

Acknowledgment

The authors express their appreciation for the financial support given by the Asahi Glass Foundation for Industrial Technology and to N. Hamada for his careful efforts in the measurements.

Nomenclature

a	= activity	[—]
f	= fugacity	[MPa]
H	= enthalpy	[kJ·mol ⁻¹]
P	= pressure	[MPa]
R	= gas constant	[J·K ⁻¹ ·mol ⁻¹]
T	= temperature	[K]
V	= volume	[m ³]
Z	= compressibility factor	[—]

Literature Cited

- 1) Barduhn, A. J., H. E. Towson and Y. C. Hu: *A.I.Ch.E. Journal*, **8**, 176 (1962).
- 2) Claussen, W. F.: *J. Chem. Phys.*, **19**, 259, 1425 (1951).
- 3) Dymond, J. H. and E. B. Smith: "The Virial Coefficients of

- Pure Gases and Mixtures," Clarendon Press, Oxford, England (1980).
- 4) Frost, E. M., Jr. and W. M. Deaton: *The Oil and Gas Journal*, **45**, 170 (1946).
 - 5) Miller, B. and E. R. Strong: *Am. Gas Ass'n. Monthly*, **28**, 63 (1946).
 - 6) Piroen, A. P.: *Rec. Trav. Chem.*, **74**, 995 (1955).
 - 7) Reamer, H., F. T. Selleck and B. H. Sage: *Petroleum Transactions, AIME*, **195**, 197 (1952).
 - 8) Von Stackelberg, M.: *Naturwiss.*, **36**, 327, 359 (1949).
 - 9) Von Stackelberg, M. and H. R. Müller: *Z. Elektrochem.*, **58**, 25 (1954).
 - 10) Weast, R. C. (Ed.): "Handbook of Chemistry and Physics," 56th ed., CRC Press Inc., Ohio, U.S.A. (1976).
 - 11) Wilcox, W. I., D. B. Carson and D. L. Katz: *Ind. Eng. Chem.*, **33**, 662 (1941).

(A part of this paper was presented at the 15th Autumn Meeting of The Society of Chemical Engineers, Japan, at Kanazawa, October 15, 1981 and at the 23rd High Pressure Conference of Japan at Kyoto, November 18, 1982.)

LONGITUDINAL CONCENTRATION DISTRIBUTION OF DROPLETS IN MULTI-STAGE BUBBLE COLUMNS FOR GAS-LIQUID-LIQUID SYSTEMS

YASUO KATO, TOKIHIRO KAGO AND SHIGEHARU MOROOKA

Department of Applied Chemistry, Kyushu University, Fukuoka 812

Key Words: Chemical Reactor, Three Phase, Bubble Column, Droplet, Holdup, Dispersion, Slip Velocity, Multiphase Flow

The longitudinal concentration distribution of droplets dispersed in multi-stage bubble columns was analyzed by the one-dimensional dispersion model with an effective slip velocity of droplets. The variables of the model—mean gas holdup, longitudinal dispersion coefficient of droplet phase and droplet concentration at the top of the column in 4- and 8-stage bubble columns of 6.6 and 12.2 cm i.d.—were measured and correlated. The effective slip velocity was determined by comparing observed mean droplet concentrations with theoretical ones and was correlated in an experimental equation. The concentration distributions of droplets calculated with the above parameters are in good agreement with the observed ones. Mean droplet diameter was also correlated as a function of gas velocity, total liquid velocity and free area of horizontal baffle plates.

Introduction

A bubble column containing a dispersed organic liquid phase is useful as an efficient contactor for gas-liquid-liquid systems (G-L-L systems). The droplet phase may be a liquid catalyst, an extracting solvent, and so on. Though many studies have been reported on the droplet size in agitated vessels,^{1,5)} information on the droplets in bubble columns is quite

limited.^{2,3,17)}

Yoshida and Yamada¹⁷⁾ measured the average diameter of kerosene droplets dispersed in bubble columns which were operated batchwise with respect to liquid. Hatate *et al.*²⁾ measured gas holdup, longitudinal dispersion coefficient of droplet phase, effective slip velocity between droplet and continuous liquid, and longitudinal concentration distribution of droplets in bubble columns. Hatate *et al.*³⁾ also correlated the average diameter of droplets as a function of gas velocity, interfacial tension and bubble column

Received December 1, 1983. Correspondence concerning this article should be addressed to S. Morooka.

diameter. All these works were carried out by using single-stage bubble columns where the degree of longitudinal liquid-phase dispersion was essentially large.⁴⁾ The back-mixing of liquid can be reduced by dividing the bubble column with horizontal baffle plates.¹³⁾ However, we have no information on the behavior of droplets in multi-stage bubble columns to date.

