

Connection Between Liquid Distribution and Gas-Liquid Mass Transfer in Monolithic Bed*

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Abstract With a particular focus on the connection between liquid flow distribution and gas-liquid mass transfer in monolithic beds in the Taylor flow regime, hydrodynamic and gas-liquid mass transfer experiments were carried out in a column with a monolithic bed of cell density of 50 cpsi with two different distributors (nozzle and packed bed distributors). Liquid saturation in individual channels was measured by using self-made micro-conductivity probes. A mal-distribution factor was used to evaluate uniform degree of phase distribution in monoliths. Overall bed pressure drop and mass transfer coefficients were measured. For liquid flow distribution and gas-liquid mass transfer, it is found that the superficial liquid velocity is a crucial factor and the packed bed distributor is better than the nozzle distributor. A semi-theoretical analysis using single channel models shows that the packed bed distributor always yields shorter and uniformly distributed liquid slugs compared to the nozzle distributor, which in turn ensures a better mass transfer performance. A bed scale mass transfer model is proposed by employing the single channel models in individual channels and incorporating effects of non-uniform liquid distribution along the bed cross-section. The model predicts the overall gas-liquid mass transfer coefficient with a relative error within $\pm 30\%$.

Keywords monoliths, flow distribution, gas-liquid mass transfer, Taylor flow, single-channel

1 INTRODUCTION

A monolithic bed consists of a bundle of separated channels in parallel with slit sizes of 1–3 mm, in which the slit wall is coated with catalytic or active materials to form a functional layer of 20–150 μm . In multiphase applications, gas and liquid flow through the channels upwards or downwards co-currently or counter-currently, and the reactants transform in the presence of the functional layer. It has been reported that monolithic beds are promising functional packings suitable for catalytic distillation [1–3] and catalytic multiphase reaction [4, 5]. In such applications, understanding gas-liquid mass transfer is important because it is closely related to separation and/or reaction performance of the process.

Gas-liquid mass transfer has been investigated [6–9] in a single channel or capillary scale mainly in the Taylor flow regime. Berčić and Pintar [6] measured liquid-side volumetric mass transfer coefficients ($k_L a$) for capillary gas-liquid flows and presented a correlation between the transfer coefficient and liquid slug lengths. van Baten and Krishna [8] studied gas-liquid mass transfer in capillaries by performing computational fluid dynamics (CFD) simulations and considered that both the bubble caps and the film side of the bubble contribute to mass transfer for Taylor flow. Vandu *et al.* [9] modified the correlation by van Baten and Krishna [8] on the basis of their experimental data for air absorption in water in 1–3 mm diameter capillaries of circular and square cross-sections. These single capillary correlations have been used to predict

$k_L a$ in monolithic beds [10, 11]. In general, mass transfer in a single channel or capillary may reflect to some extent the overall mass transfer on a monolithic bed scale. However, it has been indicated that in a monolithic bed, non-uniform distribution of gas and liquid at the entrance of the bed is frequently encountered [12–21]. In some of these studies, the liquid distribution in monolithic beds was experimentally investigated either using a CT (Gamma-ray computed tomograph) facility [12, 13] or the liquid collecting method [14], and other reported measured data of pressure drop, liquid saturation, and Taylor flow parameters in different channels of monolithic beds [15–21]. It has been recognized that the non-uniformity of flow distribution in a monolithic bed is largely dependent on the type of liquid distributor and superficial velocities of gas and liquid. This in turn determines the mass transfer and reaction performance in a monolithic bed. It also suggests that the mass transfer in a single channel can not be directly translated to that on a monolithic bed scale. Unfortunately, to the best of our knowledge, how the flow distribution is related to mass transfer in the bed and how the single channel mass transfer is associated with the bed scale mass transfer are still questions in the literature.

The objective of the present work is to establish a relation between flow distribution and gas-liquid mass transfer in monolithic beds. We first measure the liquid saturation in selected individual channels of a monolithic bed by using micro-conductivity probes and pressure drop of the bed. Then the liquid side mass transfer coefficients of the monolithic bed are measured *via* the stationary nitrogen desorption

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method. Finally, based on the foregoing measurements, the connection between flow parameters such as liquid velocity and slug length and mass transfer are discussed semi-theoretically and a model to describe mass transfer in monolithic beds is developed. In this way, we are able to take account for the contributions of individual channels to the mass transfer on the bed scale and provide a tool for prediction of the gas-liquid mass transfer coefficient in monolithic beds.

2 EXPERIMENTAL

A schematic representation of the experimental setup is depicted in Fig. 1. It mainly consists of (1) a cold model plexiglas column, $H=0.6$ m in height and $D=10$ cm in diameter, holding two monolithic beds, each with a height of 30 cm, block diameter 9.5 cm and individual square channel side-length 3.02 mm; and (2) a measuring system for phase distribution, pressure drop and gas-liquid mass transfer.

