# THE HYDRODYNAMICS AND MASS TRANSFER <br> IN A PACKED BUBBLE BED COLUMN 

V.Pexidr ${ }^{a}$ and J.C.Charpentier ${ }^{b}$<br>${ }^{a}$ Institute of Chemical Technology, Department of Organic Technology,<br>${ }^{b}$ Laboratoire des Sciences du Génie Chimique, C.N.R.S., E.N.S.I.C. 54042 Nancy, France

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The relations are presented in the paper for pressure drop, gas hold-up and the over-all coefficient of mass transfer per unit volume $K_{\mathrm{L}} a$ for a 100 mm in diameter bubble bed coiumn filled with 6.4 and 10 mm Raschig rings. The system studied was water-air- $\mathrm{CO}_{2}$.

In numerous industrial gas-liquid operations the liquid trickles down either an active or non-active packing under simultaneous flow of gas, or bubbles of gas move through a column of liquid. A link between these two types of absorbers or reactors are packed bubble bed columns. Unlike trickle beds these columns exhibit higher liquid hold-up and larger gas-liquid interfacial area.

Few studies have been devoted to date to packed bubble bed columns. A literature survey is presented in Table I. The aim of this work is to gather additional data on the hydrodynamics and mass transfer in packed bubble beds, to obtain relations necessary for chemical engineering calculations and to compare the properties of these beds with bubble beds without packing.

Initial size of gas bubbles pumped into a column of liquid through a distributor depends on the surface tension, the density difference of the liquid and the gas and on the diameter of the distributor openings. However, the bubbles on their way through the liquid column undergo coalescence which in turn causes gradual decrease of the interfacial area. Under the presence of the fixed packing in the column the coalescence takes place simultaneously with the breaking of large bubbles on the edges of the fixed packing. Starting from a certain distance above the gas distributor the distribution function of the bubble diameter no longer varies. The mean size of the bubbles depends again on the surface tension and density difference of both phases and the size, or equivalent diameter, of the packing instead of the distributor openings. One can define the so called critical packing, or critical hydraulic diameter of the packing ${ }^{12,13}$ as

$$
\begin{equation*}
\left(d_{\mathrm{h}}\right)_{\mathrm{crit}}=2(\sigma / g \Delta \varrho)^{0.5} \tag{1}
\end{equation*}
$$

In packings larger than the critical diameter the bubbles move freely within the
interstices; for subcritical packings, however, their freedom is considerably restricted and the motion takes place rather through mutual impacts.
On their way through the packing the bubbles can follow either relatively free paths or be held up in the interstices with a limited chance to escape. The total gas hold-up can thus be divided into the dynamic part, $\alpha_{\mathrm{d}}$, i.e, a fraction of the free volume of the column occupied by mobile bubbles, and the static (quasistatic) part, $\alpha_{\mathrm{s}}$, i.e. a fraction of the free volume occupied by the bubbles moving only occasionally by the impact of other bubbles or due to liquid vibration.
Increasing gas flow rate causes the total hold-up to increase as well as the probability that gas bubbles reach new interstices in the packing with limited chance to escape. Consequently, both the dynamic and the static hold-up of gas will grow. On decreasing the gas flow rate a certain portion of the static bubbles will remain in the packing giving rise to a hysteretic curve $\alpha=\mathrm{f}\left(u_{\mathrm{G}}\right)$. It is apparent that for subcritical packings the static/dynamic hold-up ratio will be substantially greater than for packing larger than the critical diameter.

Similarly, liquid hold-up can be divided into the dynamic, $\beta_{d}$, and the static (capillary), $\beta_{s}$, pert. The static hold-up is a fraction of the free volume of the column occupied by liquid that clings to the packing after it has been left to drain. The static hold-up of both gas and liquid is practically without effect on the hydrodynamics and mass transfer. It is therefore possible to define a quantity which shall be termed the operating volume of the column

$$
\begin{equation*}
\omega_{\mathrm{op}}=\alpha_{\mathrm{d}}+\beta_{\mathrm{d}}=1-\alpha_{\mathrm{s}}-\beta_{\mathrm{s}} . \tag{2}
\end{equation*}
$$

Assuming $a$ ) the gravity force acting on the volume of liquid $\beta_{\mathrm{s}}$ to be balanced by the capillary forces holding it to the packing, $b$ ) the buoyancy force of gas $\alpha_{s}$ to be balanced by the packing, $c$ ) steady and isothermal flow, and, $d$ ) no effect of mass transfer on the physical properties of the phases, a balance of momentum about an infinitesimal volume of column may be written in liquid as

$$
\begin{equation*}
A \varepsilon \beta_{\mathrm{d}} \Delta P=A \varepsilon \beta_{\mathrm{d}} Q_{\mathrm{L}} g \Delta Z-\tau_{\mathrm{LS}} a_{\mathrm{g}}(1-\varepsilon) A \Delta Z-\tau_{\mathrm{LG}} a_{\mathrm{LG}}(1-\varepsilon) A \Delta Z \tag{3}
\end{equation*}
$$

and in gas as

$$
\begin{equation*}
A \varepsilon x_{\mathrm{d}} \Delta P=A \varepsilon \alpha_{\mathrm{d}} \varrho_{\mathrm{G}} g \Delta Z+\tau_{\mathrm{GS}} a_{\mathrm{g}}(1-\varepsilon) A \Delta Z+\tau_{\mathrm{LG}} a_{\mathrm{LG}}(1-\varepsilon) A \Delta Z . \tag{4}
\end{equation*}
$$

