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A general pressure drop model based on liquid holdup of gas–liquid flow in micro-packed beds

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ABSTRACT

The understanding of the gas–liquid flow characteristics in a micro-packed bed reactor is still immature, especially for many gas–organic working systems commonly used in industry. Accordingly, this study proposes a platform to investigate the gas–liquid flow characteristics in a micro-packed bed reactor and presents a unified expression for these characteristics of both organic and aqueous liquid phase. The influence of two-phase flow rate, working solution viscosity, and packing particle size on the liquid holdup and pressure drop were studied. The gas-organic working systems results show that the liquid holdup ranges between 0.5 and 0.8 and pressure drop ranges from 50 to 350 kPa·m^{-1}. In particular, a strong correlation between the two flow characteristics parameters (liquid holdup and pressure drop) was proposed for the first time. Finally, a general pressure drop mathematical prediction model in micro-packed bed were developed.

1. Introduction

Over the past two decades, microreactors have gained considerable attention in the field of chemical engineering due to their remarkable attributions of rapid mixing, exceptional mass transfer capabilities, exceedingly large surface area, and inherent safety [1–4]. As one of the widely developed types of microreactor, the type of micro-packed bed reactors (μPBRs) provides an effective solution to address the low mass transfer efficiency in gas-liquid-solid three-phase reaction systems [5,6]. Due to the large specific surface area of μPBRs with the packing particle less than 500 μm, the use of μPBRs has become an advisable choice in various gas-liquid-solid chemical reaction process intensification, including hydrogenation [7–9] and oxidation [10,11]. Losey *et al.* [12] first invented μPBRs and reported distinct flow patterns and characteristics compared to large-scale traditional trickle bed reactors (TBRs). The difference of hydrodynamics between μPBRs and TBRs mainly depends on the Bond number (*Bo*, the ratio of gravitational force to capillary force, $Bo = (\rho_L - \rho_G)gd_s^2/\sigma$). Because the packing particle size in the μPBR is usually smaller than 500 μm; thus, the *Bo* number is far less than 1.0, and thereby the gas-liquid microflow in the μPBR is primarily dominated by capillary force and viscous force [13]. As a result, organic solutions and aqueous solutions in micro-channel exhibit very different interfacial phenomena and viscosity characteristics [14]. It is well known that extensive researches on gas–liquid hydrodynamics in TBRs have been reported [15–17]. However, there is a limited amount of literature available on the hydrodynamics (*i.e.*, liquid holdup, pressure drop, residence time distribution, *etc.*) of micro-packed bed reactors using gas-organic working solution systems. This gap significantly hinders the reliable design of μPBRs for industrial reactions. Therefore, the gas–liquid hydrodynamics in the μPBR is highly required to be studied, especially description of gas-organic system.

For the liquid holdup in μPBRs, van Herk *et al.* [18] tested the residence time distribution in the μPBR to obtain the liquid holdup, and they found that the flow profiles at the microscale is familiar with the velocity profiles of plug flow, which exhibits a narrow residence time distribution in μPBRs. Márquez *et al.* [19] measured the liquid holdup in the μPBR and found that the liquid holdup ranges from 0.65 to 0.85, which is significantly higher than the range of 0.05–0.25 in TBRs. The higher liquid holdup in the μPBR signifies the non-ideality of the gas–liquid flow, which leads to the liquid holdup being one of the important flow parameters in the μPBR. Based on a mathematic model proposed by Atton *et al.* [20] for the large-scale trickle beds, Zhang *et al.* [21] chosen the water-air working system to derive the predictive formula for the liquid holdup in the μPBR, which contains the two-phase capillary

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number (*Ca*). Furthermore, Wang *et al.* [22] used the glycerol–water and air as the working system and promoted the predictive model for the liquid holdup. However, based on the above-described works, it is evident that investigations on liquid holdup primarily focus on air-water systems, with studies on gas-organic working systems being rarely reported.

For the pressure drop in the μPBR, the most notable characteristic is that it is two orders of magnitude higher than that of traditional trickle bed reactors (TBRs) [22]. Márquez *et al.* [23] discovered that the high pressure drop in the μPBR primarily results from its large value of the liquid holdup. However, there has been no in-depth research conducted to establish a correlation between these two parameters. Sang *et al.* [24] found that replacing the packing material with foam metal in the μPBR can effectively reduce the flow pressure drop. Faridkhou *et al.* [25] reported that when the liquid flow velocity exceeds 25 $mm \cdot s^{-1}$, the interaction between the gas-liquid two phases will become much stronger and the pressure drop in the μPBR significantly increases, Therefore, in practical applications of micro-packed bed reactors, such high liquid phase velocities would not be considered. Furthermore, based on glycerol-water and air working system, Wang *et al.* [22] developed a predictive model for the pressure drop in the μPBR, which predicts the darcy friction factor (*f*) to get pressure drop, and the model includes Reynolds numbers (*Re*) of two phases, liquid-phase Weber number (*We*), particle size and contact angle.

From the literature review, it is evident that the research on the liquid holdup and pressure drop in the μPBR has been extensively studied *via* water and air working system as shown in Table 1. However, the study of the organic solution as the continue phase has been rarely reported. In microchannel gas–liquid flow research, Liu *et al.* [27] reported the different flow regimes between air–water and air–oil mixture experiment system. The study demonstrated that the experimental system with the organic solution as the continuous phase exhibited clear distinctions compared to the system with the water phase as the continuous phase. Besides, there are different experimental phenomena observed in the gas-organic working system in the μPBR, and the pressure drop and liquid holdup for methane as the continuous phase are both lower than those for water as the continue phase [21]. The result also showed the different gas–liquid flow characteristics between organic and water as the continuous phase in the μPBR. However, the study was not comprehensive and requires further exploration. Unfortunately, there is a noticeable lack of research on hydrodynamics for the gas-organic working systems at present in the μPBR, despite these systems being widely encountered in many chemical processes [28,29]. Besides, at the micro-scale, Sheng *et al.* [30] found the strong correlation between phase fraction and gas–liquid flow pressure drop in the microchannel; however, the relationship between the hydrodynamics of the liquid holdup and pressure drop in the μPBR remains unclear, and whether there exits some correlations between liquid holdup and pressure drop in the μPBR also need further exploration.

Herein, we explored the gas–liquid hydrodynamics in the μPBR, focusing on liquid holdup and pressure drop with a gas-organic system based on the Reidl–Pfleiderer process as the working system. This system includes various benzene and lipid substances, exhibiting rich characteristics such as viscosity, density, and interfacial tension. Various influencing factors on the two hydrodynamics characteristics parameters were comprehensively studied, including two-phase flow rate, initial gas–liquid pre-dispersion state, particle size, and liquid phase properties. The relationship and correlation equation between these two parameters were explored. Finally, a general mathematical model for predicting pressure drop in micro-packed beds was developed.

