PAA 수용액에서 이산화탄소의 물질전달

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Mass Transfer of Carbon Dioxide in Aqueous PAA Solution

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INTRODUCTION

Gas-liquid mass transfer in non-Newtonian liquid is an important example of gas absorption in pseudoplastic flow relevant to industrial process such as a fermentation broth, slurry, and fluidized bed, et al. Variation of the volumetric liquid-phase mass transfer coefficient(k_L) in gas-dispersed systems consists of the variation of the mass transfer coefficient(k_L) and that of the specific gas-liquid interfacial area(a). The former could be correlated with the Reynolds and Schmidt numbers, which include 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.

Mere use of the apparent viscosity of non-Newtonian fluids was not sufficient to obtain a unified correlation[1-5] 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 carboxymethylcellulose(CMC), polyacrylate(PA), polyethylene oxide(PEO), polyacrylamide(PAA), and polyisobutylene (PIB) solutions. If some substance is to be given to the suggestion that the 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. One of the dimensionless numbers, which relate the elastic properties with the process parameters, is the Deborah number defined as ratio of the material's characteristic relaxation time to the characteristic flow time.

In this study, carbon dioxide was absorbed into aqueous polyacrylamide(PAA) solution in a flat-stirred vessel to get the volumetric mass transfer coefficient(k_La) of CO₂, and the effect of viscosity, viscoelasticity, and speed and size of impeller on the gas-liquid mass transfer using rheological property such as Deborah number have been investigated.

EXPERIMENTAL

Absorption experiments were carried out in an agitated vessel constructed of glass of 0.102 m inside diameter and of 0.157 m in height. Four equally spaced vertical baffles, each one-tenth of 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 CO_2 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 rev/min. The absorption rate of CO_2 was measured in

the aqueous solution of PAA of $0\sim100 \text{ kg/m}^3$ and AMP of $0\sim2 \text{ kmol/m}^3$ 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.

RESULTS AND DISCUSSIONS

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 PAA solutions.

$$\mathbf{t} = \mathbf{K} \boldsymbol{\gamma}^n \tag{1}$$

$$\mu = K\gamma^{n-1} \tag{2}$$

$$\mathbf{N}_{1} = \mathbf{A}\boldsymbol{\gamma}^{\mathrm{D}} \tag{3}$$

where τ , γ , μ , and N₁ are shear stress, shear rate, viscosity, and primary normal stress difference, respectively, and n, K, b, and A are material parameters.

These parameters were obtained from the dependence of τ and N₁ on γ .

In order to observe the dependence of τ and N₁ on γ , τ and N₁ of the aqueous PAA solution were measured according to the change of γ by a rheometer.

Figure 1 shows the typical logarithmic plot of shear stress vs shear rate for the aqueous PAA solutions in the range of $0 \sim 100 \text{ kg/m}^3$. 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.

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(λ) to the



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



characteristic flow time(t). From the definition of λ as $N_1/\mu\gamma^2$ and relation of t with the reciprocal of the impeller speed in case of stirred tanks, De is derived as follows:

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$$De = \lambda / t = \frac{A}{K} \gamma^{b-n-1} N$$
(4)

where γ is obtained in case of agitation of liquid in a cylindrical vessel as $4\pi N/n[6]$.

In analyzing the relationship between k_La 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.

In order 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) defined as $k_L a d^2 / D_A$ and Reynolds number(Re) as $d^2 N \rho / \mu$ are used.

Figure 3 shows logarithmic plots of Sh against Re in case of water as absorbent of CO_2 . 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:

$$Sh = 8.2 \times 10^{-3} \text{ Re}^{1.17}$$
(5)

The calculated values of Sh from Eq. (5) approached to the measured Sh very well with a standard deviation(SD) of 0.81% and a mean deviation(MD) of 6.37%.

Figure 4 shows logarithmic plots of Sh against Re for the aqueous PAA solutions, and the solid line in Fig. 4presents the Sh calculated from Eq. (5). As shown in Fig. 4, 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 were used to lessen the deviation of the plots for the PAA solution from those for water as shown in Fig. 4. A simple multiple regression exercise was used for the plots of Sh combined with $(\mu/\mu_w)^{c_1}(1+c_2De)^{c_3}$ 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. 5.





Figure 3. Sh vs. Re in water at various impeller size.

Figure 4. Dimensionless correlation of volumetric mass transfer coefficient of CO₂ in PAA aqueous solution at various impeller size.

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

$$k_{\rm L}ad^2/D_{\rm A} = 8.2 \times 10^{-3} (d^2 N\rho/\mu)^{1.17} (\mu/\mu_{\rm w})^{0.68} (1+54.7 \text{De})^{-0.45}$$
 (6)

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Figure 5. Dimensionless correlation of volumetric mass transfer coefficient of CO₂ in PAA aqueous solution with corrected specific viscosity and Deborah number at various impeller size.

CONCLUSIONS

Rates of the chemical absorption of CO_2 in the aqueous solution of PAA at 0.1~100 kg/m³ were measured in a flat-stirred vessel to get 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 rev/min at 25 °C and 0.101 MPa. The elastic property such as Deborah number of the aqueous PAA solution was considered to get an empirical correlation of the volumetric mass transfer coefficient in the non-Newtonian liquid.

ACKNOWLEDGEMENTS

This work was supported by research grant from the Korea Science and Engineering Foundation(KOSEF) through the Applied Rheology Center(ARC), Brain Korea 21 Project in 2003, and Brain Busan 21 Program.

REFERENCES

- 1. Yagi, H.; Yoshida, F. Gas absorption by Newtonian and non-Newtonian fluids in sparged agitated vessel. Ind. Eng. Chem. Process Des. Dev., **14** (4), 488-493 (1975)
- 2. Ranade, V. R.; Ulbrecht, J.J. Influence of polymer additives on the gas-liquid mass transfer in stirred tanks. AIChE J., **24** (5), 796-803 (1978).
- 3. Nakanoh, M.; Yoshida, F. Gas absorption by Newtonian and non-Newtonian liquids in a bubble column. Ind. Eng. Chem. Process Des. Dev., **19** (1), 190-195 (1980).
- Park, S. W.; Sohn, I. J.; Park, D. W.; Oh, K. J. Absorption of carbon dioxide into non-Newtonian liquid. I. Effect of Viscoelasticity. Sep. Sci. Technol., 38(6), 1361-1384 (2003).
- 5. Park, S. W.; Sohn, I. J.; Sohn, S. G.; Kumazawa, H. Absorption of carbon dioxide into non-Newtonian liquid. II. Effect of w/o Emulsion. Sep. Sci. Technol., **38**(6), 3983-4007 (2003).
- 6. Metzner, A. B.; Otter, R. E. Agitation of non-Newtomian fluids. AIChE J., 3, 3-10 (1957).