In the present paper, a model for the longitudinal concentration distribution of droplets is newly developed by modifying the model for the longitudinal concentration distribution of solid particles suspended in multi-stage columns for gas-liquid-solid systems (G-L-S systems).⁷⁾ The hydrodynamic properties such as mean gas holdup, longitudinal dispersion coefficient of droplet phase, effective slip velocity of droplets with respect to total liquid flow, droplet concentration at the outlet and mean droplet diameter are measured and discussed.

1. Mathematical Model

Figure 1 shows a model for longitudinal concentration distribution of droplets dispersed in a multi-stage bubble column. Two immiscible liquids for droplet and continuous phase are introduced at the bottom of the column and allowed to flow upward cocurrently with gas bubbles and to overflow from the top of the column. The column is partitioned into N equal stages of height L/N . To simplify the equation, the following assumptions are made.

(1) Concentration of droplets is maintained at low levels.

(2) Concentration of droplets is laterally uniform.

(3) Gas holdup is uniform longitudinally and laterally.

(4) Time-averaged motion of droplets in each stage can be expressed by the one-dimensional dispersion model with an effective slip velocity of droplets.^{1,2,6-8,10,12,14)} The values of the dispersion coefficient of droplet phase, E_p , and the effective slip velocity of droplet in each stage, v_p , are longitudinally uniform.

(5) The intermixing between adjacent stages is brought on by the back-flow velocity of liquid, u_B , through baffle plates.

The mass balance of suspended droplets in the i -th stage is expressed as follows.

$$(i-1)L/N < z < iL/N;$$

$$E_p \frac{d^2 C_i}{dz^2} - (v_p + u_t) \frac{dC_i}{dz} = 0 \quad (1)$$

where $i=1, 2, 3, \dots, N$. The concentration of droplets, C_i , is defined as the volume fraction of droplets per unit volume of liquid-liquid mixtures, and u_t is the total velocity of liquid-liquid mixtures. The values of

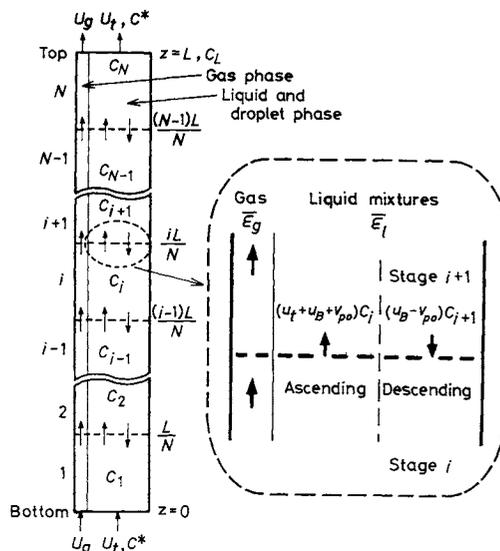


Fig. 1. Flow model of multi-stage bubble column.

E_p and u_t are based on the volume fraction of liquid-liquid mixtures, $\bar{\epsilon}_l$ or $(1 - \bar{\epsilon}_g)$. The effective slip velocity is a parameter which expresses relative motions of droplets with respect to u_t . The effect of droplet concentration on v_p must be studied experimentally.

As shown in Fig. 1, the mass balance of droplets crossing the i -th baffle plate can be given as follows.

$$(u_t + u_B + v_{po})C_i - (u_B - v_{po})C_{i+1} - u_t C^* = 0 \quad (2)$$

where $i=1, 2, 3, \dots, N-1$. v_{po} is the effective slip velocity of droplet in the holes of baffle plates and is expressed as

$$v_{po} = \gamma A_r v_p \quad (3)$$

where γ is the correction factor for the flow area of liquid mixture in the holes of baffle plates.

The other boundary condition is the mass balance of droplets at $z=0$ and L .

$$\left. \begin{aligned} z=0: & E_p \frac{dC_1}{dz} - (v_p + u_t)C_1 + u_t C^* = 0 \\ z=L: & E_p \frac{dC_N}{dz} - (v_p + u_t)C_N + u_t C^* = 0 \end{aligned} \right\} \quad (4)$$

The concentration of droplets at the top of the column, $C_L (= C_N |_{z=L})$, is not necessarily equal to the feed or the effluent concentration, C^* . Equations (1), (2) and (4) hold approximately in the range of $C^* \leq 0.3$ as shown in the following experiments.