2. Experimental

2.1. Reagents and materials

The liquid phase used in this experiment was an anthraquinone organic solution based on the Reidl–Pfleiderer process. The composition of the working fluid in this process reflects practical industrial reaction systems, thus making the study of this system more practically significant. The solute, 2-enthylanthraquinone (EAQ), was purchased from Shanghai Titan Scientific Co., Ltd. The solvent consisted of 1,3,5-trimethylbenzene (TMB, Shanghai Titan Scientific Co., Ltd., A.R.) and trioctyl phosphate (TOP, Shanghai Titan scientific Co., Ltd., A.R.). The viscosity and surface tension of the organic solution with different volume ratio of TMP and TOP were measured by an electronic viscometer (DV-IIa + P, Brookfield, WI, USA) and interface tensiometer (OCAH200, DataPhysics Instruments, Germany), respectively. The physical properties of the organic solutions have been listed in Table 2. H_2 and N_2 (Beijing Praxair Inc., 99.995%) were used as the gas phase. All chemicals were used without any further purification.

2.2. Experimental setup and procedure

Fig. 1 shows the experimental procedure. Hydrogen gas and the organic solution were introduced into a stainless steel T-junction (Tee 1/16″, SS, VICI VALCO, USA) by a gas mass flowmeter (HORIBA Precision Instruments Beijing Co., Ltd) and a high-pressure syringe pump (Fusion 6000, Chemyx, America), respectively. Then the gas-liquid micro-dispersed flow system was transported into the μPBR. When achieving the stable condition, the flow direction of the two three-ball valves were simultaneously changed to allow high-pressure nitrogen gas to sweep through the μPBR. This process is typically performed every 30 min, with a usual stabilization time of 5 min. Longer periods of stable conditions help ensure gas-liquid flow reliability. The liquid phase was swept from the rear end and collected for subsequent weighting and analysis. The experimental setup utilizes a stainless steel μPBR, as shown in Fig. 2. The μPBR has an inner diameter of 4.6 mm and a length of 10 cm, and it was connected to a tube at the front and rear ends, each with an outer diameter of 1.6 mm and an inner diameter 1.0 mm. Commercially available frit (Applied Porous Technologies) with a pore size of 100 μm and a thickness of 1.0 mm was fixed at the front and rear of the μPBR to support the fixation of bed

Table 1
Hydrodynamic of gas–liquid flow in the μPBR

Literature	Type	Packing material	Liquid	Gas
Iliuta *et al.* [26]	Particle	Glass	Water	Air
Faridkhou *et al.* [25]	Particle	Glass	Water	Air
Zhang *et al.* [21]	Particle	Glass	Water/methanol	Nitrogen
Sang *et al.* [24]	Foam	Copper	Glycerol–water	Nitrogen
Wang *et al.* [22]	Particle	Glass/stainless	Glycerol–water	Nitrogen

Table 2
Physical properties of the organic solutions (25 °C)

Fluids	TMB:TOP	EAQ/g·L^{-1}	Density/kg·m^{-3}	Viscosity/mPa·s	Interface tension/mN·m^{-1}
1	3:1	20	881	1.56	29.32
2	3:2	20	889	2.45	28.73
3	3:3	20	892	3.22	28.35
4	3:4	20	901	4.16	29.14

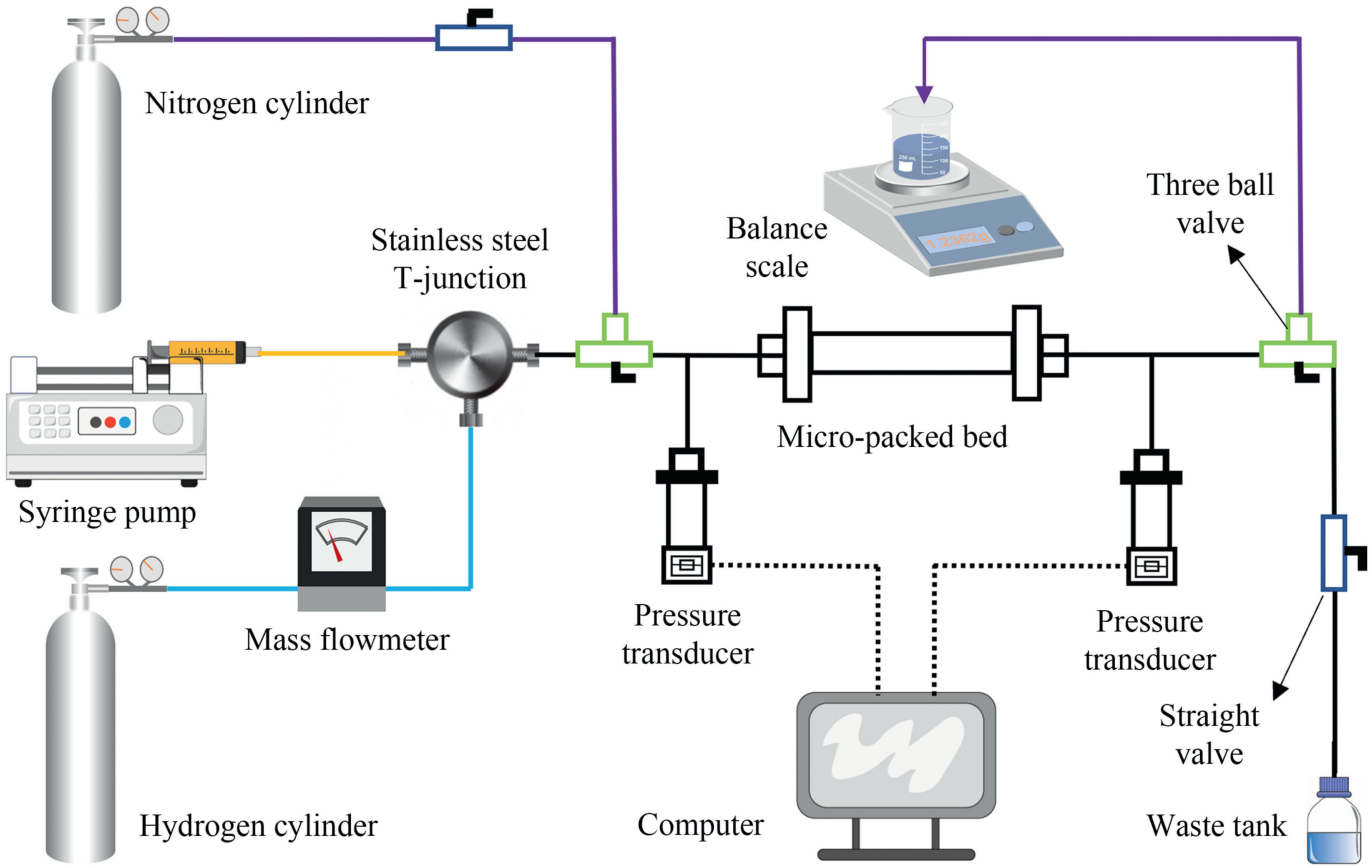

Fig. 1. Schematic of the experiment setup.