Summing up the last two equations, factoring the result by $\varepsilon \omega_{o p} \varrho_{L} g A \Delta Z$ and substituting for $\beta_{\mathrm{d}}$ from Eq . (2), one obtains

$$
\begin{equation*}
\frac{\Delta P}{\varrho_{\mathrm{L}} g \Delta Z} \equiv \frac{\Delta H}{\Delta Z}=1-\frac{\alpha_{\mathrm{d}}}{\omega_{\mathrm{op}}}-\left(\tau_{\mathrm{LS}}-\tau_{\mathrm{GS}}\right) \frac{a_{\mathrm{g}}(1-\varepsilon)}{\varepsilon \omega_{\mathrm{od}} \varrho_{\mathrm{L}} g} . \tag{5}
\end{equation*}
$$

Table I
Literature Survey on Raschig Ring Packed Bubble Columns

$$
\text { Authot } \quad d, \mathrm{~cm} \quad \varepsilon \quad Z, \mathrm{~m}
$$

| Mashelkar and coworkers | $1-2.5$ | $0.61-0.77$ | $0.7-1.2$ |
| :--- | :---: | :---: | :---: |
| Chen and coworkers ${ }^{2}$ | mesh Raschig <br> rings | 0.97 | $0.7-1.3$ |
| Musil and coworkers ${ }^{3}$ | 1 | 0.67 | 1.75 |
| L'Homme and coworkers $^{4}$ | 1 | 0.69 | 0.8 |
| Carleton and coworkers $^{5-6}$ | $0.64-3.8$ | $0.71-0.75$ | 1.5 |
| Hoogendorn and coworkers $^{7}$ | 1.3 | 0.63 | $1.6-3.2$ |
| Hofmann $^{8}$ | 0.64 | 0.71 | - |
| Kunugita and coworkers ${ }^{9}$ | $1-2$ | - | -7 |
| Blyakhman and coworkers ${ }^{10}$ | $0.6-2.5$ | - | $8-25$ |
| Voyer and coworkers ${ }^{11}$ | mesh Raschig | $0.92-0.95$ | $0.22-2.6$ |
| Sahay, Sharma ${ }^{20}$ | rings | $1-5$ | $0.53-0.94$ |
| This work | $0.64-1$ | $0.69-0.70$ | 1.75 |

" + Results given; -- no resulis.
$\tau_{\text {LS }}$ in Eqs (3)-(5) stands for the vertical component of liquid packing shear stress which takes positive (negative) values if the liquid descend (ascends) due to the effect of the buoyancy force of gas. $\tau_{G S}$ stands for the vertical component of gas packing shear stress. The hydrostatic resistance of the liquid column is represented by the term $1-\alpha_{d} / \omega_{o p}$.

The mass flux across the interface is given by the driving force and the over-all coefficient of mass transfer per unit volume $K_{\mathrm{L}} a$, or $K_{\mathrm{C}} a$. The interfacial area, $a$, may be computed provided the distribution function of the bubble size is known. For the water-air system it is assumed that the bubbles are oblide spheroids. For the calculation of the transfer area it is preferable to characterize the mean bubble size by the Sauter's average diameter defined by

$$
\begin{equation*}
d_{\mathrm{vS}}=\sum_{\mathrm{i}} n_{\mathrm{i}} \pi d_{\mathrm{ei}}^{3} / \sum_{\mathrm{i}} n_{\mathrm{i}} \pi d_{\mathrm{e} i}^{2} \tag{6}
\end{equation*}
$$

The equivalent diameter of the bubble is

$$
\begin{equation*}
d_{\mathrm{e}}=\left(d_{\mathrm{a}}^{2} \cdot d_{\mathrm{b}}\right)^{1 / 3} \tag{7}
\end{equation*}
$$

Table I
(continued)

| Column | Arrange- | Flowrate |  |  |
| :---: | :---: | :---: | :---: | :---: |
| diameter | ment | Liquid | gas | quantity |
| cm | of flows | $\mathrm{kg} \mathrm{m}^{-2} \mathrm{~S}^{-1}$ | $\mathrm{~kg} \mathrm{~m}^{-2} \mathrm{~s}^{-1}$ | $\alpha$ |


| 6.6-20 | $\rightleftarrows$ | $1-5$ | 0.05-0.35 |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
| 6.6-20 | $\rightarrow$ | $1-5$ | $0 \cdot 05-0.35$ | $\cdots$ | - |


