



# Pressure drop and liquid hold-up in multiphase monolithic reactor with different distributors

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## ABSTRACT

Total pressure drop and liquid hold-up in two different monolith packings have been investigated experimentally in the co-current downward flow mode with a reactor diameter of 0.1 m and 0.6 m high monolith packing operating in the Taylor flow regime. The effect of distributor design on the flow distribution was investigated using two different types of distributor (nozzle distributor and packed bed distributor with 1 mm glass beads). A close relation of the distributor design to the hydrodynamics in monolith beds was observed to exist, evidenced by the larger pressure drop and liquid hold-up values for the monolith packings with an accompanying use of the packed bed distributor, as compared with the use of the nozzle distributor. An explanation for this phenomenon is the better gas and liquid distribution capability the packed bed has. By taking account of the effect of the liquid slug lengths, correlations were derived for predicting the two-phase friction factor and liquid hold-up and satisfactorily describe the experimental data. Finally an analysis of the unstable flow phenomenon characterized by negative pressure drops within bed shows that the unstable region is not only dependent on the operating conditions and properties of monolith but also on the distributor design.

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## 1. Introduction

In multiphase monolith reactors, gas and liquid flow concurrently down- or up-ward over monolith blocks composed of an array of uniformly structured parallel channels. In view of the advantages of such a reactor configuration over the conventionally used trickle beds and slurry bubble columns, efforts have been paid to extend the application of monoliths to a host of multiphase catalytic reaction systems. During the course, it is stressed by many researchers that an even gas–liquid distribution across the channels of monolith is crucial for the effective gas–liquid dispersion and contacting, thereby ensuring a good reactor performance and facilitating reactor scale-up. Satterfield and Ozel [1] studied the effect of three different distributors on the pressure drop characteristics of monolith beds; the results showed that the bed pressure drop is sensitive to the design of distributor. Grolman et al. [2] measured the liquid hold-ups of monolith beds, on the top of which two types of distributor, i.e., a showerhead and a foam layer, were mounted; they found that a height of 15 cm high foam layer leads to good phase distribution at higher liquid hold-ups ( $>0.65$ ). Roy and Al-Dahhan [3] investigated the liquid distribution in a monolith bed by comparing three different types of distributor (nozzle, showerhead and foam) in the

Taylor flow regime; it was demonstrated that the nozzle-type distributor was the most suitable for monolith reactors, and the range of operation conditions was identified in which the flow distribution approached to be uniform for the 400 cpsi monolith. Heibel et al. [4,5] studied the gas and liquid distribution and their effect on the performance of the monolith film flow reactor; they illustrated the importance of the selection of the distributor for a uniform distribution of the liquid across the monolith.

Despite these previous studies, still it is not quite clear what is a good homogeneous flow distributor design for a monolith reactor. To address this issue, we carried out experimental studies to investigate the link between the multiphase hydrodynamics of the monolith packing and the configuration of distributors. In what follows, we present measuring results of pressure drop and liquid hold-up in monolith packings with two different types of distributors (nozzle and 30 cm high packed bed distributors), under different operation conditions. Then we discuss the quantitative relations derived from the measurements, including correlations for estimating the friction pressure drop and liquid hold-up, and an analysis of the unstable flow phenomena characterized by negative pressure drops over the packing.

## 2. Experimental section

A schematic representation of the experimental setup is depicted in Fig. 1. It mainly consists of (1) a cold model monolith

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## Nomenclature

$d_h$	diameter of monolith (mm)
Eo	Eötvös number $((\rho_L - \rho_G)d_h^2 g / \sigma_L)$
$f$	frictional factor
cpsi	cells per square inch (in. <sup>-2</sup> )
$g$	gravity (m <sup>2</sup> /s)
$\Delta p$	pressure drop (Pa)
$L$	height of monolith (m)
$L_b$	length of a bubble (m)
$L_{b0}$	initial length of a bubble (m)
$L_{slug}$	liquid slug length
Re	Reynolds number
$Re'_G$	gas phase Reynolds number $(u_G d_h \rho_L / \mu_L)$
$Re_{TP}$	two-phase Reynolds number $(u_{TP} d_h \rho_L / \mu_L)$
$u$	velocity (m/s)
$u_s$	superficial velocity (m/s)
$C_0$	distribution parameters in drift-flux model
$v_G$	drift velocity in drift-flux model (m/s)

## Greek symbols

$\rho$	density (kg/m <sup>3</sup> )
$\varepsilon_L$	liquid hold-up
$\varepsilon_G$	gas hold-up
$\varepsilon_{gG}$	volume fraction of bubbles
$\mu$	viscosity (Pa s)
$\sigma$	surface tension (N/m)
$\psi_{slug}$	dimensionless liquid slug length

## Subscripts

L	liquid
G	gas
TP	two-phase

reactor ( $D = 10$  cm;  $H = 0.6$  m) and (2) a pressure drop and liquid hold-up measurement system.

