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Correlations for dynamic liquid holdup under pulsing flow in a trickle-bed reactor

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Abstract

Based on theoretical analysis and experimental results, a new attempt has been made to characterize the dynamics of the fluid flowing under conditions of pulsing flow in a trickle-bed reactor (TBR). Two kinds of correlation are proposed for the dynamic liquid holdup under pulsing flow, which can predict the dynamic liquid holdup for a given packing type and given operating conditions.

Keywords: Dynamic liquid holdup; Pulsing flow; Trickle-bed reactors

1. Introduction

Trickle-bed reactors (TBRs) of industrial dimensions generally are operated in the trickling regime, but more frequently for petrochemical reactions like hydrodesulphurization and hydrotreating pulsing flow conditions are reached. Many researchers [1–3,5,6,8,12] have studied the inception of pulsing flow through hydrodynamic experiments and mathematical models. A variety of factors, such as packing shape, surface roughness, dimension and wettability as well as gas–liquid interface tension and velocity affect the inception of a pulse. The dynamic liquid holdup is an important parameter in designing a TBR. It is not only related to the pressure drop and the mean residence time of liquid phase, but is also an important parameter for safety when there is a strong exothermic reaction in the TBR. Its value reflects the degree of wetting of the catalyst particles to some extent, and is related to the liquid film thickness around a particle, which affects the mass transfer of the gaseous reactant through the liquid and into the catalyst. It has been shown by various authors [4,7,10] that the liquid holdup is dependent on the flow regime, but few correlations except those of Blok and coworkers [11,12], Ellman et al. [13] and Tsochatizidis and Karabelas [14] are concerned with the dynamic liquid holdup under pulsing flow (so-called high interaction flow regime). Although actually the local liquid holdup in a TBR varies with time and position, to obtain the bed averaged liquid holdup is necessary for engineering application. The purpose of this paper is to provide more experimental holdup data and correlations based on consideration of the fluid flowing inside the narrow channels.

2. Experimental setup and procedure

A diagram of the experimental installation is shown in Fig. 1. The dynamic liquid holdup is determined by the draining method. The reactor has an inner diameter of 0.10 m and is packed to a height of 1.0 m. Packing materials are non-porous ceramic and glass beads with a diameter of 0.002–0.003 m. Magnetic valves are positioned both at the entrance and the exit of the reactor, which can be shut or opened simultaneously so as to collect the whole bed's liquid holdup, with a draining time more than 30 min. The experiments are specially designed to minimize the entrance effect. Prior to each experimental run a similar procedure to that of Blok and coworkers [11,12] and Wammes et al. [9] is adopted — the packing materials are prewetted. Operating the reactor at high liquid flow rate, ensures that the flow regime is that of pulsing flow. For each kind of packing material, we varied the liquid viscosity and measured the dynamic liquid holdup at different gas and liquid superficial velocities. A point conductance probe is set at a certain axial position in the column and the local conductance which is related to the local liquid holdup is recorded. The measured conductance divided by the maximum conductance

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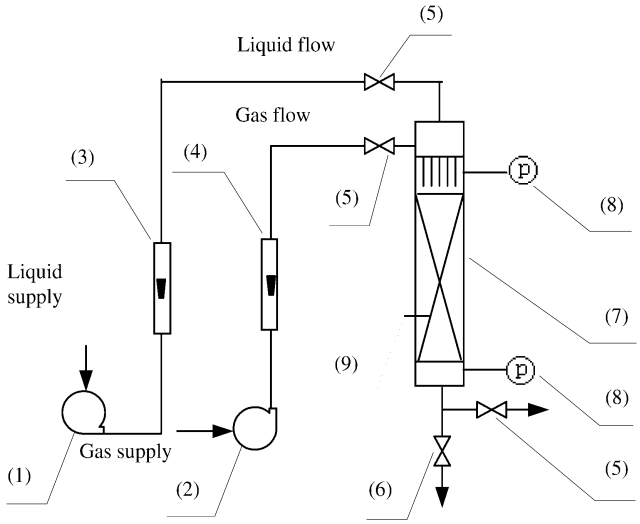

(1) Liquid pump (2) Gas compressor (3) Liquid rotameter

(4) Gas rotameter (5) Electro-magnetic valves ; (6) Drainge valve
(7) Trickle-bed reactor (8) Pressure manometer (9) Local probe

Fig. 1. Schematic diagram of dynamic liquid holdup measuring apparatus.

measured at liquid full condition represents the fraction of the channel area occupied by the flowing liquid [14]. The range of the liquid superficial velocity is 0.008–0.016 m/s and the gas superficial velocity varies between 0.07 and 0.36 m/s in our experiments.

