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LIQUID CIRCULATION IN BUBBLE COLUMN WITH DRAUGHT TUBE

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The flow rate Q_L of circulating liquid was measured by the heat-pulse method. It was found that Q_L increased with increasing liquid viscosity. In a liquid where bubble coalescence was not hindered, the gas holdup in the annulus decreased and therefore Q_L increased.

Empirical equations for gas holdups in the draught tube and the pressure drop due to flow reversals, necessary for estimating Q_L , were proposed.

Introduction

The bubble column with draught tube has recently drawn attention in relation to fermentation processes. In such a column the gas is dispersed into the draught tube and a stable circulating liquid flow is induced by the density difference between the aerated liquids in the draught tube and in the annulus. The circulating liquid flow enhances the heat transfer and makes the liquid properties homogeneous in the column.

The flow rate Q_L of the circulating liquid is measured by the tracer method, using an inorganic electrolyte^{1,4,12)} or a dye,⁵⁾ the electrochemical method¹⁰⁾ or the heat-pulse method.^{2,9)} In the first and second methods where an inorganic electrolyte or a dye is used, additions of these substances to the liquid hinder bubble coalescence, and this might change the Q_L value. The last method has no effect on bubble coalescence and might be better than the first and the second methods for measuring the Q_L value.

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The purpose of this work is to clarify experimentally the effects of gas velocity \bar{U}_G , geometrical dimensions of apparatus and properties of the liquid on the flow rate of circulating liquid by using the heat-pulse method.

1. Fundamentals

Macroscopic mass, momentum and energy balances in the column shown in **Fig. 1** give inherent

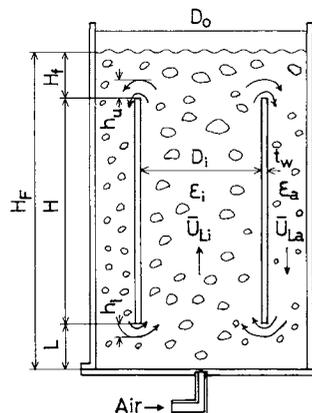


Fig. 1. Scheme of bubble column with draught tube and with gas dispersion into it

relationships among the flow rate Q_L of circulating liquid, liquid velocities, gas holdups and reversal losses due to reversing the flow direction.¹⁰⁾

$$Q_L = S_a \bar{U}_{La} = S_i \bar{U}_{Li} \quad (1)$$

$$\rho_L g H (\varepsilon_i - \varepsilon_a) = \Delta P_l + \Delta P_u + \Delta P_{fa} + \Delta P_{fi} \quad (2)$$

ε_a , ε_i , ΔP_l , ΔP_u , ΔP_{fa} and ΔP_{fi} in Eq. (2) must be given as functions of the average gas velocity \bar{U}_G , Q_L , the properties of the liquid and the geometrical dimensions of the column so that Q_L , which is related to \bar{U}_{Li} and \bar{U}_{La} through Eq. (1), can be calculated by Eq. (2).

2. Experimental

2.1 Experimental apparatus and conditions

The experimental apparatus used in this work is shown in Fig. 2. Three kinds of plexiglass columns with flat bottom were used. Their dimensions are 0.1 m dia., 2.3 m in height, 0.14 m dia., 2.3 m and 3 m in height, and 0.3 m dia., 2 m in height. Table 1 shows the dimensions of the draught tube and its height from the base plate of the column. Single orifice plates and perforated plates were used as gas distributors, details of which are shown in Table 1. The gas

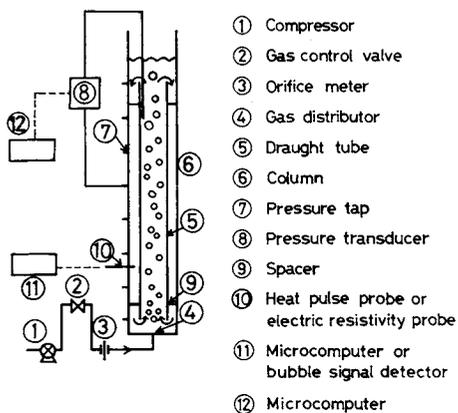


Fig. 2. Experimental apparatus

Table 1. Experimental apparatus

D_o [m]	D_i^* [m]	H [m]	H_L [m]	L/D_i [—]	Gas distributor**
0.100	0.060	1.40	1.50	1/2	S1
0.140	0.066	1.40	1.54	1/2	S2
0.140	0.082	0.70	0.84	1/2	S2
0.140	0.082	1.40	1.40–1.64	1/8–1	S2, P1, P2
0.140	0.082	2.10	2.24	1/2	S2
0.140	0.094	1.40	1.54	1/2	S2
0.140	0.104	1.40	1.54	1/2	S2
0.300	0.190	1.40	1.54	1/2	S2

* $t_w = 3$ mm for $D_i \leq 0.104$ m and 5 mm for $D_i = 0.190$ m.

