Interfacial Turbulence Accompanying Chemical Absorption into Falling Liquid Film

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The interfacial turbulence induced by the Marangoni instability may easily be established in the entrance region of any absorber, because the surface of the liquid absorbent is not contaminated shortly after the surface is formed. Existing experimental data on the rate of absorption of carbon dioxide into aqueous monoethanolamine solutions in laminar liquid-jet and wetted-wall column absorbers were increased substantially over those predicted by the conventional penetration-theory. Brian et al. (1967) first paid attention to this contradiction and performed experiments on the desorption of inert gas (propylene) from aqueous monoethanolamine solutions saturated with propylene into carbon dioxide and nitrogen streams in a short wetted-wall column in order to detect the presence of the interfacial turbulence. The substantial increase in the propylene desorption coefficient into carbon dioxide stream indicated the presence of the interfacial turbulence accompanying absorption of carbon dioxide into aqueous monoethanolamine solutions.

In our previous work (Sada et al., 1977a, 1977b), we also detected the interfacial turbulence accompanying carbon dioxide absorption into aqueous solutions of various amines (monoethanolamine, ethylenediamine and monoisopropanolamine) using a liquid-jet column. The variation of the interfacial turbulence with gas-liquid contact time and amine concentration was measured by desorbing the inert tracer gas (ethylene) simultaneously with the absorption of carbon dioxide into aqueous amine solutions. In a series of work (Brian et al., 1967; Sada et al., 1977a, 1977b), however, the bulk liquid phase was maintained at a laminar flow condition. Thus, no mention has been made of the correlation of the liquid flow condition to the Marangoni instability. As the liquid flow rate increases, a minute turbulence such as rippling is induced on the liquid surface. The minute turbulence may superimpose on the interfacial turbulence induced by the Marangoni instability. Little work has been done on superposition of these two kinds of turbulence.

It is the purpose of this note to investigate the superposition of these two kinds of turbulence in gas-liquid reactions. To this end, the variation of the interfacial turbulence during carbon dioxide absorption into aqueous monoethanolamine solutions in a wetted-wall column, with the gas-liquid contact time was followed by the tracer gas (ethylene) desorption technique.

EXPERIMENTAL

Absorption of Carbon Dioxide into Aqueous Monoethanolamine Solutions

Prior to the desorption experiment, absorption of pure carbon dioxide into aqueous monoethanolamine (MEA) solutions was carried out at 288 K. The gas-liquid contactor used was a wetted-wall column (Sada et al., 1976). The liquid film was formed on the outer surface of glass tube of 25 mm in diameter. The flow rate of aqueous MEA solution was varied from 3 to 12 cm^3/s ($Re_L = 130 \sim 540$). The height of the wetted-wall ranged from 5 to 14 cm.

Desorption of Ethylene from MEA Solutions into Carbon Dioxide and Nitrogen Streams

Experiments on desorption of ethylene from aqueous MEA solutions into carbon dioxide and nitrogen streams were conducted in the same wetted-column at 298 K. The liquid phase was saturated in advance with ethylene by bubbling for a sufficient time. The flow rate of aqueous MEA solution was varied from 3.6 to 20.4 cm^3/s ($Re_L = 160 \sim 930$). The desorption rate of ethylene was calculated from the ethylene concentration in the effluent stream and the total gas flow rate. The concentration of ethylene was determined by gas chromatography and the gas flow rate by a soap-film meter. In one run of the desorption into carbon dioxide stream, 0.10 vol. % of surface active agent, Tween 80, was added to aqueous MEA solution.

EXPERIMENTAL RESULTS AND DISCUSSION

Experimental results on absorption of carbon dioxide into aqueous MEA solutions were shown in Figure 1 as a plot of the enhancement factor ϕ against the diffusion-reaction modulus $\sqrt{M}$. The value of the rate constant of the second-order reaction between carbon dioxide and MEA which is required for calculation of the modulus $\sqrt{M}$ was taken as 3,300 L/mol·s at 288 K (Hikita et al., 1977). The solid lines represent the penetration-theory solutions for gas absorption with the second-order reaction for carbon dioxide-MEA system. The experimental enhancement factors fall much above the solid curves. The deviation increases with increasing the liquid flow rate as well as the MEA concentration.

