k_La	=	volumetric mass transfer coefficient	[s^{-1}]
L_F	=	aerated liquid height in column	[m]
L_s	=	quiescent liquid height in column	[m]
Sc	=	Schmidt number $=\mu_L/D_L\rho_L$	[—]
Sh	=	Sherwood number $=k_LD_T/D_L$	[—]
t	=	time	[s]
u_G	=	superficial gas velocity	[$m \cdot s^{-1}$]
V_{gel}	=	volume of gels in column	[m^3]
V_L	=	volume of liquid in column	[m^3]
ε_G	=	gas-phase holdup	[—]
ε_L	=	liquid-phase holdup	[—]
$\varepsilon_{S/L}$	=	$V_{gel}/(V_L+V_{gel})$=particle concentration	[—]
μ_L	=	liquid viscosity	[Pa·s]
ρ_L	=	liquid density	[$kg \cdot m^{-3}$]
σ	=	surface tension	[$kg \cdot s^{-2}$]

Literature Cited

1) Akita, K.: *Kagaku Kogaku Ronbunshu*, **13**, 181 (1987).
2) Akita, K. and F. Yoshida: *Ind. Eng. Chem. Proc. Des. Develop.*, **12**, 76 (1973).
3) Berk, D., L. A. Behie, A. Jones, B. H. Lesser and G. M. Gaucher: *Can. J. Chem. Eng.*, **62**, 112 (1984a).
4) Berk, D., L. A. Behie, A. Jones, B. H. Lesser and G. M. Gaucher: presented at the 34th Can. Chem. Eng. Conf., Quebec City, Quebec (Oct., 1984b).
5) Chibata, I. and T. Tosa: *Ann. Rev. Biophys. Bioeng.*, **10**, 197 (1981).
6) Dhanuka, V. R. and J. B. Stepanek: *AIChE J.*, **26**, 1029 (1980).
7) Fan, L. T. and M. P. Newcomer: *Adv. Biotechnol.* (Proc. Int. Ferment. Symp., 6th, 1980) **1**, 643 (1981).
8) Jones, A., D. N. Wood, T. Razniewska and G. M. Gaucher: *Can. J. Chem. Eng.*, **64**, 547 (1986).
9) Koide, K., A. Takazawa, M. Koumura and H. Matsunaga: *J. Chem. Eng. Japan.*, **17**, 459 (1984).
10) Nakanoh, M. and F. Yoshida: *Ind. Eng. Chem. Proc. Des. Develop.*, **19**, 190 (1980).
11) Ostergaard, K.: *AIChE J. Symposium series*, **74**, No. 176, 82 (1978).
12) Sato, K.: "Busseiteisu Suisanhou," p. 307, 3rd ed., Maruzen (1975).
13) Sun, Y.: M.S. Thesis, University of Tokyo (1987).
14) Sun, Y. and S. Furusaki: *J. Chem. Eng. Japan*, **21**, 20 (1988).

(Presented at the 52nd Annual Meeting of the Society of Chemical Engineers, Japan, at Nagoya, April 1987.)

MEAN BUBBLE DIAMETER AND OXYGEN TRANSFER COEFFICIENT IN A THREE-PHASE FLUIDIZED BED BIOREACTOR

YAN SUN AND SHINTARO FURUSAKI

Department of Chemical Engineering, University of Tokyo, Tokyo 113

Key Words: Mass Transfer, Mean Bubble Diameter, Interfacial Area, Mass Transfer Coefficient, Electroresistivity Probe, Three Phase Fluidized Bed, Bioreactor, Calcium Alginate Gel

The volume-surface mean diameter of bubbles d_{vs} was measured using the electroresistivity probe method. From the mean bubble diameter and volumetric mass transfer coefficient k_La, interfacial area a and mass transfer coefficient k_L were obtained. The solid phase, Ca-alginate gels, had almost no effect on bubble breakup, and a decrease in k_La with decreasing gel diameter was found to be caused by the corresponding decrease in k_L. The mean bubble diameter increased significantly with increasing particle concentration. The value of k_L was not affected by particle concentration. Thus, a decrease in k_La with increasing particle concentration was mainly attributed to the corresponding decrease in interfacial area. Dimensionless correlations were proposed for d_{vs}, a and k_L.