| 7-15 | $\rightleftarrows$ | 1-50 | $0.04-0.2$ | - | - |
| :---: | :---: | :---: | :---: | :---: | :---: |
| 10 | $\longrightarrow$ | 1-15.5 | 0-1 | - | 1 |
| $7 \cdot 2$ | $\rightleftarrows$ | $0 \cdot 6-2 \cdot 2$ | $0 \cdot 007-0 \cdot 1$ | +- | - |
| 7.6-30.5 | $\rightleftarrows$ | 2-3--10 | 0-0.26 | -- | + |
| 41 | $\rightleftarrows$ | 1-6 | 0.15-1.5 | $+$ | - |
| 5 | $\xrightarrow{\rightarrow}$ | 0.3-3.7 | $0.001-0.03$ | $\pm$ | - |
| 10 | $\rightleftarrows$ | 1-30 | $0-0.037$ | $+$ | - |
| 8-25 | $\rightleftarrows$ | 0-5 | 0-0.8 | + | + |
| 14 | $\xrightarrow{\rightarrow}$ | 5-30 | 0.12-1.1 | $\because$ | - |
| 10-38.5 | $\cdots$ | $1.7 \cdots-5$ | $0 \quad 0.3$ | -- | -- |
| 10 | $\stackrel{\square}{*}$ | 0-17.5 | $0-0 \cdot 2$ | $+$ | $+$ |

where $d_{\mathrm{a}}$ and $d_{\mathrm{b}}$ are respectively the length of the principal and the conjugate axis of the oblide spheroide. Assuming that the mass transport takes place only across that part of the interface corresponding to the dynamic hold-up of gas, one obtains from the expression for the volume of the dynamic hold-up

$$
\begin{equation*}
\sum_{i} 1 / 6 n_{i} \pi d_{\mathrm{ei}}^{3}=\varepsilon \alpha_{\mathrm{d}} V \tag{8}
\end{equation*}
$$

and for its surface area

$$
\begin{equation*}
\sum_{i} n_{i} \pi d_{\mathrm{e} i}^{2}=a V \tag{9}
\end{equation*}
$$

Using Eq. (6) an expression for the specific interfacial area results

$$
\begin{equation*}
a=6 \varepsilon x_{\mathrm{d}} / d_{\mathrm{vs}} . \tag{10}
\end{equation*}
$$

From numerous papers on mass transfer in a plate column ${ }^{14}$, bubble column with or without packing ${ }^{1,15,16}$, and in a mixed reactor ${ }^{14,17}$ it follows that the coefficient $K_{\mathrm{L}}$ is practically independent of the hydrodynamic condition and varies only with
the physical properties of both phases. For systems with the equivalent dameter of the bubble exceeding 2.5 mm Calderbank ${ }^{14}$ proposed the following relation

$$
\begin{equation*}
K_{\mathrm{L}}=0 \cdot 42(\mathrm{Sc})^{-1 / 2}\left(g \Delta \varrho \eta_{\mathrm{L}} / \rho_{\mathrm{L}}^{2}\right)^{1 / 3} . \tag{11}
\end{equation*}
$$

Higbie ${ }^{18}$ derived using his theory

$$
\begin{equation*}
K_{\mathrm{L}}=2\left(D / \pi t_{\mathrm{c}}\right)^{1 / 2}, \tag{12}
\end{equation*}
$$

where the contact time, $t_{c}$, is most often defined as the ratio of the mean equivalent diameter of the bubble and the terminal velocity of bubble rise ${ }^{14,19}$. This velocity corresponds to $u_{\mathrm{L}}=0$ and $u_{\mathrm{G}} \rightarrow 0$. In liquid-liquid systems the terminal velocity of bubble rise is given by ${ }^{13}$

$$
\begin{equation*}
u_{0}=k\left(d_{\mathrm{L}} \boldsymbol{g} \Delta \varrho / \varrho_{\mathrm{L}}\right)^{1 / 2} \tag{13}
\end{equation*}
$$

with $k=0.5$ for a packing of Raschig rings smaller than 12 mm , or $k=0.64$ for the same packing larger than 12 mm . According to these relations the coefficient $K_{\mathrm{L}}$ depends only on the parameters of the packing and physical properties of both phases.

## EXPERIMENTAL

The experiments were designated to find relations between the flow rates of phases, hoid-ups, pressure drop and the mass transfer coefficient. The bubble bed was a glass column 10 cm in diameter 1.75 m high filled ether with 6.4 or 10 mm Raschig rings. Additional characteristics of the packings used are summarized in Tabie II. The measurements were carried out with the water-air- $\mathrm{CO}_{2}$ system.

The sketch of the experimental set-up is shown in Fig. 1. The water used for experiments was deionized in a system consisting of a filter (1a), a cation exchange column (1b), an anion exchange column (1c) and a purification column (1d). The flow rate of water was metered by a set of rotameters (2) and fed at the column top through a distributor (3).