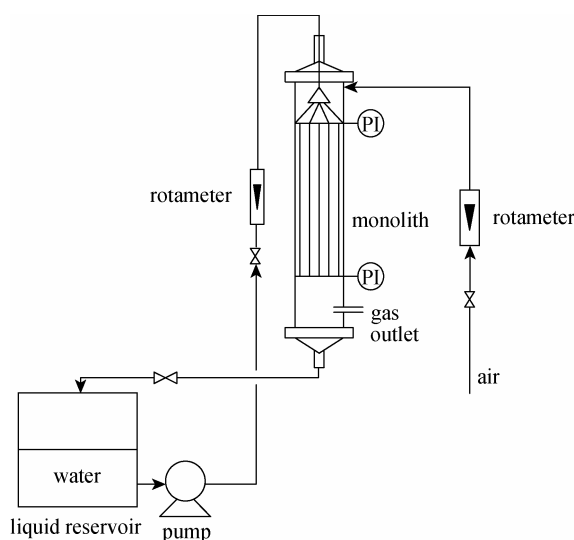


Figure 1 Schematic diagram of experimental setup

The cold model set up was operated in the co-current down-flow and continuous mode, using air as the gas phase and water as the liquid phase introduced at the top through a distributor. Two types of distributors were adopted for a comparative study for their effects on the hydrodynamics and mass transfer of the monolithic bed, including a commercial spray nozzle (Lechler, type 461.126AM) and a bed packed with 1 mm glass beads of 30 cm height. Fig. 2 shows the schematic representation of the nozzle and packed bed distributors, the installation of which were described in detail in our previous work [21]. Below the distributors, two blocks of monolith were stacked as the column bed. The monoliths were sealed inside the column by Teflon packing, which prevented by-pass flow of gas or liquid near the column wall.

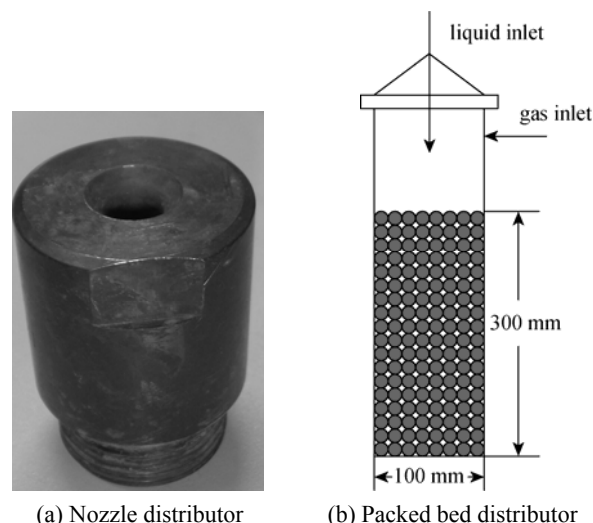


Figure 2 Liquid distributors: nozzle distributor (a) and packed bed distributor (b)

The overall pressure drop, gas liquid mass transfer, and liquid saturation in selected nine individual channels of the monolithic bed were measured with the superficial gas and liquid velocities of $0.065\text{--}0.445\text{ m}\cdot\text{s}^{-1}$ and $0.046\text{--}0.115\text{ m}\cdot\text{s}^{-1}$, respectively.

The pressure drops between the top and the bottom of the monoliths were measured by differential pressure transducers (Micro sensor, model MDM4951 DP3E) with a measurement range from 0.7 kPa to 6 kPa. During k_La measurements, the following procedure was followed. First, the k_La coefficients in the reactor as a whole, including the contribution of distributor, separator and monolithic bed, were measured by stationary oxygen absorption technique. Luminescent dissolve oxygen (LDO) probe (Hach, model sc100TM) was used to measure the amount of oxygen dissolved in the water at the inlet and exit of the monoliths. Then a blank experiment, in which only one block of monolith was stacked within the column instead of two blocks in the previous case, was implemented in the similar manner. The net k_La coefficients in monoliths were obtained by subtracting the k_La value obtained in the blank experiment from the previous one. Correspondingly, the contributions of the distributor and separator were excluded. The oxygen concentrations at the exit of the column, derived from the two cases, are used to calculate the experimental values of k_La :

$$k_La = \frac{Q_L / A}{\Delta Z} \ln \left(\frac{(C_{L,O_2})_e}{(C_{L,O_2})'_e} \right) \quad (1)$$

where Q_L is the volumetric liquid flow rate, A is the cross sectional area of the column, ΔZ is the height of one block of monolith, $(C_{L,O_2})_e$ and $(C_{L,O_2})'_e$ are the oxygen concentrations at the exit of the column

packed with two blocks and one block of monolith, respectively. The detail of the derivation of Eq. (1) is given in Appendix. The $k_L a$ values obtained are converted to a value at a reference state of 20°C by the following formula [22]

$$(k_L a)_{(20\text{ }^{\circ}\text{C})} = k_L a \times \theta^{(20-T)} \quad (2)$$

where θ is the temperature coefficient (here $\theta = 1.02$), and T is the temperature in the experiment. In what follows, all the reported $k_L a$ values in this work are the corrected ones.