** Single orifice plates, S1: $\delta = 3$ mm; S2: $\delta = 3$ mm; S3: $\delta = 15$ mm. Perforated plates, P1: $\delta = 0.5$ mm, $n = 19$; P2: $\delta = 1$ mm, $n = 13$.

velocity \bar{U}_G of air used was 0.002 – 0.169 $\text{m} \cdot \text{s}^{-1}$.

The liquids used were demineralized water and aqueous solutions of glycerol and glycol. The value of the clear liquid height H_L is shown in Table 1. During each run, liquid was neither fed nor discharged. The operating temperature of liquid was kept at 298.2 ± 0.5 K. Table 2 shows the properties of the liquid at operating temperature.

2.2 Measurement of liquid flow rate

Q_L was obtained by measuring the liquid velocities U_{La} in the annulus by the heat-pulse method.⁹⁾ The heat-pulse probe shown in Fig. 3 was installed on a radially moving device fitted to the column. The liquid heated by the heater for 10 ms flows, contacting two thermocouples one after another, revealing the temperature rise of the liquid as shown in Fig. 3. The difference Δt of the times when each thermocouple detects the temperature rise is measured, and U_{La} is calculated by Eq. (3):

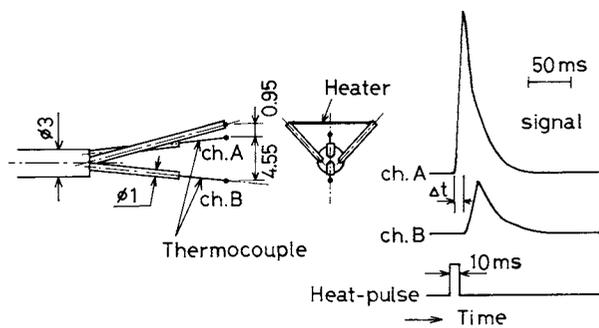
$$U_{La} = k(\Delta l / \Delta t) \quad (3)$$

where k is a correction factor and Δl the distance between two thermocouples. Calibration of the heat-pulse probe was done by measuring the velocity of a liquid jet in a tank both with the probe and with a standard pitot tube and by comparing values of the jet velocity measured by the two probes. The values of correction factors k were 1.17 for water, 1.16 for 35GL, 1.54 for 60GL and 1.38 for 70EG. 100 data of U_{La} were sampled by a microcomputer and the average value of U_{La} was obtained. U_{La} obtained at

Table 2. Properties of liquids at 298.2 K

Liquid*	ρ_L [$\text{kg} \cdot \text{m}^{-3}$]	$\mu_L \times 10^3$ [$\text{Pa} \cdot \text{s}$]	$\sigma_L \times 10^3$ [$\text{N} \cdot \text{m}^{-1}$]
W	997	0.894	72.0
35GL	1095	2.60	70.8
60GL	1179	16.81	66.13
70EG	1088	5.64	53.0

* W: water; 35GL and 60GL: 35 and 60 vol% glycerol aq. solns; 70EG: 70 vol% ethylene glycol aq. soln.



(a) Detail of heat-pulse probe (b) Signals from probe

Fig. 3. Heat-pulse probe and detected signals

seven positions in the radial direction of the annulus were multiplied by $(1 - \varepsilon_a')$ and used to evaluate Q_L by graphical integration, where ε_a' was measured by the electric resistivity method.⁷⁾

2.3 Gas holdup and reversal losses

Axial distributions of static pressures in the draught tube and the annulus were measured to evaluate ε_a , ε_i , ΔP_l and ΔP_u in Eq. (2), by a method of which details were shown elsewhere.¹⁰⁾ ΔP_{fi} and ΔP_{fa} in Eq. (2) were respectively calculated by using Takamatsu's results^{10,11)} and the friction factor for single-phase liquid flow.³⁾

3. Results and Discussion

3.1 Pressure drops

Figure 4 shows that ΔP_l increases with increasing $\bar{U}_{La}/(1 - \varepsilon_a)$ and liquid viscosity, and with decreasing distance L between the lower end of the draught tube and the base plate of the column. The effect of D_i/D_o on ΔP_l was similar to that reported in the previous paper.¹⁰⁾