Compressed air was humidified in a Raschig-ring packed water column and mixed with carbon dioxide supplied from a pressure cylinder (6). Mixing of both gases to a predetermined concentra-

## Table II

Characteristics of the Packing Used

tion (max. $4 \% \mathrm{CO}_{2}$ by volumc) was controlled by a system of valves and rotameters (7) and the mixture proceeded into the column at the bottom through a circular perforated tube (8). The over-pressure of gas was checked by U-manometer (13). The diameter of the openings of the tube was 1 mm . Volume flow rate of gas was metered by a dry gasometer (10) at the column exit. Constant head of liquid was maintained in the column by a valve (9) located in the bottom part of the column. The experimental set-up enabled the following quantities to be measured:
dyamic hold-up of liquid, $\beta_{\mathrm{d}}$, by wighing the liquid trapped in the column after turning off simultancously the feed of water and the regulation valve (9);
static hold-up of liquid was obtained as a difference of the weight of water poured into the column and that drained from the column within 15 minutes;
static hold-up of gas, $\alpha_{s}$, by measuring the difference of the beight of liquid level during the passage of the gas and with the gas valve shut afier the gas had been left to bubble through the column for some time at zero flow rate of liquid;
dinamic holitip of gas from the balance $\gamma_{\mathrm{d}}=1-\beta_{\mathrm{d}}-\gamma_{\mathrm{s}}-\beta_{\mathrm{s}}$ or from the measured dificrence of the liquid ievel above the packing before and after closing the feed of the gas. The values of $x_{d}$ obtained by these two methods differed by less than $5 \%$;


Fig. 1
Scheme of Experimental Set-Up
1 waser deionization, 1a filter, 1b cation exchange column, 1c anion exchange column, 1d purification column. 2 set of rotameters, 3 distributor of iquid, 4 celumn, 5 gas humidification column, $6 \mathrm{CO}_{2}$ pressure cylinder, 7 gas flow meters, 8 gas distributor, 9 regulation valve, 10 gasometer, 11 mercury manometer, 12 piezometric tube, 13 manomecr, 14 membrane pump, $15 \mathrm{CaCl}_{2}$ column, $16 \mathrm{MgClO}_{4}, 17$ analyzer column, 18 NaOH sofution column, 19 solid NaOH column, $20 \mathrm{CaCl}_{2}$ column.
pressure drop of the bubble bed by two piezomeiric tubes (12) and of the trickle bed by a mercury manometer (11) with pressure taps located below and above the layer of Raschig rings;
concentration of $\mathrm{CO}_{2}$ continuously in the air entering $\left(y_{1}\right)$ and exiting from the column $\left(y_{2}\right)$. The gas was sampled by a membrane pump (14), dried in two columns filled with $\mathrm{CaCl}_{2}$ (15) and $\mathrm{MgClO}_{4}$ (16) and analyzed by an infra-red ONERA analyzer (17). Prior to each experimental run the analyzer was calibrated by an air $\mathrm{CO}_{2}$ mixture of known composition. Zero position of the analyzer was set by pure air stripped of $\mathrm{CO}_{2}$ by scrubbing in a NaOH solution (18) and dried by solid NaOH (19) and $\mathrm{CaCl}_{2}$ (20);
temperature within the column by thermocouples located at the inlet and the outlet of the column.
The measured inlet and outlet concentrations of $\mathrm{CO}_{2}$ scrved to calculate the number of gas phase transfer units from

$$
\begin{equation*}
\mathrm{NTU}=[1 /(1-r)]\left[\log (1-r)\left(y_{1} / y_{2}\right)+r\right] . \tag{14}
\end{equation*}
$$

The over-all coefficient of mass transfer per unit volume was computed from

$$
\begin{equation*}
K_{\mathbf{L}} a=m u_{\mathbf{L}} . \mathrm{NTU} / \Delta Z, \tag{15}
\end{equation*}
$$

i.e. assuming plug flow in both phases, constant molar flow rate of gas, $G_{m}$, aiong the column height, $\Delta Z$, and negligible gas-side mass transier resistance. The measurement of the mass transfer coefficient was carried out in the range $0.5<r<2$, i.e. far below the flooding point.


Fig. 2
Plot of Total Gas Hold-Up versus Gas Flow Rate

| $\mu_{\mathrm{L}}, \mathrm{cm} / \mathrm{s}$ | Packing |  | Without |
| :---: | :---: | :---: | :---: |
|  | 6.4 mm | 10 mm | packing |
| 0.65 | 0 | 0 | $\bigcirc$ |
| 1.11 |  | 0 | 0 |
| 1.60 | - | 0 | 0 |



Fig. 3
Plot of ced versus Gas Flow Rate

- This work, packing 6.4 mm ; O This work, packing 10 mm ; Hogendorn and Lips ${ }^{7}$; Weber ${ }^{8}$; © L'Homme, Villers ${ }^{4}$; - Carleton and coworkers ${ }^{5}$.

The measurement of the bubble size distribution was made for both packings used. 5 photographs of the two-phase mixture were shot for each pair of $u_{G}$, $u_{\mathrm{L}}$ values just above the packing. Each photograph was evaluated to give the number of the bubbles of a given principal $\left(d_{a}\right)$ and conjugate $\left(d_{b}\right)$ diameter. These data served 10 calculate the equivalent diameter of the bubble $d_{e}$, the mean equivalent bubble diameter $\bar{d}_{\mathrm{c}}$ and the Sauter's mean bubble diameter.

## RESULTS AND DISCUSSION

## Gas Hold-Up and Interfacial Area

In the theoretical part it was noted that the $\alpha_{d}=f\left(u_{\mathrm{G}}\right)$ function may display hysterezis. To ensure reproducible operating conditions all experiments were carried out after flooding the column. The gas flow rate was then decreased to the initial required value and gradually increased. The static hold-up of gas given in Table II thus corresponds to this method of measurement.