A typical example of experimental results on desorption of ethylene from aqueous MEA solutions into carbon dioxide and nitrogen streams was shown in Figure 2 where the liquid-side mass transfer coefficient (i.e., desorption coefficient) was expressed as a function of the gas-liquid contact time. The chain line represents the corresponding penetration-theory prediction for ethylene desorption. The values of liquid-side mass transfer coefficient for the desorption into nitrogen stream are in good agreement with the corresponding theoretical values in low gas-liquid contact times ($t_c < 0.1$). As the contact time increases, the experimental desorption coefficient is slightly higher than that predicted by penetration theory.

On the other hand, the ethylene desorption coefficient into carbon dioxide stream was substantially increased over that into nitrogen stream. Figure 2 also shows that the rates of desorption into carbon dioxide stream are effectively decreased due to the addition of 0.10 vol. % of surface active agent and coincide well with those into nitrogen stream. The dependence of the desorption coefficient on the gas-liquid contact time is rather low and as the contact time decreases, the desorption coefficient is apt to approach a constant value. Let the contact time which gives such a constant

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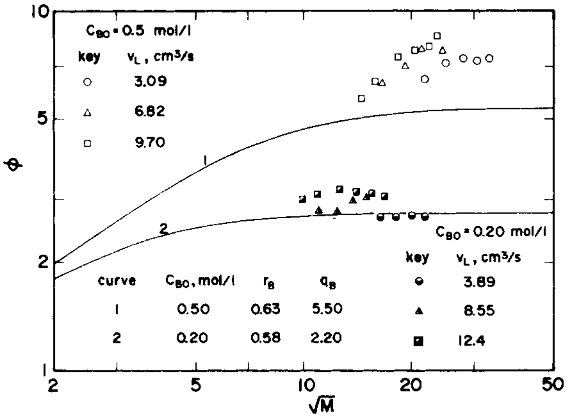

Figure 1. Enhancement factor as a function of reaction-diffusion modulus for absorption of CO_2 into aqueous MEA solutions.

desorption coefficient in the penetration-theory be referred to $t_{c,\text{crit.}}$. Then, $t_{c,\text{crit.}}$ corresponds to the time elapsing before the interfacial turbulence occurs. The increase in the liquid flow rate results in an increase in the desorption coefficient, but there seems to exist a certain upper-bound on the desorption coefficient with respect to the liquid flow rate.

To clearly show the promotion in ethylene desorption induced only by the Marangoni instability, the ratio of the desorption coefficient into carbon dioxide stream to that into nitrogen stream was plotted against the dimensionless gas-liquid contact time relative to $t_{c,\text{crit.}}$ in Figure 3. The quantity ϕ_M, in which the desorption coefficient into carbon dioxide stream is not divided by that predicted by the penetration-theory but that into nitrogen stream, does not involve the effect of mass transfer promotion due to the rippling on the surface of the falling liquid film, and may be reflected by the Marangoni effect alone. ϕ_M should be termed as the Marangoni-effect coefficient. When the abscissa is unity, the Marangoni-effect coefficient should be equal to unity. If the Marangoni instability does not diminish at all with an increase in the contact time, then the relationship between ϕ_M and $t_c/t_{c,\text{crit.}}$ should follow the chain line with a slope of 0.5. The observed coefficient in short contact times has a trend to approach the chain line, while in long contact times it becomes almost constant irrespective of the contact time. The Marangoni-effect coefficient reaches an upper-bound value when the liquid flow rate is increased to a certain value (~6.8 cm³/s). The rippling on the surface of the falling liquid film can depress some damping of the interfacial flow induced by the Marangoni instability.