3. Mechanics of pulsing flow in TBR

Ng's trickle-pulse transition model [6] postulates the fact that the critical gas interstitial velocity needed for flow regime changing from trickling flow to pulsing flow is a certain value, which is related to the gas–liquid surface tension, liquid phase density, gas phase density and structure of the fluid channel. A lot of experiments reveal the phenomenon that the flow regime transition between trickling flow and pulsing flow always takes place firstly at the bottom of the reactor when increasing the gas and liquid flow rates. The relatively higher gas velocity at the bottom of the reactor owing to the pressure decrease can account this for. As the flow rates increase the inception zone moves up the column. At higher gas velocity 'waves' form on the interface of the liquid film, liquid blocks the flow channel, and pulsing arises [8]. We assume that the occurrence of pulsing in TBRs can be accounted for by two necessary conditions: sufficient liquid flux and compressibility of the gas phase. For a given packing structure and liquid–solid surface properties, the ratio of gas to liquid interstitial velocity is the main factor that affects the characteristics of pulsing flow. Based on this assumption the liquid holdup is correlated to the ratio of the gas to liquid interstitial velocity.

4. Experimental results and discussion

We obtain the characteristics of liquid holdup in the time domain using the conductance method. One trace of the time series of the liquid holdup is shown in Fig. 2. In Fig. 2 the liquid holdup varies rapidly with time. The liquid arriving at the probe leads to an increase in the conductance, but conductance decreases rapidly when a gas bubble penetrates the probe.

The influence of the liquid viscosity, packing material, gas superficial velocity and liquid superficial velocity on the liquid holdup have been investigated. The results are shown in Figs. 3(a)–(d), 4(a) and (b). Fig. 3 shows the effect of both gas superficial velocity and liquid superficial velocity on dynamic liquid holdup. The gas superficial velocity has a strong effect on the liquid holdup in the pulsing flow regime,

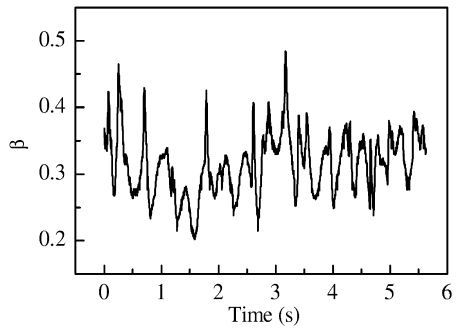

Fig. 2. Time series of liquid holdup measured using a point conductance probe. U_g=0.2832 m/s, U_l=0.00885 m/s.

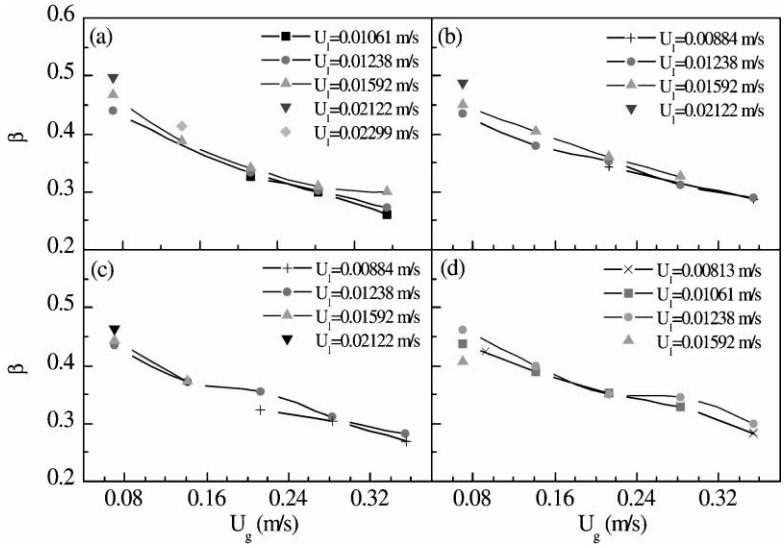

Fig. 3. Dependence of dynamic liquid holdup on the gas and liquid superficial velocity. The trickle-bed reactor (TBR) is packed with 2–3 mm ceramic beads. (a) Water–air system; (b) 0.15% CMC solution–air system; (c) 0.22% CMC solution–air system; (d) 0.30% CMC solution–air system.