Introduction

The volumetric mass transfer coefficient, k_La, has been investigated in a three-phase fluidized bed of large and light particles.[5] k_La depends on the mass transfer coefficient k_L and the interfacial area a, and there is enough evidence in the literature to suggest that k_L and a depend upon completely different variables of the system. k_L depends mainly on the diffusivity of gas solute in the liquid and the viscosity of the liquid. However, a depends mainly on the surface tension and ionic strength of the solution and on the solid particles. Fan and Newcomer[2] studied the gas-liquid mass transfer properties in three-phase fluidized beds of glass and metal beads and suggested that larger and denser particles broke up bubbles. Ostergaard[3] reported that the interfacial area in a bed of 6-mm glass beads was much greater than that in a

Received April 21, 1987. Correspondence concerning this article should be addressed to S. Furusaki.

bubble column. Hence, to understand fully the mass transfer properties in the reactor, k_L and a should be investigated separately.

Thus the present work attempts to study systematically the mean bubble diameter in the three-phase fluidized bed. The electroresistivity probe method was employed to measure the mean bubble diameter d_{vs}, and the effects of liquid properties, particle concentration, and gel diameter on d_{vs}, a and k_L were studied.

1. Experimental

1.1 Apparatus

The experimental setup was the same as in the previous work[5] except for an electric probe instead of the oxygen electrode. **Figure 1** shows the details of the measuring circuits. The probe consisted of two 0.5 mm dia. enamel-covered copper wires as electrodes which were aligned vertically 3 mm apart. The electrodes were insulated except for a distance of about 0.2 mm from the tip, which was cut sharply. The electrodes were connected with shield wire and contained and fixed with epoxy resin in stainless steel tubes. Then they were inserted through the column wall at a height of 0.27 m above the bottom using a silicon stopper as seal. The electrodes could be positioned at any radial point within the bed. The wall electrode was made using a copper wire coil positioned at the bottom of the column.

The output from each electrode was adjusted by a DC amplifier to give a voltage drop fluctuating between 0 and 1 volt when the circuit between the tip and the wall electrode was opened and closed by the bubble and liquid phase, respectively. The output from the two electrodes was recorded on an oscillograph.

1.2 Determination of mean bubble diameter

Measuring time of one experiment was about 30–40 s and about 100–150 bubble data were obtained. All the experimental data were used to determine the volume-surface mean diameter d_{vs} of bubbles. Since no radial distribution of the mean bubble diameter was present,[4] the bubble data were obtained by positioning the tip electrodes at the central axis of the column. Moreover, because the bubble size was visually uniform above a height of 5 cm from the gas distributor, the bubble data obtained as above were used as the average values for the whole column.

The data were analysed according to Ueyama and Miyauchi.[6,7] From the values of the mean bubble diameter one obtains

$$a=\frac{6\varepsilon_G}{d_{vs}} \tag{1}$$

and

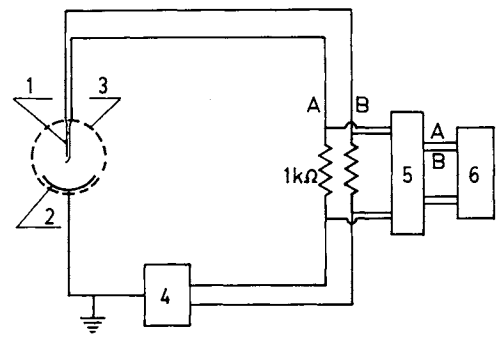


Fig. 1. Measuring circuits: 1, tip electrodes; 2, wall electrode; 3, column; 4, oscillator; 5, DC amplifier; 6, oscillograph.