The cold model set up is operated in the co-current down-flow and continuous mode, using air as the gas phase and water as the liquid phase, respectively. Two types of distributors are adopted

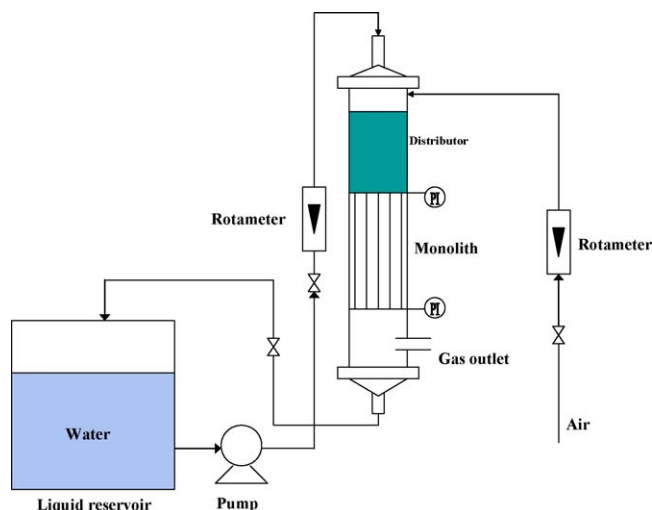


Fig. 1. Schematic of the co-current down-flow monolith reactor.

Table 1

Geometrical specifications of two different monoliths used in experiments.

Cell density (cpsi)	Diameter (cm)	Length (cm)	Hydraulic diameter (mm)	Skeleton thickness (mm)	Cell spacing (mm)
400	9.5	15	1.1	0.17	1.27
100	9.5	15	2.1	0.45	2.55

for a comparative study of their effect on the hydrodynamics of monolith beds, including a commercially available spray nozzle (Lechler, Germany), and a bed packed with 1 mm glass beads of 30 cm high. The nozzle-type distributor was positioned at a specified distance of 5 cm above the monolith packing, such that the expanding cone just hits the column wall, and this distance is the appropriate height pointed out by Roy and Al-Dahhan [3]. The packed bed distributor was placed straightly on the top of the monolith packings, using a close wire netting to prevent glass beads entering into the channels of the monoliths. On the top of the packed distributor, a plate of 100 mm diameter, containing 210 holes (2 mm diameter) was placed to make sure uniform liquid spreading over the packed bed. Below the distributors, four blocks of monolith ( $\phi = 9.5$  cm;  $L = 15$  m; channel diameters = 1.1 mm and 2.1 mm, respectively) were stacked as the reactor bed. The monoliths were sealed inside the column using Teflon tapes, which prevented by-pass flows of gas or liquid. The geometrical specifications of the two different monoliths used are list in Table 1.

Pressure drop and liquid hold-up measurements were made with the superficial gas and liquid velocities covering a span of 0.055–0.388 m/s and 0.039–0.118 m/s, respectively. This range of velocities corresponds to the slug flow regime for a capillary two-phase flow as depicted by the flow regime map developed by Mishima and Hibikisome [6], and the pulsing flow regime for the packed bed as depicted by flow regime map developed by Fukushima et al. [7]. Pressure drops between the top and the bottom of the monolithic packing were measured by various differential pressure transducers (MDM4951 DP3E  $\pm 0.5\%$ ; MICRO SENSOR Co. Ltd., China) capable of pressure measurements ranged from 0.7 kPa to 6 kPa. During the liquid hold-up measurements, the following procedure was followed. First, the liquid hold-up in the reactor as a whole, including the contribution of the amounts of liquid before (on top of the bed) and after the bed, was measured by collecting and weighting the liquid after simultaneous shutdown of the liquid and gas feeds. Then a so-called blank experiment, in which case only one block of monolith was stacked within the reactor in contrast to the three blocks of monolith in the previous case, was implemented in the similar way. A net weight value of liquid was obtained by subtracting the weight obtained in the blank experiment from the previous one. Correspondingly, the net value indicates the part of liquid retained in the blocks of monolith, excluding the contribution before and after the bed. In each experiment, three measurements of hold-up were made, and the repetition was found to be within  $\pm 5\%$ .

