
01 Jan 1998

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M. (Muthanna) H. Al-Dahhan

Missouri University of Science and Technology, aldahhanm@mst.edu

M. R. Khadilkar

Y. Wu

M. P. Duduković

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Recommended Citation

M. H. Al-Dahhan et al., "Prediction of Pressure Drop and Liquid Holdup in High-Pressure Trickle-Bed Reactors," *Industrial and Engineering Chemistry Research*, vol. 37, no. 3, pp. 793 - 798, American Chemical Society, Jan 1998.

The definitive version is available at <https://doi.org/10.1021/ie970460+>

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Prediction of Pressure Drop and Liquid Holdup in High-Pressure Trickle-Bed Reactors

M. H. Al-Dahhan,* M. R. Khadilkar, Y. Wu, and M. P. Duduković

Chemical Reaction Engineering Laboratory, Department of Chemical Engineering, Washington University, Saint Louis, Missouri 63130

The Holub et al. (1992, 1993) phenomenological model for pressure drop and liquid holdup in trickle flow regime at atmospheric pressure was noted by Al-Dahhan and Duduković (1994) to systematically underpredict pressure drop at high pressure and high gas flow rates. In this study, the Holub et al. (1992, 1993) model has been extended to account for the interaction between the gas and liquid phases by incorporating the velocity and the shear slip factors between the phases. As a result, the prediction of pressure drop at the operating conditions of industrial interest (high pressure) has been improved noticeably without any significant loss in predictability of liquid holdup. The extended model and the comparison between its prediction and experimental high pressure and high gas flow rate data are presented and discussed.

Introduction

A trickle-bed reactor (TBR) is a fixed bed of catalyst contacted by cocurrent downflow of gas and liquid. It is used widely in petroleum, petrochemical, and chemical industry. Most of the industrial trickle beds operate at high pressure, up to ~20–30 MPa (~3000–4500 psi) to improve the solubility of the gaseous reactants, achieve better heat and mass transfer, and slow down catalyst deactivation. Two broad flow regimes are observed in TBRs based on the superficial mass velocities of the two phases, fluid properties and bed characteristics: a low gas–liquid interaction regime (LIR, trickle flow regime) and high gas–liquid interaction regimes (pulse, spray, wavy, bubble, and dispersed bubble flow regimes). The two phase flow fluid dynamics changes from one regime to another and, hence, the operating, design and scale-up parameters are affected differently in each flow regime. The trickle flow regime, the pulse flow regime, and the transition between the two are of particular interest to industry (Al-Dahhan and Duduković, 1994; Charpentier and Favier, 1975; Holub, 1990; Wammes and Westerterp, 1991). Although the trickle flow regime is described as low gas–liquid interaction, strictly speaking, low gas–liquid interaction is true only at very low superficial mass velocities. However, as the superficial velocities are increased towards the pulsing regime, the interaction between the gas and liquid is enhanced, particularly at high-pressure operation (Al-Dahhan and Duduković, 1994; Al-Dahhan et al., 1996). This enhanced interaction can affect liquid holdup, pressure drop, transport parameters, and the resultant performance of the reactor. Thus, a basic understanding of the hydrodynamics of trickle bed reactors at the operating conditions of interest is essential to their design, scale-up, scale-down, and performance prediction.

Most of the previous studies on quantifying holdup and pressure drop have been conducted under atmospheric pressure, whereas the desired conditions of investigation are industrial operating pressures of 20–

30 MPa. Recently, a few investigations have been performed to study the influence of reactor pressure and gas flow rate on pressure drop and liquid holdup (Al-Dahhan et al., 1997; Al-Dahhan and Duduković, 1994; Wammes et al., 1991; Larachi et al., 1991a,b). Their experimental observations show that at high pressure and high gas flow rate, for a given liquid superficial velocity, liquid holdup decreases and pressure drop increases significantly compared with that at low pressure operation. The effect of pressure arises because of the increase in gas density and, hence, when the pressures of gases of different molecular weights are set to have equal densities (e.g., if pressure of He is about seven times that of N₂ pressure) at constant liquid mass velocities, the pressure drops and liquid holdups are about identical (Wammes et al., 1991; Larachi et al., 1991a,b, 1994).

Al-Dahhan and Duduković (1994, 1995) have proposed a phenomenological analysis for five limiting cases to describe the effect of reactor pressure and gas flow rate on the hydrodynamic parameters, such as pressure drop, liquid holdup, catalyst wetting efficiency, gas–liquid interfacial area, etc. These cases can be summarized as follows (Al-Dahhan et al., 1997).