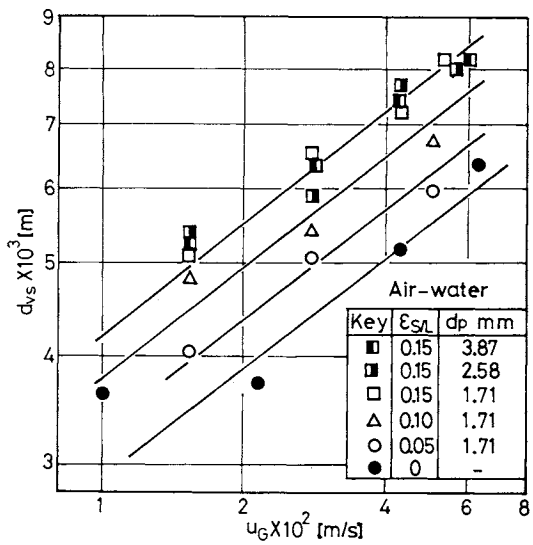


Fig. 2. Effect of gel diameter and particle concentration on d_{vs}

$$k_L=\frac{k_La}{a} \tag{2}$$

2. Results and Discussion

2.1 Volume-surface mean diameter and interfacial area

1) Effect of variables **Figure 2** shows the volume-surface mean diameter of bubbles as a function of the superficial gas velocity with the gel diameter and the particle concentration as parameters. The bubble size increased significantly with increasing superficial gas velocity. This is considered to be due to the fact that bubble density in the column became higher as superficial gas velocity was increased. Consequently, more frequent bubble coalescence occurred. No effect of gel diameter on d_{vs} was found in the case of such low-density gels. Since the gas holdup was also independent of gel diameter,[5] the interfacial area is independent of gel diameter.

The relationship between volumetric mass transfer coefficient and particle concentration has been discussed.[5] It was pointed out that large bubbles were

observed visually in three-phase fluidized beds, especially in beds of $\varepsilon_{S/L}=0.15$. Here the dependence of the bubble diameter on the particle concentration was determined and is also shown in Fig. 2. The figure verifies the visual observation mentioned above. Because of the presence of the solid phase, the free space for bubble rising becomes smaller in three-phase fluidized beds than in bubble columns. This effect was greater at higher particle concentrations. Hence the bubbles contact or collide with each other more frequently in three-phase fluidized beds.

The decrease in a with increasing particle concentration can be derived in terms of Eq. (1). The greatest interfacial area was found in the bubble column ($\varepsilon_{S/L}=0$).

The mean diameter of bubbles was much smaller in the sucrose solutions, compared with that in water, as shown in **Fig. 3**. This may be attributed to the higher viscosity and density of the sucrose solutions, which makes coalescence of bubbles more difficult. However, the reason why an insignificant difference was found between 10 and 30% sucrose solutions is not clear. In the same figure, the fact that the mean bubble diameter was much smaller in the ethanol solutions than in water at lower u_G values is shown, affirming the visual observation mentioned in the previous paper.[5] The higher increasing rate of mean bubble diameter with increasing u_G was considered to be due to the very small size of the bubbles at lower u_G, and much more significant bubble coalescence occurred at higher u_G values.

2) Correlations for d_{vs} and a According to the results discussed above, dimensional analysis yields

$$\frac{d_{vs}}{D_T}=f_1(Bo,\ Ga,\ Fr,\ \varepsilon_{S/L}) \tag{3}$$

Assuming that

$$\frac{d_{vs}}{D_T}=c_1 Bo^p Ga^q Fr^r \exp(c_2\varepsilon_{S/L}) \tag{4}$$

the least square fitting the experimental data gives

$$\frac{d_{vs}}{D_T}=0.87Bo^{-0.44}Ga^{0.10}Fr^{0.39}\exp(2.0\varepsilon_{S/L}) \tag{5}$$

Rewriting Eq. (5) gives

$$d_{vs}=0.14u_G^{0.39}\rho_L^{-0.22}\mu_L^{-0.20}\sigma^{0.44}\times\exp(2.0\varepsilon_{S/L}) \tag{6}$$

The standard deviation of Eq. (5) is 6.3%.

Figure 4 shows the plot of Eq. (5) on log–log coordinates with the Froude number as abscissa. The results for the bubble column are presented in the figure. Eq. (5) well correlates the values of d_{vs} in the bubble column. The result reported by Akita and Yoshida[1] for bubble columns is also shown. They

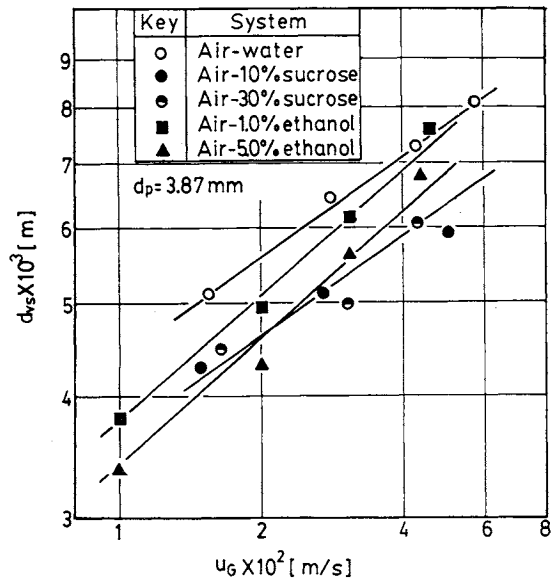


Fig. 3. Dependence of d_{vs} on liquid property ($\varepsilon_{S/L}=0.15$)