## 3. Results and discussion

### 3.1. Basic features of liquid hold-up and pressure drop in monolith packing

Fig. 2 shows the measured liquid-ups for various superficial gas and liquid velocities with the use of the two different distributors. Obviously, the hold-up values are not sensitive to change in the gas velocity particularly at higher gas velocities, a trend in contrast to that commonly observed in a bubble column where the phase hold-up varies significantly with an increase in gas velocity. And in

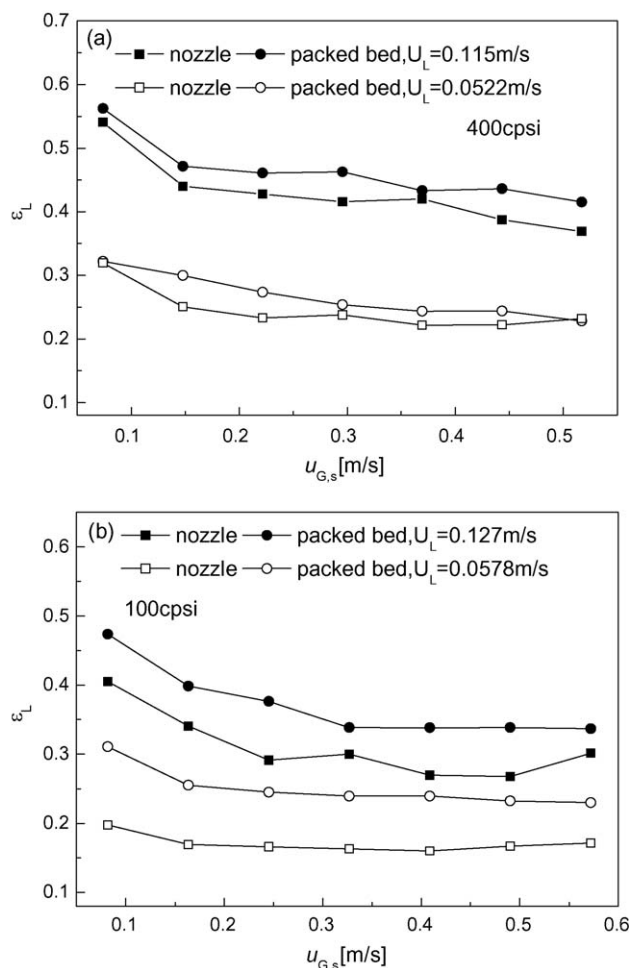


Fig. 2. Liquid hold-up as a function of the superficial gas velocity for the two monolith packings: (a) 400 cpsi; (b) 100 cpsi.

general, liquid retention in the 400 cpsi monolith packing is higher than that in the 100 cpsi bed. It is noticeable that for both the 400 cpsi and 100 cpsi monolith packings, the hold-up values for the packed bed distributor are higher than that for the nozzle distributor, especially for the 100 cpsi monolith packings the difference is more pronounced. To seek an explanation for the observations, we carried out further measurements by inserting a video probe (6 mm in diameter) of a VIDEOPROBE system (VP300, Everest VIT Inc., America) to the bottom of the packings, to monitor and take dynamic pictures there. We found that in the case of the

nozzle distributor, there existed gas-dominant channels for both types of monolith packings; shown in Fig. 3 are the typical images indicating the existence of the gas-dominant channels. And at a specified liquid velocity, a higher gas velocity ( $u_{G,s} > 0.2$  m/s) leads to an increased number of gas-dominant channels. On the contrary, such phenomena were not observed with the use of the packed bed distributor. For the gas and liquid velocities adopted in the present work, the packed bed distributor was operated in the pulsing flow regime that is characterized by the spontaneous formation of alternative gas- and liquid-rich regions which travel down the packed bed, which leads to uniform phase distribution on a monolith cross-section. This is the reason for the absence of the gas-dominant channels when the packed bed was used as a distributor.