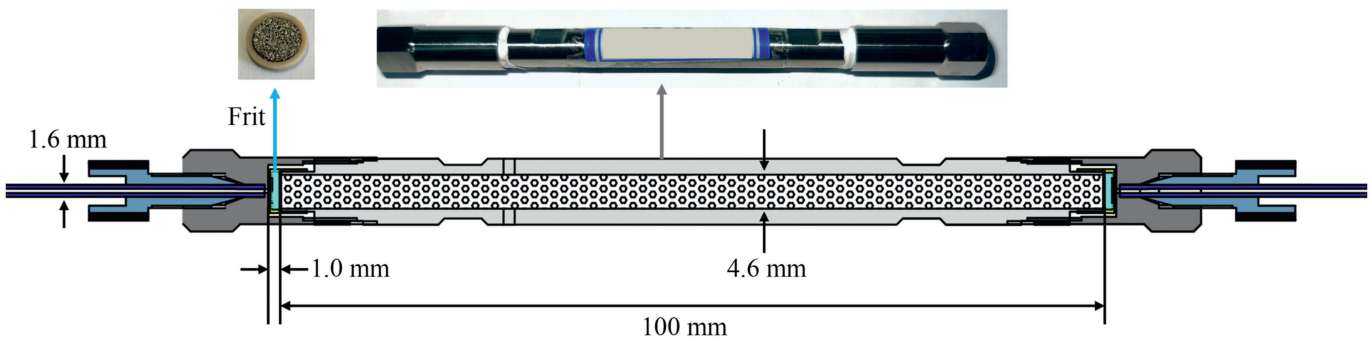

Fig. 2. Schematic of the micro-packed bed reactor.

particles. The glass microspheres (ASONE, Japanese) were used as the packing material in the μPBR and have average diameter of 400 μm (350–500 μm), 200 μm (177–250 μm), and 100 μm (95–115 μm), respectively. If no specifically indication, it means that the experimental liquid phase is TMB:TOP = 3:1, and the front-end dispersion size is 500 μm.

3. Results and Discussion

3.1. Liquid holdup and pressure drop in micro-packed bed

First, the definition and methods for obtaining of the liquid holdup and pressure drop parameters within the micro-packed bed

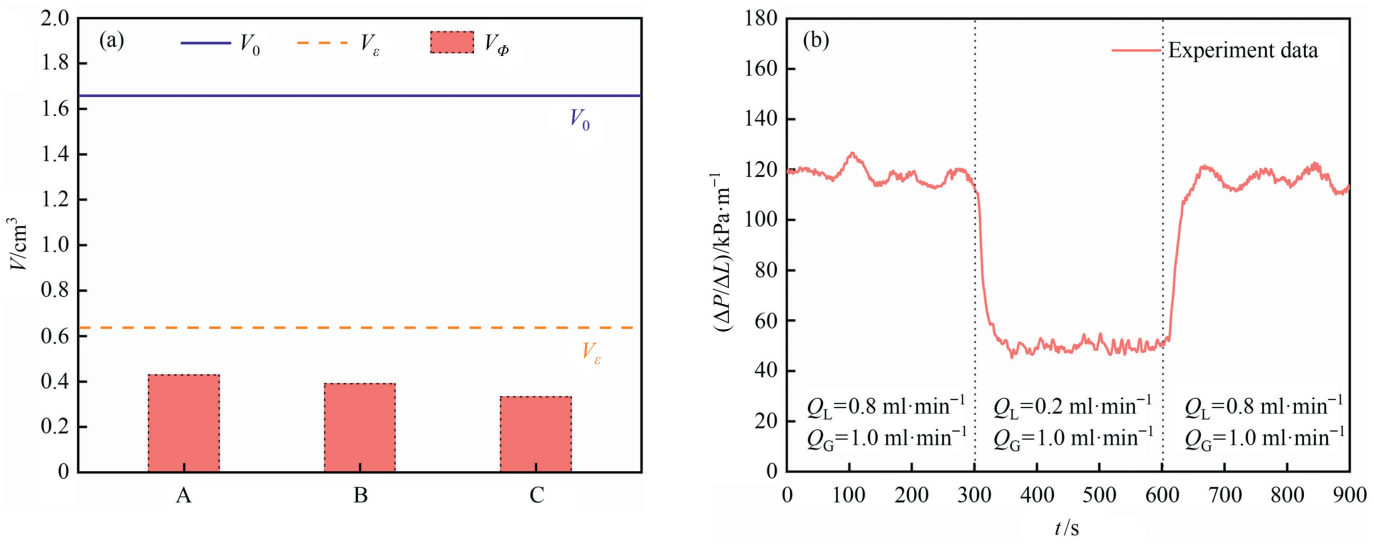

Fig. 3. Original data of liquid holdup and pressure drop in the μPBR: d_s = 400 μm, D = 4.6 mm. (a) Column volume, pore volume and liquid volume in different condition (A: Q_G = 1 ml·min^{-1}, Q_L = 1 ml·min^{-1}; B: Q_G = 3 ml·min^{-1}, Q_L = 0.6 ml·min^{-1}; C: Q_G = 6 ml·min^{-1}, Q_L = 0.2 ml·min^{-1}); (b) the pressure drop over time in different conditions.

need to be clearly specified. In the three different gas–liquid operating conditions, the empty column volume (V_0), pore volume (V_ε) and liquid in pore volume (V_Φ) are shown in Fig. 3(a). In the same μPBR, the empty column volume and pore volume remain constant. However, the volume of liquid within the packed bed changes under different experimental conditions. In this study, we first explored the liquid holdup in the μPBR by the direct measurement method, by weighting the liquid in the μPBR and getting the liquid volume (V_Φ) in the μPBR at various experiment condition according to its density. The parament of liquid holdup (Φ) equals the ratio between the liquid volume (V_Φ) to the pore volume of the μPBR (V_ε), The pore volume (V_ε) represents the effective gas-liquid flow volume within the μPBR. Compared with the indirect measurement method [21,22], which involves calculating the two-phase flow rate in the μPBR by RTDs. But the direct measurement method can get the value of liquid holdup more intuitively and eliminate interference caused by the gas–liquid flow instability. For example, when calculating liquid holdup using residence time distribution, it is easy to encounter distortions in the residence time distribution curve due to back-mixing flow. This can introduce systematic errors in the calculation of the mean residence time [21]. Moreover, the pressure drop values in the μPBR during this experiment were collected by a pressure sensor every second as shown in Fig. 3(b), and the mean pressure drop remains consistent under the same experimental conditions. It can also be confirmed that within 5 min of changing the conditions, the flow and pressure stabilize. Therefore, taking samples 30 min after changing conditions in the experiment is accurate.

3.2. Effect of variables on liquid holdup and pressure drop

3.2.1. Influence of flow rates of two phases

The average liquid holdup in the μPBR varies in the range of 0.5–0.8 and the average pressure drop varies between 50 and 300 kPa·m^{-1} in different two-phase flow velocities. The liquid holdup and pressure drop of the gas–liquid two-phase flow in the μPBR are primarily influenced by the capillary force, viscous force, and inertial force. The gravitational force could be ignored [19,22]. The viscous force and inertial force are mainly determined by two-phase flow velocity and the liquid physical properties, and the interfacial interaction between gas–liquid, liquid–solid, and gas–solid also is an important influence factor on the viscous force.