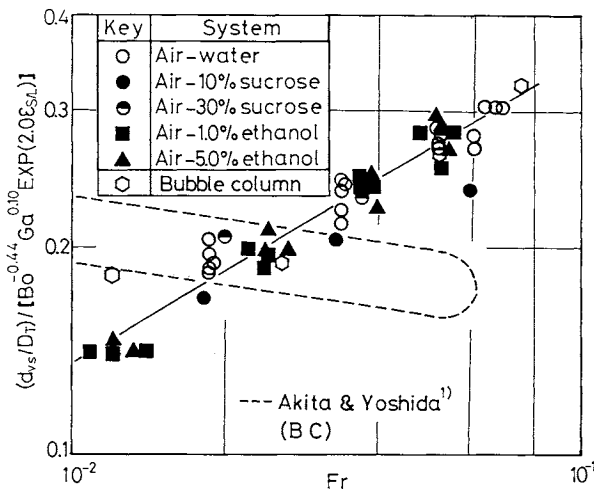


Fig. 4. Dimensionless correlation for d_{vs} (BC, bubble column)

suggested that d_{vs} was proportional to Fr to the -0.12 power, which is different from the present result. The different gas distributors employed were considered to be the main reason, as discussed in the previous paper.[5]

The correlation for the gas holdup was given as follows:[5]

$$\varepsilon_G=2.1\times10^{-3}Fr^{0.82}Bo^{0.80}Ga^{0.10} \tag{7}$$

Combining Eqs. (1), (5) and (7) and simplifying the exponents, the following relationship is obtained:

$$aD_T=1.4\times10^{-2}Fr^{0.43}Bo^{1.24}\exp(-2.0\varepsilon_{S/L}) \tag{8}$$

The dependence of the interfacial area a on particle diameter and concentration, discussed above, is shown in Eq. (8).

Ostergaard[3] measured the specific interfacial area by a chemical method in beds of 1, 3 and 6-mm glass

ballotini and reported that the greatest interfacial area was formed in the bed of 6-mm beads and the smallest in that of 1-mm beads. The relationship of the interfacial area in the three-phase fluidized beds and the bubble column was

$$a_6 : a_3 : a_1 : a_{BC} = 105 : 56 : 15 : 49 \qquad (9)$$

The a in the bed of 6-mm particles was 7 times as great as that of 1-mm particles, and over twice that in the bubble column. Fan and Newcomer[2] reported that the bubbles broke in beds of large particles. In this work, however, the gels of low density were considered not to cause the bubbles to split, even if the gels were large particles.

2.2 Mass transfer coefficient

1) Effect of variables It was noted in the previous section that the interfacial area was not affected by gel diameter. Calculated by Eq. (2) with the experimental data of k_La,[5] k_L decreased with decreasing gel diameter, as shown in **Fig. 5**. The result obtained in the bubble column is also shown in the figure. It is found that the value of k_L in the bubble column was approximately the same as that in the bed of 3.87-mm beads. Since the independence of d_{vs} (or a) from the gel diameter was found in the previous section, the decrease in k_La with decreasing gel diameter mentioned in the previous paper[5] was determined by the k_L dependence on d_p.

For gels with such a low density, the smaller ones were easy to adhere to the bubbles. From this we can conclude that two phenomena will occur for the smaller gels: (1) The effective interfacial area for mass transfer decreases. Thus the apparent decrease in the mass transfer coefficient may be attributed to a decrease in the effective interfacial area. (2) The renewal of gas-liquid interface was impeded. Thus the gas-liquid mass transfer coefficient decreases. It can be said that it is inappropriate to use very small gels to immobilize aerobe, as mentioned by Sun *et al.*[5]

Figure 6 shows that the particle concentration had negligible effect on k_L. Therefore, the decrease in k_La with increasing particle concentration was mainly attributed to the decrease in interfacial area.

In the sucrose solutions, k_L was much lower than that in water and decreased with increasing sucrose concentration, as shown in **Fig. 7**. The decrease in k_L owing to the addition of ethanol, as shown in the same figure, was considered partly due to the effect of ethanol impeding the renewal of gas-liquid interface.