Fig. 4 shows the measured total pressure drop for various gas and liquid velocities using the two different distributors. It is known that the measured total pressure drop consists of three parts, i.e., the wall frictional pressure drop, the static head, and the acceleration contribution. For the present case, the static head is dominant, which is in direct relation with the liquid hold-up in the bed. So the trends shown in Fig. 4 are easily explained on account of those shown in Fig. 3. On the other hand, neglecting the acceleration contribution and knowing the hold-up values, we can calculate the two-phase frictional pressure drop with the following formula

$$\frac{\Delta p_f}{L} = f_{TP} \frac{1}{2} \rho_L u_{TP}^2 \frac{4}{d_h} \epsilon_L = \frac{\Delta p_T}{L} + \rho_L g \epsilon_L \quad (1)$$

where  $f_{TP}$  is the two-phase friction factor, and  $u_{TP}$  is the sum of the superficial gas and liquid velocities.

Fig. 5 shows the friction factor calculated by Eq. (1) as a function of the two-phase Reynolds number for the two distributors and two monoliths. It is noted that the dependency of the friction factor on the Reynolds number is different for the distributors and monoliths, and the friction factors are larger for the 100 cpsi monolith and packed bed distributor than that for 400 cpsi monoliths and nozzle at the same Reynolds number.

### 3.2. Correlations for estimating pressure drop and liquid hold-up

Grolman et al. [2] considered the effect of gas-induced excess pressure drop, and correlated their data by the following relation

$$\frac{\Delta p_T}{L} = \epsilon_L \frac{64}{Re_{TP}} \frac{1}{d_h} \frac{1}{2} \rho_L (u_{L,s} + u_{G,s})^2 + 45000 u_{G,s} \quad (2)$$

Mewes et al. [8] considered that the total pressure drop consists of additionally the friction pressure drop of gas phase and the pressure drop caused by liquid vortices in the head of the liquid

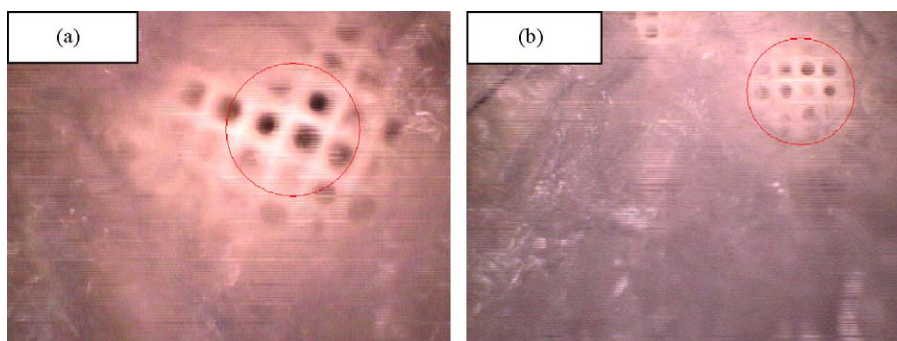
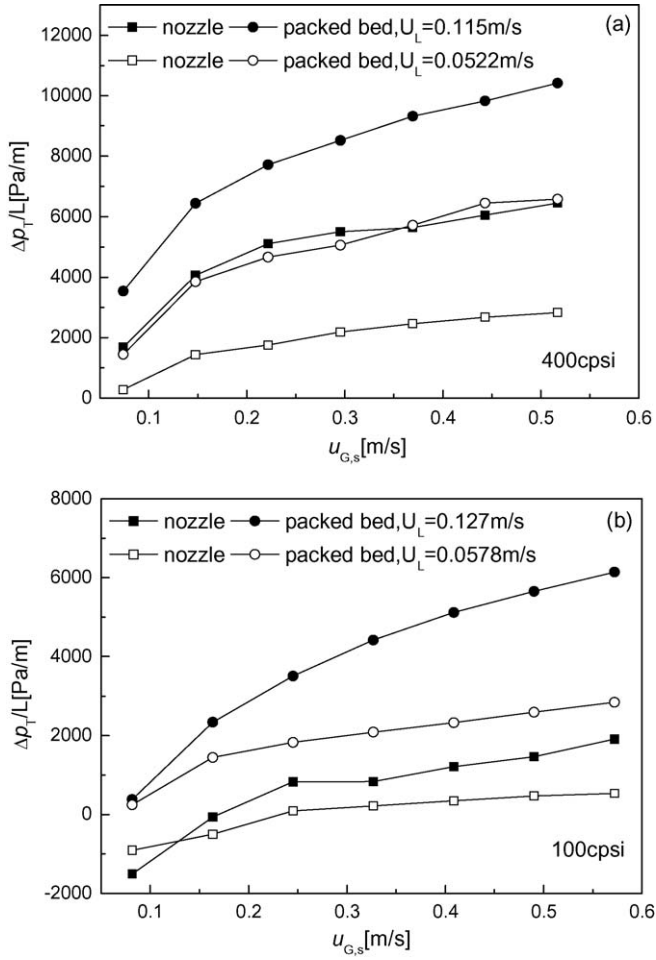
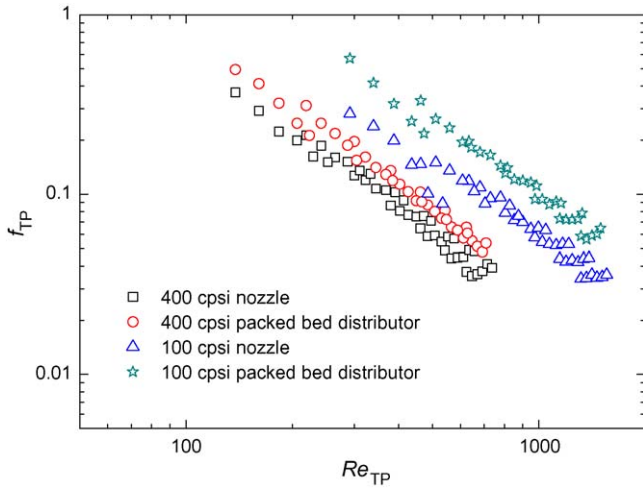