By solving Eq. (1) under the boundary conditions of Eqs. (2) and (4), the longitudinal concentration distribution of droplets is obtained as follows.

$$C_{i,z} = B_i \exp\left(\frac{v_p + u_t}{E_p} z\right) + \frac{u_t}{v_p + u_t} C^* \quad (5)$$

where $i=1, 2, 3, \dots, N$, and $(i-1)L/N < z < iL/N$. The parameter, B_i , in Eq. (5) is given by

$$\left. \begin{aligned}
 B_i &= B_N a^{i-N} + \frac{b e^{\alpha N}}{e^{\alpha} - a} (e^{\alpha(i-N)} - a^{i-N}) \\
 B_N &= \left(C_L - \frac{u_t}{v_p + u_t} C^* \right) \exp \left(-\frac{v_p + u_t}{E_p} L \right) \\
 \alpha &= -\frac{v_p + u_t}{E_p} \frac{L}{N} \\
 a &= \frac{u_t + u_B + v_{p0}}{u_B - v_{p0}} \\
 b &= -\frac{v_p - 2v_{p0}}{u_B - v_{p0}} \frac{u_t}{v_p + u_t} C^*
 \end{aligned} \right\} \quad (6)$$

The mean droplet concentration in the column can be obtained from Eqs. (5) and (6).

$$\bar{C} = -\frac{E_p (e^{\alpha} - 1)}{(v_p + u_t) L} \sum_{i=1}^N B_i e^{-\alpha i} + \frac{u_t}{v_p + u_t} C^* \quad (7)$$

Equations (5)–(7) are analogous to those of Kato *et al.*,⁷⁾ who analyzed the concentration distribution of solid particles in bubble columns for G-L-S systems. The sign of v_p and v_{p0} in Eqs. (5)–(7) is, however, different from that of the equations of Kato *et al.*⁷⁾ To calculate the concentration distribution of droplets from Eqs. (5) and (6), the values of C_L , $\bar{\epsilon}_g$, E_p , u_B , v_p and v_{p0} are needed.

2. Experimental Apparatus and Procedure

2.1 Experimental apparatus

A schematic diagram of the experimental apparatus is shown in Fig. 2. Bubble columns of 6.6 and 12.2 cm i.d. were made of transparent acrylic resin. The columns were 200 to 220 cm in height and were divided into 1, 4 and 8 stages. The horizontal baffles were 1.0 mm-thick perforated brass plate with holes drilled in an equilateral triangular arrangement. The dimensions of the bubble columns and the baffle plates are summarized in Table 1.

The gas distributors of both the columns were 3.0 mm-thick acrylic plates with 2-mm holes in an equilateral triangular arrangement. The number of holes was 7 and 19 for the 6.6 and 12.2 cm i.d. column, respectively. To determine the longitudinal concentration distribution of droplets, sixteen sampling taps were installed on the column wall at different heights.

Air, tap water and kerosene were used as gas, continuous liquid and dispersed liquid, respectively. Density, interfacial tension and viscosity of kerosene were $800 \text{ kg} \cdot \text{m}^{-3}$, $33 \pm 3 \text{ mN} \cdot \text{m}^{-1}$ and $1.6 \text{ mPa} \cdot \text{s}$, respectively. Water and kerosene were fed into the column independently through a single pipe just above the gas distributor. The diameter of the kerosene feed pipe was 0.8 and 0.7 cm for the 6.6 and 12.2 cm i.d. column, respectively. For the single-stage

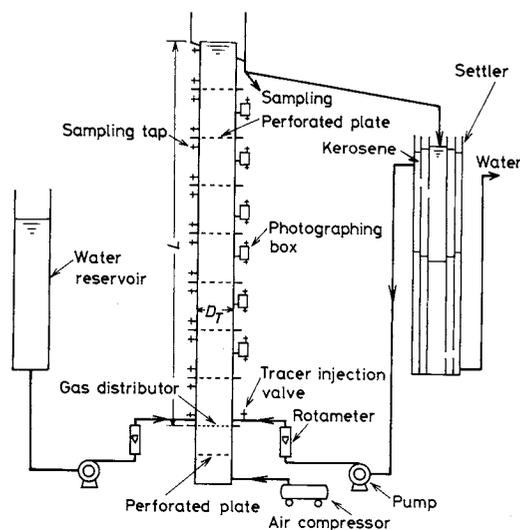


Fig. 2. Experimental apparatus.

Table 1. Dimensions of bubble column and horizontal baffle plate

D_T [cm]	N	AL [cm]	D_p [cm]	Number of holes	A_r
6.6	1	210–220	—	—	1.0
	4	50	0.65	7	0.068
	4	50	0.90	7	0.130
	4	50	1.20	7	0.231
	8	25	0.65	7	0.068
	8	25	0.90	7	0.130
	8	25	1.20	7	0.231
12.2	1	200–220	—	—	1.0
	4	50	1.0	9	0.0605
	4	50	1.0	19	0.128
	4	50	1.0	43	0.289
	8	25	1.0	9	0.0605
	8	25	1.0	19	0.128
	8	25	1.0	43	0.289

column of 12.2 cm i.d., kerosene was also fed through a 0.4 and 1.4 cm i.d. pipe. The experiments were carried out mainly under the following conditions.

$$U_g = 1.5\text{--}13 \text{ cm} \cdot \text{s}^{-1}$$

$$U_t = 0.1\text{--}1.0 \text{ cm} \cdot \text{s}^{-1}$$

$$T = 287\text{--}298 \text{ K}$$

$$C^* = 0.05\text{--}0.3$$

For measurement of the mean gas holdup only, the range of C^* was 0.05–0.5.