Both the static and the dynamic portion of the gas hold-up increase with decreasing size of the packing. As it is apparent from Fig. 2 the dynamic hold-up depends on the size of the packing only little. A more significant correlation appears in case of the static part of the hold-up. Gas hold-up is independent of the flow rate of liquid. The dependence on the square root of gas velocity is almost linear. The data for both


Fig. 4
Plot of Mean Equivalent and Sauter's Mean Bubble Diameter versus Gas Flow Rate

Packing 6.4 mm : o $u_{\mathrm{L}}=0.65 \mathrm{~cm} / \mathrm{s}$, $u_{\mathrm{L}}=1 \cdot 11$, $\bullet u_{\mathrm{L}}=1 \cdot 6 ;$ packing 10 mm : क $u_{L}=1 \cdot 11$.


Fig. 5
Plot of Pressure Drop versus Dynamic Gas Hold-Up

Packing $6.4 \mathrm{~mm}: \circ u_{\mathrm{L}}=0 \mathrm{cms}^{-1}, 0.65$,

- $1 \cdot 11,-1 \cdot 6$; packing $10 \mathrm{~mm}:-\cdots-u_{\mathrm{L}}=$ $=0.65, \cdots-u_{\mathrm{L}}=1 \cdot 11$.
packings may be correlated by the following equation

$$
\begin{equation*}
\alpha_{d}=1.24 u_{G}^{0.5} \tag{16}
\end{equation*}
$$

Fig. 3 is a graphical comparison of our experimental dependence of the product $\varepsilon x_{d}$ on $u_{\mathrm{G}}^{\mathrm{n} .5}$ with the results of other authors. All data correspond to the packing of Raschig rings and the water-air system. The data can be correlated by the following equation

$$
\begin{equation*}
\varepsilon x d=0.085 u_{\mathrm{G}}^{0.5}, \quad u_{G}=\mathrm{cm} . \tag{17}
\end{equation*}
$$

The scatter of the data $( \pm 25 \%)$ is due to the different method of measuring hold-up by various authors, the effect of hysteresis as well as the conditions of measurement (temperature, wettability of packing) and the wall effect. The correlation corfrms the logical assumption that total hold-up decreases withincreasing diameter of the packing. The amount of data, however, is not sufficient to determine more precisely the exponent of the correlation.

The independence of the mean equivalent bubble diameter, $\bar{d}_{\mathrm{e}}$, on gas flow rate (Fig. 4) confirms the concept of simultaneous coalescence and bubble breaking within the packing. The two processes result in a certain equilibrium size of the bubbles. Their mean size will depend on the size of the packing although the presented fgure


Fig. 6
Plot of Pressure Drop Due to Liquid-Packing Friction versus Liquid Flow Rate

- Packing 6.4 mm , o packing 10 mm .


Fig. 7
Plot of Sum of Experimental Pressure Drop and Loss Due to Liquid-Packing Friction versus Dynamic Gas Hold-Up for 6.4 mm Packing
$u_{\mathrm{L}}=0 \mathrm{~cm} / \mathrm{s}, 00.65 \mathrm{~cm} / \mathrm{s}, 1.11 \mathrm{~cm} / \mathrm{s}$, - $1.60 \mathrm{~cm} / \mathrm{s}, \cdots \cdots(\Delta H / \Delta Z)_{H}$.
does not sufficiently evidence this hypothesis owing to a relatively small difference of the two packings used. The independence of the Sauter's mean bubble diameter on gas flow rate suggests that the area of gas-liquid interface is proportional to the gas velocity raised to 0.5 power similarly as the gas hold-up.

## Pressure Drop

The momentum balance indicates that the pressure drop of the column is a sum of two resistances: hydrostatic head of liquid $(\Delta H \mid \Delta Z)_{\mathrm{H}}=1-\alpha_{\mathrm{d}} / \omega_{\text {op }}$; friction liquid--packing and gas-packing

$$
(\Delta H / \Delta Z)_{\mathrm{F}}=\left(\tau_{\mathrm{LS}}-\tau_{\mathrm{GS}}\right) \frac{a_{\mathrm{g}}(1-\varepsilon)}{\varepsilon \omega_{\mathrm{op}} Q_{\mathrm{L}} g} .
$$

The principal resistance is concentrated in the hydrostatic head of the column of liquid. The weight of liquid corresponding to the static hold-up is counterbalanced by the capillary forces and therefore does not enter the expression for the hydrostatic pressure. Also the static gas hold-up enters the balance of forces only through the diminished total volume of the column, i.e. diminished weight of liquid by a certain constant value. Accordingly, the pressure drop is plotted in Fig. 5 as a function of


Fig. 8
Plot of Sum Experimental Pressure Drop and Loss Due to Liquid-Packing Friction versus Dynamic Gas Hold-Up for 10 mm Packing
$u_{\mathrm{L}}=0.65 \mathrm{~cm} / \mathrm{s}, \oplus 1.11 \mathrm{~cm} / \mathrm{s}, \bullet 1.6 \mathrm{~cm} / \mathrm{s}$,