We have used the micro-conductivity probe to identify flow regimes and measure gas-liquid flow parameters such as bubble rising velocity and its lengths in capillary gas-liquid flows [23]. Here, in order to characterize the liquid flow distribution, we extend the use of the probe for measuring liquid saturation in individual channels of the monolithic bed. In this case, there are 25 square channels along the bed diameter. Due to geometrical symmetry of the bed cross-section, nine copper microprobes were positioned along the bed diameter, with equal intervals between two adjacent microprobes and the no. 5 located in the axis of the bed diameter. Some other symmetric points within the cross-section of the bed were selected to install micro-probes so as to yield liquid saturation data under the same operating conditions, and the deviation of the different saturation data at different symmetrical positions was within 5%. During data acquisition, a sampling frequency of 1000 Hz and total sampling time of 10 s were implemented to ensure time resolution and statistical averaging. Typical signals yielded by one of the conductivity probes are shown in Fig. 3. It shows that gas bubble and liquid slug pass through the microprobe in turn, showing a typical Taylor flow in a channel. Over the range of the operating conditions in this work, the detected flow regime in the nine channels was found to be the Taylor flow, while with the nozzle distributor and at high gas velocities, flows in some channels may be gas-dominant. In what follows, we report data in the Taylor flow regime, in which the time averaged liquid saturation in channel i can be calculated by

$$\beta_{L,i} = \frac{L_{\text{slug}}}{L_{\text{slug}} + L_{\text{bubble}}} = \frac{t_{\text{slug}}}{t_{\text{slug}} + t_{\text{bubble}}} \quad (3)$$

where L_{slug} and L_{bubble} represent the liquid slug and gas bubble length, respectively; t_{slug} and t_{bubble} represent the time intervals during which a liquid slug and a gas bubble pass the probe, respectively.

To quantify the degree of liquid flow non-uniformity, the following mal-distributor factor [14] is calculated based on the measured liquid saturation data

$$\sigma^2 = \frac{1}{9} \sum_i \left(\frac{\beta_{L,i} - \beta_{L,\text{average}}}{\beta_{L,\text{average}}} \right)^2 \quad (4)$$

where $\beta_{L,i}$ is the liquid saturation in channel i , and $\beta_{L,\text{average}}$ is the average liquid saturation in the nine channels,

$$\beta_{L,\text{average}} = \frac{1}{9} \sum_i \beta_{L,i} \quad (5)$$

Obviously, smaller value of $\beta_{L,\text{average}}$ corresponds to more uniform distribution of the liquid flow.

3 RESULTS AND DISCUSSION

3.1 Liquid distribution in monolithic bed

Figure 4 shows the cross-sectional profiles of the time averaged liquid saturation at two superficial gas velocities with the nozzle [Fig. 2 (a)] as the liquid distributor. At the low gas velocity ($0.064 \text{ m}\cdot\text{s}^{-1}$), nearly uniform liquid distribution is observed; at the high velocity ($0.440 \text{ m}\cdot\text{s}^{-1}$) the distribution shows a “W” shape along the cross-section, with two channels (no. 3 and no. 8) having saturation values close to zero. It was visually observed that (1) in the former case, a thick gas-liquid suspension formed just at the top of the bed but more liquid spread to the column wall region due to the liquid jet effect; and (2) in the latter case, the gas-liquid suspension was thinner, reducing liquid retention on the top of the bed and resulting in

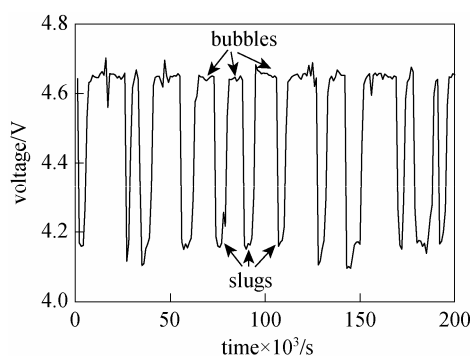


Figure 3 Typical signals of the conductivity probe

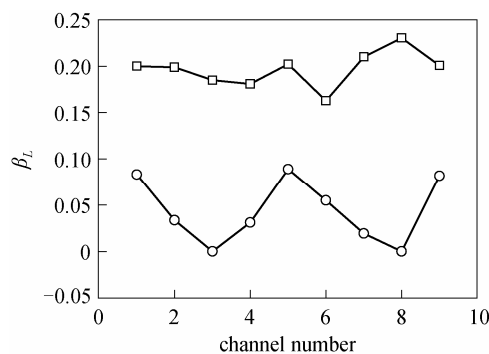


Figure 4 Liquid saturation in different channels at different gas velocities with nozzle distributor ($u_{L,s} = 0.046 \text{ m}\cdot\text{s}^{-1}$)
 $u_{G,s}/\text{m}\cdot\text{s}^{-1}$: $\square$ 0.064; $\circ$ 0.440

the formation of zero saturation or gas-dominant channels (channels No. 3 and No. 8). The occurrence of the gas-dominant channels was observed in our previous work [21]. This phenomenon suggests that the liquid flow distribution is sensitive to the change in superficial gas velocities.