Fig. 3. Photos taken at the bottoms of the monolith packings with the nozzle distributor: (a) 400 cpsi,  $u_{L,s} = 0.073$  m/s,  $u_{G,s} = 0.443$  m/s; and (b) 100 cpsi,  $u_{L,s} = 0.081$  m/s,  $u_{G,s} = 0.409$  m/s. The red circle areas are corresponding to the gas-dominant channels. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)



**Fig. 4.** Effect of distributor type on pressure drop in two monolith packings: (a) 400 cpsi; and (b) 100 cpsi.



**Fig. 5.** The friction factor as a function of the two-phase Reynolds number.

slugs. Thus the pressure drop can be estimated from

$$\frac{\Delta p_T}{L} = -\varepsilon_L \rho_L g + 32 \mu_L \frac{u_{L,s}}{d_h^2} + 32 \mu_G \frac{u_{G,s}}{d_h^2} + \frac{\rho_L}{2} u_{TP}^2 \frac{\varepsilon_{gG}}{L_b} \quad (3)$$

$$\frac{\varepsilon_{gG}}{L_b} = \frac{(1 - \varepsilon_G) 0.15}{L_{b0}(1 - 0.15)} \quad \varepsilon_G \geq 30\% \quad (4)$$

$$\frac{\varepsilon_{gG}}{L_b} = \frac{\varepsilon_G}{L_{b0}} \quad \varepsilon_G \leq 30\% \quad (5)$$

where  $L_b$  is the length of a bubble,  $L_{b0}$  is the initial length of the bubble (0.011 m for 2 mm diameter capillary), and  $\varepsilon_{gG}$  is the volume fraction of bubbles.

Heiszwolf et al. [9] studied the hydrodynamics of the monolith loop reactor and correlated pressure drop data using the following formula

$$\frac{\Delta p_T}{L} = f_{TP} \frac{1}{2} \rho_L u_{TP}^2 \frac{4}{d_h} \varepsilon_L - \varepsilon_L \rho_L g \quad (6)$$

where  $f_{TP} = F/Re_{TP}$ , with  $F = 18$  for 200 cpsi,  $F = 22$  for 400 cpsi and  $F = 28$  for 600 cpsi. Fig. 6 shows a comparison of the total pressure drop calculated by Eqs. (2)–(6) and our measured data for the two monoliths and distributors. It is noted that all these three correlations cannot predict our measured data. In what follows, we will develop a correlation for pressure drop prediction by taking account of the effect of the liquid slug length, an important hydrodynamic parameter reported to affect significantly the gas–liquid flow and mass transfer in monolith packings [10].