2.2 Experimental procedure

Droplet concentration and mean holdups After steady state was attained, samples of liquid mixture were taken from the sampling taps at the column wall and from the effluent stream. The droplet concentration was determined from the settled volume of kerosene in each sample. The value of C_L was ob-

tained by extrapolating the observed droplet concentration curve to $z=L$. At the end of each run the feeds of gas, water and kerosene were stopped simultaneously, and the mean holdups of gas and continuous liquid and the mean droplet concentration were determined from the settled volume of each phase.

Longitudinal dispersion coefficient The residence time distribution curve of the droplet phase in single-stage bubble columns was measured by the impulse response method.²⁾ The tracer was a kerosene solution of an oleophilic dye (Fat orange R, Hoechst). The longitudinal dispersion coefficient of the droplet phase, E_p , was calculated by applying the one-dimensional dispersion model.^{2,9,16)}

Mean droplet diameter The droplet diameter was determined by the photographic method. In the range of higher gas velocities, gas-liquid mixtures in the column were very cloudy, and there was no distinction between small gas bubbles and droplets. Therefore, small rectangular boxes (4 cm wide, 2 cm thick and 6 cm high) were connected to the column, and gas-liquid mixtures were quickly introduced into the box by opening a release valve. Very shortly after the valve was closed again, gas bubbles in the box went upward. A photograph of droplets was taken at that moment. The mean droplet diameter was calculated from the following equation.

$$\bar{d}_p = \left(\frac{\sum_i n_i d_{pi}^3}{\sum_i n_i} \right)^{1/3} \quad (8)$$

Equation (8) was adopted because the droplet concentration was closely related to the volume of each droplet.

3. Results and Discussion

3.1 Evaluation of variables

Mean gas holdup Figure 3 shows the mean holdup in multi-stage bubble columns. The value of $\bar{\epsilon}_g$ was nearly independent of the free area of baffle plates, the number of stages and the column diameter, and was correlated as follows.

$$\bar{\epsilon}_g = U_g / (30 + 3.3 U_g^{0.8}) \quad (9)$$

where U_g is expressed in the unit of $\text{cm} \cdot \text{s}^{-1}$. Kato *et al.*⁶⁾ and Nakamura *et al.*¹⁰⁾ found that the gas holdup in bubble columns with suspended solid particles (G-L-S systems) was smaller than that in gas-liquid systems (G-L systems). Their results are in agreement with the present ones.

Longitudinal dispersion coefficient Figure 4 shows the longitudinal dispersion coefficient of droplet phase in single-stage bubble columns. The value of E_p increased with increasing gas velocity and column diameter, and was independent of total liquid velocity in the range of $0.05\text{--}1.0 \text{ cm} \cdot \text{s}^{-1}$. The data of E_p were

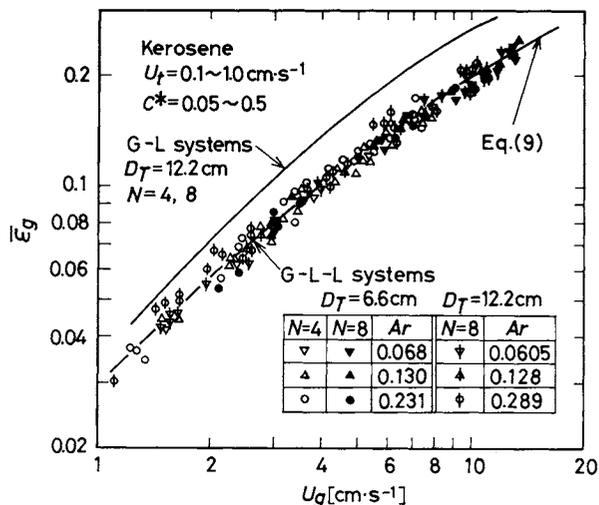


Fig. 3. Mean gas holdup in multi-stage bubble columns.

