Characteristics of Gas-liquid Mass Transfer and Interfacial Area in a Bubble Column

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Abstract – Characteristics of gas-liquid mass transfer and interfacial area were investigated in a bubble column of diameter and height of 0.102 m and 2.5 m, respectively. Effects of gas and liquid velocities on the volumetric gas-liquid mass transfer coefficient ($k_L a$), interfacial area (a) and liquid side true mass transfer coefficient (k_L) were examined. The interfacial area and volumetric gas-liquid mass transfer coefficient were determined directly by adopting the simultaneous physical desorption of O_2 and chemical absorption of CO_2 in the column. The values of $k_L a$ and a increased with increasing gas velocity but decreased with increasing liquid velocity in the bubble column which was operated in the churn turbulent flow regime. The value of k_L increased with increasing gas velocity but did not change considerably with increasing liquid velocity. The liquid side mass transfer was found to be related closely to the liquid circulation as well as the effective contacting frequency between the bubbles and liquid phases.

Key words: Bubble Column, Volume Mass Transfer Coefficient, Interfacial Area, Liquid Side Mass Transfer Coefficient, Liquid Circulation

1. Introduction

Simplicity, low operating and maintenance costs, and high heat and mass transfer rates in the column have enabled the bubble column to be employed widely in the fields of biochemical, food, energy, environmental and medical engineering processes [1-3]. Numerous investigations, therefore, have been conducted to utilize the unique features of bubble columns, including the phase holdups, mixing, heat and mess transfer, bubble properties etc. [1-3].

To design, scale-up and operate the bubble column reactors or contactors, the information on the mass transfer between the continuous liquid and the discrete bubble phase has been an essential factor, since the information is necessary to analyze and estimate the performance of the reactors or contactors adopting the bubble columns. For the analysis of mass transfer of gas phase such as O_2, CO_2, SO_X and NO_X from the flowing bubbles to the liquid phase, the interfacial area between the two phases has to be determined. In addition, the liquid side true mass transfer coefficient should be verified, since the resistance in the gas phase for the mass transfer has been understood to be negligible [1-4]. However, various kinds of investigations have focused on the overall volumetric mass transfer coefficients, and the values of interfacial area between the contacting two phases have been estimated from a knowledge of the bubble size and its holdup indirectly [5-9]. In the present study, the interfacial area and liquid side true mass transfer coefficient in addition to the overall volumetric mass

transfer coefficient were determined directly by resorting to the simultaneous physical desorption of O_2 and chemical absorption of CO_2 [10-13]. Characteristics of the gas-liquid interfacial area and the liquid side and volumetric mass transfer coefficients were also discussed.

2. Experiments

The experiments were in a bubble column of diameter and height of 0.102 m and 2.5 m, respectively, as can be seen in Fig. 1. A perforated plate served as a gas and liquid distributor. The plate, which contained 120 evenly spaced holes of 2.0×10^{-3} m in diameter, was installed between the main column section and a 0.2 m high stainless steel distributor box into which the liquid phase was introduced through a 0.025 m pipe from a liquid reservoir. Oil-free compressed gas was fed to the column through a pressure regulator, filter and a calibrated gas flowmeter. The gas was admitted to the column through 3.0×10^{-3} m ID perforated pipes with 88 holes of 1.0×10^{-3} m ID drilled horizontally in the plate. The details of gas and liquid distributor can be found elsewhere [14-16]. The gas holdup was determined by means of static pressure drop method [2,3], by measuring the pressure drop variations in the column. Five pressure taps were mounted flush with the wall of the column with 0.2 m height intervals, 0.5 m apart from the gas and liquid distributor. The signals of pressure drops were processed to produce the digital data [17,18]. Compressed O_2 and CO_2 were used as the gas phase, and tap water and sodium carbonate/sodium bicarbonate solutions were used as the continuous liquid phase, of which temperature was kept at 20 ± 2 °C. When a steady state was reached at a given operating condition, five liquid samples were obtained simultaneously by means of solenoid valves and sampling ports located axially at the wall of the column at an interval of 0.1 m

Fig. 1. Experimental apparatus.

