



Effect of pressure on volumetric mass transfer coefficient and gas holdup in bubble column

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Abstract—Volumetric mass transfer coefficient and gas holdup in bubble column was studied experimentally under pressurized condition with variation of liquid phase, diameter of single nozzle used as gas disperser. The pressure in the bubble column was in the range 0.1–1.1 MPa. Gas holdup and volumetric mass transfer coefficient increased with pressure. The effect of pressure on gas holdup and volumetric mass transfer coefficient appeared significantly in the case where the single nozzle with smaller diameter were used as gas disperser. Experimental equations for both gas holdup and volumetric mass transfer coefficients under small volume of gas chamber were presented incorporating the effect of pressure, diameter of single nozzle used as gas disperser and gas velocity. © 1997 Elsevier Science Ltd

Keywords: Bubble column; high system pressure; gas holdup; volumetric mass transfer coefficient; enzyme solution.

1. INTRODUCTION

Bubble columns under pressurized condition are used as reactors for liquefaction of coal or hydrogenation of heavy oil, and may be useful as reactors for other reactions, for example, oxidation of glucose, because the increased solubility of the reactant gas with pressure is expected to bring the enhancement of the reaction rate. Gas holdup and liquid-phase volumetric mass transfer coefficient are the important design parameters of the bubble column reactor.

The effect of pressure on gas holdup has been studied by several workers. In these works, gas holdup has been found to increase with the operating pressure. The increase of gas holdup at the elevated operating pressure has been attributed to the smaller bubble size resulting from the decrease of coalescence rate (Sagert and Quinn, 1976), the enhancement of bubble break-up (Wilkinson *et al.*, 1990), or the decreased size of bubbles formed at the gas disperser (Kling, 1962; LaNauze and Harris, 1974; Tsuge and Hibino, 1980).

When gas dispersers have holes larger than about 1 mm and the column diameter is greater than about 0.15 m, the average size of bubble swarm in the bubble column is said to be determined by the coalescence or break-up process. For this case, Wilkinson *et al.* (1992) have presented the semi-empirical

equation for gas holdup that agrees well with the pilot-plant-scale data (Tarmy *et al.*, 1984; Yamaguchi *et al.*, 1991).

The data on the design parameters in the bubble column with diameter less than about 0.1 m are also important for the scale-up or for the analysis of laboratory data. In this case, gas holdup seems to be influenced by the bubble size formed at the gas disperser. In the bubble column with sintered porous plate used as gas disperser, gas holdup is not influenced by the increase of pressure and the homogeneous bubbly flow prevailed up to the relatively higher gas velocity (Kölbel *et al.*, 1962; Deckwer *et al.*, 1980). Having used a perforated plate with holes of 1 mm as a gas disperser, Idogawa *et al.* (1985) have found that gas holdup increased with pressure and presented the experimentally correlated equation. Kojima *et al.* (1991) have reported that gas holdup increased with pressure in the bubble column equipped with a single-nozzle gas disperser whose diameter is less than 2 mm and no effect of pressure was observed in the bubble column with a single nozzle of 3 mm.

Papers on the volumetric mass transfer coefficient under elevated operating pressure are far less numerous. Wilkinson and Haringa (1994) have found that volumetric mass transfer coefficient has increased with pressure in the bubble column with diameter of 0.158 m using a ring sparger with 19 holes of 10 mm. They have measured the chemical absorption rate of oxygen to determine the volumetric mass transfer coefficient.

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The objectives of this work are to determine the influence of pressure on volumetric mass transfer coefficient in the bubble column with the single-nozzle gas dispersers of the diameter, 1.4–4 mm with variation of liquid phase.