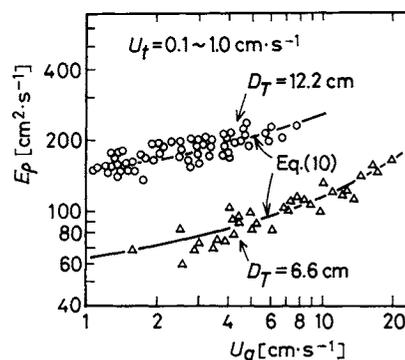


Fig. 4. Longitudinal dispersion coefficient of droplet phase in single-stage bubble columns.

correlated as follows.

$$U_g D_T / E_p = 10 (U_g / \sqrt{g D_T}) / \{1 + 6.5 (U_g / \sqrt{g D_T})^{0.8}\} \quad (10)$$

Equation (10) was derived analogously to the correlation for the longitudinal dispersion coefficient of continuous liquid for G-L systems.⁵⁾ The longitudinal dispersion coefficient of droplet phase in each stage of the multi-stage column was assumed to be equal to E_p in the single-stage column.

Droplet concentration at outlet As shown in Figure 5, the concentration ratio at the outlet, C_L/C^* , was equal to unity.

$$C_L/C^* = 1 \pm 0.15 \quad (11)$$

This equation holds good also for single-stage columns.

Mean droplet diameter Figure 6 shows examples of the droplet diameter distribution measured at fixed heights. Figure 7 shows the longitudinal distribution of \bar{d}_p in the eight-stage bubble column. The mean droplet diameter became smaller at higher axial levels, and was affected by U_g , U_t and A_r . However, \bar{d}_p was not much influenced by the number of stages or the

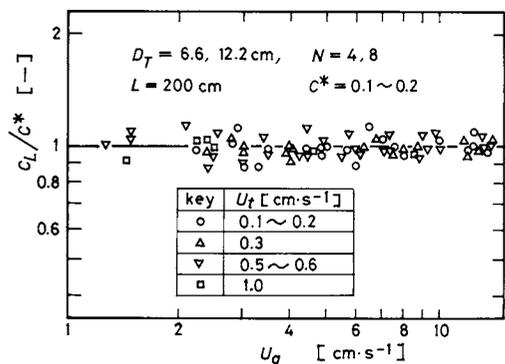


Fig. 5. Droplet concentration at outlet.

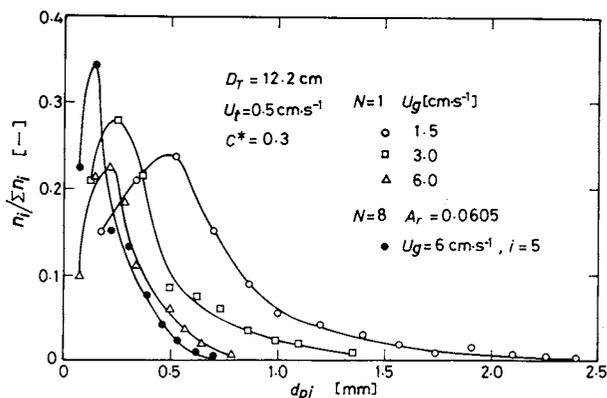


Fig. 6. Example data of droplet size distribution.

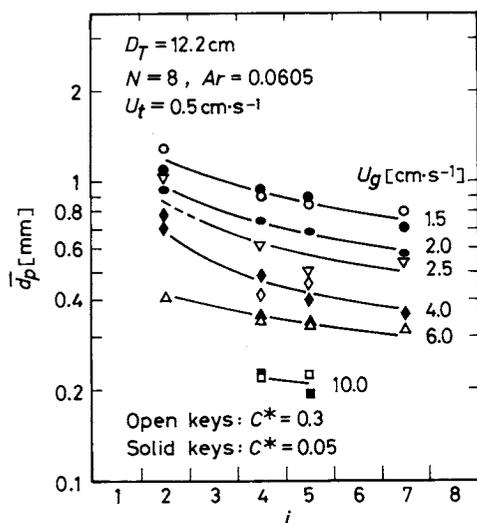


Fig. 7. Longitudinal distribution of mean droplet diameter in eight-stage bubble column of 12.2 cm i.d.

droplet concentration. When kerosene was dispersed in the 12.2 cm i.d. column, \bar{d}_p near the middle of the column was correlated by the following equation as shown in Fig. 8.

$$\bar{d}_p = 2.6 U_g^{-0.78} U_t^{0.33} A_r^{0.15} \quad (12)$$

where \bar{d}_p , U_g and U_t are expressed in mm, $\text{cm} \cdot \text{s}^{-1}$ and $\text{cm} \cdot \text{s}^{-1}$, respectively. The mean droplet diameter in