1. Main column	15. G/L distributor
2. Gas/liquid separator	16. CO_2 gas bomb
3. Liquid reservoir	17. O_2 gas bomb
4. Control valve	18. N_2 gas bomb
5. Liquid flowmeter	19. Liquid sample analyzer
6. Gas flowmeter	20. Liquid pump
7. Sampling tap	21. Gas filter & regulator
8. Pressure tap	22. Calming section
9. Pressure sensor	23. Liquid supplying tank
10. Amplifier	24. N_2 sparger
11. Low-pass filter	25. Thermocouple
12. Data acquisition system	26. Solenoid valve
13. A/D converter	27. Dissolved gas analyzer
14. Computer	

and 0.6 m from the distributor. The sampling was conducted at three radial coordinators, r/R, 0, 0.5 and 1.0 for a given axial position. The sample was passed through a tube containing an oxygen probe (membrane type) to measure the dissolved oxygen concentration in the sample and was then analyzed to measure the carbonate/bicarbonate concentration by a titration method for the determination of CO_2 concentration in the sample [12,13].

The concentrations of desorbing O_2 and chemically absorbing CO_2 gas were determined from the mean values of the experimentally measured values at the axial and radial positions of the column. The values of the interfacial area and volumetric mass transfer coefficient were obtained, based on the following equations [10-13], that is, after measuring of oxygen concentration in the liquid sample by using oxygen probe and of CO_2 concentration by means of titration method, the values of a and $k_L a$ were calculated by using Eqs. (1)-(5).

The absorption can be assumed to be a fast pseudo first-order reaction in the liquid phase as [4].

$$R_{CO_2}a = aC^*_{CO_2}(k^2_{L,CO_2} + D_{CO_2}k_2C_O)^{1/2} \tag{1}$$

Since the desorption rate coefficient of oxygen can be written as Eq. (2).

$$k_{L,O_2} = k_{L,CO_2}(D_{O_2}/D_{CO_2})^{1/2} \tag{2}$$

and the physical desorption rate of O_2 can be written as Eq.(3),

$$R_{O_2}a = k_{L,O_2}a\Delta C_{O_{2,m}} \tag{3}$$

the desorption rate of oxygen can be expressed as Eq. (4).

$$R_{O_2}a = k_{L,CO_2}a(D_{O_2}/D_{CO_2})^{1/2}\Delta C_{O_{2,m}} \tag{4}$$

Therefore, the interfacial area, a, can be determined from Eqs. (1) ~(4) as Eq. (5).

$$a = [\{(R_{CO_2}a/C^*_{CO_2})^2 - k_{L,CO_2}a^2\}/D_{CO_2}k_2C_O]^{1/2} \tag{5}$$

The liquid-side mass transfer coefficient was determined by using Eq. (6) with the assumption that the gas phase resistance can be negligible.

$$k_L = K_L = (k_L a)/a \tag{6}$$

3. Results and Discussion

Gas-liquid mass transfer is closely related to the bubble holdup in the column, since the bubbles are flowing as a dispersed phase in the continuous liquid medium. Effects of gas and liquid velocities on the gas holdup can be seen in Fig. 2. The gas holdup increases proportional to the increase in the gas velocity, but it decreases with an increase in the liquid velocity. The increase in the gas velocity leads to the increase in the amount of gas per unit cross sectional area of the column, which consequently results in the increase in the gas holdup. However, the increase in the liquid velocity results in the increase in the liquid holdup, which causes to the decrease in the gas
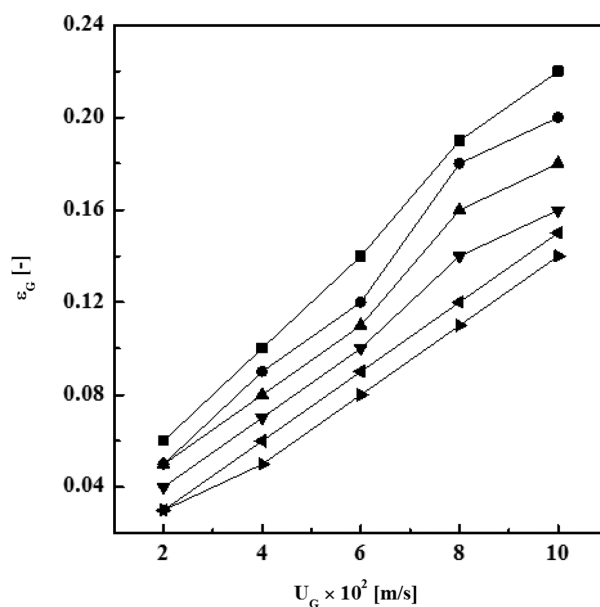