Fig. 4(a) shows the change in liquid holdup as the two-phase flow velocity varies. The liquid holdup increases with an increase in liquid flow velocity and decreases with an increase in gas flow velocity. Increasing the liquid flow rate increases the liquid flow inside the bed, occupying a larger flow space and thus increasing the liquid holdup. Conversely, increasing the gas flow rate allows gas to occupy more flow space inside the bed, thereby decreasing the liquid holdup. However, due to the significant difference in density between gas and liquid phases, the liquid holdup is more sensitive to changes in liquid flow rate. Furthermore, the gas and liquid flow velocities can be calculated by dividing the flow rate by the effective cross-sectional area in the μPBR. The calculation of gas flow velocity is based on dividing the volume flow rate measured by the gas mass flow meter at atmospheric pressure by the effective gas void cross-sectional area inside the micro-packed bed. The liquid holdup ranges from 0.5 to 0.8 in the μPBR, which is higher than the liquid holdup in TPBs ranged from 0.05 to 0.2 [23]. This can be attributed to the greater specific surface area of the particles in the μPBR and the presence of a liquid film adhering to the particle surfaces will be more than that in TPBs. Besides, the pressure drop is increased with the increase of the liquid flow velocity and gas flow velocity as shown in Fig. 4(b). It is evident that the influence of liquid flow on the pressure drop in the μPBR is more significant than that of gas flow, which is primarily due to the considerably higher density and viscosity of the liquid phase. Márquez *et al.* [23] also reported the pressure drop in the μPBR correlates strongly with the liquid-flow rate: for Q_L = 0.02 ml·min^{-1}, ΔP ~50 kPa·m^{-1}, and for Q_L = 0.1 ml·min^{-1}, ΔP ~250 kPa·m^{-1} and regardless the gas-flow rate. There is order of magnitude difference of pressure drop result of the same amount change in gas and liquid rate in the μPBR.

3.2.2. Effect of the initial two-phase dispersion states

For the previous research on the hydrodynamic in micro-channels, Sheng *et al.* [30] reported the dispersion state of the gas–liquid two-phase flow would directly affect the phase ratio and pressure drop in microchannel. Therefore, it is worth to explore whether the same phenomenon can be observed in the μPBR. The liquid holdup and pressure drop experimental date are shown in

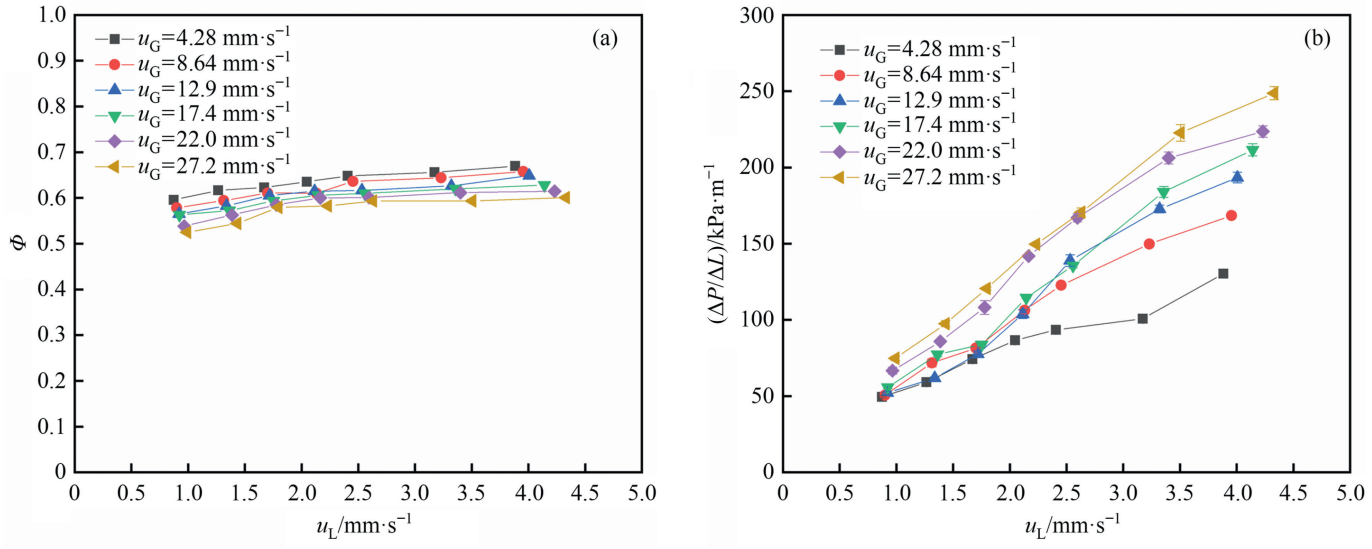

Fig. 4. Influence of the flow rates of the two phases in the μPBR: d_s = 400 μm, D = 4.6 mm, d = 500 μm, and solvent composition is TMB:TOP = 3:1. (a) The liquid holdup is affected by the two-phases flow rates; (b) the pressure drop is affected by the two-phases flow rates.