Figure 5 shows the cross-sectional profiles of the time averaged liquid saturation at two superficial gas

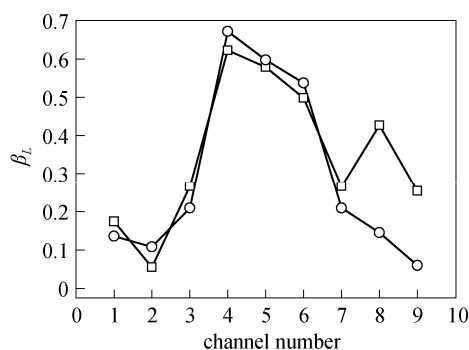
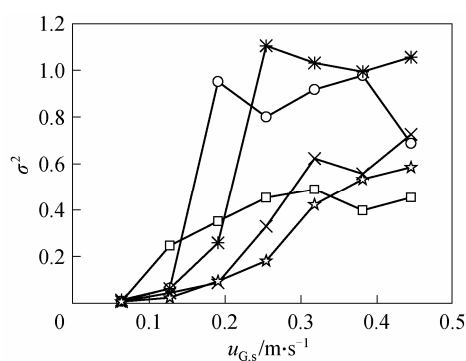
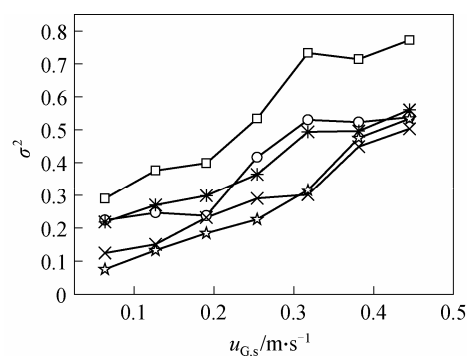


Figure 5 Liquid saturation in different channels at different gas velocities with packed bed distributor
 $u_{G,s}/\text{m}\cdot\text{s}^{-1}$: $\square$ 0.064; $\circ$ 0.440



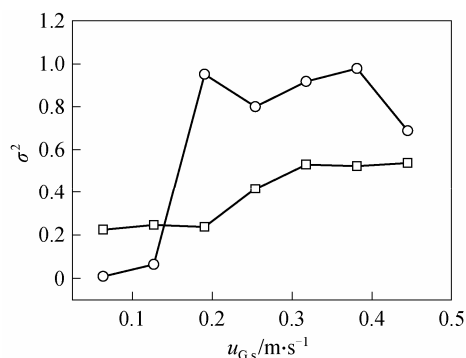
(a) Nozzle distributor



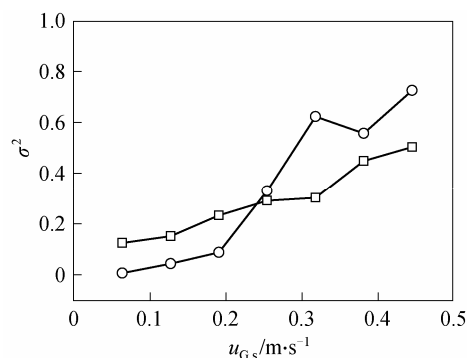
(b) Packed bed distributor

Figure 6 Mal-distribution factor as a function of gas and liquid superficial velocities for nozzle distributor (a) and packed bed distributor (b)

$u_{L,s}/\text{m}\cdot\text{s}^{-1}$: $\square$ 0.046; $\circ$ 0.064; $*$ 0.083; $\times$ 0.101; $\triangle$ 0.115



(a) $u_{L,s} = 0.064 \text{ m}\cdot\text{s}^{-1}$



(b) $u_{L,s} = 0.101 \text{ m}\cdot\text{s}^{-1}$

Figure 7 Comparison of mal-distribution factors for nozzle and packed bed distributors

$\square$ packed bed; $\circ$ nozzle

velocities with the packed bed [Fig. 2 (b)] as the liquid distributor. At the two gas velocities, the distribution shows an inverse “V” shape along the cross-section because the liquid is introduced through the center of the packed bed distributor, which brings more liquid in the center of the monolithic bed, and the profiles are close to each other and have similar shape characterized by a high saturation domain in the central part of the bed. In contrast to the nozzle distributor, the packed bed distributor presents less varying profiles with changing superficial gas velocities.

Figure 6 shows the calculated values of mal-distribution factor from the measured saturation data at different superficial gas and liquid velocities for the two distributors. For the nozzle distributor (a), at low superficial gas velocities the mal-distribution factor has small values, but the liquid distribution is not really uniform due to the existence of unstable flow, which will be explained later, while at relatively low superficial liquid velocities, the mal-distribution factor increases sharply in accompany with the formation of gas-dominant channels in the bed. For the packed bed distributor (b), the mal-distribution factor decreases with increasing liquid velocities and decreasing gas velocities.

Figure 7 compares the effects of the two distributors on liquid distribution in terms of the mal-distribution factor. At relatively low gas velocities ($u_{G,s} < 0.2 \text{ m}\cdot\text{s}^{-1}$), the nozzle distributor is better, while at relatively high gas velocities the packed bed distributor yields a more uniform liquid distribution. The reason is clear if we look at the measured bed pressure drop data at different superficial gas and liquid velocities as shown in Fig. 8. For the nozzle distributor (a), when $u_{G,s} < 0.2 \text{ m}\cdot\text{s}^{-1}$, all the measured pressure drop data are negative. This operating domain is called the unstable region for flow in monoliths [17, 19, 20], in which the bed pressure drop abnormally decreases as liquid velocity increases and the liquid and gas are automatically redistributed. This leads to reversed liquid flow in some channels and higher flow rates in others, showing a bad liquid distribution, although the mal-distribution factor measured is relatively small. For the packed bed distributor (b), the unstable region is negligible.