Fig. 2. Effects of gas and liquid velocities on the gas holdup in bubble columns.

	■	●	▲	▼	◄	►
$U_L \times 10^2$ [m/s]:	2.0	3.0	4.0	6.0	8.0	10.0

Fig. 3. Plot of drift flux model for analysis of the bubble flow in bubble columns.

Fig. 4. Effects of gas and liquid velocities on the volumetric gas-liquid mass transfer coefficient in bubble columns.

$U_L \times 10^2$ [m/s] :	■ 2.0	● 4.0	▲ 8.0	▼ 10.0

holdup in the column. Those trends agree well with the results of other investigations [8,9,18].

The complicated flow behavior of bubbles in the bubble column can be described by means of the drift flux model which was suggested by Zuber and Findley[19]. The interstitial velocity of bubbles in the column can be written as Eq. (7) based on the model.

$$\frac{U_G}{\varepsilon_G} = C(U_G + U_L) + U_0 \qquad (7)$$

In Eq. (7), C is a constant which can be utilized to anticipate the non-uniformity of the bubbles and liquid flow in the radial direction in the column, and U_0 can be the mean rising velocity of a single bubble in the column. As can be seen in Fig. 3, the plot of drift flux model is a straight line with a slope of 2.45 and an intercept of 27.40, respectively. The value of slope, 2.45, implies that the bubbling behavior in the column could belong to churn turbulent flow regime, due to vigorous contacting and flowing of bubbles in the column [6,7,20]. The rising velocity of a single bubble in the continuous liquid medium can also be estimated from Eq. (8), with the properties of liquid medium [7,21].

$$U_0 = 1.53(\sigma_L g / \rho_L)^{0.25} \qquad (8)$$

The estimated rising velocity of a single bubble in the experimental condition of this study was 0.25 m/s, since filtered O_2 or CO_2 and tap water were used as gas and liquid medium, respectively. The rising velocity of a single bubble predicted from Fig. 3 is 0.27 m/s, which is comparable to that estimated from Eq. (8) within a deviation of 8%.

Effects of gas and liquid velocities on the overall volumetric gas-liquid mass transfer coefficient, $k_L a$, can be seen in Fig. 4. The value of $k_L a$ increases gradually with increasing gas velocity, but it decreases with increasing liquid velocity. The increase of $k_L a$ with U_G is due to
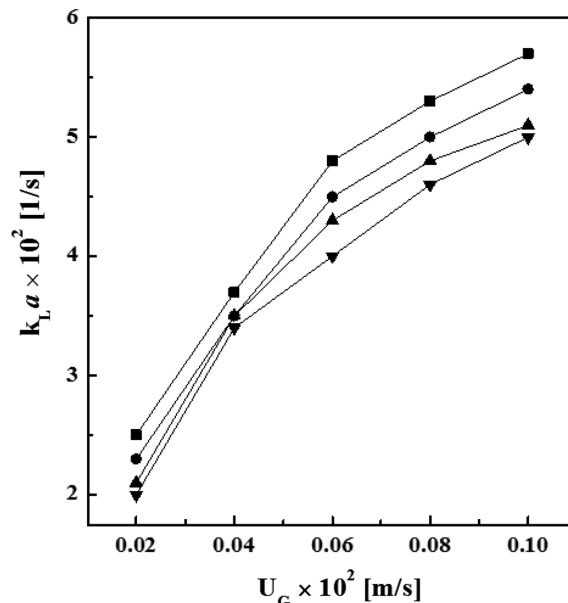
the increases of gas holdup and turbulence in the column, with increasing U_G. However, the residence time of bubbles in the column could decrease with increasing liquid velocity, since the drag force acting on the rising bubbles increases with increasing upward U_L. This means that the contacting efficiency between the gas and liquid medium could decrease and thus the overall volumetric mass transfer between them could also decrease, with increasing liquid velocity.