2) Correlation for k_L Dimensional analysis of k_L results in

$$Sh' = f_2\left(Sc,\ Bo',\ Ga',\ \frac{d_p}{D_T}\right) \qquad (10)$$

where

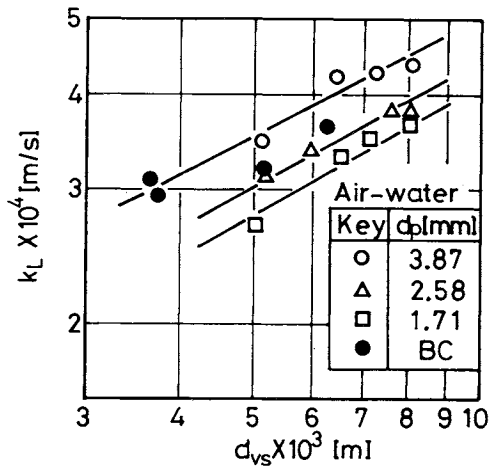


Fig. 5. Dependence of k_L on d_{vs} and d_p ($\varepsilon_{S/L}=0.15$)

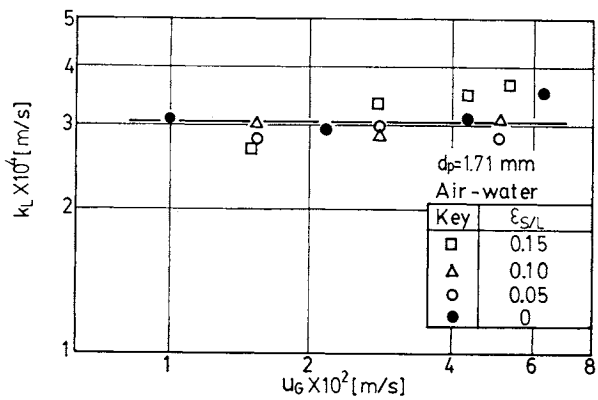


Fig. 6. Effect of particle concentration on k_L

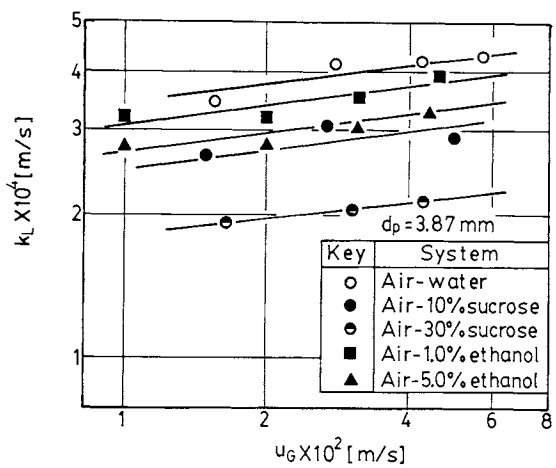


Fig. 7. Dependence of k_L on liquid property ($\varepsilon_{S/L}=0.15$)

$$Sh' = \frac{k_L d_{vs}}{D_L} \qquad (11)$$

$$Sc = \frac{\mu_L}{D_L \rho_L} \qquad (12)$$

$$Bo' = \frac{g d_{vs}^2 \rho_L}{\sigma} \qquad (13)$$

$$Ga' = \frac{g d_{vs}^3 \rho_L^2}{\mu_L^2} \qquad (14)$$

Referring to Higbie's penetration theory, the exponent of the Schmidt number should be 1/2. Hence, the least square fitting gives

$$Sh' = 0.13 Sc^{1/2} Bo'^{0.10} Ga'^{0.43} \left(\frac{d_p}{D_T}\right)^{0.20} \quad (15)$$

From Eq. (15), k_L is proportional to $d_{vs}^{0.49}$.

Akita and Yoshida[1] reported a correlation of the same type as Eq. (15) for k_L in the bubble column and noted that the exponent of d_{vs} was 0.5. **Figure 8** shows a plot for Eq. (15). For comparison, Akita and Yoshida's result is shown in the figure with the term $(d_p/D_T)^{0.20}$ being excluded in the oridinate. The present results obtained in the bubble column are also shown in Fig. 8, and are in agreement with the correlation of Akita and Yoshida.[1] To make Eq. (15) hold for the bubble column, we may let the term $(d_p/D_T)^{0.20}$ be a constant and the coefficient be 0.067 (equal to the average value of 0.13 $(d_p/D_T)^{0.20}$). The exponents of the other dimensionless numbers are the same as those of the equation.

Conclusions

1. Ca-alginate gel was considered not to split bubbles, regardless of gel diameter. A decrease in k_La with decreasing gel diameter was found to be the corresponding decrease in k_L.