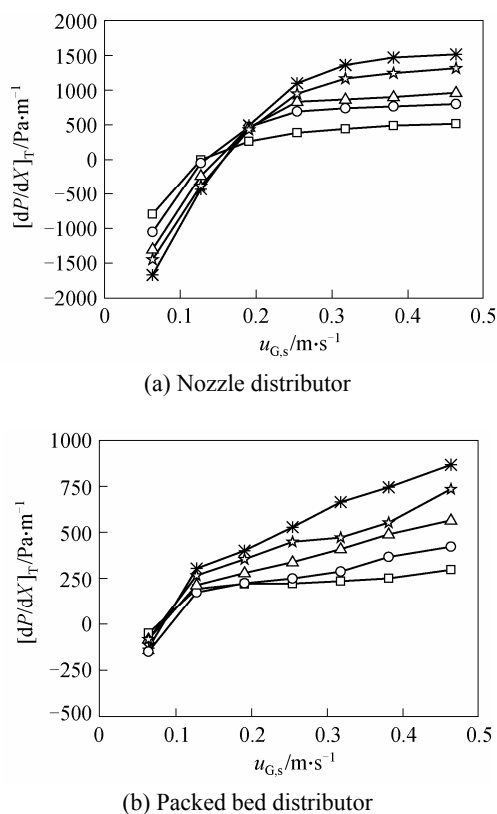


Figure 8 Pressure drop as a function of superficial gas and liquid velocities for nozzle distributor (a) and packed bed distributor (b)
 $u_{L,s} / \text{m}\cdot\text{s}^{-1}$: $\square$ 0.046; $\circ$ 0.064; $\triangle$ 0.083; $\star$ 0.101; $*$ 0.115

3.2 Gas-liquid mass transfer in monolithic bed and its connection with liquid flow distribution

Figure 9 shows the measured liquid-side mass transfer coefficients, $k_L a$, as a function of superficial gas and liquid velocities for the two distributors. In

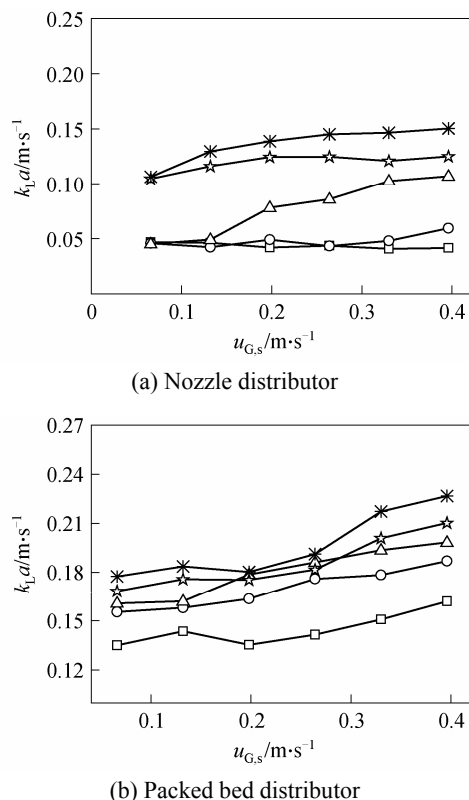


Figure 9 Effect of superficial liquid and gas velocities on $k_L a$ in monolith with nozzle distributor (a) and packed bed distributor (b)
 $u_{L,s} / \text{m}\cdot\text{s}^{-1}$: $\square$ 0.046; $\circ$ 0.064; $\triangle$ 0.083; $\star$ 0.101; $*$ 0.115

both cases, $k_L a$ increases with liquid velocity, while the gas velocity has a slight effect on the transfer coefficient. In comparison, we find that the packed bed distributor outperforms the nozzle distributor by a factor up to 3 in terms of the mass transfer coefficient.

In what follows, we will investigate the above mass transfer performance in the monolithic bed and the flow parameters in individual channels of the bed so as to ascertain the connection between the flow distribution and gas-liquid mass transfer in monolithic beds.

It has been demonstrated in previous mass transfer investigations [6–9] that flow parameters such as liquid slug lengths and phase velocities have pronounced effects on mass transfer in the two phase flow through a single capillary. A single capillary is one of many building elements for a monolithic bed, so the bed performance could be attributed to the contribution of individual channels. Thus we derive values of the liquid slug length and velocity from the measured pressure drop and liquid saturation data by using the following single channel models.

For computation of the liquid velocities in a single channel, we adopt the model proposed by Roy [24]. With experimental data of pressure drop of the bed and liquid saturation in a channel, the frictional pressure drop in channel i is calculated by

$$[dP/dX]_F = [dP/dX]_T + \rho_L g \beta_{L,i} \quad (6)$$

where $[dP/dX]_F$ and $[dP/dX]_T$ represent the frictional drop and total pressure drop of the bed ($\text{Pa} \cdot \text{m}^{-1}$), respectively.