The interfacial area between the dispersed bubbles and the continuous liquid medium has been estimated from the data of gas holdup and bubble size indirectly [1-3]. However, in this study, the value of interfacial area was obtained directly by means of gas absorption and desorption method as written in Eq. (5). Effects of gas and liquid velocities on the interfacial area, a, for gas-liquid mass transfer can be seen in Fig. 5. The value of gas-liquid interfacial area increases with increasing gas velocity but slightly decreases with increasing liquid velocity. The results are closely related to the gas holdup in the bubble column, as mentioned before. That is, the gas-liquid interfacial area increases due to the increase in the gas amount in the column, with increasing gas velocity. But, the interfacial area decreases as the decrease in the gas holdup, with increasing liquid velocity.

Effects of gas and liquid velocities on the liquid side true mass transfer coefficient, k_L, can be seen in Fig. 6. The value of k_L was obtained by Eq. (6) from the measured values of $k_L a$ and interfacial area, a, in a given operating condition. The value of liquid side mass transfer coefficient increases with increasing gas velocity, however, it does not change considerably with increasing liquid velocity. Note that the value of k_L sometimes increases slightly but sometimes
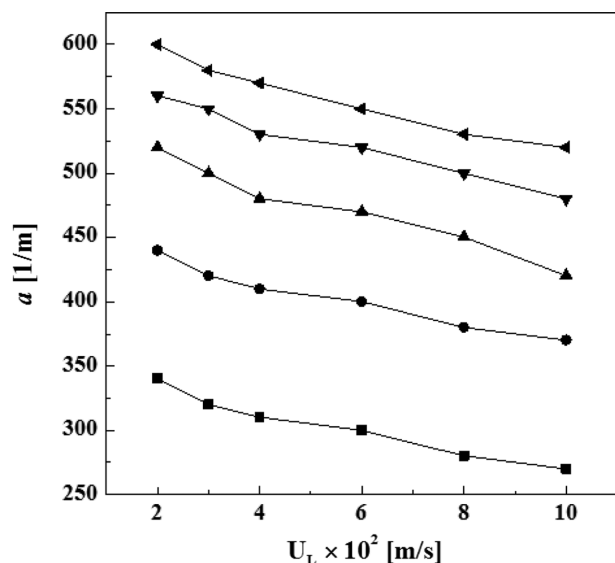
Fig. 5. Effects of gas and liquid velocities on the gas-liquid interfacial area in bubble columns.

$U_G \times 10^2$ [m/s]:   ■ 2.0   ● 4.0   ▲ 6.0   ▼ 8.0   ◄ 10.0
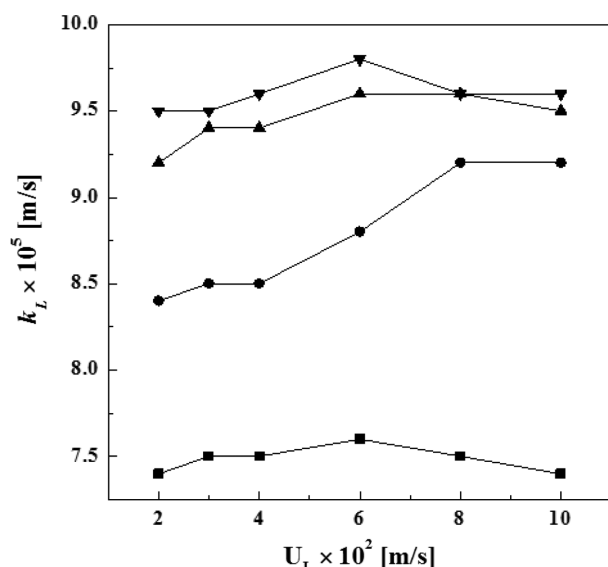

Fig. 6. Effects of gas and liquid velocities on the liquid side mass transfer coefficient in bubble columns.