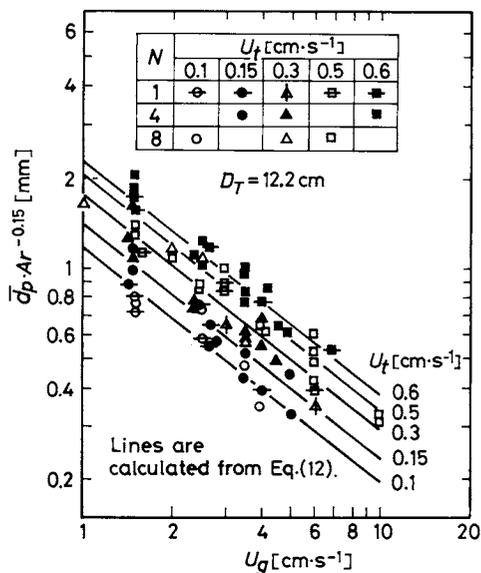


Fig. 8. Correlation of mean droplet diameter in 12.2 cm i.d. bubble column.

the multi-stage bubble columns of 6.6 cm i.d. was nearly expressed by Eq. (12). The effect of the kerosene feed pipe diameter on \bar{d}_p was determined under the conditions of $N=1$, $D_T=12.2$ cm, $U_g=3$ $\text{cm} \cdot \text{s}^{-1}$, $U_t=0.5$ $\text{cm} \cdot \text{s}^{-1}$ and $C^*=0.05$. The data for pipe diameters of 0.4, 0.7 and 1.4 cm coincided.

Back-flow velocity The back-flow velocity through baffle plates for G-L-L systems is approximated by the back-flow velocity for G-L systems, where the following correlations have been obtained over a wide range of experimental conditions.¹¹⁾

At lower gas velocities of 1.5–4.5 $\text{cm} \cdot \text{s}^{-1}$:

$$\begin{aligned} U_B/U_t &= 4.7 / [(U_t A_r^{-1.2}) \{1 + 0.055 (U_t A_r^{-1.2})^{1.6}\}] \\ &= \beta_L \end{aligned} \quad (13)$$

At higher gas velocities of 13–20 $\text{cm} \cdot \text{s}^{-1}$:

$$\begin{aligned} U_B/U_t &= 13 / [(U_t A_r^{-1.5}) \{1 + 0.011 (U_t A_r^{-1.5})^{1.5}\}] \\ &= \beta_H \end{aligned} \quad (14)$$

At intermediate gas velocities of 4.5–13 $\text{cm} \cdot \text{s}^{-1}$:

$$U_B/U_t = \beta_L (U_g/4.4)^{-2.17 \log_{10}(\beta_L/\beta_H)} \quad (15)$$

where U_g and U_t are expressed in $\text{cm} \cdot \text{s}^{-1}$. The back-flow velocity based on the liquid holdup is given by $U_B/\bar{\epsilon}_L$.

Effective slip velocity By assuming the value of γ in Eq. (3) and substituting measured values of \bar{C} and the other variables into Eq. (7), the only unknown variable, v_p , can be obtained. The value of v_p decreased with increase in droplet concentration. According to Kato *et al.*,⁷⁾ v_p for G-L-S systems is expressed by

$$v_p = k(1 - \bar{C})^{2.5} \quad (16)$$

The coefficient, k , is a function of single-particle

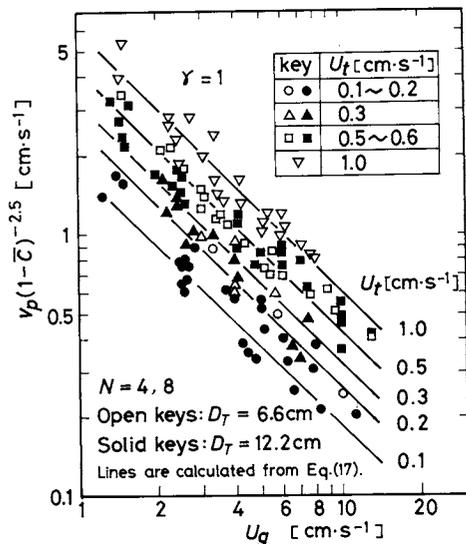


Fig. 9. Correlation of effective slip velocity in multi-stage bubble columns.

terminal velocity and gas velocity. We assume that Eq. (16) can be applied to the present experiments. In G-L-L systems, however, droplet diameter changes with gas velocity and other experimental conditions. Figure 9 shows that the values of $v_p(1-\bar{C})^{-2.5}$ in the multi-stage bubble columns decrease with increasing gas velocity and with decreasing total liquid velocity. No effect of A_r on v_p is observed in Fig. 9. With $\gamma=1$, the data were correlated as follows.

$$v_p = 6.0 U_g^{-1.0} U_t^{0.52} (1-\bar{C})^{2.5} \quad (17)$$

where v_p , U_g and U_t are expressed in $\text{cm}\cdot\text{s}^{-1}$.