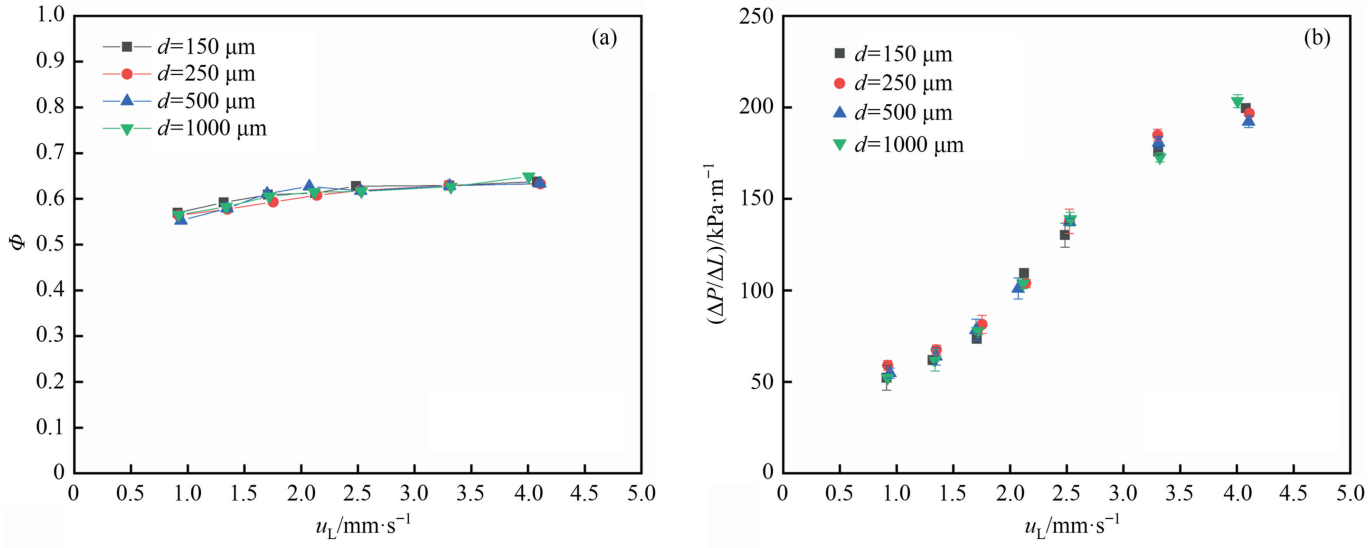

Fig. 5. Influence of the initial dispersion states at the front end of the μPBR: Q_G = 12.9 mm·s^{-1}, d_s = 400 μm, D = 4.6 mm and solvent composition is TMB:TOP = 3:1. (a) The liquid holdup is affected by the initial dispersion states; (b) the pressure drop is affected by the initial dispersion states.

Fig. 5. Our previous work [14] have systematically summarized the dispersion state of gas–liquid two-phase flow in T-junction and reported a mathematical prediction model to predict the bubble size after microdispersion. We found that the initial dispersion state of the gas–liquid two-phase flow has a minor effect on the hydrodynamics in the μPBR. The reason for this phenomenon can be explained by the redistribution of the gas–liquid two-phase flow upon entering the μPBR, and the non-ideal flow behavior within the μPBR could be the main contributing factor to this redistribution. Liu *et al.* [31] have verified the redistribution of gas-liquid two-phase flow using photographic method, and have identified four typical gas–liquid flow regimes and their characteristics, which remain consistent regardless of the initial dispersion. Therefore, for the future design of the μPBR, this finding can provide the guidance that the initial dispersion states of the gas–liquid two-phase flow is a minor factor in the μPBR hydrodynamic. Importantly, the choice of the larger tube size for gas–liquid dispersion at the front end of the μPBR can help the reduction of overall pressure drop. It should be noted that the conclusions are made based on the μPBR with a considerable length along the flow direction, and the influence of the pre-dispersion on μPBR with shorter length needs further study.

3.2.3. *Effect of the packed particle size*

The packed particle size as the primarily factor in the μPBR directly determines the hydrodynamics and mass transfer performance for gas–liquid–solid multi-phase system. The structure changes with different packed particles in the μPBR, and the significant variations in the microchannel size among the particles would obviously affect the gas–liquid flow. The porosity of the μPBR with different packed particle size was studied, and the results are shown in Fig. 6. The results show only a slight variation of

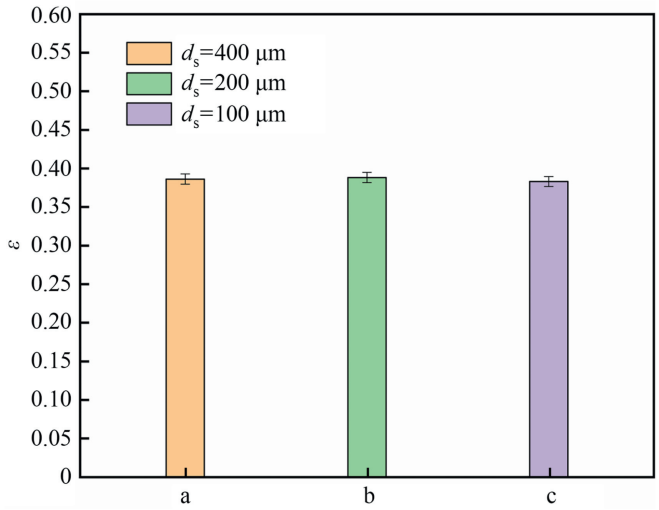

Fig. 6. The porosity for different packed particle size in the μPBR.

porosity among different particle size in the μPBR, and the porosity value is consistently around 0.38. This is attributed to the similar stacking mode, which remains consistent even when the particle size is changed. This result is also consistent with what has been reported in Ref. [21].

The variation of liquid holdup with different packed particle size in the μPBR is displayed in Fig. 7(a), which indicates that the value of liquid holdup is increased with the particle size decreases. This may be caused that the specific surface area in the μPBR increases when smaller particles are used. Furthermore, this packing state creates numerous finer pores and tortuous flow paths by using smaller particle. As a result, more liquid phase adheres to the surface of particles, and higher liquid holdup will occur. Sang *et al.* [24] reported a similar trend in the change of liquid holdup in gas–liquid flow within the μPBR, which was packed with metal foam with different pore size. Moreover, Fig. 7(b) presents the effect of the particle size on the pressure drop. The particle size significantly influences the pressure drop, and there is an observable increase in pressure drop when using smaller particles. This phenomenon may be explained as that the flow path size seriously affects the resistance of the gas–liquid flow in the μPBR. This results in the increase in liquid restriction within the small pores, leading to an enhanced capillary effect and greater resistance to gas-liquid flow through these narrow channels. Therefore, the pressure drop would be increased with the smaller particle packing. Zhang *et al.* [21] also emphasized the significant influence on the pressure drop resulting from the variation in particle size, which is attributed to the noticeable changes in interfacial tension and viscous force. Although the energy demanded in the process would be higher with the decrease of particle size, and the gas–liquid–solid mass transfer and the reaction rate would be higher to achieve process intensification [32]. It is interesting that the liquid holdup has directly proportional relationship with the pressure drop. Furthermore, Iliuta *et al.* [26] demonstrated through simulations that the high-pressure drop in μPBRs is influenced by both high liquid holdup and significant capillary forces in gas–liquid flow. This finding corroborates the relationship between liquid holdup and pressure drop. This literature also explains that pressure drop and liquid holdup are internally connected, but the details of this connection have rarely been reported. Therefore, we would like to discuss the internal connection between pressure drop and liquid holdup in Section 3.3.