The frictional pressure drop mainly arises from the liquid slug rather than the liquid film [25]. According to the Hagen-Poiseuille equation, one has

$$[dP/dX]_F = 32\mu_L u_{L,i} / d^2 \quad (7)$$

For constant fluid property and channel dimension, Eq. (7) can be written as

$$[dP/dX]_F = k_e u_{L,i} \quad (8)$$

The following mass balance equation is used to determine the proportional constant in Eq. (8)

$$u_{L,s} = \varepsilon_b \sum_i^9 u_{L,i} \quad (9)$$

where $u_{L,s}$ is the superficial liquid velocity in the column, ε_b is the bed void fraction, and $u_{L,i}$ is the superficial liquid velocity in channel i . Solving Eqs. (8) and (9) gives the nine liquid velocities in the individual channels.

The dimensionless liquid slug length is calculated by the following equation

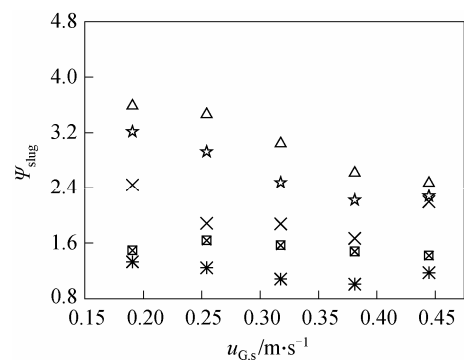
$$\psi_{\text{slug},i} = L_{\text{slug},i} / d_h = u_{\text{TP},i} t_{\text{slug},i} / d_h \quad (10)$$

where $L_{\text{slug},i}$ represents the liquid slug length in channel i , $t_{\text{slug},i}$ represents the time in which the liquid slug passes the electrode in channel i , $u_{\text{TP},i}$ is the two-phase velocity in channel i , which is given by

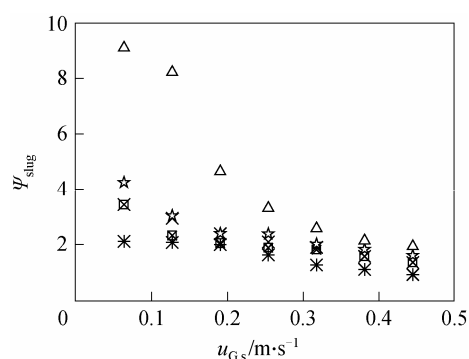
$$u_{\text{TP},i} = \frac{u_{L,i}}{\beta_{L,i}} \quad (11)$$

where $u_{L,i}$ and $\beta_{L,i}$ are the superficial liquid velocity and liquid saturation in channel i , respectively.

Typically, Fig. 10 shows the calculated liquid slug lengths in channel 5 as a function of superficial gas and liquid velocities for the two distributors. For the nozzle distributor (a), the liquid slug lengths are not shown for $u_{G,s} < 0.18 \text{ m} \cdot \text{s}^{-1}$, which correspond to the unstable or reversed flows in the bed. For both distributors, liquid slug length increases with liquid velocity and decreases as gas velocity increases. This is an expected fact and analogous to the single capillary results [26]. A comparison for the slug length shows that at the same phase velocities, the packed bed distributor always yields shorter liquid slugs than the nozzle distributor. In particular, when $u_{G,s} > 0.2 \text{ m} \cdot \text{s}^{-1}$ most of the slug lengths are less than 2. This suggests that the liquid is dispersed efficiently and uniformly by the packed bed of glass beads before entering the monolithic bed. This is why the packed bed distributor leads to a better mass transfer performance in the bed compared to the nozzle distributor (refer to Fig. 9).



(a) Nozzle distributor



(b) Packed bed distributor

Figure 10 Dimensionless liquid slug lengths as a function of gas and liquid velocity in channel 5 for nozzle distributor (a) and packed bed distributor (b)

$u_{L,s}/\text{m} \cdot \text{s}^{-1}$: * 0.046; □ 0.064; × 0.083; ☆ 0.101; △ 0.115

With the values of liquid saturation, velocity and slug length in each channel, we can extend the single channel models to predict the overall mass transfer performance of the monolithic bed. The procedure is as follows [27].

In each channel, the mass transfer coefficient can be expressed as

$$(k_L a)_i = \frac{u_{L,i}}{L} \ln \left(\frac{(C_{O_2,e})_i}{C_{O_2,f}} \right) \quad (12)$$

where $(k_L a)_i$ is the mass transfer coefficient for channel i , $u_{L,i}$ is the superficial liquid velocity in channel i , L is the height of channel i , and $C_{O_2,f}$ is the oxygen concentration at the inlet of channel i , which is considered to be the same as that measured at the inlet of the column. Therefore, the exit oxygen concentration in channel i , $(C_{O_2,e})_i$, is

$$(C_{O_2,e})_i = C_{O_2,f} \exp \left(\frac{(k_L a)_i L}{u_{L,i}} \right) \quad (13)$$

With the non-uniform distribution of liquid, the oxygen concentration at the bed exit varies. The mixing cup concentration of the nine channel exit concentrations is

$$\begin{aligned} \overline{(C_{O_2,e})} &= \sum_{i=1}^9 u_{L,i} (C_{O_2,e})_i / \sum_{i=1}^9 u_{L,i} (C_{O_2,e})_i \\ &= C_{O_2,f} \sum_{i=1}^9 u_{L,i} \exp\left(\frac{(k_L a)_i L}{u_{L,i}}\right) / \sum_{i=1}^9 u_{L,i} \quad (14) \end{aligned}$$