Case 1: No gas flow, all pressures. This case represents pure trickle flow regime where the gas is stagnant. Dimensionless pressure gradient ($\Delta P/Z\rho g$) is zero and the liquid is exclusively driven by its weight. Hence, at a given liquid superficial velocity, liquid holdup is the largest, whereas catalyst wetting efficiency and gas–liquid interfacial area are the smallest because liquid fills the major pore spaces readily but does not spread uniformly across the reactor section and over the external surface of the catalyst.

Case 2: Low pressure and low gas superficial velocity ($P < 0.35$ MPa and $U_G < 2$ cm/s) for nitrogen and for gases with equivalent density. The dimensionless pressure drop ($\Delta P/Z\rho_L g$) is small and changes only slightly with variation in gas velocity and can be neglected. TBR fluid dynamics in this case can, to a good approximation, be seen as gravity driven and gas phase independent. Hence, the effect of pressure and gas flow rate is negligible.

Case 3: Low pressure and high superficial gas velocity

* Corresponding author. Telephone: (314)935-7187. Fax: (314)935-4832. E-mail: muthanna@wuche.wustl.edu.

($P < 0.35$ MPa and $U_G > 7$ cm/s) for nitrogen and for gases with equivalent density. The pressure gradient increases in comparison with the gravitational force. Consequently, the dimensionless pressure gradient ($\Delta P/Z\rho_L g$) increases, which causes a decrease in liquid holdup and an increase in the catalyst wetting efficiency and gas-liquid interfacial area. This result is due to the increase in the liquid spreading across the reactor section and over the external particle surface caused by larger gas flow rate. The effect of gas velocity in this case is more noticeable at high liquid flow rate than that at low liquid flow rate.

Case 4: High pressure and low gas superficial velocity ($P > 3.5$ MPa and $U_G < 2$ cm/s) for nitrogen and for gases with equivalent density. As a result of the increased gas density, the pressure drop increases and so does the dimensionless pressure drop ($\Delta P/Z\rho_L g$). This causes liquid holdup, wetting efficiency, and gas-liquid interfacial area to increase in a less pronounced manner compared with Case 3 because the pressure gradient is more sensitive to velocity changes than to gas density changes.

Case 5: High pressure and high gas superficial velocity ($P > 3.5$ MPa and $U_G > 7$ cm/s) for nitrogen and for gases of equivalent density. This case is the most important one in terms of the sensitivity of TBR fluid dynamics to pressure in which gas-liquid interaction becomes noticeable. Dimensionless pressure drop ($\Delta P/Z\rho_L g$) increases dramatically and liquid holdup decreases significantly. Hence, liquid film thickness at a constant liquid flow rate decreases, whereas the shear stress on the gas-liquid interface increases, resulting in a better spreading of the liquid film across the reactor section and over the external packing area. Therefore, catalyst wetting efficiency and gas-liquid interfacial area improve noticeably. The effects of high pressure and high gas velocity at higher liquid flow rates are more significant than those at low flow rates.

The correlations developed to predict pressure drop and liquid holdup under aforementioned flows and pressures are entirely empirical (Larachi et al., 1991a; Wammes et al., 1991; Ellman et al., 1988, 1990). In the absence of any fundamental approach due to the complex interaction between the flowing fluids and the stationary packing, a phenomenological (mechanistic) model based on a simple physical picture of the phenomena involved is preferred to strictly empirical correlations. One such model was developed by Holub et al. (1992, 1993), and its extended version as applied to high pressure and high gas flow rates is presented and discussed in this study.

Phenomenological Model

Holub et al. (1992, 1993) proposed a phenomenological model in the form of a modified Ergun equation based on representation of the complex geometry of the actual void space in a packed bed of particles at the pore level by an inclined slit (Figure 1). In trickle flow regime, the liquid flows as films or rivulets over the catalyst bed while the gas flows as a continuous phase through the remaining voids, so the liquid in the representative slit is assumed to be completely wetting the wall of the slit with a film of uniform thickness while the gas flows in the central core. The two phase momentum balance equations in the slit model are mapped to the actual bed model, which yields the dimensionless equations for

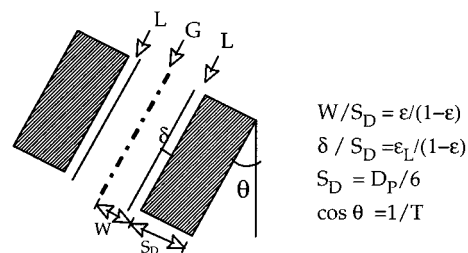


Figure 1. Slit model representation of two-phase flow in trickle-bed reactor.