3.2.4. *Influence of the liquid phase physical property*

To investigate the influence of liquid phase physical properties on the hydrodynamics, three different organic solutions and aqueous were examined as shown in Table 1. It can be observed that the significant differences in viscosity, interfacial tension, and density between aqueous and organic solutions lead to different behaviors in terms of liquid holdup and pressure drop. Therefore, hydrodynamic studies of organic solutions are essential. Besides, Zhang's experimental results indicate that in the flow of μPBR with water as the liquid phase, liquid viscosity and interfacial tension are the main influencing factors [21]. Therefore, it is necessary to study the flow patterns using viscosity and interfacial tension as parameters. The three liquid phases (Fluids 1,2,3) with different components are commonly used in actual chemical production processes. It is obvious that the viscosity has changed, but the surface tensions remain almost unchanged for the organic solutions with various

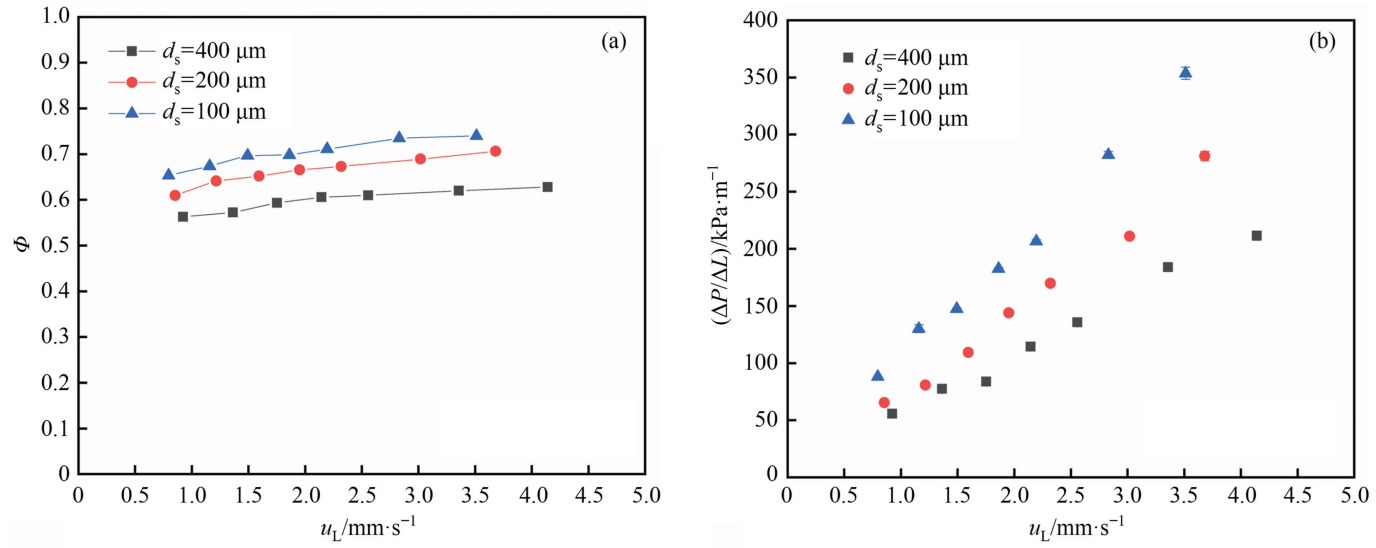

Fig. 7. Influence of the packed particle size in the μPBR: Q_G = 12.9 mm·s^{-1}, d_s = 100, 200, 400 μm, D = 4.6 mm. (a) The liquid holdup is affected by the packed particle size; (b) the pressure drop is affected by the packed particle size.

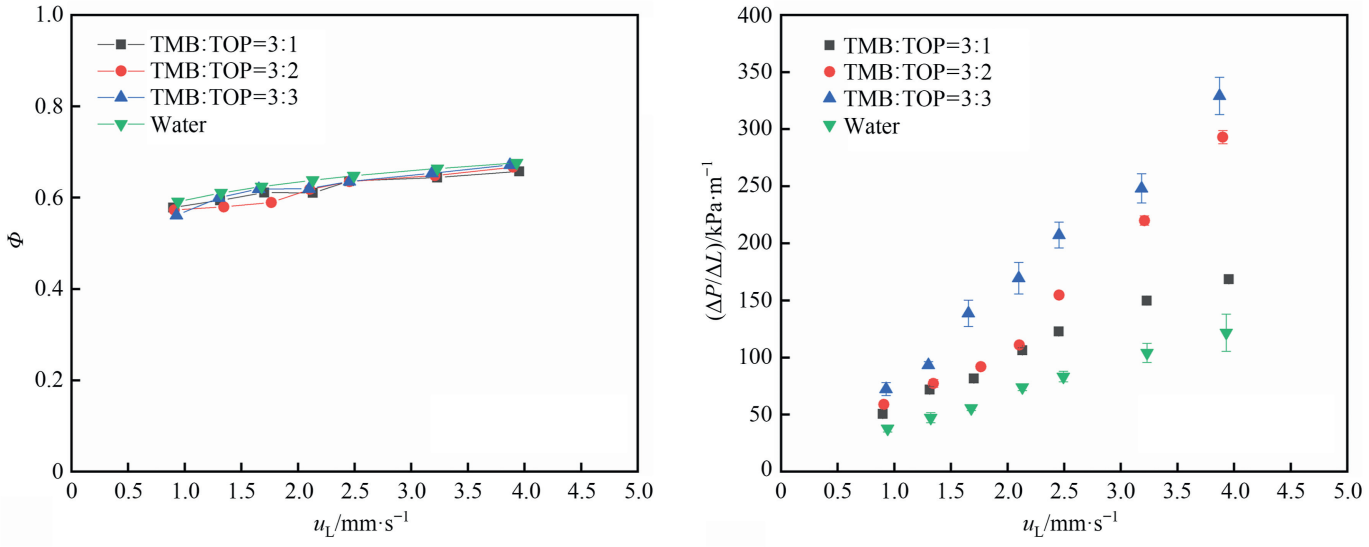

Fig. 8. Influence of the liquid physical property in the μPBR: Q_G = 12.9 mm·s^{-1}, d_s = 400 μm, D = 4.6 mm, d = 500 μm. (a) The liquid holdup is affected by the liquid physical property; (b) the pressure drop is affected by the liquid physical property.

component of TMB and TOP. Fig. 8(a) shows the effect of different component of TMB and TOP on the liquid holdup. We can find that under the same gas–liquid operating conditions, the values of liquid holdup in the μPBR almost are the same for the different components of TMB and TOP. For TBRs, the liquid holdup is almost unchanged with the increase of liquid viscosity ranging from 1.0 to 3.0 mPa·s [33]. Besides, Wang *et al.* [22] reported the liquid holdup of two types of beads increases with a decrease of liquid surface tension, for the effect of liquid surface tension, when liquid viscosity nearly remains constant at the same gas and liquid superficial velocity. Therefore, this phenomenon may be caused by the fact that the viscosity does not significantly affect the liquid spreading on the particles when the viscosity is low (in the range of 1.0–3.0 mPa·s). To further verify this result, the water with a low viscosity of 1.0 mPa·s was used as continuous phase to study the variation of the liquid holdup, and the green line in Fig. 8(a) indicates that the viscosity effect could be ignored to some extent. However, the pressure drop is obviously affected by the different component of TMB and TOP and water as shown in Fig. 8(b). The pressure drop increases with a higher TOP content in the organic solution, indicating a higher viscosity. Simultaneously, we chose the water to reduce the viscosity of the continuous phase and obtained a much smaller pressure drop value compared with the three organic solutions with higher viscosities. This result illustrates that viscosity significantly affects the pressure drop in the μPBR, which confirms that the liquid viscous force is the main influence factor on the flow resistance of gas–liquid system in the μPBR [21].