In the previous paper¹⁰⁾ ΔP_l was assumed to be expressed in the form of loss due to the convergence-divergence flow model where the effect of flow reversal on ΔP_l is included in an effective width h_l of the gas-liquid flow path under the lower end of the draught tube.

$$\Delta P_l = 0.45 \left[\frac{\rho_L}{2} \left\{ \frac{S_a \bar{U}_{La}}{\pi D_1 h_l (1 - \varepsilon_a)} \right\}^2 \right] \left(1 - \frac{\pi D_1 h_l}{S_a} \right) + \frac{\rho_L}{2} \left(\frac{\bar{U}_{Li}}{1 - \varepsilon_i} \right)^2 \left\{ \frac{S_i (1 - \varepsilon_i)}{\pi D_1 h_l (1 - \varepsilon_a)} - 1 \right\}^2 \quad (4)$$

where $(\pi D_1 h_l)$ is the minimum cross-sectional area of gas-liquid flow under the lower end of the draught

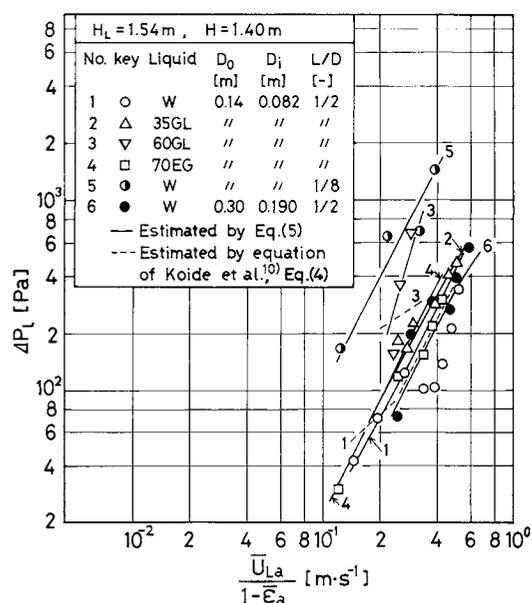


Fig. 4. Effect of $\bar{U}_{La}/(1 - \varepsilon_a)$, D_i , D_o and liquid properties on ΔP_l

tube. Figure 4 shows that Eq. (4) agrees relatively well with the observed value of ΔP_l for demineralized water but underestimates the ΔP_l value for 60 vol.% glycerol aqueous solution as $\bar{U}_{La}/(1 - \varepsilon_a)$ increases d.h. \bar{U}_G increases. Therefore, an empirical equation of ΔP_l in the form of Eq. (5) and one of h_l in the form of Eq. (6) were assumed. The numerical constant multiplied by the term $(\mu_L \bar{U}_{Gi}/D_i)$ in Eq. (5) and the numerical constants in Eq. (6) were decided by the direct search method,⁶⁾ using data observed in this work.

$$\Delta P_l = 0.45 \left[\frac{\rho_L}{2} \left\{ \frac{S_a \bar{U}_{La}}{\pi D_1 h_l (1 - \varepsilon_a)} \right\}^2 \right] \left(1 - \frac{\pi D_1 h_l}{S_a} \right) + \frac{\rho_L}{2} \left(\frac{\bar{U}_{Li}}{1 - \varepsilon_i} \right)^2 \left\{ \frac{S_i (1 - \varepsilon_i)}{\pi D_1 h_l (1 - \varepsilon_a)} - 1 \right\}^2 + 4510 \left(\frac{\mu_L \bar{U}_{Gi}}{D_i} \right) \quad (5)$$

$$\frac{h_l}{D_o} = 0.434 \left(\frac{L}{D_i} \right)^{0.401} \left(\frac{S_i}{S_o} \right)^{0.659} \left(\frac{S_a}{S_o} \right)^{1.26} \quad (6)$$

The average error of estimations of ΔP_l with Eqs. (5) and (6) was within 31% for 109 data. For the estimation of ΔP_l in columns of $(L/D_i) > 0.5$, it is recommended to estimate h_l with Eq. (6) by assuming $(L/D_i) = 0.5$. Figure 4 shows that ΔP_l values estimated by these equations agree relatively well with those observed.