Table 1. Phenomenological Model of Holub et al. (1992, 1993) in the Form of a Modified Ergun Equation for No Gas-Liquid Interaction

equation	no.
$\Psi_L = \frac{\Delta P/Z}{\rho_L g} + 1 = \left(\frac{\epsilon_B}{\epsilon_L} \right)^3 \left[\frac{E_1 R e_L}{G a_L} + \frac{E_2 R e_L^2}{G a_L} \right]$	(1)
$\Psi_G = \frac{\Delta P/Z}{\rho_G g} + 1 = \left(\frac{\epsilon_B}{\epsilon_B - \epsilon_L} \right)^3 \left[\frac{E_1 R e_G}{G a_G} + \frac{E_2 R e_G^2}{G a_G} \right]$	(2)
$\Psi_L = 1 + \frac{\rho_G}{\rho_L} (\Psi_G - 1)$	(3)

^a E_1 and E_2 are Erguns constants that represent the bed characteristics are evaluated from single phase gas flow (dry bed) experiments.

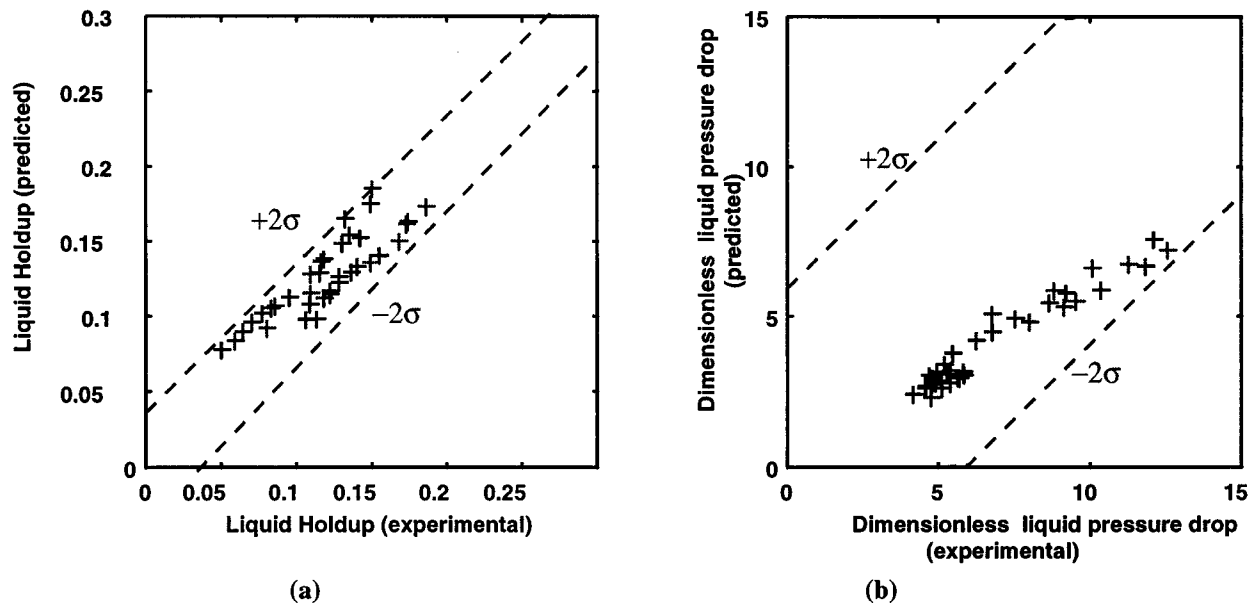
the trickle flow in the form of modified Ergun equations, as shown in Table 1 (Holub et al., 1992, 1993). These equations also tie up pressure drop and holdup in the trickle flow regime. Parameters E_1 and E_2 are the Erguns constants that characterize the bed ($E_1 = 72 T^2$, $E_2 = 6 T f_{\text{wall cat.}}$) and are determined from single-phase gas flow (dry bed) experiments in the bed of interest. By substituting eqs 1 and 2 into eq 3 and by equating the dimensional pressure gradients ($\Delta P/Z$) on the gas and liquid sides, the nonlinear implicit equation for liquid holdup can be solved by direct or Newton iteration. Knowing the liquid holdup, pressure drop can be evaluated by either eqs 1 or 2.