Fig. 9
Plot of $K_{\mathrm{L}}$ a versus Dynamic Gas Hold-Up

|  | packing |  | without |
| :---: | :---: | :---: | :---: |
| $u_{\mathrm{L}}, \mathrm{cm} / \mathrm{s}$ | 6.4 mm | 10 mm | packing |
| 0.65 | 0 | 0 | 0 |
| 1.11 | - | 0 | 0 |
| 1.60 | $\ominus$ | $\ominus$ | $\otimes$ |

the dynamic hold-up of gas. The ordinate $u_{G}=0$ represents the decrease of the total liquid packing friction resistance. In accord with the Farning's equation this contribution is a quadratic function of the velocity of liquid namely $\tau_{\text {LS }}=k u_{\mathrm{L}}^{2}$. It turns out that pressure drop (Fig. 6) due to liquid-packing friction may be well correlated by the following equation

$$
\begin{equation*}
\left(\frac{\Delta H}{\Delta Z}\right)_{\mathrm{LS}}=k_{2} \frac{a_{\mathrm{g}}(1-\varepsilon)}{\varepsilon \omega_{\mathrm{op}} \varrho_{\mathrm{L}} g} u_{\mathrm{LI}}^{2} \tag{18}
\end{equation*}
$$

where $k_{2}$ equals respectively 990 and $1080 \mathrm{~kg} \mathrm{~m}^{-3}$ for the packing of 6.4 and 10 mm Raschig rings. For $u_{\mathrm{G}} \neq 0$ too though one must use the interstitial velocity $u_{\mathrm{LI}}=$ $=u_{\mathrm{L}}\left(\varepsilon \beta_{\mathrm{d}}\right)$ since on adding the contribution $(\Delta H / \Delta Z)_{\mathrm{Ls}}$, computed from Eq. (18), to the experimental pressure drop, $\Delta H / \Delta Z$, and ploting this sum versus the dynamic hold-up of gas (Fig. 7 and 8) one obtains a single straight line for all liquid flow rates. The effect of liquid flow is thus compensated by this contribution and the contribution of gas-packing friction may be understood as the difference of the straight lines $(\Delta H / \Delta Z)_{\text {LS }}+\Delta H / \Delta Z$ versus $(\Delta H / \Delta Z)_{\mathrm{H}}$. From Figs 7 and 8 we thus have

$$
\begin{equation*}
\left(\frac{\Delta H}{\Delta Z}\right)_{\mathrm{GS}}=k_{1} \frac{\alpha_{\mathrm{d}}}{\omega_{\mathrm{op}}} \tag{19}
\end{equation*}
$$

where $k_{1}$ equals respectively 0.231 and 0.237 for 6.4 and 10 mm Raschig rings. The total pressure drop may be expressed as

$$
\begin{equation*}
\frac{\Delta H}{\Delta Z}=1-\left(1-k_{1}\right) \frac{\alpha_{\mathrm{d}}}{\omega_{\mathrm{op}}}-k_{2} \frac{a_{\mathrm{g}}(1-\varepsilon)}{\varepsilon \omega_{\mathrm{op}} \varrho_{\mathrm{L}} g} u_{\mathrm{L}, 1}^{2} \tag{20}
\end{equation*}
$$

It seems that the constants $k_{1}$ and $k_{2}$ will be affected by the size of the packing only very little. For packings used in this work the differences are within experimental error and the correlation between these constants and the size of the packing must be rated as insignificant. To decide whether such correlation really exists one would need experiments covering a wider range of the packing size.

Considering Eqs (16) and (2) one obtains for the pressure drop the following correlation

$$
\begin{equation*}
\frac{\Delta H}{\Delta Z}=1-0.95 u_{\mathrm{G}}^{0.5}-1040 \frac{a_{\mathrm{g}}(1-\varepsilon)}{\varepsilon^{3} \omega_{\mathrm{op}} \varrho_{\mathrm{L}} g} \frac{u_{\mathrm{L}}^{2}}{\left(\omega_{\mathrm{op}}-1 \cdot 24 u_{\mathrm{G}}^{0.5}\right)^{2}} \tag{21}
\end{equation*}
$$

## Mass Transfer

As it is apparent from Fig. 9 the dependence of the over-all transfer coefficient per unit volume, $K_{\mathrm{L}} a$, on the dynamic hold-up is virtually a straigh line. Only in region
just below flooding the coefficient grows more slowly than the hold-up mainly due to more favourable conditions for coalescence. The dependence shown in Fig. 9 may be fitted by the following straight line

$$
\begin{equation*}
K_{\mathrm{L}} a=0.11 \alpha_{\mathrm{d}} . \tag{22}
\end{equation*}
$$

Substituting for $a$ from Eq. (10) and considering that $d_{\mathrm{vs}}$ is a constant independent of the flow rates of both phases as well as the size of the packing (from Fig. $4 d_{\mathrm{vs}}=$ $=9 \mathrm{~mm}$ ) and further that $\varepsilon$ for both packings differs only very little, one obtains for the coefficient $K_{\mathrm{L}}$ the value $0.025 \mathrm{~cm} / \mathrm{s}$. $K_{\mathrm{L}}$, thus depends neither on the liquid nor gas velocity which is in accord with the results obtained for this type of column also with other systems. Sharma ${ }^{1}$ has found a somewhat higher value of $K_{\mathbf{L}}=0.034 \mathrm{~cm} / \mathrm{s}$. The correlation of Calderbank and Moo-Young ${ }^{14}$ yield for freely moving bubbles greater than 2.5 mm (Eq. (11)) $0.031 \mathrm{~cm} / \mathrm{s}$.