2. EXPERIMENTAL

2.1. Experimental apparatus

A schematic diagram of the experimental apparatus is shown in Fig. 1. The bubble column is a stainless-steel tube with inner diameter of 45 mm and has a cone-shaped bottom. The height of the multiphase contacting section was varied from 0.9 to 1.2 m. The gas was sparged into the column through a single nozzle with inner diameter of 1.38, 2.10, 2.90, and 4.03 mm. The top surface of the single nozzle was at the lowest level in the cone-shaped bottom. The volume of gas chamber under the single nozzle was about 35 cm³. The mixture gas of N₂ and O₂ was used as gas phase and the flow rate was measured by an orifice flow meter after reducing the pressure to atmospheric pressure. As the liquid phase, tap water, aqueous buffered solution (pH = 5.5, 0.2 mol dm⁻³ NaH₂PO₄–0.1 mol dm⁻³ citric acid solution), and aqueous solution of enzyme (glucose oxidase, Merk 24586; Catalase, Merk 5186) were used. The pH of the enzyme solution was adjusted to 5.5 by using the buffered solution, and the concentration of glucose was in the range 24⁻³ (3)–1.3 × 10³ U dm⁻³ (163 mg dm⁻³). The concentration of catalase was made to be 1000 times of the concentration of glucose oxidase.

2.2. Method of measurement

(a) *Gas holdup.* The gas holdup was obtained by measuring the height of the multiphase section expanded by gas sparging and the clear liquid height without gas sparging. These heights were measured by a sensing device for the liquid level, made of

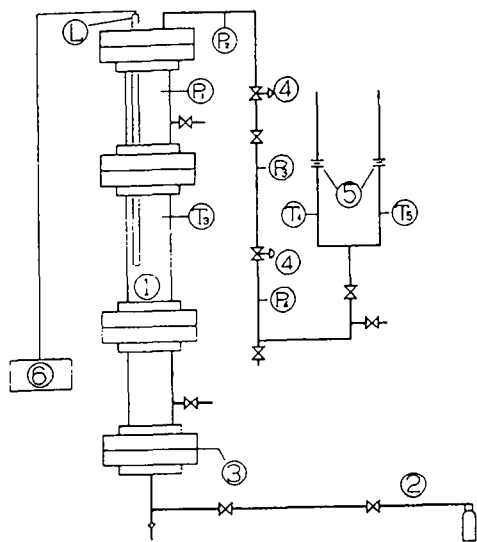


Fig. 1. Schematic diagram of experimental apparatus.

a stainless-steel pipe (4 mm in outer diameter) having 40 point-electrodes at interval between 1 and 2 cm.

(b) *Volumetric mass transfer coefficient.* After the liquid in the column was sparged with a sufficient amount of nitrogen from a cylinder until the dissolved oxygen concentration in the liquid became negligible, N₂-O₂ mixture gas with a certain composition was supplied to the column. The increasing concentration of dissolved oxygen with time was measured by the oxygen electrode (Oxi-96WTW). The electrode can be available under pressures up to 1.1 MPa.

As well known, the overall mass transfer coefficient, K , is the function of mass transfer coefficients in the gas and liquid phase, k_G , k_L , respectively, and Henry's constant, H , as shown in eq. (1):

$$1/Ka = 1/(Hk_Ga) + 1/(k_La). \quad (1)$$

Volumetric mass transfer coefficient in gas phase can be estimated by the equation of Cho and Wakao (1988). It was found that the estimated values of the volumetric mass-transfer coefficient were 6–24 times larger than those in the liquid phase. Henry's constants, H , at 25°C for the oxygen–water system is 32 kmol m⁻³ gas/(kmol m⁻³ liquid). Hence, the overall mass transfer coefficient becomes nearly equal to the liquid-phase mass transfer coefficient.

Nakonoh and Yoshida (1980) have presented the following equation that gives the relationship between the electrode signal, S_E and time, t , incorporating the time constant of the oxygen electrode, T_E .

$$\ln(S^* - S_E) = \ln\{\exp(-k_Lat) - k_LaT_E \exp(-t/T_E)\} + \ln\{S^*/(1 - k_LaT_E)\}. \quad (2)$$

The time constant, T_E of the oxygen electrode used in this work was determined experimentally. Its values were from 4 to 6 s. If the following condition, eq. (3) is fulfilled, eq. (2) is reduced to eq. (4).

$$t(1/T_E - k_La) > \ln(k_LaT_E) \quad (3)$$

$$\ln(S^* - S_E) = -k_Lat + \text{const.} \quad (4)$$

Under the experimental conditions of this work, eq. (3) is satisfied if time, t is greater than about 1 s. Then volumetric mass transfer coefficient with respect to the clear liquid volume can be obtained by fitting the experimentally observed electrode signal, S_E to eq. (4)

2.3. Experimental condition

The range of experimental condition is summarized in Table 1.