$U_G \times 10^2$ [m/s]:   ■ 2.0   ● 4.0   ▲ 6.0   ▼ 10.0

shows a local maximum value with an increase in the liquid velocity. The effect of gas velocity on the value of k_L is larger than that of liquid velocity. This implies that the liquid side mass transfer coefficient is dependent upon the turbulence in the column. In other words, the increase of gas velocity could lead to the increase of turbulence in the bubble column, which results in the increase of contacting and mass transfer efficiency between the bubbles and the continuous liquid medium. Actually, the interfacial velocities of gas and liquid medium
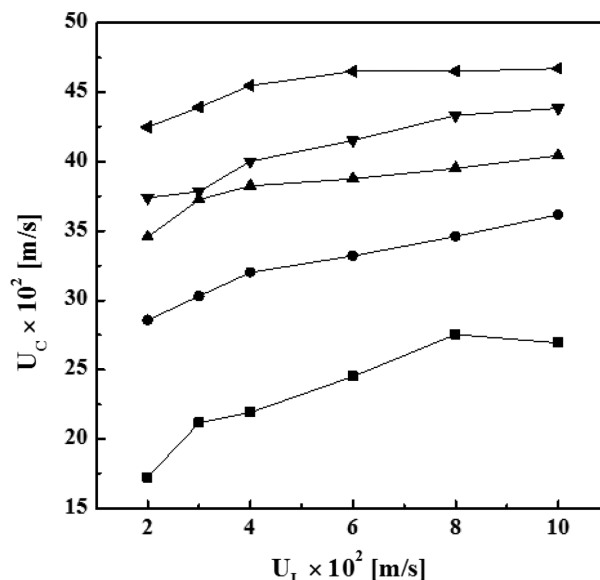
Fig. 7. Effects of gas and liquid velocities on the liquid circulation rate in bubble columns.

$U_G \times 10^2$ [m/s]:   ■ 2.0   ● 4.0   ▲ 6.0   ▼ 8.0   ◄ 10.0

in the bubble column are different. The circulation motion of liquid medium can be generated due to this velocity difference between the two phases in the column. The liquid circulation velocity in the bubble column was suggested by Joshi [22] as Eq. (9).

$$U_C = 1.31 \left\{ gD \left(U_G - \frac{\varepsilon_G}{\varepsilon_L} U_L - \varepsilon_G U_0 \right) \right\}^{1/3} \tag{9}$$

As can be seen in Fig. 7, the circulation velocity of liquid medium (U_C) increases with increasing gas velocity, but it increases only slightly with increasing liquid velocity. The liquid circulation in the column could generate turbulence, which means that the increase of liquid circulation could enhance the turbulence in the column and thus increase in the liquid side mass transfer at the gas-liquid interface. Based on the concept of surface renewal theory [1,4], the liquid side mass transfer coefficient can be written as Eq. (10), where k is a constant, D and S are the molecular diffusivity and the surface renewal rate, respectively. That is, the liquid side mass transfer coefficient is strongly influenced by the eddies with high energy, which can increase the surface renewal rate at the gas-liquid interface. In the bubble column, the surface renewal rate at the gas-liquid interface can be replaced by the effective contacting frequency between the gas and liquid phases, F_b, as Eq. (11), since the effective contacting between the gas and liquid medium causes to the mass transfer at the gas-liquid interface.

$$k_L = kD^{1/2}S^{1/2} \tag{10}$$

$$k_L = kD^{1/2}F_b^{1/2} \tag{11}$$

The interfacial area between the bubbles and continuous liquid

medium, volumetric mass transfer coefficient and liquid side true mass transfer coefficient were well correlated in terms of gas and liquid velocities as Eq's (12)~(14), with correlation coefficients of 0.993, 0.979 and 0.901, respectively.