In Eq. (14), the mass transfer coefficient for channel i is calculated by [9]

$$(k_L a)_i = \frac{8}{\sqrt{\pi}} \sqrt{\frac{D_L u_{TP,i} (1 - \beta_{L,i})}{L_{UC,i}}} \frac{1}{d_h} \quad (15)$$

where $L_{UC,i}$ is the unit cell length in channel i ($L_{UC,i} = \frac{L_{slug,i}}{\beta_{L,i}}$). This correlation was obtained by

Vandu *et al.* [9] on the basis of their mass transfer experiments in single capillaries and is applicable in the range of $[(u_{G,s} + u_{L,s})/L_{slug}]^{0.5} > 3s^{-0.5}$. We checked our data and found that most of them meet the criterion except those corresponding to the occurrence of gas-dominant channels. However, these gas-dominant channels have negligibly small contributions to the bed scale mass transfer.

With the exit concentration of the bed from Eq. (14), we can calculate the bed scale mass transfer coefficient by

$$k_L a = \frac{u_{L,s}}{L} \ln \left(\frac{\overline{(C_{O_2,e})}}{C_{O_2,f}} \right) \quad (16)$$

Figure 11 shows the calculated mass transfer coefficients using Eqs. (12)–(16) and the experimental ones. The deviation between the results is within $\pm 30\%$. In view of that the present bed scale model incorporates the complicated effects of the non-uniform liquid distribution along the bed cross-section and its prediction is based on the single channel models, this result is satisfactory.

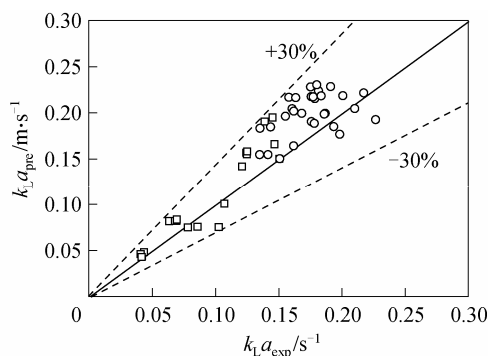


Figure 11 Comparison of measured $k_L a$ and predicted $k_L a$ with the two distributors
□ nozzle; ○ packed bed

4 CONCLUSIONS

Focusing on the connection between liquid flow distribution and gas-liquid mass transfer in monolithic beds, we carried out experiments for two-phase flow hydrodynamics and gas-liquid mass transfer in a cold model column with two 50 cpsi monoliths. Liquid saturation in individual channels of the bed and the overall bed pressure drop and mass transfer coefficients were measured with a nozzle distributor and a packed bed distributor at different phase velocities. The connection between flow parameters such as liquid velocity and slug length and mass transfer was explored semi-theoretically and a bed scale model describing gas-liquid mass transfer in monolithic beds was developed. The following major conclusions can be drawn.

(1) Within the superficial gas and liquid velocity range used, increasing the liquid velocity improves liquid flow distribution along the cross-section of the bed, while the gas velocity has a negative effect on the distribution. For liquid distribution, the packed bed distributor is superior to the nozzle one. With the nozzle distributor, liquid distribution is sensitive to changes in gas velocity and gas-dominant channels may occur within a monolithic bed.

(2) The liquid side mass transfer coefficient increases with liquid velocity, while the gas velocity has a slight effect on mass transfer. The mass transfer performance in the monolithic bed is better when the packed bed is used as liquid distributor, in which the transfer coefficient is higher by a factor up to 3.

(3) A semi-theoretical analysis using the single channel models shows that the packed bed distributor always yields shorter liquid slugs compared to the nozzle distributor, suggesting that the liquid flow is dispersed efficiently and uniformly by the packed bed of glass beads. This is its reason to present better mass transfer performance.

(4) The bed scale mass transfer model proposed by employing the single channel models in individual channels and incorporating effects of the non-uniform liquid distribution along the bed cross-section predicts the overall gas-liquid mass transfer coefficient with a relative error within $\pm 30\%$.

NOMENCLATURE

A	cross section area, m^2
C_{L,O_2}	oxygen concentration in water, $g \cdot m^{-3}$
cpsi	cells per square inch
D_L	diffusion coefficient, $m^2 \cdot s^{-1}$
d_h	diameter of monolith, mm
g	gravitational acceleration, $m^2 \cdot s^{-1}$
$k_L a$	liquid-side volumetric mass transfer coefficient, s^{-1}
k_e	correlation constant
L	height of channel, m
L_{bubble}	gas bubble length, m
L_{slug}	liquid slug length, m

L_{UC}	unit cell length, m
Q_L	volumetric liquid flow rate, $m^3 \cdot s^{-1}$
T	temperature, $^{\circ}C$
t_{slug}	time interval a liquid slug experiences at the probe, s
t_{bubble}	time interval a gas bubble experiences at the probe, s
u	velocity, $m \cdot s^{-1}$
Z	monolith bed length, m
β_L	liquid saturation
ε_b	bed void fraction
θ	temperature coefficient
μ	viscosity, $N \cdot s \cdot m^{-2}$
ρ	density, $kg \cdot m^{-3}$
σ^2	mal-distribution factor
ψ_{slug}	dimensionless liquid slug length