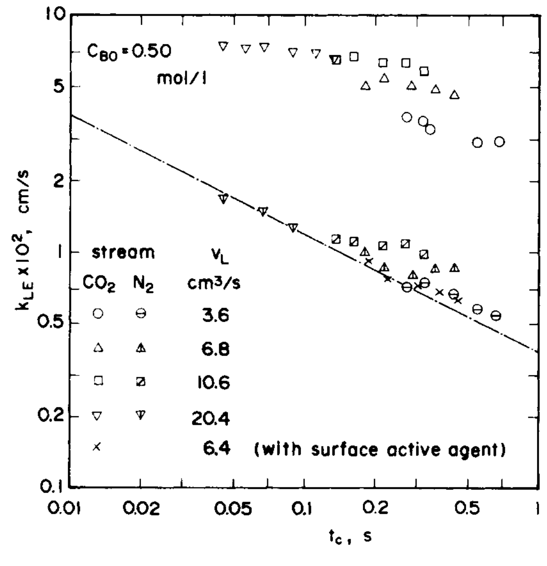

Figure 2. Variation of liquid-side mass transfer coefficient with gas-liquid contact time.

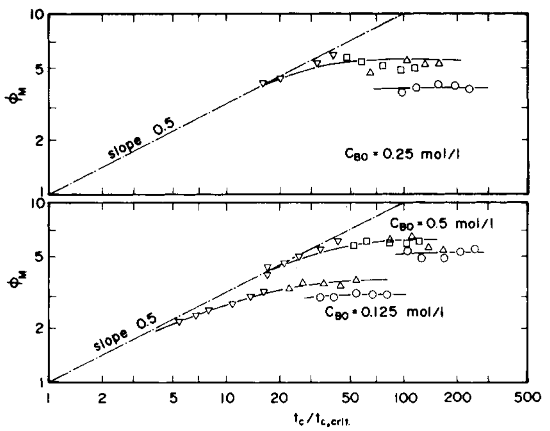

Figure 3. Variation of Marangoni-effect coefficient with dimensionless gas-liquid contact time. Keys as in Figure 2.

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NOTATION

C = concentration in liquid phase, mol/L
D = diffusivity in liquid phase, cm²/s
k = second-order reaction rate constant, L/mol·s
k_L = liquid-side mass transfer coefficient, cm/s
$\sqrt{M}$ = reaction-diffusion modulus, $\sqrt{\pi k C_{B0} t_c}/2$
q_B = $C_{B0}/\nu C_{Ai}$
r_B = D_B/D_A
t_c = gas-liquid contact time, s
V_L = volumetric liquid flow rate, cm³/s
ν = stoichiometric coefficient for the reaction between carbon dioxide and MEA
ϕ = enhancement factor
ϕ_M = Marangoni-effect coefficient

Subscripts

A = dissolved gas (carbon dioxide)
B = liquid-phase reactant (MEA)
E = tracer gas (ethylene)
i = gas-liquid interface
L = liquid phase
0 = initial value or bulk liquid phase

Superscript

o = without Marangoni instability

LITERATURE CITED

Brian, P. L. T., J. E. Vivian, and D. C. Matiatos, "Interfacial Turbulence during the Absorption of Carbon Dioxide into Monoethanolamine," *AIChE J.*, **13**, 28 (1967).

Hikita, H., S. Asai, H. Ishikawa, and M. Honda, "The Kinetics of Reactions of Carbon Dioxide with Monoethanolamine, Diethanolamine and Triethanolamine by a Rapid Mixing Method," *Chem. Eng. J.*, **13**, 7 (1977).

Sada, E., H. Kumazawa, and M. A. Butt, "Chemical Absorption Kinetics over a Wide Range of Contact Time," *AIChE J.*, **22**, 196 (1976).

Sada, E., H. Kumazawa, M. A. Butt, and J. E. Lozano, "Interfacial Turbulence Accompanying Chemical Absorption," *Can. J. Chem. Eng.*, **55**, 293 (1977a).

Sada, E., H. Kumazawa, M. A. Butt, and J. E. Lozano, "Interfacial Turbulence during Absorption with Reaction," *J. Chem. Eng. Japan*, **10**, 487 (1977b).

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