Figure 5 shows that ΔP_u is roughly proportional to $\rho_L \{ \bar{U}_{Li}/(1 - \varepsilon_i) \}^2 / 2$ in the column of $D_i/D_o = 0.47$. However, this relation does not hold if the value of D_i/D_o is increased further or the liquid used is more viscous than water. In the previous paper¹⁰⁾ an empirical equation of ΔP_u based on the data for 10 mol·m⁻³ KCl aqueous solution and 100 mol·m⁻³ KCl-50wt% glycerol aqueous solution was proposed. This

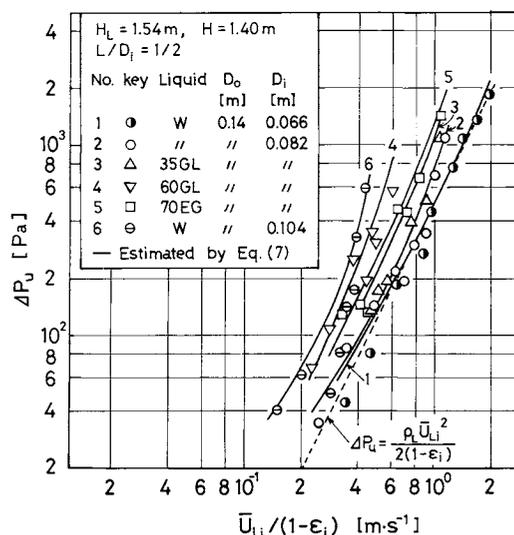


Fig. 5. Effect of $\bar{U}_{Li}/(1 - \varepsilon_i)$, D_i/D_o and liquid properties on ΔP_u

equation underestimates ΔP_u values in this work too low except for the ΔP_u value observed in the column with $D_i/D_o \leq 0.47$. One reason for this discrepancy is due to the facts that bubble coalescence in the draught tube is more frequent in the liquid used in this work than that in KCl aqueous solution, thus increasing the bubble size, and it becomes difficult for the bubbles over the upper end of the draught tube to be carried into the annulus by the circulating liquid flow. As a result, the gas holdup over the upper end of the draught tube increases. This effect of bubble coalescence on ΔP_u becomes remarkable in the range of $\bar{U}_G > 0.06 \text{ m} \cdot \text{s}^{-1}$. The second reason for the poor prediction of ΔP_u in highly viscous liquid by the previous equation is that the effect of viscosity is not taken into consideration. To correlate the experimental data of ΔP_u , a convergence-divergence flow model similar to Eq. (4) was employed instead of the divergence-convergence flow model, because a much better correlation of the experimental data was obtained thereby.

$$\Delta P_u = 0.45 \left[\frac{\rho_L}{2} \left\{ \frac{S_i \bar{U}_{Li}}{\pi D_1 h_u (1 - \varepsilon_f)} \right\}^2 \right] \left(1 - \frac{\pi D_1 h_u}{S_i} \right) + \frac{\rho_L}{2} \left(\frac{\bar{U}_{La}}{1 - \varepsilon_a} \right)^2 \left\{ \frac{S_a (1 - \varepsilon_a)}{\pi D_1 h_u (1 - \varepsilon_f)} - 1 \right\}^2 \quad (7)$$

where $(\pi D_1 h_u)$ and ε_f , respectively, are the minimum cross-sectional area of gas-liquid flow and the gas holdup over the upper end of the draught tube, and

$$\varepsilon_f = \{ D_1^2 \varepsilon_i + (D_o^2 - D_1^2) \varepsilon_a \} / D_o^2 \quad (8)$$

h_u was evaluated by Eq. (7) using measured values of ΔP_u , \bar{U}_{Li} , \bar{U}_{La} , ε_i and ε_a and was correlated with column dimensions, flow conditions and liquid properties;

$$\frac{h_u}{D_o} = 0.391 Fr_{Li}^{0.0924} \left(\frac{S_i}{S_o} \right)^{0.818} \left(\frac{S_a}{S_o} \right)^{1.36} M^{-0.00392} \times (1 - \varepsilon_i)^{1.08} \quad (9)$$

The average error of estimation of ΔP_u with Eqs. (7) and (9) was within 29% for 119 data. Figure 5 shows that ΔP_u values estimated by these equation agree relatively well with those observed.

Figure 6 shows that $(\Delta P_i + \Delta P_u)$ is about 65–85% of the total pressure drop, and ΔP_u is about 25–60% of the total pressure drop. The sharp increase in $(\Delta P_u / \Delta P_T)$ value at high gas velocity is due to the bubble coalescence mentioned above.