Holub et al. (1992, 1993) have demonstrated that the model predicts pressure drop and liquid holdup at atmospheric pressure better than current correlations developed based on atmospheric pressure data. Al-Dahhan and Duduković (1994) reported that Holub et al. model also predicts properly at high pressure the trends of effects of reactor pressure, gas flow rate, liquid flow rate, physical properties, and bed characteristics (Al-Dahhan and Duduković, 1994). However, Al-Dahhan and Duduković (1994) showed that although the model predicts pressure drop and liquid holdup better than recently reported high-pressure correlations (Larachi, 1991a,b; Wammes et al., 1991; Wammes and Westerterp, 1991; Ellman et al., 1988, 1990), as shown in Table 2, it systematically underpredicts them at high pressure and high gas flow rates.

Figure 2b (for the entire data set) and Figures 4a and 5a (for two specific cases) show the comparison of the Holub et al. model prediction for pressure drop and holdup and experimental observations as a function of liquid mass velocity. It is obvious that the agreement between the model and the data is very good at lower pressure and over the range of liquid mass velocity at all levels of gas velocity (lower cluster of points in Figure 2b). However, at high pressure and high gas velocity, the model consistently underpredicts the data. Under these conditions, the relative error in pressure drop

Table 2. Comparison of Predictions of Recent Pressure Drop and Holdup Correlations with Al-Dahhan's Data (1993)

correlation	expression	prediction error (%)	holdup pressure drop
Ellman et al. (1988)	$\frac{(\Delta P/Z)d_h\rho_G}{2G^2} = 200(X_G\delta_2)^{-1.2} + 85(X_G\delta_2)^{-0.5}$	21.5	65
(empirical correlation)	$X_G = \frac{G}{L}\sqrt{\frac{\rho_L}{\rho_G}} \quad \delta_2 = \frac{Re_L^2}{(0.001 + Re_L^{1.5})}$		
Larachi et al. (1991a)	$\frac{(\Delta P/Z)d_h\rho_G}{2G^2} = \frac{1}{[(Re_L We_L)^{0.25} X_G]^{1.5}} \left[31.3 + \frac{17.3}{[(Re_L We_L)^{0.25} X_G]^{0.5}} \right]$	14.5	89
(empirical correlation)	$X_G = \frac{G}{L}\sqrt{\frac{\rho_L}{\rho_G}} \quad We_L = \frac{L^2 d_p}{\rho_L \sigma_L}$		
Wammes et al. (1991)	$\frac{\Delta P}{0.5\rho_G U_G^2 Z} \frac{d_p}{Z} = 155 \left[\frac{\rho_G U_G d_p \epsilon_B}{\mu_G (1 - \epsilon_B)} \right]^{-0.37} \left[\frac{1 - \epsilon_B}{\epsilon_B (1 - \beta_V)} \right]$	41	88
(empirical correlation)			
Holub et al. (1992, 1993)	$\Psi_L = \frac{\Delta P/Z}{\rho_L g} + 1 = \left(\frac{\epsilon_B}{\epsilon_L} \right)^3 \left[\frac{E_1 Re_L}{Ga_L} + \frac{E_2 Re_L^2}{Ga_L} \right]$	9.7	40
(phenomenological model)	$\Psi_G = \frac{\Delta P/Z}{\rho_G g} + 1 = \left(\frac{\epsilon_B}{\epsilon_B - \epsilon_L} \right)^3 \left[\frac{E_1 Re_G}{Ga_G} + \frac{E_2 Re_G^2}{Ga_G} \right]$		

**Figure 2.** Prediction of holdup and pressure drop (Case I: $f_s = f_v = 0$, only high pressure, high gas flow data; $\sigma_X = (\sum (X_{pred} - X_{exp})^2 / N)^{1/2}$).

prediction (~48%) is more noticeable compared with that of holdup prediction (9%). This larger relative error is because the interaction between gas and liquid phases increases at high pressure and high gas flow rate, as discussed in the earlier cases, and this is not accounted for in the original form of the Holub's model as reviewed in the assumptions used to develop its simplest form. In the detailed derivation of the governing equations for the model, Holub et al. (1992, 1993) neglected the interaction at the gas-liquid interface and assumed a discontinuity in both shear and velocity at the interface. Essentially zero-velocity gradient and no shear were assumed in the model at the free liquid film surface (i.e., both velocity slip factor, f_v , and shear slip factor, f_s , are considered to be equal to zero), which is not the case at high pressure and high gas velocity (Al-Dahhan and Duduković, 1994). Accordingly, in this work, the Holub et al. (1992, 1993) model has been extended to account for the interaction between the gas and the liquid

phases in the attempt to improve pressure drop predictions in high pressure and high gas flow rate operations.