which has also been observed by other groups [11,12,14]. The dependence of liquid holdup on liquid superficial velocity is also shown in Fig. 3. For a given gas superficial velocity, the liquid holdup increases slightly on increasing the liquid superficial velocity. From Fig. 3(a)–(d), at a relatively low gas superficial velocity the dynamic liquid holdup is more sensitive to the liquid superficial velocity than at higher gas velocity. With relatively low interaction between the two flowing phases, the liquid viscosity has more effect on the dynamic liquid holdup, especially under trickling flow [7]. Fig. 4 indicates that the liquid viscosity has little effect on the liquid holdup under pulsing flow conditions compared with the effect of gas superficial velocity. This is because with strong interaction the inertial force becomes the major factor affecting the behavior of fluid traversing the channels between particles. Increasing the liquid viscosity by a factor of two, the liquid holdup increases less than 4% in our experiments. Data plotted in Fig. 5 shows that the packed materials affect the dynamic liquid holdup significantly, where the abscissa is the ratio of the interstitial velocity of gas to that of liquid. This may be accounted for by the difference of the surface roughness of the two kinds of materials, which makes the bed porosity different. The ceramic beads packed bed has higher bed porosity.

5. Correlations of dynamic liquid holdup under pulsing regime

There are a great number of empirical equations correlating the dynamic liquid holdup with some dimensionless parameters for the reactor operated at atmospheric pressure in the trickling regime [7]. In the trickling flow regime the dynamic liquid holdup is strongly affected by the liquid superficial velocity, but the gas superficial velocity has little influence on the liquid holdup. Some researchers correlated

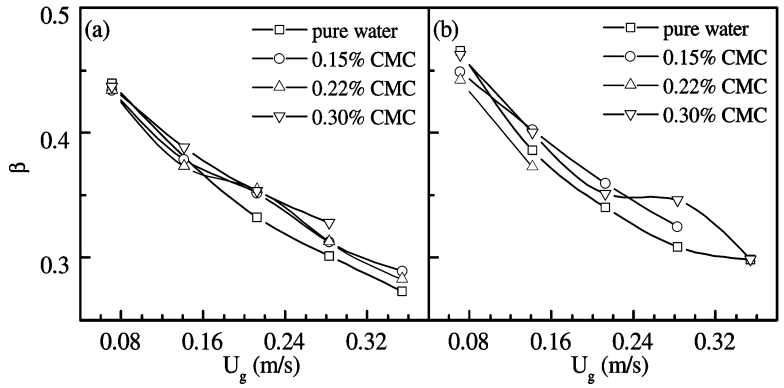

Fig. 4. Dependence of the dynamic liquid holdup on the liquid viscosity. (a) U_l=0.01238 m/s; (b) U_l=0.01592 m/s. 0.15% CMC, viscosity=1.565 cP; 0.22% CMC, viscosity=1.842 cP; 0.30% CMC, viscosity=2.628 cP; viscosity of pure water is 1.0 cP.

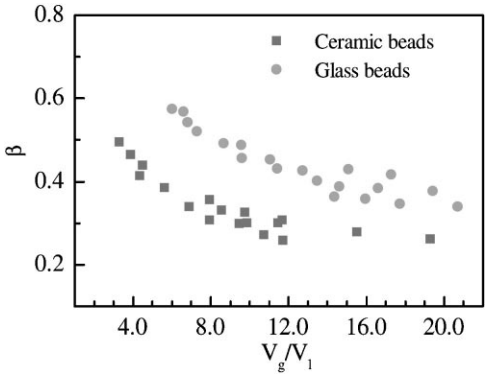

Fig. 5. Effect of packed materials on the dynamic liquid holdup.