3.2 Longitudinal concentration distribution of droplets

Figure 10a and 10b show example data of the longitudinal concentration distribution of droplets in the four-stage bubble columns. The solid lines are calculated ones from Eqs. (5) and (6) with the correlations of Eqs. (9)–(11), (13)–(15) and (17) and with $\gamma=1$. The broken lines indicate the calculated droplet concentration with $\gamma=0.7$. This value of $\gamma=0.7$ was deduced from the experiments of Kato *et al.*⁷⁾ for G-L-S systems. The effect of γ on the concentration of droplets was very small in the range of higher gas velocities.

Figure 11 shows sample data of the longitudinal concentration distribution of droplets in the eight-stage bubble columns. The calculated values of C agree with the observed ones within 10–20%.

Conclusion

The longitudinal concentration distribution of droplets in multi-stage bubble columns was analyzed on the basis of the one-dimensional dispersion model with an effective slip velocity of droplet with respect

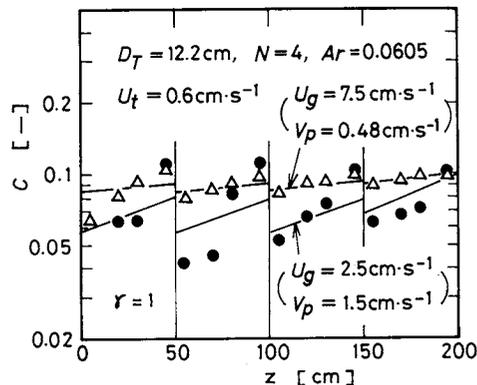


Fig. 10a. Examples of longitudinal concentration distribution of droplets in four-stage bubble column of 12.2 cm i.d. ($C^*=0.1$, $A_r=0.0605$).

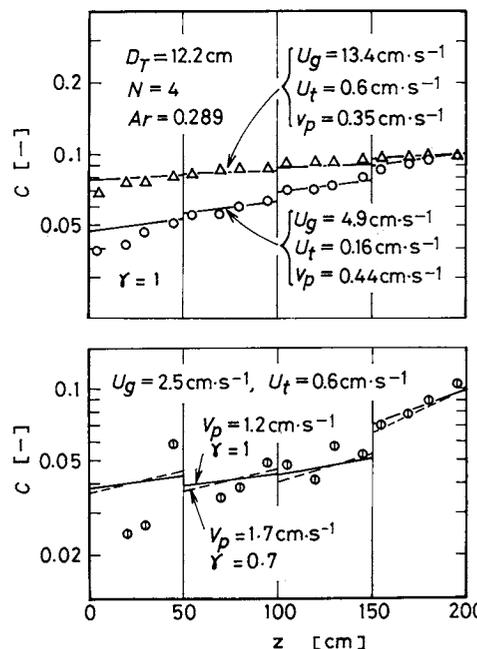


Fig. 10b. Examples of longitudinal concentration distribution of droplets in four-stage bubble column of 12.2 cm i.d. ($C^*=0.1$, $A_r=0.289$).

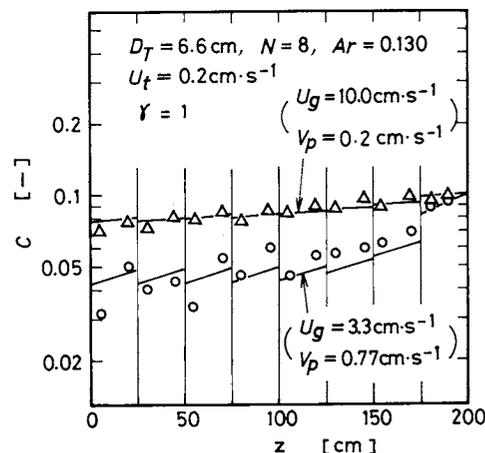


Fig. 11. Examples of longitudinal concentration distribution of droplets in eight-stage bubble column of 6.6 cm i.d. ($C^*=0.1$, $A_r=0.130$).

to u_t . The applicability of the model was confirmed for four- and eight-stage bubble columns of 6.6 and 12.2 cm i.d. The variables of the model were correlated as Eqs. (9)–(11), (13)–(15) and (17).

The mean droplet diameter was affected by various variables as expressed by Eq. (12). The dependence of \bar{d}_p on the physical properties of liquid remains to be studied in future work.

Acknowledgment

The authors are grateful to Mr. Kazuo Kai, Mr. Akira Ninomiya, Mr. Kan-ichiro Korematsu, Mr. Kenko Tsuji and Mr. Satoshi Ohshima for their experimental assistance. Useful discussions with Dr. Akio Nishiwaki are also acknowledged.