3.2 Gas holdup in draught tube, ε_i

Figure 7 shows that ε_i increases with increasing liquid viscosity and (D_i/D_o) value at the same gas velocity. Figure 7 shows that ε_i values observed in water agree relatively well with those in KCl aqueous solution,¹⁰⁾ although the previous equation of ε_i ¹⁰⁾

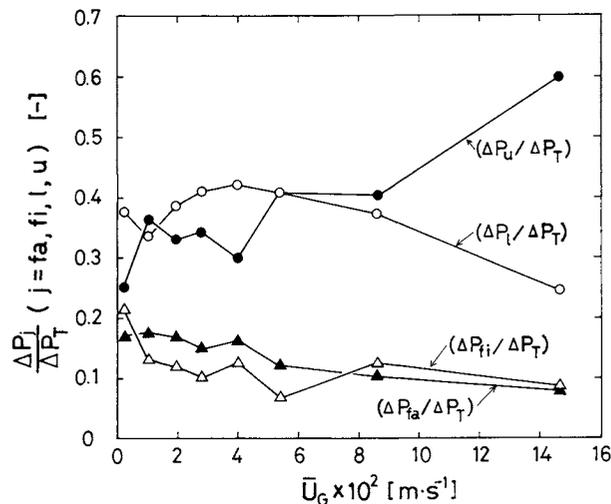


Fig. 6. Effect of \bar{U}_G on pressure drops, and comparison of pressure drops due to friction and flow reversal ($D_o=0.14 \text{ m}$, $D_i=0.094 \text{ m}$, $H_L=1.54 \text{ m}$, $H=1.40 \text{ m}$, $L/D_i=1/2$ and $\delta=3 \text{ mm}$)

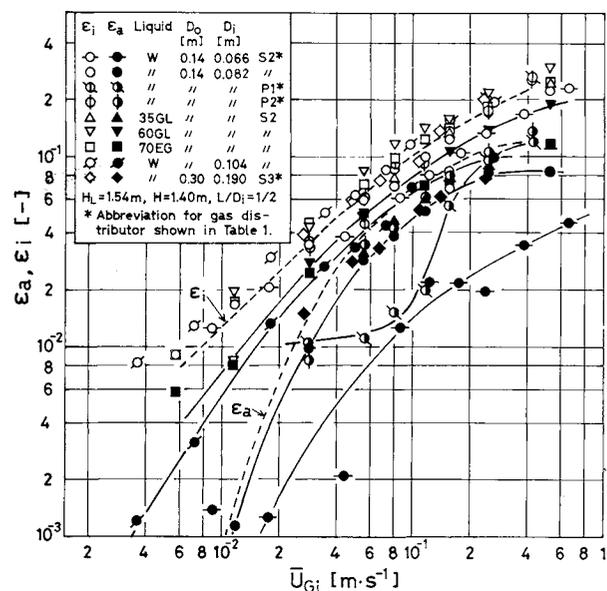


Fig. 7. Effect of \bar{U}_{Gi} , D_o , D_i , gas distributor and liquid properties on gas holdup (Dotted lines: Observed values in $10 \text{ mol} \cdot \text{m}^{-3}$ KCl aq. soln.¹⁰⁾ $D_o=1.14 \text{ m}$, $D_i=0.082 \text{ m}$ and $\delta=3 \text{ mm}$ and 3.6 mm)

based on the data in KCl aqueous solutions underestimated ε_i values in 60 wt% glycerol aqueous solution. So the Zuber-Findley plot $(\bar{U}_{Gi}/\varepsilon_i)$ vs. $(\bar{U}_{Gi} + \bar{U}_{Li})^{13}$ was tried by using observed data, but a linear relationship between $(\bar{U}_G/\varepsilon_i)$ and $(\bar{U}_{Gi} + \bar{U}_{Li})$ held only for the column with $D_i \leq 0.066 \text{ m}$. Therefore, the Zuber-Findley equation was modified by considering the effects of D_i , D_o and the liquid properties on ε_i :

$$\varepsilon_i = \frac{Fr_{Gi}}{0.415 + 4.27 Fr_i Bo^{-0.188} + 1.13 Fr_{Gi}^{1.22} M^{0.0386}} \quad (10)$$

Eq. (10) is applicable to both the column with a single

orifice plate and that with a perforated plate. The average error of estimations of ε_i with Eq. (10) was within 12% for 140 data.

3.3 Gas holdup in annulus, ε_a

Figure 7 shows that ε_a increases with increasing \bar{U}_{Gi} ; (D_i/D_o) and liquid viscosity. ε_a value in the 0.14 m ϕ column with the perforated plate ($\delta=1$ mm) was higher than that in the column with the single orifice plate S2, as more small bubbles existed in the former column. However, in the column with the perforated plate ($\delta=0.5$ mm) bubbles of relatively uniform size ($=5-6$ mm) were generated and rose in the draught tube in the range of $\bar{U}_{Gi} < 0.09$ m \cdot s $^{-1}$. In this case bubbles were not entrained into the annulus by the circulating liquid flow and so the ε_a value was low compared to that in the column with a single orifice plate, as shown in Fig. 7. Further increase in \bar{U}_{Gi} resulted in the increase of Q_L and bubble entrainment, and so the ε_a value increased.