3. EXPERIMENTAL RESULTS AND THE CORRELATION

3.1. Gas holdup

Gas holdup data for the case where tap water was used as the liquid phase and a single nozzle with diameter of 1.38, 2.1, 2.9, 4.03 mm was used as the gas disperser are shown in Fig. 2. As shown in this figure, gas holdup in the nozzle column with a single nozzle with inner diameter of 4.03 mm under atmospheric

Table 1. Experimental condition

Column diameter (D_T): 45×10^{-3} m
Height of dispersion: 0.9–1.2 m
Gas distributor: Single nozzle with diameter of 1.38×10^{-3} , 2.1×10^{-3} , 2.9×10^{-3} , 4.03×10^{-3} m
Gas phase: N_2 , N_2 - O_2 mixed gas (mole fraction of oxygen: 0.03–0.97)
Liquid phase: Tap water, Buffered solution (pH = 5.5 at 25°C 0.2 mol dm^{-3} $\text{Na}_2 \text{HPO}_4$ - 0.1 mol dm^{-3} citric acid), buffered solution of enzyme (Glucose oxidase Merk 24586; Catalase Merk 5186)
Concentration of enzymes:
Glucose oxidase (C_E): 23.9, 662, $1.3 \times 10^3 \text{ U dm}^{-3}$ (2.99, 82.8, 163 mg dm^{-3})
Catalase: 2.4×10^4 , 6.6×10^5 , $1.3 \times 10^6 \text{ U dm}^{-3}$
Superficial gas velocity (v_G): 0.005 – 0.15 m s^{-1}
Operating pressure (P): 0.1–1.1 MPa
Temperature in the column: 17–27 $^\circ\text{C}$

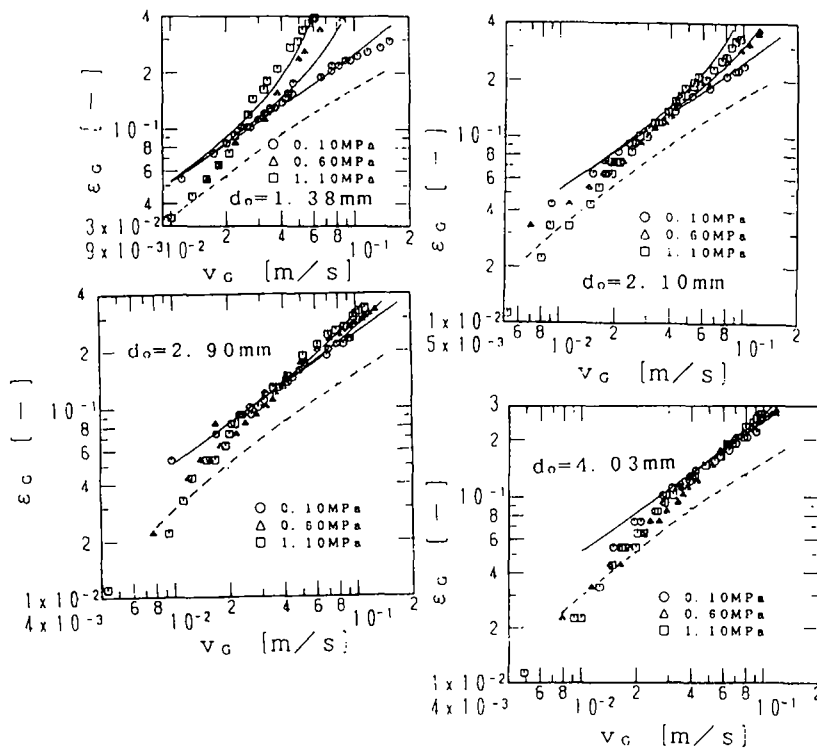


Fig. 2. Observed effect of pressure on gas holdup with variation of the diameter of single nozzle and the comparison with the correlation (N_2 -tap water system). (—) calculated by eq. (6); (---) predicted by the equation of Akita-Yoshida for gas holdup.

condition agreed approximately with the predicted values by the equation of Akita and Yoshida (1973). Gas holdup tends to increase with pressure and the tendency becomes more significant with decrease of the diameter of single-nozzle gas disperser.