Kreutzer et al. [11] studied the two-phase pressure drop under Taylor flow conditions in a capillary, and found that the friction factor was a function of liquid slug length. The correlation was expressed as

$$f_{TP} = \frac{16}{Re_{TP}} \left[ 1 + \frac{0.17}{\psi_{slug}} \left( \frac{Re_{TP}}{Ca} \right)^{0.33} \right] \quad (7)$$

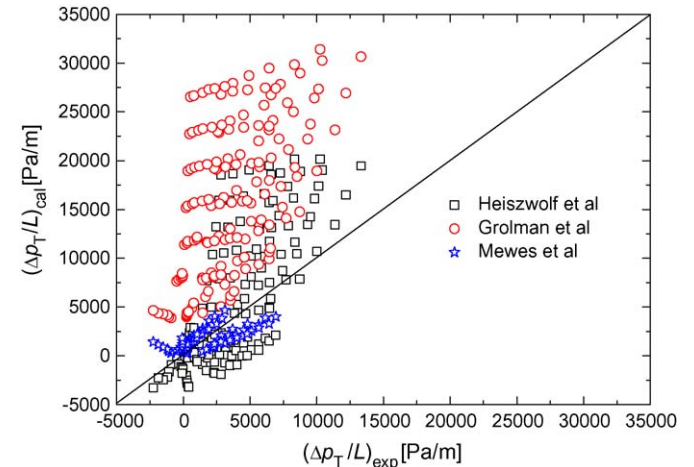
where  $\psi_{slug}$  is the dimensionless liquid slug length ( $\psi_{slug} = L_{slug}/d_h$ ) and  $Ca$  is the Capillary number ( $Ca = \mu_L u_{TP}/\sigma_L$ ). The determination of slug lengths from pressure drop by Eq. (7) in monoliths was also confirmed upon a basis of their work [12]. Kreutzer [13] described the slug length data of Heiszwolf et al. [14] in 200 cpsi monoliths using the following correlation

$$\psi_{slug} = \frac{\varepsilon_L}{-0.00141 - 1.556 \varepsilon_L^2 \ln(\varepsilon_L)} \quad (8)$$

Laborie et al. [15] studied gas–liquid flow in vertical capillaries and correlated liquid slug length data using the following formula

$$\psi_{slug} = 3451 \left( \frac{1}{Re'_G Eo} \right)^{1.2688} \quad (9)$$

where  $Re'_G$  is the gas-phase Reynolds number and  $Eo$  is the Eötvös number. Liu et al. [16] measured liquid slug length in vertical



**Fig. 6.** Comparison of the measured total pressure drop and the estimated values by various correlations.



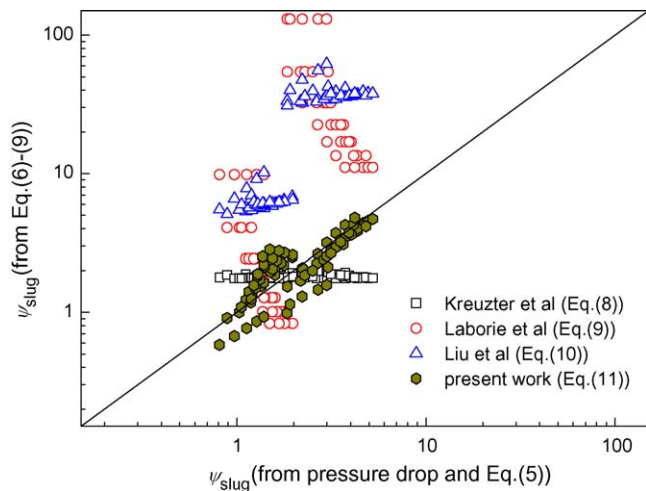


Fig. 7. Comparison of liquid slug lengths derived from the measured pressure drop and Eq. (7) and predicted slug lengths by various correlations.

capillaries and gave an empirical correlation to evaluate the liquid slug length

$$\frac{u_{TP}}{\sqrt{L_{slug}}} = 0.088 Re_G^{0.72} Re_L^{0.19} \quad (10)$$

Based on the liquid slug lengths derived from the pressure drop data for the packed bed distributor using Eq. (7) (substituting a factor 14.2 for 16 for the present square channels), the following formula is obtained:

$$\psi_{slug} = 1.449 \times 10^{-3} \left( \frac{Re_{TP}}{Eo} \right)^{0.9676} \quad (11)$$

Fig. 7 shows a comparison of the liquid slug lengths calculated with the present pressure drop data and Eq. (7) with the predicted slug lengths using Eqs. (8)–(11). Remarkably, an enormous amount of scatter is observed for the literature correlations. This suggests that the packed bed distributor with 1 mm glass beads leads to different liquid slug lengths. The friction factor with the packed bed distributor can also be evaluated using Eqs. (11) and (7) if the values of liquid slug length are known.