Subscripts

average	average value
exp	experiment
F	friction
G	gas
i	channel i
L	liquid
pre	predicted
s	superficial
T	total
TP	two phase

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APPENDIX DERIVATION OF EQ. (1)

Figure A1 shows the experimental mass transfer model for interpreting mass transfer measurements in this work. Fig. A1 (a) is the whole model system (Model A) including all the four mass transfer regions within the column, namely, the distributor, monolith blocks 1 and 2, and gas-liquid separation region. Fig. A1 (b) shows the blank model system (Model B) with only one monolith block.

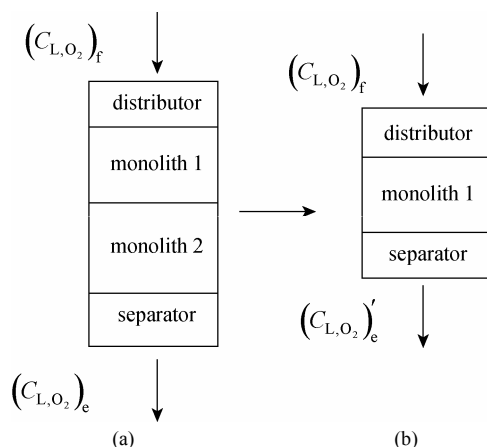


Figure A1 Schematic diagram of experimental mass transfer models

For the four regions of Model A, the mass balance equations of oxygen are as follows

$$Q_L dC_{L,O_2} = -(k_L a)_{\text{top}} (C_{L,O_2} - 0) AdZ \quad (\text{A1})$$

$$Q_L dC_{L,O_2} = -(k_L a)_1 (C_{L,O_2} - 0) AdZ \quad (\text{A2})$$

$$Q_L dC_{L,O_2} = -(k_L a)_2 (C_{L,O_2} - 0) AdZ \quad (\text{A3})$$

$$Q_L dC_{L,O_2} = -(k_L a)_{\text{bottom}} (C_{L,O_2} - 0) AdZ \quad (\text{A4})$$

where Q_L is the liquid flow rate in $\text{m}^3 \cdot \text{s}^{-1}$; C_{L,O_2} is the dissolved oxygen concentration in the bulk liquid phase in $\text{g} \cdot \text{m}^{-3}$; A is the cross-section area of column bed in m^2 ; and Z is the distance from the top of column bed in m. In the range of gas flow rates used in this work, the oxygen concentration in gas phase is negligibly small. Therefore, the equilibrium concentration of oxygen at the interface is zero. The following boundary conditions are used to integrate the above mass balance equations

$$C_{L,O_2} = (C_{L,O_2})_f, Z = 0 \quad (\text{A5})$$

$$C_{L,O_2} = (C_{L,O_2})_1, Z = Z_{\text{top}} \quad (\text{A6})$$

$$C_{L,O_2} = (C_{L,O_2})_2, Z = Z_{\text{top}} + Z_1 \quad (\text{A7})$$

$$C_{L,O_2} = (C_{L,O_2})_3, Z = Z_{\text{top}} + Z_1 + Z_2 \quad (\text{A8})$$

$$C_{L,O_2} = (C_{L,O_2})_e, Z = Z_{\text{top}} + Z_1 + Z_2 + Z_{\text{bottom}} \quad (\text{A9})$$

Therefore,

$$(k_L a)_{\text{top}} Z_{\text{top}} + (k_L a)_1 Z_1 + (k_L a)_2 Z_2 + (k_L a)_{\text{bottom}} Z_{\text{bottom}} = \frac{Q_L}{A} \ln \left(\frac{(C_{L,O_2})_e}{(C_{L,O_2})_f} \right) \quad (\text{A10})$$

where $(k_L a)_{\text{top}}$ is the mass transfer coefficient in the distributor region and Z_{top} is the corresponding length; $(k_L a)_1$ and $(k_L a)_2$ are the mass transfer coefficients in the two monolithic regions, and Z_1 and Z_2 are the corresponding lengths; $(k_L a)_{\text{bottom}}$ is the mass transfer coefficient in the separation region and Z_{bottom} is the corresponding length; $(C_{L,O_2})_f$ and $(C_{L,O_2})_e$ are the inlet and exit concentrations of oxygen, respectively.

In a similar manner, one can integrate the mass balance equations for the three regions of Model B and obtain the following result

$$(k_L a)_{\text{top}} Z_{\text{top}} + (k_L a)_1 Z_1 + (k_L a)_{\text{bottom}} Z_{\text{bottom}} = \frac{Q_L}{A} \ln \left(\frac{(C_{L,O_2})'_e}{(C_{L,O_2})_f} \right) \quad (\text{A11})$$

where $(C_{L,O_2})'_e$ is the exit concentration of oxygen in a blank mass transfer experiment.

Subtracting Eqs. (A10) and (A11) yields the equation for calculating the liquid-side mass transfer coefficient, *i.e.*, Eq. (1) in the paper.