Figure 7 shows that ε_a value in KCl aqueous solution¹⁰⁾ is higher than that in water and this tendency increases at high gas velocity, since bubble coalescence is hindered and so more small bubbles exist in KCl aqueous solution than in water. Since the previous equation of ε_a ¹⁰⁾ based on data observed in KCl aqueous solutions cannot be applied to ε_a data observed in this work, an empirical equation of ε_a observed in the bubble column with a single orifice plate is proposed:

$$\frac{\varepsilon_a}{(1-\varepsilon_a)^5} = 279 e_i^{1.50} \left(\frac{S_i}{S_o}\right)^{2.98} \left(\frac{S_a}{S_o}\right)^{2.59} \left(\frac{H}{D_o}\right)^{0.352} \times M^{0.0529} (1 + 0.433 Fr_{Gi})^{-1} \quad (11)$$

The average error of estimations of ε_i with Eq. (11) was within 24% for 109 data.

3.4 Flow rate of circulating liquid, Q_L

Figure 8 shows that Q_L increases with increasing \bar{U}_G and D_o and with decreasing liquid viscosity. Q_L in KCl aqueous solution is smaller than that in water, since ε_a in the solution is larger than that in water, and so the driving force of liquid circulation (the left-hand side of Eq. (2)) decreases.

Figure 9 shows that the maximum value of Q_L for constant value of \bar{U}_G and D_o is observed at $D_i/D_o \approx 0.6$ in the range of $\bar{U}_G \leq 0.08$ m \cdot s $^{-1}$, but the value of D_i/D_o where this maximum is observed shifts to about 0.5 in the range of $\bar{U}_G > 0.08$ m \cdot s $^{-1}$. This is due to the fact that ΔP_u increases steeply with increasing \bar{U}_G as shown in Fig. 6, and the ΔP_u value is lower in the column with lower D_i/D_o value as shown in Fig. 5.

Figure 10 shows that Q_L increases with increasing L/D_i in the range of $L/D_i \leq 1/2$. However, no effect of L/D_i on Q_L was observed in the range of $1/2 < L/D_i \leq 1$. Q_L increases with increasing hole diameter of the gas distributor, as ε_a decreases. From Eq. (2) Q_L

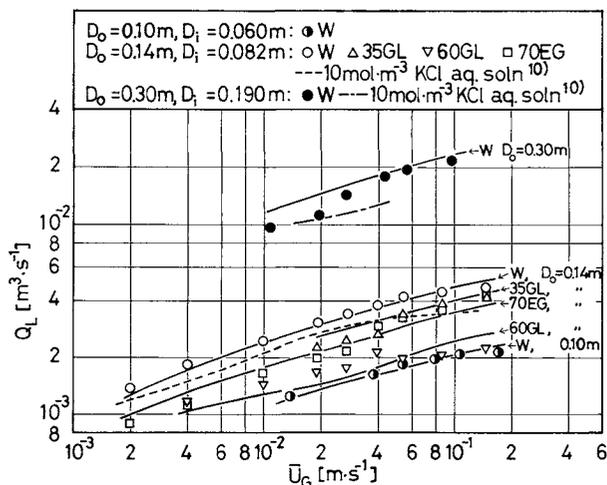


Fig. 8. Effect of \bar{U}_G , D_o and liquid properties on Q_L ($H_L=1.54$ m, $H=1.40$ m and $L/D_i=1/2$). Solid lines: estimated by Eq. (2)

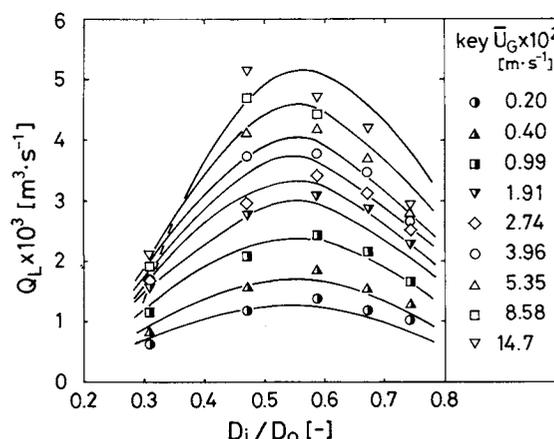


Fig. 9. Effect of D_i/D_o ratio and \bar{U}_G on Q_L (Liquid: water. $D_o=0.14$ m, $H_L=1.54$ m and $L/D_i=1/2$). Solid lines: estimated by Eq. (2)