Because of the presence of some gas-dominant channels in the case of the nozzle distributor, the determination of liquid slug lengths would be not exact, so we only give empirical correlations of friction factor for both monoliths as a function of two-phase Reynolds number as follows

$$\text{For the 400 cpsi monoliths } f_{TP} = 399.7 Re_{TP}^{-1.411} \quad (12)$$

$$\text{For the 100 cpsi monoliths } f_{TP} = 309.5 Re_{TP}^{-1.243} \quad (13)$$

For prediction of the liquid hold-up data obtained, we adopt the drift-flux model [17] used to estimate the void fraction in single channel flows, which was used also by Roy [20] and Roy and co-workers [21] for monolith packings. The liquid hold-up correlation is given by

$$\varepsilon_L = 1 - \frac{u_{G,s}}{C_0(u_{G,s} + u_{L,s}) + v_G} \quad (14)$$

$$C_0 = 1.2 - 0.2 \sqrt{\frac{\rho_G}{\rho_L}} \quad \text{and} \quad v_G = 0.35 \sqrt{\frac{\Delta \rho g d_h}{\rho_L}} \quad (15)$$

Fig. 8 shows the comparison of the measured and the estimated liquid hold-up. We see most deviations for liquid hold-up prediction are below 20%.

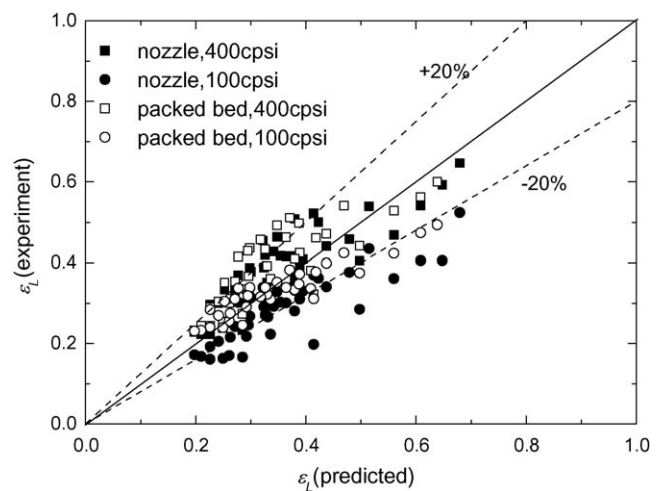


Fig. 8. The measured liquid hold-up as a function of the estimated liquid hold-up from Eqs. (14) and (15).

### 3.3. Analysis of negative pressure drop and unstable phenomena

Fig. 9 shows the measured total pressure drop for the 100 cpsi monolith with the nozzle distributor. It is noted that at velocities  $u_{G,s} < 0.2$  m/s,  $0.0578 \text{ m/s} < u_{L,s} < 0.1733$  m/s, the total pressure drop is negative, in which region, the pressure drop decreases with increasing liquid flow rates ( $\partial(\Delta p_T/L)/\partial u_{L,s} < 0$ ). This is the so-called unstable region for operation of monolith packings, in which case the liquid will continue to accelerate upon a slight increase in the superficial velocities, the perturbation will grow, and the flow will be unstable. This could be an indication of maldistribution for the two-phase flows within the monolith packings. In the present work, negative pressure drop was not observed for the 100 cpsi monolith with the packed bed distributor.

There were some reports of negative pressure drop (the pressure at the top of bed is below the pressure at bottom of the bed) in the literature [2,18,19] both for single channels and monolith packings at low gas and liquid flow rates. The static pressure drop is dominant at these conditions. Fig. 10 shows lines of constant pressure gradient calculated by Eqs. (1), (7), (11), (13) and (14) on a map of  $u_{L,s}$  versus  $u_{G,s}$  for the 100 cpsi monolith with