Nomenclature

A_r	= fractional free area of baffle plate	[—]
C	= droplet concentration defined by volume of droplet per unit volume of liquid-liquid mixture (a function of z)	[—]
\bar{C}	= mean droplet concentration	[—]
C^*	= droplet concentration in feed or effluent flow	[—]
C_L	= droplet concentration extrapolated to $z=L$	[—]
D_p	= hole diameter of baffle plate	[cm]
D_T	= diameter of bubble column	[cm]
\bar{d}_p	= mean droplet diameter defined by Eq. (8)	[mm]
d_{pi}	= diameter of i -th droplet	[mm]
E_p	= longitudinal dispersion coefficient of droplet phase	[cm ² ·s ⁻¹]
g	= gravitational acceleration	[cm·s ⁻²]
i	= number of stage counted from bottom	[—]
k	= coefficient in Eq. (16)	[cm·s ⁻¹]
L	= height of bubble column	[cm]
ΔL	= height of stage	[cm]
N	= total number of stage	[—]
n_i	= number of droplets of size d_{pi}	[—]
T	= temperature	[K]
U_B	= superficial velocity of back-flow through baffle plate	[cm·s ⁻¹]
U_g	= superficial velocity of gas	[cm·s ⁻¹]
U_t	= superficial total velocity of liquid mixture	[cm·s ⁻¹]
u_B	= $U_B/\bar{\epsilon}_t$, linear velocity of back-flow through baffle plates	[cm·s ⁻¹]
u_t	= $U_t/\bar{\epsilon}_t$, linear total velocity of liquid mixture	[cm·s ⁻¹]

v_p	= effective slip velocity of droplet with respect to u_t , defined by Eq. (1)	[cm·s ⁻¹]
v_{po}	= effective slip velocity of droplet with respect to u_t in holes of baffle plates	[cm·s ⁻¹]
z	= axial coordinate	[cm]
β	= U_B/U_t , back-flow velocity ratio	[—]
γ	= correction factor for flow area of liquid mixture in holes of baffle plates	[—]
$\bar{\epsilon}_g$	= mean gas holdup	[—]
$\bar{\epsilon}_l$	= mean holdup of liquid mixture	[—]

Literature Cited

- 1) Fukuda, T., S. Tanaka, H. Haneda and Y. Kato: *Kagaku Kogaku Ronbunshu*, **3**, 103 (1976).
- 2) Hatate, Y., S. Okuma and Y. Kato: *Kagaku Kogaku Ronbunshu*, **1**, 577 (1975).
- 3) Hatate, Y., S. Mori, S. Okuma and Y. Kato: *Kagaku Kogaku Ronbunshu*, **2**, 133 (1976).
- 4) Kato, Y. and A. Nishiwaki: *Kagaku Kōgaku*, **35**, 912 (1971).
- 5) Kato, Y. and A. Nishiwaki: *Kagaku Kōgaku*, **35**, 912 (1971); also *Int. Chem. Eng.*, **12**, 1 (1972).
- 6) Kato, Y., A. Nishiwaki, T. Fukuda and S. Tanaka: *J. Chem. Eng. Japan*, **5**, 112 (1972).
- 7) Kato, Y., A. Nishiwaki, S. Tanaka and T. Fukuda: *J. Chem. Eng. Japan*, **15**, 376 (1982).
- 8) Kubota, H. and T. Sekizawa: "Fluidization Technology," Keairns, D. L. (ed.), Vol. I, p. 431, Hemisphere Pub., Washington (1975).
- 9) Levenspiel, O.: "Chemical Reaction Engineering," p. 277, John Wiley & Sons, New York (1972).
- 10) Nakamura, M., K. Hioki, A. Takahashi, H. Tanahashi and S. Watari: *Kagaku Kogaku Ronbunshu*, **5**, 473 (1978).
- 11) Nishiwaki, A. and Y. Kato: *Kagaku Kōgaku*, **36**, 1112 (1972).
- 12) Nishiwaki, A. and Y. Kato: *Kagaku Kōgaku*, **36**, 1147 (1972).
- 13) Nishiwaki, A., T. Kago, Y. Kato, S. Tanaka and T. Fukuda: *Kagaku Kogaku Ronbunshu*, **1**, 86 (1975).
- 14) Saganuma, T. and T. Yamanishi: *Kagaku Kōgaku*, **30**, 1136 (1966).
- 15) Tavlarides, L. L. and M. Stamatoudis: "Advances in Chemical Engineering," Drew, T. B. et al. (eds.), Vol. 11, p. 200, Academic Press, New York (1981).
- 16) Yagi, S. and T. Miyauchi: *Kagaku Kōgaku*, **17**, 382 (1953).
- 17) Yoshida, F. and T. Yamada: *J. Ferment. Technol.*, **49**, 235 (1971).