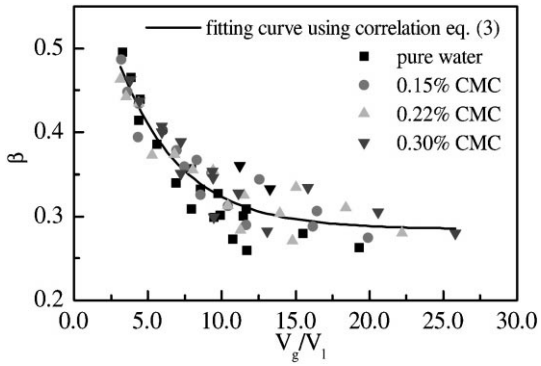

Fig. 7. Empirical correlation of dynamic liquid holdup to the ratio of gas to liquid interstitial velocity according to Eq. (3).

dynamic liquid holdup with the liquid Reynolds number [9]. In view of the mechanism of pulsing flow in a packed bed, both the gas and liquid flowing velocities have an obvious effect on the dynamic liquid holdup. It seems reasonable to correlate β with the ratio Re_g/Re_l using the following equation:

$$\beta = a + b\left(\frac{Re_g}{Re_l}\right)^c \quad (1)$$

In Eq. (1) the interstitial velocity is used to calculate the gas and liquid Reynolds number.

Modifying Eq. (1) we obtain

$$\beta = A_1 + A_2\left(\frac{V_g}{V_l}\right)^{A_3} \quad (2)$$

where A_1, A_2, A_3 are the fitting parameters. We use a non-linear estimation method to correlate the experimental data in the form of Eq. (2) and the correlated curves are also plotted in Fig. 6, where we have used the parameters A_1=0.2277, A_2=0.6766, A_3=−0.8358. 86.27% of the variance is accounted for and the correlation coefficient R=0.9288. The data points in Fig. 6 indicate that β decreases with V_g/V_l as an exponential function. We deduce the following correlation for which the correlation curve is plotted in Fig. 7.

$$\beta = y_0 + B_1 e^{-(V_g/V_l - x_0)/t_1} \quad (3)$$

where y_0, B_1, x_0, t_1 are fitting parameters. Parameter y_0=0.2847, B_1=0.3732, x_0=03758, t_1=4.2413. The proportion of the variance accounted for is 86.79% and the correlation coefficient R=0.9316.

Blok et al. considered that only the gas interstitial velocity and the solid specific area affected the liquid holdup [12]. The correlation equation was given as:

$$\frac{\beta_{Blok}}{\varepsilon} = 4.48 \times 10^{-2} \times \left(\frac{S}{V_g}\right)^{0.265} \quad (4)$$

In their papers the liquid holdup β_{Blok} is based on the whole bed volume. We have to reduce β_{Blok} by the bed porosity to find the liquid holdup which has the same meaning as β used in this paper. For non-porous spheres the specific area $S=(6/D_p)\times(1-\varepsilon)\,m^2/m^3$ bed volume, then Eq. (4) can be expressed as

$$\beta = a_1(V_g)^{-b_1} \quad (5)$$

where a_1=0.32, b_1=0.265. The dotted line in Fig. 8 is the calculated curve using these two parameters. To evaluate our experimental data the correlation form Eq. (5) is used and the solid line is the best-fit curve. The correlated parameters a_1=0.30, b_1=0.318.

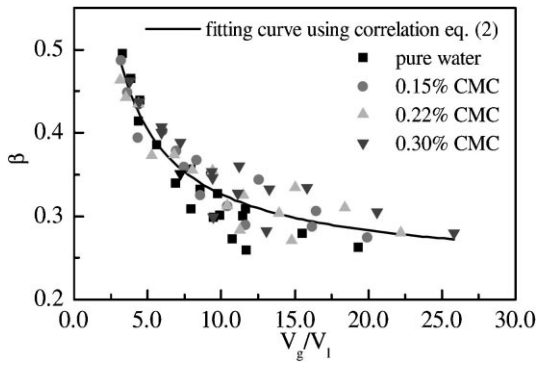

Fig. 6. Empirical correlation of dynamic liquid holdup to the ratio of gas to liquid interstitial velocity according to Eq. (2).