Figure 3 shows the observed gas holdup for aqueous solution of enzyme with concentration of $163 \text{ mg (glucose oxidase) dm}^{-3}$. As shown in this figure, the increase of gas holdup with pressure in the enzyme solution is more significant than that in tap water.

The conceivable factors affecting the gas holdup under the condition in the present work seem to be

the operating pressure, P , the diameter of single nozzle, d_o , interfacial surface tension, γ , and superficial gas velocity based on the operating pressure, v_G . The diameter of the bubble formed at the single nozzle may be a function of modified Weber number, $\rho Q^2 d_o^3 \gamma^{-1}$ and the operating pressure, P . The function of these factors was assumed to be

$$v_G \varepsilon_{G_0} = \exp\{A(\rho Q^2 d_o^3 \gamma^{-1})(P/P_0)^B\}$$

where Q and P_0 refer to volumetric flow rate of gas and atmospheric pressure, respectively, and A and B are constants to be determined experimentally. Gas holdup at atmospheric pressure was correlated by the

following equation in the region where the values of gas holdup data were larger than 0.05.

$$\varepsilon_{G_0} = 1.18v_G^{0.679}(\gamma/\gamma_0)^{-0.546} \quad (5)$$

Equation (6) was obtained for the empirical correlation of gas holdup for the individual liquid phase used in this work. The constants, A and B , determined are

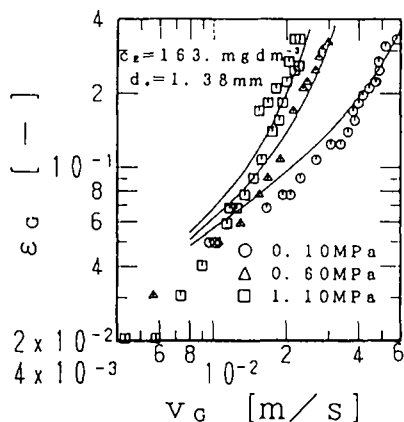


Fig. 3. Observed effect of pressure on gas holdup for the system N_2 -enzyme solution and the comparison with the correlation.

listed in Table 2. As shown in this table, the coefficients, A and B depend on the concentration of enzyme.

$$\varepsilon_G = 1.18v_G^{0.679}(\gamma/\gamma_0)^{-0.546} \times \exp\{A(\rho Q^2 d_s^{-3} \gamma^{-1})(P/P_0)^B\} \quad (6)$$

γ_0 in eq. (6) is surface tension of water at 20°C. Physical properties of the liquid phase under the present experimental condition are listed in Table 3. Solid lines in Figs (2) and (3) represent that calculated by eq. (6).

3.2. Volumetric mass transfer coefficient

Volumetric mass transfer coefficient with respect to the clear liquid volume, $k_L a$, also increased with pressure showing the same tendency as the effect of pressure on gas holdup. Figures 4 and 5 show examples of the observed volumetric mass transfer coefficient for water and enzyme solution, respectively, using a single nozzle with diameter of 1.38×10^{-3} m as the gas disperser. The increasing rate of volumetric mass transfer coefficient due to pressure increase seems to be larger than that of gas holdup as shown in these figures.

The data of $k_L a$ were correlated by eq. (7), where C , D , E and F are constant to be determined

Table 2. Constants in eq. (6)

	Water	Buffered solution of Enzymes (glucose oxidase concentration, C_E (mg dm^{-3}))		
		2.99	82.8	163
A	1.27×10^{-4}	1.27×10^{-4}	4.96×10^{-3}	1.07×10^{-2}
B	1.00	1.31	0.336	0.0