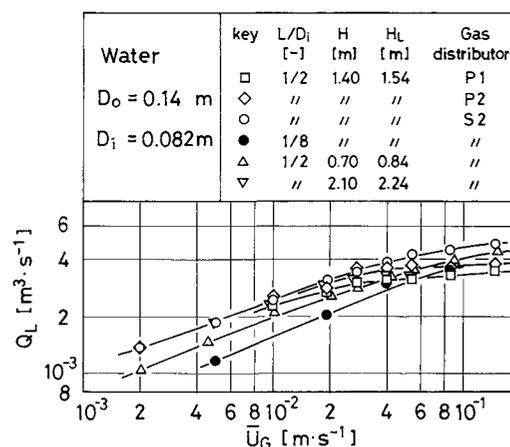


Fig. 10. Effect of \bar{U}_G , draught tube length, L/D ratio and gas distributor on Q_L

seems to increase with increasing H , but Fig. 10 shows that the Q_L value at $H=2.1$ m is lower than that at $H=1.4$ m. This is due to the fact that bubble coales-

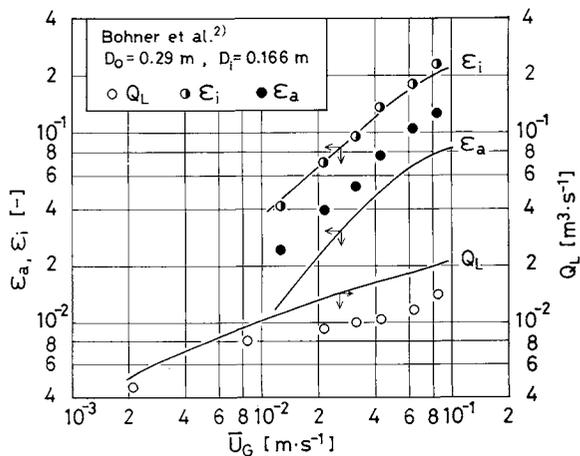


Fig. 11. Comparison of Q_L , ε_i and ε_a observed by Bohner *et al.*²⁾ in air-water system with those estimated, respectively, by Eqs. (2), (10) and (11) (Solid lines: estimated values)

cence in the draught tube occurs more frequently and ΔP_u increases more in a longer tube, when water is used as liquid.

3.5 Estimation of Q_L and comparison with previous works

Q_L at a given gas velocity can be estimated by solving Eq. (2) numerically, as each term in Eq. (2) is expressed as a function of Q_L , \bar{U}_G , the column dimensions, H_L and the liquid properties. ΔP_l , ΔP_u , ε_i and ε_a are expressed, respectively, by Eqs. (5), (7), (10) and (11). τ_{wi} and τ_{wa} , respectively, can be calculated by using Takamatsu's results^{10,11)} and the friction factor for single-phase liquid flow.³⁾ As Eq. (11) is only valid for estimating ε_a in the column with a single orifice plate, the method proposed here for estimating Q_L cannot be applied to the column with a perforated plate, unless a correlation of ε_a in such a column is available. Estimated values of Q_L agree relatively well with those observed, as shown in Figs. 8 and 9, and the average error of estimating Q_L was within 12% for 141 data. For estimating Q_L in the column with a perforated plate, the previous equation for ε_a in KCl aqueous solution was used.

Figure 11 shows Q_L , ε_i and ε_a observed by Bohner *et al.*²⁾ and those estimated by Eqs. (2), (10) and (11). ε_a was underestimated and Q_L overestimated. This discrepancy might be due to the fact that Bohner *et al.* used a ring nozzle and that it might generate finer bubbles than a single orifice plate. Q_L values observed in tap water by Jones⁸⁾ ($D_o=0.25$ m and perforated plate) were smaller than those estimated by the method proposed here, but if the previous equation of ε_a ¹⁰⁾ was used instead of Eq. (11), agreement between Q_L values observed and those estimated was fairly good. This might be because tap water is more contaminated than demineralized water and so the ε_a value in tap water is higher.

Conclusions

1) A comparison of this work with the previous ones indicated that if bubble coalescence was hindered, ε_a was increased and Q_L decreased, and vice versa, and that the degree of the bubble coalescence was determined by the liquid properties.

2) Empirical equations of gas holdup in the draught tube and annulus and the pressure drops due to flow reversals, necessary for estimating Q_L in the bubble column with a single orifice plate were proposed.