The Extended Model

The degree of interaction between the gas and liquid phases in trickle bed reactors can be accounted for by incorporating the velocity and shear slip factors between the phases as suggested and derived by Holub and co-workers (1990, 1992, 1993). This incorporation of velocity and shear slip factors is an extension of the original general model (Holub, 1990) based on the two-phase flow momentum balance for the slit, which additionally incorporates the velocity slip factor ($f_v = V_{IG}/V_{IL}$) and the shear slip factor ($f_s = \tau_L/\tau_G$) to relate the velocity and shear stress in the gas and liquid phase under increasing degree of phase interaction observed at higher gas densities and velocities. The detailed derivation is an extension of the original model deriva-

Table 3. Extended Model Equations for Pressure Drop and Liquid Holdup

equation	no.
$\Psi_G = \left(\frac{\epsilon_B}{\epsilon_B - \epsilon_L} \right)^3 \left[\frac{E_1(Re_G - f_v \epsilon_G Re_L)}{Ga_G} + \frac{E_2(Re_G - f_v \epsilon_G Re_L)^2}{Ga_G} \right]$	(4)
$Re_L = \frac{V_{iL} D_p}{\nu_L (1 - \epsilon_B)}$	(5)
$\Psi_L = \left(\frac{\epsilon_B}{\epsilon_L} \right)^3 \left[\frac{E_1 Re_L}{Ga_L} + \frac{E_2 Re_L^2}{Ga_L} \right] + f_s \frac{\epsilon_G}{\epsilon_L} \left(1 - \frac{\rho_G}{\rho_L} - \Psi_L \right)$	(6)
$Re_L = \Phi \eta_L$	$0 < \eta_L < 5$ (7)
$Re_L = \Phi(-3.05 + 5 \ln(\eta_L))$	$5 < \eta_L < 30$ (8)
$Re_L = \Phi(5.5 + 2.5 \ln(\eta_L))$	$\eta_L > 30$ (9)
where	
$\Phi_L = \frac{10}{(E_1)^{0.75}} \frac{\nu_L}{\nu_G} \sqrt{\Psi_L Ga_L \frac{\epsilon_L}{\epsilon_B^3} \left(1 + f_s \frac{\epsilon_G \rho_G \Psi_G}{\epsilon_L \rho_L \Psi_L} \right)}$	(10)
$\eta_L = \frac{1}{5(E_1)^{0.25}} \sqrt{\Psi_L Ga_L \left(\frac{\epsilon_L}{\epsilon_B} \right)^3 \left(1 + f_s \frac{\epsilon_G \rho_G \Psi_G}{\epsilon_L \rho_L \Psi_L} \right)}$	(11)
and	
$\Psi_L = 1 + \frac{\rho_G}{\rho_L} (\Psi_G - 1)$	(12)

tion (Holub, 1990), and only the final form of the equations are presented in Table 3 (eqs 4–12).

The shear and velocity slip correction factors, f_s and f_v , respectively, characterize the degree of phase interaction at the gas–liquid interface. Hence, when $f_s = f_v = 0$ (i.e., no interaction occurs), the model (eqs 4–12) simplifies to the original Holub et al. model represented by eqs 1–3 (Table 1). The rationale behind assuming $f_s = f_v = 0$ is that for atmospheric pressure data, Holub et al. (1993) have shown that f_v and f_s can both be zero (no interaction) with only a small increase in the overall average error over the observed minimum error. However, this is not the case when interaction between the phases occurs at high pressure and high gas flow rate while still in the trickle flow regime (Al-Dahhan and Duduković, 1994, Al-Dahhan et al., 1996). Accordingly, the model represented by eqs 4–12 is suggested as a two phase flow form of the Ergun equation containing the two phase interaction parameters, f_s and f_v , which must be determined from two phase flow experimental data as discussed in the following section. Ergun's constants, E_1 and E_2 , characterize the bed and are still determined from single (gas) phase flow experiments. Equation 12 is an implicit equation in liquid holdup formed by equating the dimensional pressure gradient in the gas and liquid phases, and is solved for liquid holdup (as done for eq 3) from which pressure drop is then evaluated.