Fig. 10
Plot of Relative Velocity of Phases versus Dynamic Gas Hold-Up

Packing 6.4 mm : $\odot u_{\mathrm{L}}=0.65 \mathrm{cms}^{-1}$, $\bigcirc u_{\mathrm{L}}=1 \cdot 11, \otimes u_{\mathrm{L}}=1 \cdot 6$; packing 10 mm : (1) $u_{L}=0.65, \bullet u_{L}=1 \cdot 11, \ominus u_{\mathrm{L}}=1 \cdot 6$.


Fig. 11
Comparison of the Dependence of $K_{\mathrm{L}} a$ on Gas Velocity with Data from Literature

1 This work; 2 Carleton and coworkers ${ }^{6}$, counter-current flow, 10 mm packing, 7.6 cm column; 3 this work, empty column; 4 Sharma and coworkers ${ }^{1}$, empty column; 5 Sharma and coworkers ${ }^{1}$, 10 mm packing, 10 cm column, co-current flow. Packing 6.4 mm : o $u_{\mathrm{L}}=0.65 \mathrm{~cm} / \mathrm{s}, \circ u_{\mathrm{L}}=1.11, \bigcirc u_{\mathrm{L}}=1.6$; packing $10 \mathrm{~mm}:\left(u_{\mathrm{L}}-0.65, \bullet u_{\mathrm{L}}=1 \cdot 11\right.$, $\ominus u_{\mathrm{L}}=1 \cdot 6$.

According to the Higbie's theory ${ }^{18}$ (Eq. (12)) the calculation of $K_{\mathrm{L}}$ requires the knowledge of the conctact time, which may be defined as $t_{\mathrm{c}}=\bar{d}_{\mathrm{e}} / u_{0}$. The terminal velocity may be understood as the relative velocity of gas with respect to liquid for $\alpha_{d} \rightarrow 0$

$$
\begin{equation*}
u_{\mathrm{r}}=u_{\mathrm{LI}}+u_{\mathrm{GI}} . \tag{23}
\end{equation*}
$$

Fig. 10 plots the relative velocity as a function of $\alpha_{d}$. The values of $u_{0}$ according to Eq. (13) are 9.0 and $12.7 \mathrm{~cm} / \mathrm{s}$ for 6.4 and 10.3 mm Raschig rings, respectively. The values of $K_{\mathrm{L}}$ computed from Eq. (12) are in excellent agreement with the experimental ones. The value of $u_{0}$ used for the calculation though was calculated from Eq. (13); experimental values of $u_{0}$ yield rather low values of $K_{\mathrm{L}}$ for 6.4 mm Raschig rings. The mass transfer coefficient thus does not correspond to the true velocity $u_{0}$ but rather a fictious velocity of the motion of the bubble which would exist if it were not for the effect of deccelaration due to the presence of the packing. According to Eq. (1) the 6.4 mm packing is subcritical and the deceleration of the bubbles is thus markedly stronger than in the case of 10 mm packing.

On combining Eqs (12) and (13) the mass transfer coefficient may be expressed by

$$
\begin{equation*}
K_{\mathrm{L}}=0.798\left(D / d_{\mathrm{c}}\right)^{1 / 2}\left(d_{\mathrm{h}} g \Delta \varrho / \varrho_{\mathrm{L}}\right)^{1 / 4} \tag{24}
\end{equation*}
$$

As the calculations for the given packings as well as the experimental data have shown the coefficient $K_{\mathrm{L}}$ to be independent of the packing size and because Eq. (24) contains, apart from $d_{\mathrm{n}}$ and $\vec{d}_{\mathrm{e}}$, only the quantities characterizing the given gas-liquid system it can be inferred that the equivalent bubble diameter, $\bar{d}_{\mathrm{c}}$, and the hydraulic radius of the packing are related by

$$
\begin{equation*}
\bar{d}_{\mathrm{e}}=k d_{\mathrm{h}}^{1 / 2} \tag{25}
\end{equation*}
$$

The validity of the last relation, however, should be still tested experimentally in in a wider range of the packing size.
On combining Eqs (17) and (22) one can formulate the following correlation for the examined system

$$
\begin{equation*}
K_{\mathrm{L}} a=0 \cdot 136 u_{\mathrm{G}}^{1 / 2} . \tag{26}
\end{equation*}
$$

This correlation is compared with the results of other authors in Fig. 11.