2. Increasing particle concentration caused bubble coalescence to increase the bubble size, but had almost no effect on the mass transfer coefficient. A decrease in k_La with increasing particle concentration was mainly attributed to the corresponding decrease in the interfacial area.

3. Dimensionless correlations for d_{vs}, a and k_L were proposed.

Acknowledgment

The authors wish to gratefully acknowledge the assistance of Dr. K. Ueyama with the construction of the electroresistivity probe used in the study. Valuable advice from Dr. T. Nozawa is also greatly appreciated.

Nomenclature

a	=	gas-liquid interfacial area	[m^{-1}]
a_{BC}, a_1, a_3, a_6	=	interfacial area in bubble column and beds of 1-, 3- and 6-mm beads, respectively	[m^{-1}]
Bo	=	Bond number $= gD_T^2\rho_L/\sigma$	[—]
Bo'	=	Bond number $= gd_{vs}^2\rho_L/\sigma$	[—]
D_L	=	molecular diffusivity of O_2 in liquid phase	[$m^2 \cdot s^{-1}$]
D_T	=	inside column diameter	[m]
d_p	=	particle diameter	[m]
d_{vs}	=	volume-surface mean diameter of bubbles	[m]
Fr	=	Froude number $= u_G/(gD_T)^{1/2}$	[—]
Ga	=	Galileo number $= gD_T^3/\nu_L^2$	[—]
Ga'	=	Galileo number $= gd_{vs}^3/\nu_L^2$	[—]
g	=	gravitational constant	[$m \cdot s^{-2}$]
k_L	=	gas-liquid mass transfer coefficient	[$m \cdot s^{-1}$]
k_La	=	volumetric mass transfer coefficient	[s^{-1}]
Sc	=	Schmidt number $= \mu_L/D_L\rho_L$	[—]
Sh'	=	Sherwood number $= k_Ld_{vs}/D_L$	[—]
u_G	=	superficial gas velocity	[$m \cdot s^{-1}$]
V_{gel}	=	volume of gels in column	[m^3]
V_L	=	volume of liquid in column	[m^3]
ε_G	=	gas-phase holdup	[—]
$\varepsilon_{S/L}$	=	$V_{gel}/(V_{gel}+V_L)$ = particle concentration	[—]
μ_L	=	liquid viscosity	[Pa·s]
ρ_L	=	liquid density	[$kg \cdot m^{-3}$]
σ	=	surface tension	[$kg \cdot s^{-2}$]

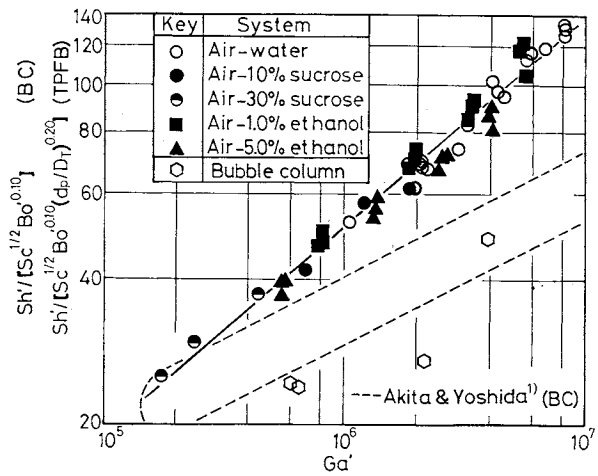


Fig. 8. Dimensionless correlation for k_L (BC, bubble column; TPFB, three-phase fluidized bed)

Literature Cited

1) Akita, K. and F. Yoshida: *Ind. Eng. Chem. Proc. Des. Develop.*, **13**, 84 (1974).
2) Fan, L. T. and M. P. Newcomer: *Adv. Biotechnol.* (Proc. Int. Ferment. Symp., 6th, 1980), **1**, 643 (1981).
3) Ostergaard, K.: *AIChE Symp. Series*, **74**, No. 176, 82 (1978).
4) Sun, Y.: M. S. Thesis, University of Tokyo (1987).
5) Sun, Y., T. Nozawa and S. Furusaki: *J. Chem. Eng. Japan*, **21**, 15 (1988).
6) Ueyama, K. and T. Miyauchi: *Kagaku Kogaku Ronbunshu*, **2**, 430 (1976).
7) Ueyama, K. and T. Miyauchi: *Kagaku Kogaku Ronbunshu*, **3**, 19 (1977).