Results and Discussion

Although f_s and f_v are defined as the ratio of the stresses and velocities of the two phases at the interface, the exact dependence of these parameters on flow conditions is not easily determinable. In fact, at any given time or location in the reactor, different types and levels of interactions are possible resulting in varying f_s and f_v values. Thus, f_s and f_v reflect in an averaged sense the net interaction between gas and liquid in the reactor. A continuity of velocity and shear profiles indicates that both slip factors are equal to unity, whereas a zero value of the slip factors indicates no interaction. Negative values of f_v indicate the presence of recirculation cells, whereas negative values of f_s indicate the liquid exerting a shear on the gas phase as

Table 4. Range of Operating Conditions for the Data Used in Developing f_s and f_v Correlations

conditions	operating range
reactor pressure	$0.35 \leq P \leq 5.0$ MPa $30 \leq P \leq 700$ psig
gas superficial velocity, cm/s	$1 \leq U_g \leq 11.7$
gas superficial mass velocity, kg/m ² s	$6.4 \times 10^{-3} \leq G \leq 4.03$
liquid superficial velocity, cm/s	$0.042 \leq U_l \leq 0.41$
liquid superficial mass velocity, kg/m ² s	$0.42 \leq L \leq 4.1$
temperature	~298 K
liquid phase	water hexane
gas phase	helium nitrogen
solid particles	glass beads (0.11 cm) cylinders (0.157 × 0.43 cm)
reactor dimensions	diameter = 2.2 cm length = 57.2 cm

the interaction increases. These slip factors are expected to be functions of flow variables and a large amount of experimental two phase pressure drop and holdup data under moderate-to-high interaction conditions is required to quantify their dependence precisely (note that this data should include independently measured Ergun's constants E_1 and E_2 for the bed).

In absence of such an extensive database, the limited experimental pressure drop and holdup data of Al-Dahhan (1993) and Al-Dahhan and Duduković (1994) that cover low to high pressure and high gas flow rates are used to evaluate f_s and f_v using the extended model equations (eqs 4–12). Due to the limited number of data points available (see Table 4), it was not possible to observe a strong discernible dependence of f_s and f_v with either Re_L or Re_G . Therefore, correlations for f_s and f_v are developed by obtaining f_s and f_v that minimize the pressure drop prediction error. This procedure led to only a weak dependence on the gas as well as liquid Reynolds numbers. Although the powers on the Reynolds numbers have a large degree of uncertainty, they are the best estimates that the limited data set provided.