## CONCLUSION

Splitting the gas hold-up into its static a dynamic part and using the dynamic hold-up as a principal characteristic of the column has permitted analysis to be made of the basic relations for pressure drop and the mass transfer coefficient in a packed bubble
column. The size of the packing turns out to have a negligible effect on both the friction coefficients and the mass transfer coefficient as well as the interfacial area. Smaller packing exhibits only smaller static hold-up of gas which plays no role in the transfer process and diminishes only the operating volume of the column and the hydrostatic head of the liquid column. The dependence of the principal characteristic of the column, i.e. $\Delta H / \Delta Z$ and $K_{\mathrm{L}} a$, on the size of the packing and the physical properties of both phases require additional detailed study.

The results of this study indicate that the packed bubble column has numerous advantages over that without packing. Under identical velocities of gas and liquid the packed column exhibits higher hold-up of gas and hence larger interfacial area and lower pressure drop. The presence of the packing reduces the rate of coalescence of the bubbles and the interfacial area thus increases linearly with increasing gas hold-up. This permits effective operation even at higher gas flow rates and increases the mass transfer coefficient by several hundred percent over in the column without packing. As the packed bubble columns display also lower axial dispersion in both phases the column could be used effectively for a number of reactions instead of the so far used bubble columns.

## LIST OF SYMBOLS

| $a, a_{\mathrm{LG}}$ | interfacial area per unit volume of column ( $\mathrm{m}^{-1}$ ) |
| :---: | :---: |
| $a_{\mathrm{g}}$ | surface area of packing per unit volume of column ( $\mathrm{m}^{-1}$ ) |
| A | column cross section ( $\mathrm{m}^{2}$ ) |
| $d$ | diameter of Raschig rings (m) |
| $d_{\mathrm{e}}$ | equivalent bubble diameter (m) |
| $\ddot{d}_{\mathrm{e}}=\sum_{\mathrm{i}} n_{\mathrm{i}} d_{\mathrm{e}} / N$ | mean equivalent bubble diameter (m) |
| $d_{\mathrm{h}}$ | hydraulic diameter of packing (m) |
| $d_{\mathrm{vS}}$ | Sauter's mean bubble diameter (m) |
| D | diffusion coefficient ( $\mathrm{m}^{2} \mathrm{~s}^{-1}$ ) |
| $g$ | acceleration due to gravity ( $\mathrm{m} \mathrm{s}^{-2}$ ) |
| $G_{\mathrm{m}}$ | molar velocity of gas ( $\mathrm{mol} \mathrm{m} \mathrm{m}^{-2} \mathrm{~s}^{-1}$ ) |
| H | Henry's constant (Pa) |
| $\Delta H / \Delta Z$ | dimensionless pressure drop per unit height ( m of water head/m of column) |
| $k$ | constant |
| $K_{\text {L }}$ | over-all mass transfer coefficient ( $\mathrm{ms}^{-1}$ ) |
| $K_{\mathrm{L}} a$ | over-all mass transfer coefficient per unit volume ( $\mathrm{s}^{\mathbf{- 1}}$ ) |
| $L_{\mathrm{m}}$ | molar velocity of liquid ( $\mathrm{mol} \mathrm{m}^{-2} \mathrm{~s}^{-1}$ ) |
| $m=H / P$ |  |
| $N$ | total number of bubbles in a sample of two-phase mixture |
| $P$ | pressure (Pa) |
| $\Delta P$ | pressure drop (Pa) |
| $r=m G_{m} / L_{m}$ |  |
| $\mathrm{Sc}=\mu / \varrho D$ | Schmidt number |
| $u$ | superficial velocity ( $\mathrm{m} / \mathrm{s}$ ) |
| $u_{0}$ | terminal velocity of bubble rise ( $\mathrm{ms}^{-1}$ ) |


| $u_{\mathrm{LI}}=u_{\mathrm{L}} /\left(\varepsilon \beta_{\mathrm{d}}\right)$ | interstitial velocity of liquid $\left(\mathrm{ms}^{-1}\right)$ |
| :--- | :--- |
| $u_{\mathrm{GI}}=u_{\mathrm{G}} /\left(\varepsilon \alpha_{\mathrm{d}}\right)$ | interstitial velocity of gas $\left(\mathrm{ms}^{-1}\right)$ |
| $u_{\mathrm{r}}$ | relative velocity of phases $\left(\mathrm{ms}^{-1}\right)$ |
| $v$ | column volume $\left(\mathrm{m}^{3}\right)$ |
| $y_{1}, y_{2}$ | inlet and outlet mole fractions of $\mathrm{CO}_{2}$ |
| $\Delta Z$ | column height $(m)$ |
| $\alpha$ | gas hold-up $\alpha=\alpha_{\mathrm{s}}+\alpha_{\mathrm{d}}$ |
| $\beta$ | liquid hold-up $\beta=\beta_{\mathrm{s}}+\beta_{\mathrm{d}}$ |
| $\varepsilon$ | void fraction |
| $\eta$ | viscosity $(\mathrm{Pas})$ |
| $\omega_{\mathrm{op}}$ | operating volume of column |
| $\varrho$ | density $\left(\mathrm{kg} \mathrm{m}^{-3}\right)$ |
| $\sigma$ | surface tension $(\mathrm{Pa})$ |
| $\tau$ | shear stress $(\mathrm{Pa})$ |

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