Table 3. Physical properties at 25°C

	Water	Buffered solution of Enzymes (glucose oxidase concentration, C_E (mg dm^{-3}))		
		2.99	82.8	163
Density ρ (kg m^{-3})	1000	1015	1020	1025
Viscosity μ (Pa s^{-1}) $\times 10^3$	0.890	1.029	1.051	1.075
Surface tension γ (N m^{-1}) $\times 10^3$	71.96	68.1	64.74	63.36

Table 4. Constants in eq. (7)

	Water	Buffered solution of Enzymes (glucose oxidase concentration, C_E (mg dm^{-3}))		
		2.99	82.8	163
C	7.33×10^{-3}	0.303	0.175	0.151
D	0.702	1.06	0.871	0.862
E	0.122	-0.0116	-0.0496	-0.106
F	0.151	0.155	0.0339	0.0705

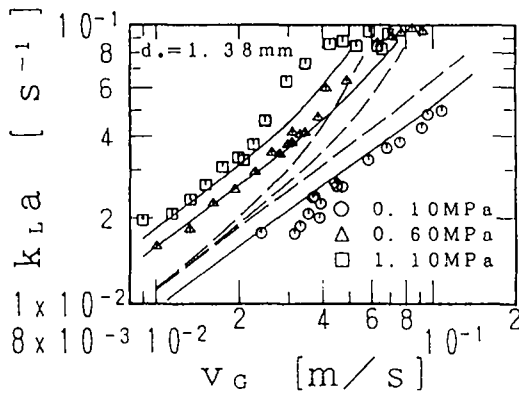


Fig. 4. Experimental result on volumetric mass transfer coefficient for the system N_2 - O_2 mixture gas-tap water: (—) calculated by eq. (7); (· · ·) calculated by the equation of Akita-Yoshida for volumetric mass transfer coefficient using gas holdup data in this work.

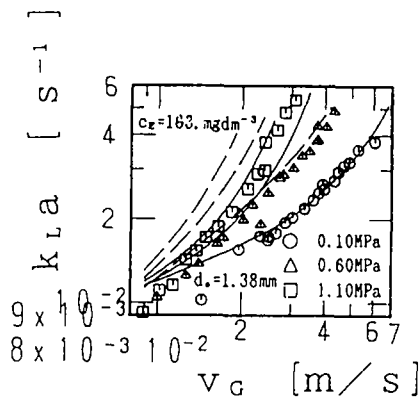


Fig. 5. Experimental result on volumetric mass transfer coefficient for the system N_2 - O_2 mixture gas-enzyme solution.

experimentally.

$$k_L a = C v_G^D (\rho Q^2 d_n^{-3} \gamma^{-1})^E (P/P_0)^F \quad (7)$$

The determined values of C , D , E and F are listed in Table 4. The experimentally determined coefficients, C , D , E , F in eq. (7) depend on the concentration of enzyme as in the case of gas holdup. This may lead to the fact that our results on gas holdup and volumetric mass transfer coefficients cannot be fully explained by the variation of surface tension in the liquid phase. The same tendency has been observed in break-up and coalescence of bubbles in electrolytes solutions (Prince and Blanch, 1990; Tarmy and Coulaloglou, 1992).

Solid lines in Figs 4 and 5 are the values calculated by eq. (7). Dotted lines in Figs 4 and 5 show the values of $k_L a$ predicted by the equation of Akita and Yoshida (1973), using the observed gas holdup in this work. Dotted lines show the same tendency on the effect of pressure as the experimental values of $k_L a$, but the difference between them cannot be negligible.

4. CONCLUSIONS

Gas holdup and volumetric mass transfer coefficient ($k_L a$) increased with pressure. The effect of pressure on gas holdup and $k_L a$ became significant in relatively higher superficial gas velocity in the case where a single nozzle with relatively smaller diameter was used as gas disperser. In the case where the solution of enzyme was used as the liquid phase, the effect of pressure appeared more significant than in the case where tap water was used.

The empirical correlations were obtained for gas holdup and volumetric mass transfer coefficient with respect to clear liquid volume, $k_L a$, respectively.