$$f_s = -4.4 \times 10^{-2} Re_G^{0.15} Re_L^{0.15} \quad (13)$$

$$f_v = -2.3 Re_G^{0.05} Re_L^{-0.05} \quad (14)$$

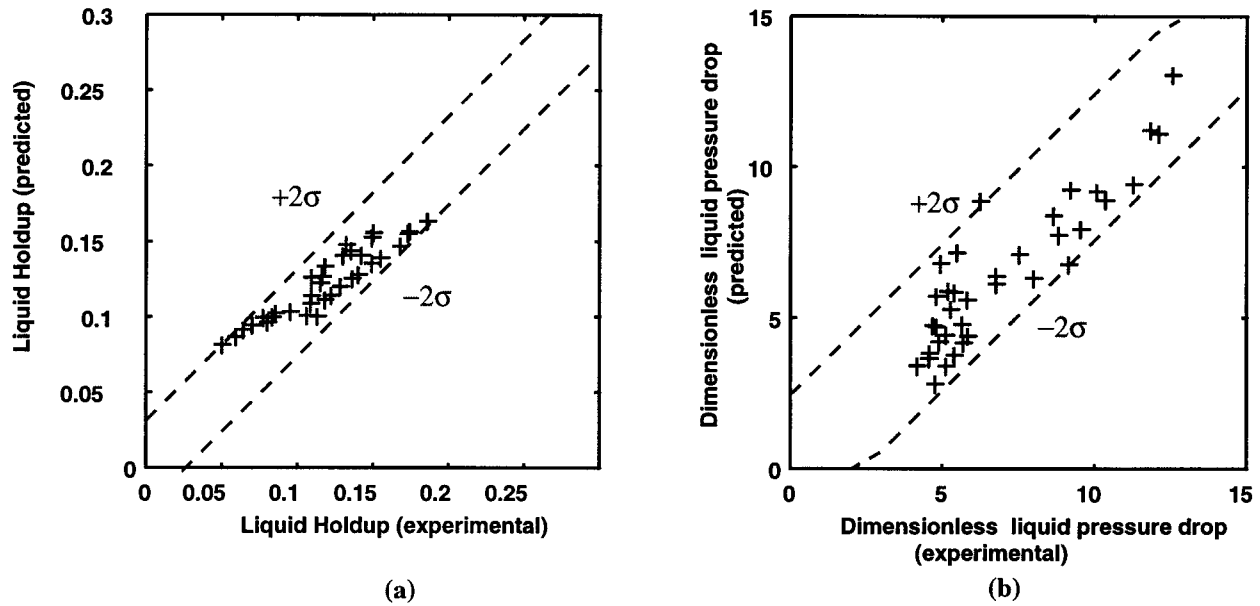


Figure 3. Prediction of holdup and pressure drop (Case II: f_s and f_v correlations; only high pressure and high gas flow data set; $\sigma_X = (\sum(X_{\text{pred}} - X_{\text{exp}})^2/N)^{1/2}$).

Equations 13 and 14 can now be used to calculate f_s and f_v as the two phase flow parameters in the model eqs 4–12. As a result, the prediction of pressure drop improved significantly compared with that of the simplified model (relative error decreased from 48% to 20%), as shown in Figure 2b (for the simplified model) and 3b (for the extended model). Liquid holdup prediction remained within the same range of predictability as that by Holub's original model (relative error is $\sim 9\%$ for both simplified and extended model), as shown in Figure 2a (for the simplified model) and Figure 3a (for the extended model). Figures 4 and 5 show the improvement in prediction for particular cases of fluids and solid particles. In both cases (Figure 4, water-nitrogen on glass beads; and Figure 5, hexane-nitrogen on glass beads), the systematic error in pressure drop prediction observed previously is reduced considerably without any significant loss in predictability of liquid holdup (Figures 4b and 5b). This result reveals that pressure drop is more affected by the interaction between phases compared with liquid holdup. It should be noted that the f_s and f_v values used for this prediction were based on the entire data set (used to obtain eqs 13 and 14). Furthermore, the trend in the experimental pressure drop data is captured correctly by incorporating the shear and velocity slip correction factors as interaction parameters.

Concluding Remarks

This study demonstrates that shear- and velocity-based correction factors are necessary for accurate prediction of pressure drop and holdup, particularly in the moderate interaction range within the trickle flow regime. It is noteworthy to mention that a large bank of high pressure and gas flow rate data is needed to develop sound correlations for the prediction of f_s and f_v , which is not available at present. Moreover, high pressure data in the literature cannot be used directly because E_1 and E_2 were not reported, and these parameters can only be obtained from single phase flow experiments. More work on correlation of f_s and f_v using data at moderate-to-high phase interaction within the

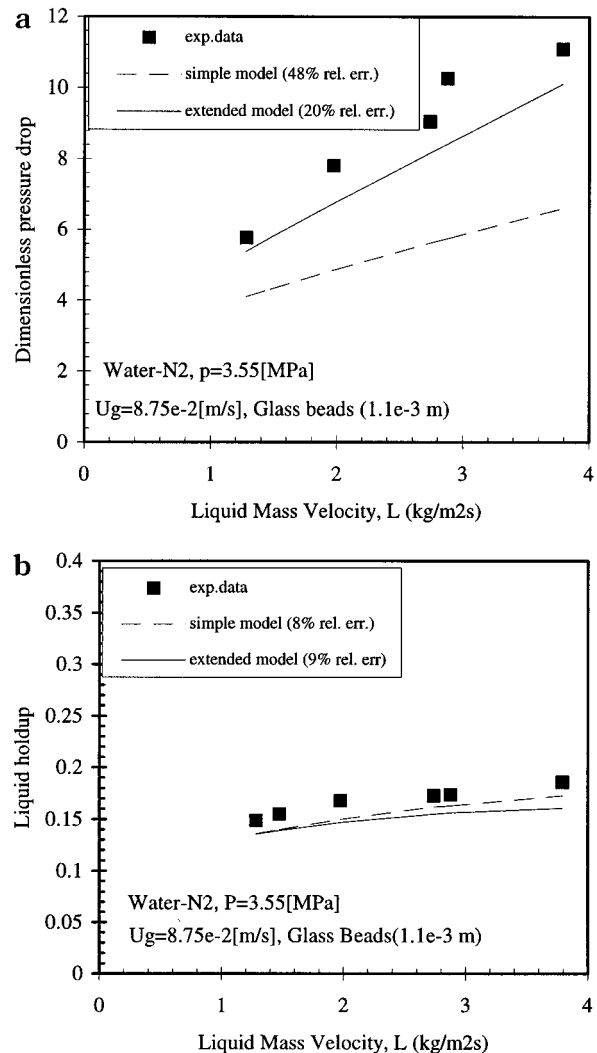


Figure 4. (a) Comparison of dimensionless pressure drop ($\Delta P/(\rho_L g Z)$) prediction by simple and extended models and experimental data for the water nitrogen system. (b) Comparison of liquid holdup prediction by simple and extended models and experimental data for the water nitrogen system.