3.3. Mathematical model correlating pressure drop with liquid holdup in the μPBR

Developing an accuracy mathematical model for the prediction of pressure drop in the μPBR has been well considered in previous research, aiming to reveal the inherent principles of gas–liquid flow hydrodynamics. Moreover, there is a significant gap in the establishment of two-phase flow models for gas-organic systems. Eq. (1) is commonly used for predicting two-phase pressure drops in chemical engineering pipelines [22,24]. The μPBR can be approximately regarded as tubes filled with packing particles. The pressure drop direct correlates with the friction factor (f) under different conditions in the μPBR. Sang *et al.* [24] reported a prediction model for the pressure drop as Eq. (2), which was carried out in the μPBR with the packing of metal foam and using a gas-aqueous system. And we chose this correlation model to predict the experimental results obtained in this work. Fig. 9 shows that the correlation is failed to predict the pressure drop by Eq. (2) and the prediction values are much lower than the experimental values. The model was developed based on experiments using metal foam packing in the μPBR, which has significantly larger ranging from 0.77 to 0.98 when compared with the bed porosity of 0.38 in particle packing. Due to the larger bed porosity resulting in lower pressure drop, its predicted results are significantly lower than the actual experimental results.

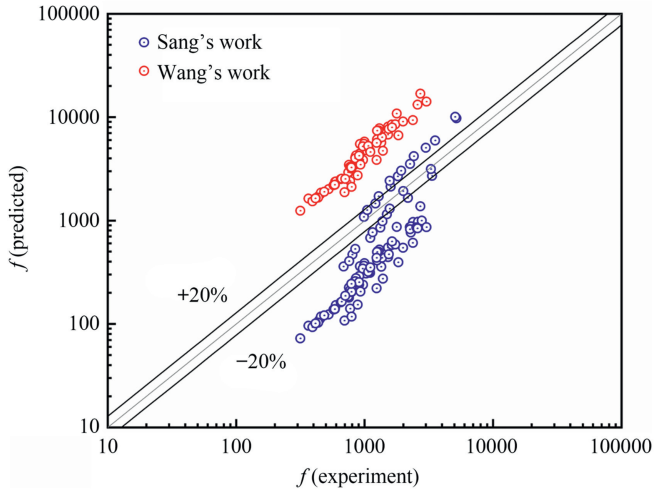

Fig. 9. Comparison of the pressure drop between the experiment data and the predicted values from literatures. (a: Sang's work based on packing of metal foam in μPBR; b: Wang's work based on glycerol-water and nitrogen system).

$$\frac{\Delta P}{L}=\frac{f\rho u_L{}^2}{2D} \tag{1}$$

$$f=1.988\times10^{-5}Re_G^{0.42}Re_L^{-0.3}We_L^{-0.59}\left(\frac{d_s}{D}\right)^{-0.99}\varepsilon^{3.12} \tag{2}$$

$$f=0.16Re_G^{0.36}Re_L^{-0.42}We_L^{-0.49}\left(\frac{d_s}{D}\right)^{-3.01}\theta^{0.6} \tag{3}$$

where $\Delta P/L$ is the pressure drop per unit length in μPBRs, u_L is the liquid flow rate in μPBRs, D is internal diameter of the micro-packed bed, d_s is the packing particle diameter, ε is the porosity in μPBRs, and θ is the contact angle between particle and liquid phase.

Besides, Wang *et al.* [22] reported a prediction model for the pressure drop as Eq. (3), which was carried out in the μPBR with the packing of particle, and the glycerol–water and nitrogen system were chosen as the working system. It can be observed that Eq. (3) also fails to accurately predict the liquid holdup values, as shown in Fig. 9. This model reflects the significant influence of high interfacial tension on the liquid holdup in We_L. Specifically, in the glycerol-water system, the gas–liquid interfacial tension is higher than that in the gas-organic phase. Therefore, Wang's prediction model overestimates the results obtained in our work. The current mathematical prediction models are unsuitable for the organic system in the μPBR. Therefore, developing a new prediction model is urgently needed.

In the previous studies, the liquid holdup and the pressure drop were discussed as two independent parameters in gas–liquid hydrodynamics in the μPBR. The liquid holdup and pressure drop exhibit a strong intrinsic correlation of gas–liquid flow within the μPBR, and this correlation has been verified through experiments and simulations [26]. Unfortunately, there are few studies that have illustrated the correlation between these two parameters. Besides, many empirical correlations for pressure drop can be found in literature for large-scale TBRs [15], but such correlations are scarce for μPBRs. Therefore, developing a general pressure drop model for micro-packed beds based on flow characteristics such as liquid holdup is urgently needed.

The primary factors influencing pressure are packing particle size, interfacial tension, inertial and viscous force, as discussed in the Section 3.2. Therefore, we use the liquid phase Reynolds number (Re_L) and gas phase Weber number (We_G) to correlate the friction factor of pressure drop. Furthermore, we also used the value of liquid holdup to correlate the friction factor due to the strong correlation. And the effect of packing particle size can also be reflected in the model through the parameter entry of liquid holdup. From the experimental results, it can be seen that the pressure drop in μPBRs, the most significant influences are the gas-liquid flow state and the channel size. The impact of channel size on the flow can be reflected in the liquid holdup. Therefore, incorporating the liquid holdup into the fitted model for pressure drop is very meaningful. Moreover, the pressure drop and the liquid holdup have been correlated through experimental data in Fig. 10(a) in our work, and we summarized the correlation in Eq. (4). The predictive correlation model exhibits a strong ability to forecast the friction factor, representing a pressure drop error within 20%. Besides, we used another liquid component, TMB:TOP = 4:3, to verify the prediction ability of the model. To investigate the universality of the model, we further compared Zhang's [21] experimental results with the model's predictions, which tests whether the model is applicable to the gas–water system. The verification results are presented in Fig. 10(b), and the prediction error remains below 20%, even when applying the gas–liquid flow data from water–gas flow in the μPBR. The correlation prediction model for liquid holdup and pressure drop can offer valuable insights for the industrial applications of μPBRs.

$$f=218.6Re_L{}^{-0.92}We_G^{0.1}\Phi^{-2.37} \tag{4}$$

where Re_L is Reynolds number of liquid phase, We_G is Weber number of gas phase, Φ is the liquid holdup in the micro-packed bed.

