG concentration of 0.05 wt% in Fig. 3. The triangle and circle are the measured values in water and XG solution.

As shown in Fig. 3, R_A increases in water, XG solution, and imaginary solution order. Same trends were obtained in the other two XG concentrations of 0.1 and 0.15 wt%. If the aqueous XG solution in this study would have only viscous behavior, R_A should be R_A represented as the dashed line. But, the actual R_A of the solid line is larger than that of the dashed line. The increase of R_A from the dashed line to the solid line might be attributed to the elasticity of the aqueous XG solution.

6. Conclusions

The physical and chemical absorption rates of carbon

dioxide were measured into the aqueous XG solution of 0-0.15 wt% with AMP of 0-2 kmol/m³ in a flat-stirred vessel with 0.05 m size and 50 rpm of agitation speeds of impeller at 25°C and 0.101 MPa.

The value of $k_L a$ was correlated with an empirical formula having the rheological behavior of XG solution such as Deborah number as follows:

$$k_L a = k_{LW} a \left(\frac{\mu_w}{\mu} \right)^{0.33} (1 + 0.541 De^{0.828})$$

The chemical absorption rate was estimated from the mass balance accompanied by chemical reaction based on the film theory using the value of $k_L a$. Reduction of $k_L a$ and the absorption rate according to increase of XG concentration was explained by the elastic properties of the aqueous XG solution.

Nomenclature

- a : specific gas-liquid area (m²/m³)
- C_i : concentration of species, i (kmol/m³)
- d : diameter of impeller (m)
- D_i : diffusivity of species, i (m²/s)
- k_2 : reaction rate constant in reaction (1) (m³/kmol·s)
- k_L : liquid-side mass transfer coefficient of CO_2 in absorbent (m/s)
- N_1 : primary normal stress difference (kg/m·s²)
- V_L : volume of the liquid phase (m³)
- r_A : reaction rate in Equation (2) (kmol/m³·s)
- R_A : chemical absorption rate of CO_2 (kmol/s)
- R_{A0} : physical absorption rate of CO_2 (kmol/s)
- T : temperature (K)
- XG : xanthan gum
- z : diffusion coordinate of CO_2 (m)
- z_L : film thickness (m)

Greek letters

- β : enhancement factor of absorption rate by the chemical reaction
- γ : shear rate (1/s)
- μ : viscosity of liquid (N·s/m²)
- μ_w : viscosity of water (N·s/m²)
- ρ : density of liquid (kg/m³)
- τ : shear stress (N/m²)

Subscripts

- A : CO_2
- B : AMP
- i : gas-liquid interface

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