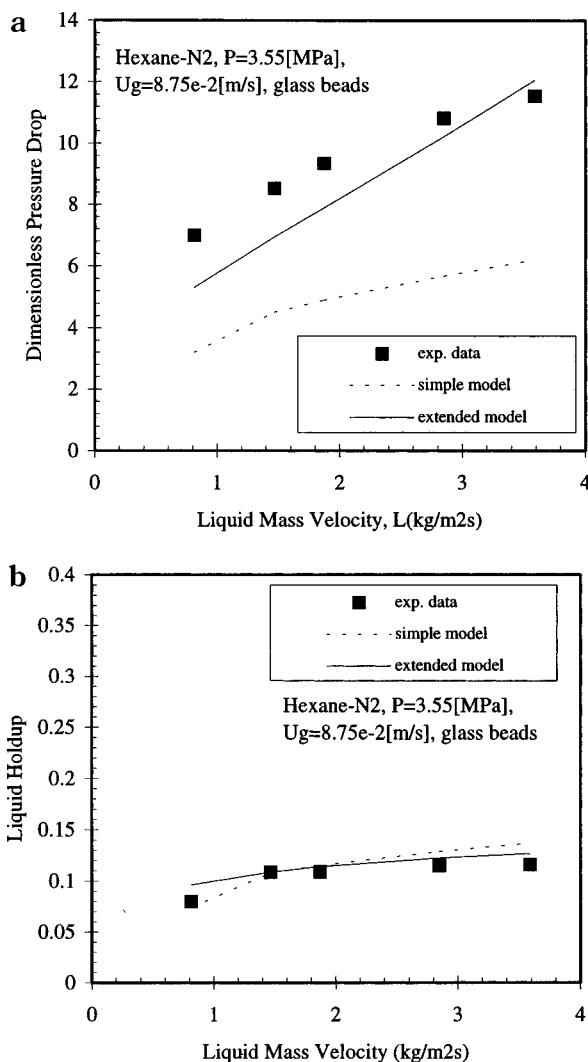


Figure 5. (a) Comparison of dimensionless pressure drop ($\Delta P / (\rho_L g Z)$) prediction by simple and extended models with experimental data for the hexane nitrogen system. (b) Comparison of liquid holdup prediction by simple and extended models with experimental data for the hexane nitrogen system.

trickle flow regime is recommended to understand and quantify their dependence on flow variables.

Acknowledgment

The authors acknowledge the financial support of industrial sponsors of the Chemical Reaction Engineering Laboratory (CREL).

Nomenclature

D_p = equivalent diameter of packing particle
 E_1, E_2 = Ergun equation constants for single phase flow
 f = phase interaction parameters
 g = gravitational acceleration
 G = gas superficial mass velocity
 $G a_\alpha$ = Galileo number ($g D_p^3 \epsilon_B^3 / \nu_\alpha^2 (1 - \epsilon_B)^2$)
 L = liquid superficial mass velocity
 P = operating pressure
 Re_α = Reynolds number of α phase ($V_\alpha D_p / \nu_\alpha (1 - \epsilon_B)$)
 S_D = half wall thickness
 T = bed tortuosity
 V_α = superficial velocity of α phase
 W = half slit width
 We_L = liquid Weber number ($U_L^2 D_p \rho_L / \sigma_L$)

X = flow factor ($(G/L) \sqrt{\rho_L / \rho_G}$)

Z = bed height

Greek Symbols

δ = film thickness

ϵ_B = bed porosity

ϵ_α = bed holdup of α phase

η_α = pseudo bed Reynolds number based on α phase

μ_α = viscosity of α phase

ν_α = kinematic viscosity of α phase

ρ_α = density of the α phase

Ψ_α = dimensionless body force on the α phase

Subscripts

α = general subscript meaning gas (G) or liquid (L) phase

G = gas phase

L = liquid phase

s = shear

v = velocity

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Received for review July 1, 1997

Revised manuscript received September 24, 1997

Accepted September 26, 1997

IE970460+