$$a = 9.86 \times 10^2 U_G^{0.38} U_L^{-0.11} \qquad (12)$$

$$k_L a = 0.15 U_G^{0.54} U_L^{-0.09} \qquad (13)$$

$$k_L = 1.53 \times 10^{-4} U_G^{0.16} U_L^{0.02} \qquad (14)$$

The values of gas-liquid interfacial area in the bubble columns are usually obtained from the measured values of bubble holdup and bubble size, indirectly [1,2,8]. However, in this study, the values were determined directly [10-13]. Thus, the values of gas-liquid interfacial area obtained in this study could not be easily compared with those in the literature, because the directly obtained data in the bubble columns have been little, although the data obtained in the three phase fluidized beds have been available [10-13]. The values obtained in this study were higher than those in the literature [1,2,8]. This can be attributed to the fact that the methods for determining the values are different, in addition to the different systems including the gas distributor.

4. Conclusion

The volumetric gas-liquid mass transfer coefficient ($k_L a$), gas-liquid interfacial area (a) and liquid side mass transfer coefficient (k_L) were measured and determined simultaneously in a bubble column. The bubbling behavior in the column belonged to the churn turbulent flow regime. The values of $k_L a$ and a increased with increasing gas velocity but decreased with increasing liquid velocity. The value of k_L increased with increasing gas velocity but did not change considerably with increasing liquid velocity in the bubble column. The liquid circulation velocity in the column increased with increasing gas velocity but increased only slightly with increasing liquid velocity. The liquid side mass transfer coefficient was closely related to the liquid circulation, which affects the effective contacting frequency between the gas and liquid phases.

Acknowledgment

Nomenclatures

a : interfacial area [1/m]
C : constant in Eq. (7) [-]
C_0 : reactant concentration in the bulk liquid [mol/l]
$\triangle C_m$: logarithmic mean of the concentration difference [mol/l]
D : molecular diffusivity [m²/s]
D_B : column diameter [m]

F_b : effective contacting frequency between bubbles and liquid medium [1/s]
g : gravity acceleration [m/s²]
H : bed height [m]
k_L : liquid-side mass transfer coefficient [m/s]
$k_L a$: volumetric mass transfer coefficient [1/s]
k : constant in Eq. (10) [-]
K_L : overall mass transfer coefficient based on the liquid-side [m/s]
r : diameter of radial dispersion [m]
R : absorption rate per unit surface area [mol/m²·s]
S : surface renewal rate [1/s]
U_C : liquid circulation rate [m/s]
U_G : gas velocity [m/s]
U_L : liquid velocity [m/s]
U_0 : mean rising velocity of a single bubble [m/s]

Greek Letters

ε : phase holdup [-]
ρ : density [kg/m³]
σ : surface tension [N/m]

Subscripts

b : bubble
G : gas phase
L : liquid phase

References

1. Deckwer, W. D., Bubble Column Reactors, *John Wiley & Sons*, England, 239-267(1992).
2. Kim, S. D. and Kang, Y., "Heat and Mass Transfer in Three-phase Fluidized-bed Reactors: An Overview," *Chem. Eng. Sci.*, **52**, 3639-3660(1997).
3. Kim, S. D. and Kang, Y., "Hydrodynamics, Heat and Mass Transfer in Inverse and Circulating Three-phase Fluidized-bed Reactors for Wastewater Treatment," *Stud. Surf. Sci. and Catal.*, **159**, 179-196(1993).
4. Danckwerts, P. V., "Significance of Liquid-film Coefficients in Gas Absorption," *Ind. Eng. Chem.*, **43**, 1460-1465(1951).
5. Kang, Y., Lee, K. I., Shin, I. S., Son, S. M., Kim, S. D. and Jung, H., "Characteristics of Hydrodynamics, Heat and Mass Transfer in Three-phase Inverse Fluidized Beds," *Korean Chem. Eng. Res.*, **46**, 451-464(2008).
6. Clark, N. N. and Flemmer, R. L., "Predicting the Holdup in Two-phase Bubble Upflow and Downflow Using the Zuber and Findlay Drift-flux Model," *AIChE J.*, **31**, 500-503(1985).
7. Clark, N. N. and Flemmer, R. L., "On Vertical Downward Two Phase Flow," *Chem. Eng. Sci.*, **39**, 170(1984).
8. Nguyen-Tien, K., Patwari, A. N., Schumpe, A. and Deckwer, W. D., "Gas-liquid Mass Transfer in Fluidized Particle Beds," *AIChE J.*, **31**, 194-201(1985).
9. Son, S. M., Kim, U. Y., Shin, I. S., Kang, Y. and Jung. H., "Enhancement of Gas Holdup and Oxygen Transfer in Three-phase Circulating Fluidized-bed Bioreactors with Swirling Flow," *J. Chem.*