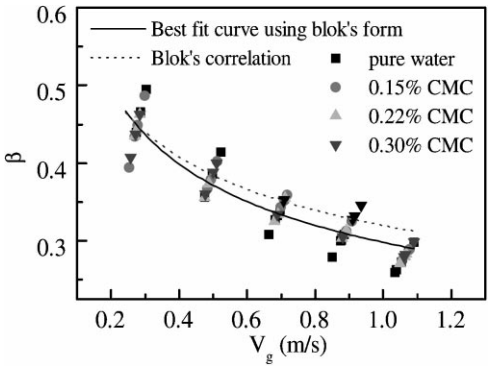

Fig. 8. Comparison of experimental data with Blok's correlation. Dot line: calculated using Eq. (4); solid line: best-fit curve using Eq. (5).

In Fig. 8 the data scatter around the best-fit curve. This can be explained by the neglect of the effect of the liquid velocity. Finally it should be noted that the gas superficial velocity in our experiments is much lower than that of Blok, so the gas–liquid interaction in our experiments may be termed mild pulsing flow. Blok's correlation equation predicted a higher liquid holdup as the liquid interstitial velocity increases. Other pulsing characteristics such as pulsing frequency, length of a pulse, ratio of liquid holdup in liquid rich zone in a pulse are also affected significantly by the ratio of the gas to liquid interstitial velocity. The relationships between these parameters with the interstitial velocity are discussed elsewhere.

6. Conclusion

Two correlations are obtained as

$$\beta = 0.2277 + 0.6766\left(\frac{V_g}{V_l}\right)^{0.8358} \quad (6)$$

and

$$\beta = 0.2847 + 0.3732e^{-(V_g/V_l - 0.3758)/4.2413} \quad (7)$$

for the ceramic beads. The first correlation equation has a more obvious physical basis but the second one correlates the experimental data better. Blok's correlation can be used to calculate the liquid holdup but it predicts higher liquid holdup than the experimental data as the gas interstitial velocity increases.

7. Nomenclature

a, b, c	fitted parameters defined in Eq. (1)
a_1, b_1	fitted parameters defined in Eq. (5)
A_1, A_2, A_3	fitted parameters defined in Eq. (2)
B_1, x_0, y_0, t_1	fitted parameters defined in Eq. (3)
D	diameter (m)
U	superficial velocity (m/s)
V	interstitial velocity (m/s)
Re	Reynolds number, based on the interstitial velocity
S	specific area of the packed materials (m^2/m^3)

Greek letters

β	bed averaged dynamic liquid holdup based on the voidge volume
β_{Blok}	symbol used by Blok to represent the liquid holdup in ref. [12]
ε	bed voidage
μ_l	viscosity (Pa s) (or cP)

Subscript

g	gas phase
l	liquid phase
p	particle

References

[1] Sicardi, G. Baldi, V. Specchia, Chem. Eng. Sci. 35 (1980) 1775.
[2] D.A. Krieg, J.A. Helwick, P.O. Dillon, M.J. McCready, AIChE J. 41 (1995) 1653.
[3] J.A. Helwick, P.O. Dillon, M.J. McCready, Chem. Eng. Sci. 47 (1992) 3257.
[4] J.C. Charpentier, M. Favier, AIChE J. 21 (1975) 1213.
[5] K. Grosser, R.G. Carbonell, S. Sundarenson, AIChE J. 34 (1988) 1850.
[6] K.M. Ng, AIChE J. 32 (1986) 115.
[7] A. Gianetto, V. Specchia, Chem. Eng. Sci. 47 (1992) 3197.
[8] Sicardi, H. Gerhard, H. Hoffman, Chem. Eng. J. 18 (1979) 173.
[9] W.J.A. Wammes, J. Middelkamp, W.J. Huisman, C.M. DeBaas, K.R. Westerterp, AIChE J. 37 (1991) 409.
[10] S. Fukushima, K. Kusaka, J. Chem. Eng. Jpn. 10 (1977) 461.
[11] J.R. Blok, A.A.H. Drinkenburg, Chem. Eng. J. 25 (1982) 89.
[12] J.R. Blok, J. Varkevisser, A.A.H. Drinkenburg, Chem. Eng. Sci. 38 (1983) 687.
[13] M.J. Ellman, N. Midoux, G. Wild, A. Laurent, J.C. Charpentier, Chem. Eng. Sci. 45 (1990) 1677.
[14] N.A. Tsochatizidis, A.J. Karabelas, AIChE J. 41 (1995) 2371.