Nomenclature

Bo	= $D_o^2 g \rho_L / \sigma_L$, Bond number	[—]
D	= inner diameter	[m]
D_e	= $(D_o - D_i - 2t_w)$	[m]
D_i	= $(D_i + t_w)$	[m]
Fr_i	= $(\bar{U}_G + \bar{U}_L) / \sqrt{g D_i}$, Froude number	[—]
Fr_{Gi}	= $\bar{U}_{Gi} / \sqrt{g D_i}$, Froude number	[—]
Fr_{Li}	= $\bar{U}_{Li} / \sqrt{g D_i}$, Froude number	[—]
g	= gravitational acceleration	[m·s ⁻²]
H	= length of draught tube	[m]
H_F, H_f	= level of aerated liquid measured respectively from base plate of column and from upper end of draught tube	[m]
H_L	= clear liquid height	[m]
h_i, h_u	= effective flow width of gas-liquid flow path shown in Fig. 1	[m]
L	= distance between lower end of draught tube and base plate of column	[m]
M	= $g \mu_L^4 / \rho_L \sigma_L^3$, Morton number	[—]
n	= number of nozzles or holes	[—]
$\Delta P_l, \Delta P_u$	= pressure drop due to flow reversal at lower end and upper end of draught tube, respectively	[Pa]
ΔP_f	= frictional pressure drop	[Pa]
ΔP_T	= $(\Delta P_l + \Delta P_u + \Delta P_{fa} + \Delta P_{fi})$, total pressure drop	[Pa]
Q_L	= flow rate of circulating liquid	[m ³ ·s ⁻¹]
S	= cross-sectional area	[m ²]
t	= time	[s]
t_w	= wall thickness of draught tube	[m]
\bar{U}_G, \bar{U}_{Gi}	= gas velocity based on cross section of column and draught tube, respectively, and based on average static pressure in column	[m·s ⁻¹]
\bar{U}_L	= average liquid velocity based on cross section	[m·s ⁻¹]
U_{La}	= liquid velocity in annulus	[m·s ⁻¹]
δ	= hole diameter	[mm]
ε	= gas holdup	[—]
ε'_a	= gas holdup in annulus measured by electric resistivity method	[—]
ε_f	= gas holdup defined by Eq. (8)	[—]
μ_f	= liquid viscosity	[Pa·s]
σ_L	= liquid surface tension	[N·m ⁻¹]
τ_w	= shear stress at wall	[Pa]
 <Subscripts>		
a	= annulus	
i	= draught tube	
o	= column	

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A MEMBRANE REACTOR USING MICROPOROUS GLASS FOR SHIFTING EQUILIBRIUM OF CYCLOHEXANE DEHYDROGENATION

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Key Words: Membrane Reactor, Microporous Glass Membrane, Reversible Reaction, Equilibrium Shift, Dehydrogenation of Cyclohexane

A membrane reactor, which is a double-tubular reactor equipped with a selective membrane tube as the inner tube, was proposed. Such a reactor makes it possible to obtain a product yield of a reversible reaction beyond its equilibrium value by continuous removal of the products during reaction.

A microporous glass tube was employed as the selective membrane, through which gases permeate almost according to Knudsen's law. Experiments in the dehydrogenation of cyclohexane to benzene as a model reaction were carried out, using the membrane reactor under atmospheric pressure in the range of 453–493 K. It was shown experimentally and theoretically that a marked increase in conversion over that at equilibrium can be achieved.

Introduction

About 20 years ago, Michaels⁶⁾ pointed out that the application of membranes in reaction engineering would produce new chemical processes. According to his suggestion, it is possible to prevent a reaction mixture from attaining equilibrium composition by using a reactor containing a membrane separation unit, which is used for continuous and selective removal of products from the reaction mixture. This system, of course, will increase product yield and decrease the energy requirement for separation. We call such a reactor a "membrane reactor".

Such an idea has been tested by several investigators. Gryaznov²⁾ and Nagamoto *et al.*^{7,8)} studied dehydrogenation and/or hydrogenation using palladium and its alloy membranes, through which only hydrogen can diffuse. Carles *et al.*¹⁾ attempted direct thermolysis of water vapor using a calcia-stabilized zirconia membrane, through which oxygen can be transferred in the form of O²⁻ ions. In these cases, the membranes simultaneously function as catalysts. On the other hand, when the membrane itself does not have catalytic activity, a catalyst must be packed in the membrane reactor to make a reaction progress at sufficient speed. Regarding such a membrane reactor, Kameyama *et al.*⁵⁾ and Shinji *et al.*¹⁰⁾ succeeded in selective separation of hydrogen from a reacting

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