Eng. Japan, **42**, 142-148(2009).

10. Robinson, C. W. and Wilke, C. R., "Simultaneous Measurement of Interfacial Area and Mass Transfer Coefficients for a Well-mixed Gas Dispersion in Aqueous Electrolyte Solutions," *AIChE J.*, **20**, 285-294(1974).

11. Dhanuka, V. R. and Stepanek, J. B., "Simultaneous Measurement of Interfacial Area and Mass Transfer Coefficient in Three Phase Fluidized Beds," *AIChE J.*, **26**, 1029-1038(1980).

12. Lee, J. S., Jin, H. R., Lim, H., Lim, D. H., Kang, Y. and Kim, S. D., "Interfacial Area and Liquid-side and Overall Mass Transfer Coefficients in a Three-phase Circulating Fluidized Bed," *Chem. Eng. Sci.*, **100**, 203-211(2013).

13. Chang, S. K., Kang, Y. and Kim, S. D., "Mass Transfer in two and Three-phase Fluidized Beds," *J. Chem. Eng. Jpn*, **18**, 524-530 (1986).

14. Kang, Y., Fan, L. T., Min, B. T. and Kim, S. D., "Promotion of Oxygen Transfer in Three Phase Fluidized-bed Bioreactors by Floating Bubble Breakers," *Biotechnol. Bioeng.*, **37**, 580-586 (1991).

15. Lim, D. H., Park, J. H., Kang, Y. and Jun, K. W., "Structure of Bubble Holdups in a Viscous Slurry Bubble Column with Low Surface Tension Media," *Fuel Process. Technol.*, **108**, 2-7 (2013).

16. Shin, I. S., Son, S. M., Kim, U. Y., Kang, Y., Kim, S. D. and Jung, H., "Multiple Effects of Operating Variables on Bubble Properties in Three-phase Slurry Bubble Columns," *Korean J. Chem. Eng.*, **26**, 587-591(2009).

17. Lee, K. I., Son, S. M., Kim, U. Y., Kang, S. H., Kang, Y. and Kim, S. D., "Particle Dispersion in Viscous Three-phase Inverse Fluidized Beds," *Chem. Eng. Sci.*, **62**, 7060-7067(2007).

18. Jin, H. R., Lim, D. H., Lim, H., Kang, Y., Jung, H. and Kim, S. D., "Demarcation of Large and Small Bubbles in Viscous Slurry Bubble Columns," *I&EC Research*, **51**, 2062-2069(2012).

19. Zuber, M. and Findley, J. A., "Average Volumetric Concentration in Two-Phase Flow Systems," *Trans. ASME J. Heat Transfer*, **87**, 453-468(1965).

20. Wallis, G. B., One Dimensional Two Phase Flow, *McGraw-Hill*, NY(1969).

21. Harmathy, T. Z., "Velocity of Large Drops and Bubbles in Media of Infinite or Restricted Extent," *AIChE J.*, **6**, 281-288(1960).

22. Joshi, J. B., "Axial Mixing in Multiphase Contactors - A Unified Correlation," *Trans. Inst. Chem. Eng.*, **58**, 155-165(1980).