NOTATION

- a gas-liquid interfacial area per unit volume of liquid phase, $m^2 m^{-3}$
 C_E enzyme concentration in the liquid phase, $mg dm^{-3}$
 d_n inner diameter of single nozzle, mm
 K over-all mass transfer coefficient, $m s^{-1}$
 k_L liquid side mass transfer coefficient, $m s^{-1}$
 k_G gas side mass transfer coefficient, $m s^{-1}$
 P pressure in the bubble column, Pa
 P_0 standard atmospheric pressure, 1.013×10^5 Pa
 Q volumetric flow rate of gas under the condition in the bubble column, $m^3 s^{-1}$
 S_E signal intensity of oxygen electrode, mV
 S^* signal intensity of oxygen electrode corresponding to saturated concentration, mV
 T_E time constant of oxygen electrode, s
 v_G superficial gas velocity under the condition in the bubble column, $m s^{-1}$

Greek letters

- γ surface tension, $N m^{-1}$
 γ_0 surface tension of water at 20°C, $0.076 N m^{-1}$
 c_G gas holdup
 $c_{G,0}$ gas holdup under atmospheric condition
 ρ liquid density, $kg m^{-3}$

REFERENCES

- Akita, K. and Yoshida, F. (1973) Gas holdup and volumetric mass transfer coefficient in bubble column. *Ind. Engng Chem. Process Des. Dev.* **12**, 76-81.
 Deckwer, W.-D., Louisi, Y., Zaidi, A. and Ralek, M. (1980) Hydrodynamic properties of the Fischer-Tropsch slurry process. *Ind. Engng Process Des. Dev.* **19**, 699-708.
 Idogawa, K., Ikeda, K., Fukuda, T. and Morooka, S. (1985) Effect of gas and liquid properties on the behavior of bubbles in a bubble column under high pressure. *Kagaku Kagaku Ronbunshu* **11**, 432-437.
 Kling, G. (1962) Über die Dynamik der Blasenbildung beim Begasen von Flüssigkeiten unter Druck. *Int. J. Heat Mass Transfer* **5**, 211-223.
 Kojima, H., Okumura, B. and Nakamura, A. (1991) Effect of pressure on gas holdup in a bubble column and a slurry bubble column. *J. Chem. Engng Japan* **24**, 115-117.

- Kölbel, H., Borchers, E. and Langemann, H. (1961) Grossenverteilung der Gasblasen in Blasensäulen. *Chemie-Ing. Technol.* **33**, 668-675.
- LaNauze, R. D. and Harris, I. J. (1974) Gas bubble formation at elevated system pressures. *Trans. Instn Chem. Engrs* **52**, 337-348.
- Prince, M. J. and Blanch, H. W. (1990) Bubble coalescence and break-up in air-sparged bubble columns. *A.I.Ch.E. J.* **36**, 1485-1499.
- Sagert, N. H. and Quinn, M. J. (1976) The coalescence of H₂S and CO₂ bubbles in water. *Can. J. Chem. Engng* **54**, 392-398.
- Tarmy, B. L., Chang, M., Coulaloglou, C. A. and Ponzi, P. R. (1984) The three phase hydrodynamics of the EDS coal liquefaction reactors: their development and use in reactor scaleup. *I.E.C. Symp. Ser.* No. 87, 303-317.
- Tarmy, B. L. and Coulaloglou, C. A. (1992) Alpha-Omega and beyond industrial view of gas/liquid/solid reactor development. *Chem. Engng Sci.* **47**, 3231-3246.
- Tsuge, H. and Hibino, S. (1980) Theoretical approach for the formation of gas bubble under the elevated system pressure. G116, Preprint of 14th Autumn Meeting of the Society of Chemical Engineers, Japan.
- Wilkinson, P. M., Spak, A. P. and van Dierendonck, L. L. (1992) Design parameters estimation for scale-up of high-pressure bubble columns. *A.I.Ch.E. J.* **38**, 544-554.
- Wilkinson, P. M. and Haringa, H. (1994) Mass transfer and bubble size in a bubble column under pressure. *Chem. Engng Sci.* **49**, 1417-1427.
- Yamaguchi, H., Ogawa, T. and Yokoyama, T. (1991) Gas holdup in a high pressure and temperature reactor of 2.4 ton/day direct coal liquefaction process. *Nenryou-Kyokai-si* **70**, 1143-1150.