However, the liquid holdup (Φ) used as a measured value in this model, whereas we aim to make it predictable through a formula by operational parameters in actual application. However, the research on predictive models for liquid holdup is limited, especially for micro-packed bed systems [24]. Therefore, developing an accuracy mathematical model for the prediction of liquid holdup in the μPBR is necessary to reveal the inherent principles of gas–liquid flow hydrodynamics. Therefore, we need to further consider establishing a suitable liquid holdup prediction model to help improve the pressure drop model. Because the interfacial tension between

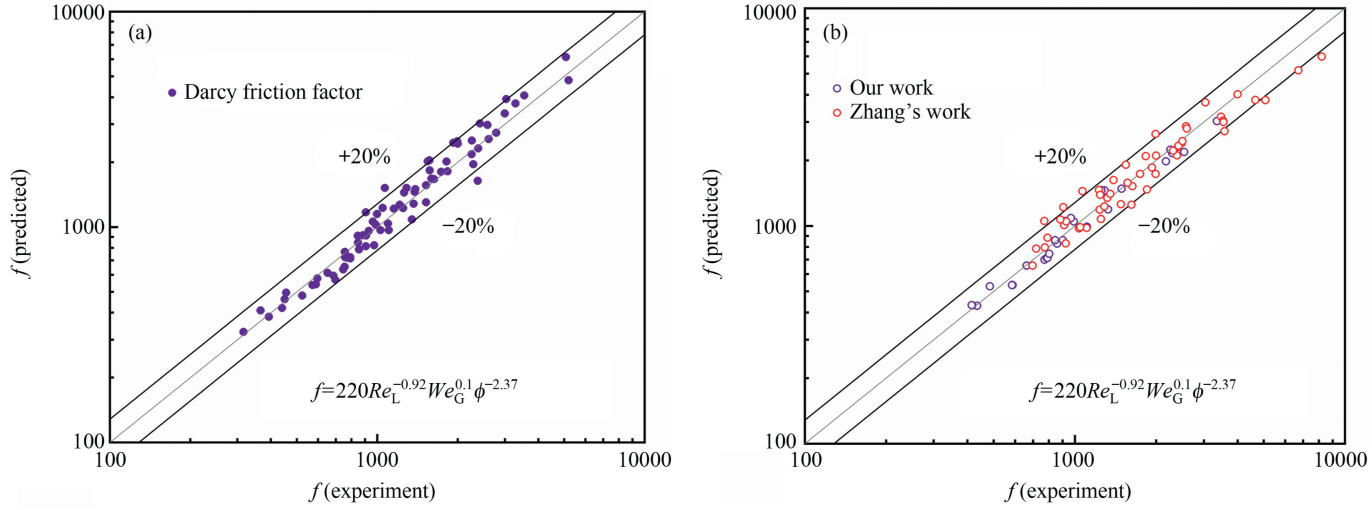

Fig. 10. Comparison of the experiment data and predicted values. (a) The predicted pressure drop based on Eq. (5), and (b) the predicted pressure drop for liquid component with TMB:TOP = 3:4 and other models from reference.

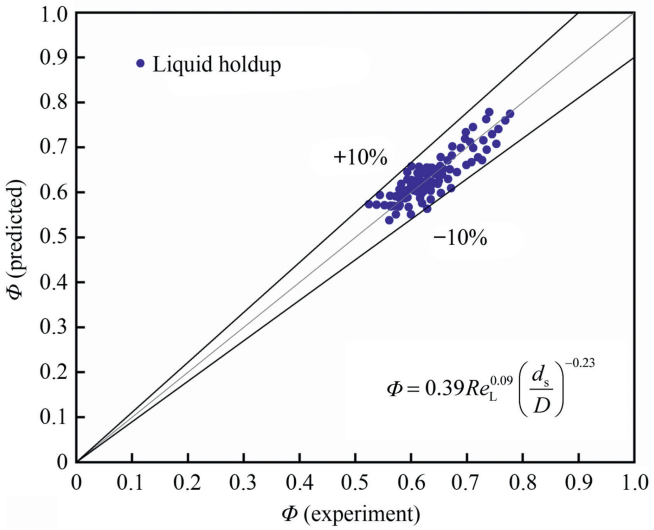

Fig. 11. Comparison of the experiment data and predicted values of liquid holdup based on Eq. (5).

different organic phase components and gas is low and the experiences minimally change, the primary factors influencing the liquid holdup are inertial and viscous forces. Therefore, we use the liquid phase Reynolds number (Re_L) to correlate the value of liquid holdup. Additionally, particle size plays a significant role in determining the liquid holdup in the μPBR and the parameter of (d_S/ D) was used as the equivalent diameter of packing particle to correlate the liquid holdup. For the liquid holdup in micro-packed beds, the scale effect produced by the channel size and the flow state influenced by the flow rate can both be reflected in the model. Hence, the correlation model presented as Eq. (5) is illustrated, and the corresponding correlation results are shown in Fig. 11. The Equation shows the good predictive ability for the liquid holdup of the gas-organic system, and the mathematical model can perfectly predict the value within ±10%. Due to the requirement of gas–liquid flow ratio in Reidl–Pfleiderer process, the model can accurately predict the liquid holdup in the μPBR and the accuracy cannot be guaranteed when gas–liquid flow ratio is greater than 30 or less than 1.

$$\Phi = 0.39 Re_L^{0.09} \left(\frac{d_s}{D}\right)^{-0.23} \quad (5)$$

where Re_L is Reynolds number of liquid phase, D is internal diameter of the micro-packed bed and d_s is the packing particle diameter.

4. Conclusions

In this work, the pressure drop and liquid holdup in micro-packed bed reactors have been investigated with the anthraquinone organic solution and hydrogen as the liquid phase and the gas phase respectively. A unified expression of gas–liquid flow characteristics, including pressure drop and liquid holdup in μPBRs, has been reported for the first time based on an organic-gas system. Furthermore, the effects of the gas–liquid two-phase flow rate, initial two-phase dispersion states, viscosity of the liquid phase, and the size of packing particle in the micro-packed bed reactor were studied. The work has filled the research gaps of gas–liquid flow characteristics in μPBRs based on organic-gas system. Based on these findings, the scaling laws for liquid holdup and pressure were established. We have found that the correlation scaling laws of the liquid holdup and pressure drop in the micro-packed beds could be summarized as a general formula, and this model can also predict the results obtained in other works. In addition, the liquid holdup could be summarized as an equation which calculate by Re_L and particle diameter. This work offers valuable design guidance for gas-liquid chemical synthesis processes in micro-packed bed reactors which based organic liquid system. In future work, microreactor design and operation can be optimized based on well-established hydrodynamics in various chemical synthesis processes.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT Authorship Contribution Statement

Junjie Wang: Writing – original draft, Methodology, Investigation, Formal analysis. Lin Sheng: Validation, Data curation. Jiang Deng: Validation. Guangsheng Luo: Writing – review & editing, Validation, Conceptualization.

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Nomenclature

a	determining factor for surfactant addition
Ca	capillary number
D	internal diameter of the micro-packed bed, μm
d	front end dispersion size, μm
d_s	packed particle diameter, μm
f	gas–liquid friction factor
L	the micro-packed bed length, m
P	the pressure drops, kPa
Q	rate of flow, ml·min^{-1}
Re	Reynolds number
t	experimental duration, s
u	total velocity in the channel, mm·s^{-1}
V_0	column volume, m^3
V_ε	pore volume, m^3
V_Φ	liquid volume, m^3
We	Weber number
μ	viscosity, mPa·s
σ	surface tension, mN·m^{-1}
ρ	density, kg·m^{-3}
θ	contact angle, (°)
ε	porosity
Φ	liquid holdup

Subscript

s	glass sphere
G	gas (hydrogen)
L	liquid (working solution)

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