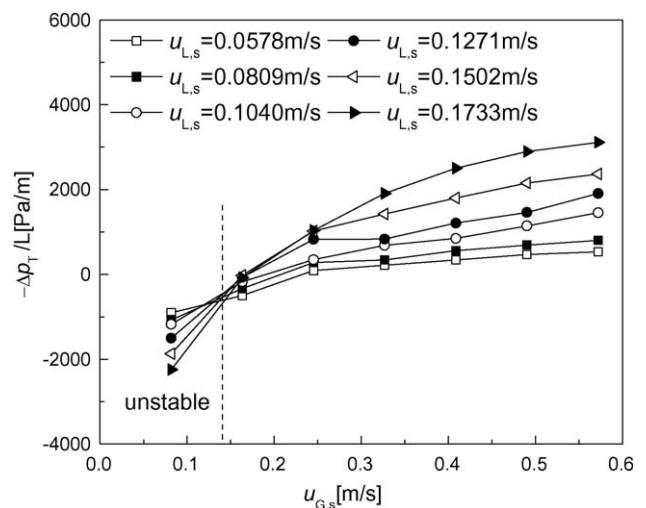
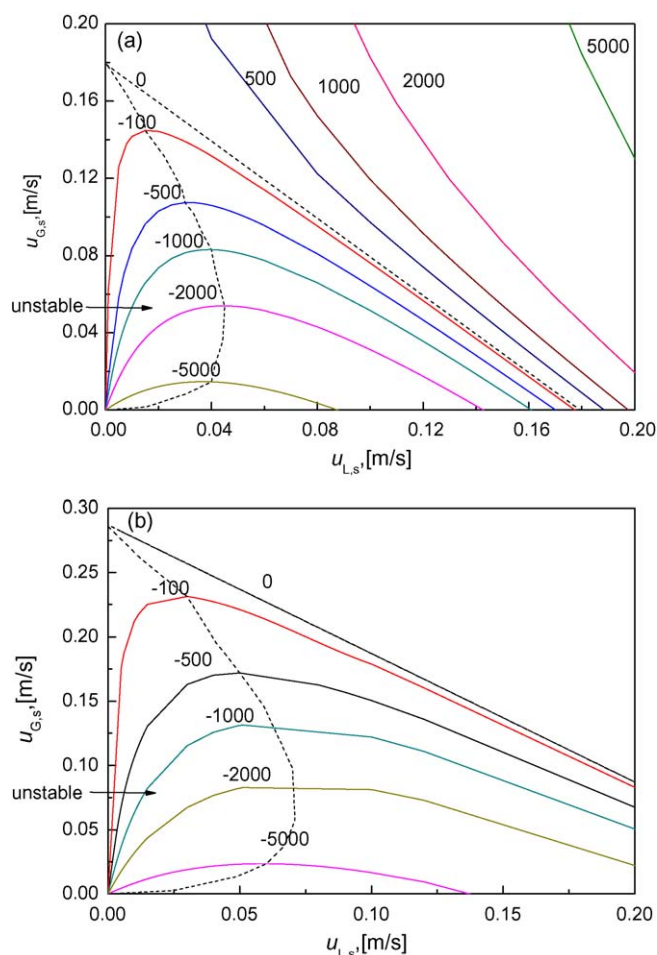


Fig. 9. The measured total pressure drop as a function of superficial gas velocity under different superficial liquid velocities, for the 100 cpsi monolith with the nozzle distributor.



**Fig. 10.** Impact of the different distributors on the unstable regime for the 100 cpsi monolith. The lines are contour lines of constant pressure drop as a function of  $u_{L,s}$  and  $u_{G,s}$ : (a) with the packed bed distributor; and (b) with the nozzle distributor.

the two types of distributors, respectively, and the unstable regions are marked. It is noted that the area of unstable region for the 100 cpsi monolith with the nozzle distributor is larger than that with the packed bed distributor due to the larger pressure drop with this device which leads to better distribution.

#### 4. Conclusions

Total pressure drop and liquid hold-up in two different monolith packings have been investigated experimentally in the co-current downward flow mode with a reactor diameter of 0.1 m and 0.6 m high monolithic packings operating in the Taylor flow regime. A comparative study of the effect of two different distributors (nozzle distributor and packed bed distributor with 1 mm glass beads) on hydrodynamics in the monoliths has been

conducted. A close relation of the distributor design to the hydrodynamics in monolith packings was observed to exist, evidenced by the larger pressure drop and liquid hold-up values for the monolith packings with the use of the packed bed distributor, when compared with the nozzle distributor. An explanation for this phenomenon is the better gas and liquid distribution capability the packed bed has. By taking account of the effect of the liquid slug lengths, correlations were derived for predicting the two-phase friction factor and liquid hold-up and satisfactorily describe the experimental data. Finally an analysis of the unstable flow phenomenon characterized by negative pressure drops within bed shows that the unstable region is not only dependent on the operating conditions and properties of monolith but also on the distributor type.

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