Necessary Condition for Pulsing Flow Inception in a Trickle Bed

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Based on the three existing forms of liquid in a trickle bed, as proposed by Charpentier et al. (1968a,b), a straight channel model is developed with a wall constituted from the packing. Within the channel, liquid and gas phases are assumed to flow concurrently downward at constant velocities, just as in an empty pipe. Since the channel wall is assumed to be formed from the packing, the capillary influence could be taken into account. Capillary and frictional forces are considered as the two principal factors which determine the stability of the liquid film covering a pellet. Evaluating the balance of these two factors gives rise to a new model, which is considered the necessary condition for pulsing flow inception corresponding to the original work by Ng (1986).

Introduction

The first problem to be solved in designing and operating an industrial trickle-bed reactor is the prediction of flow regime transition. In understanding the transition mechanism, knowledge of the fluid-flow principles discovered in an empty tube is important, since it has long been recognized that a packed bed resembles an empty tube very much, especially when only the gas phase exists. One can find such evidence from the origin of the Ergun (1952) equation and from recent research (Cheng and Yuan, 1997), which identified a coefficient of 1/3, relating the pressure drop in an empty tube to that in a packed bed. When both the gas and liquid phases are present, the two-phase frictional pressure drop correlating method proposed by Lockhart and Martinelli (1949), as well as the coordinates in flow pattern recognition introduced by Baker (1954), have also been successfully adopted in trickle-bed research. Earlier examples are contributions of Charpentier and Favier (1975) and of Larkins et al. (1961).

In view of the above similarities, there is the question of whether an identical mechanism and theory, valid in a two-phase pipe flow, can also be directly applied to a trickle bed. Sicardi et al. (1979) answered this question and proposed a straight channel model, where it was assumed that when the liquid waves were large enough to occlude the channels, pulsing flow was expected to occur. Although a large deviation was found between the prediction and experiment, for the first time, Sicardi et al. realized that the influence of the gas velocity on the pulsing inception was through the tangential stress at the liquid interface. In their second article (Sicardi and Hofmann, 1980), a constriction model was evolved as an improvement to the earlier one. The ratio of the kinematic energy of the liquid to the energy of liquid reformation was used to correlate previous experimental findings and was found to be satisfactory. Such a finding is promising; although no mathematical model was developed, it provides a phenomenological understanding of the pulsing transition mechanism, which is substantially different from that of slug flow in a pipe.

A variety of other mechanisms for pulsing flow inception have also been given by different authors. It was noted with a new abscissa of some definite physical meaning (Talmor, 1977) that flow pattern recognition could be accomplished in a different way; however, the treatment was still empirical. Later, Rao et al. (1983) considered a bubble's formation as the indication of pulsing flow, but this differs from commonly recognized observations. Recently, Holub et al. (1993) proposed a parallel slit-plate model; however, the influence of liquid surface tension, which would be important in porous

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media flow, was ignored. It appears that the most reasonable analysis until now is the method proposed by Ng (1986); nevertheless, the prediction can deviate from the actual values as much as fourfold, although the trend of analysis is good. It is obvious that more intensive research is needed to fill the gap between theory and experiment.

Model Development

As summarized above, the generation of pulsing flow in a trickle bed is different from that of slug flow in a pipe. In view of Kelvin-Helmholtz stability analysis, the stabilizing factor in two-phase pipe flow is the gravitational force exerted on the liquid phase. In comparison, it would be the surface tension that keeps the liquid film stable covering to the packing surface in a trickle bed. This deterministic difference suggests that different models should be utilized according to the specific case encountered. Since a trickle-bed model requires both capillary and pipe-flow phenomena to be considered simultaneously, it was decided to refer to Ng's (1986) model as the starting point for the present analysis. Nevertheless, examination of this model leads to the following two insufficiencies: (1) in deriving this model, the liquid was considered stagnant; (2) frictional force resulting from the gas to the liquid was neglected in the Bernoulli equation, which means that the two phases are idealized as inviscid fluids.

These two points are similar to Kelvin-Helmholtz stability analysis for two-phase pipe flow as described by Taitel and Dukler (1976). Ng's model implies that the whole kinematic energy of the gas is to be consumed to overcome the liquid capillary force covering the packing. In fact, this balancing is overestimated and certainly results in a low estimate of the gas velocity for pulsing inception. Actually, it is the gas-liquid interfacial friction which causes the occurrence of slugging or pulsing flow, as shown by Baker (1954) in two-phase pipe flow, as well as by Sicardi et al. (1979, 1980) in a trickle bed. In this regard, the Bernoulli equation, which can properly take into account interfacial frictional losses, should be established.

In a trickle bed at low gas and liquid flow rates, the two phases flow in a stratified manner. The liquid trickles freely down over the packing in droplets, films, and rivulets (Charpentier et al., 1968a,b; Charpentier, 1969). Rivulets and droplets are formed while reducing the system free energy due to the liquid-solid surface tension. According to the film-rivulet-droplet model, as summarized by Hofmann (1978), the rivulet portion decreases with the increase of gas-flow rate, and it only comprises a small fraction (about 10%) of the total liquid holdup, while the portion of the droplets remains almost unchanged with the increase of the gas-flow rate and comprises the major fraction of the static liquid holdup. Accordingly, at the moment just prior to pulsing flow, almost all the rivulets would have been pressed by the gas phase into films. A simplified physical model of this situation is shown in Figure 1.

The model shown in Figure 1 is a flow channel with the packing pellets considered as the channel wall. Such a structure combines characteristics of both pipe flow and porous media flow. In this model, liquid flows down in the form of a film over the packing, concurrently with the gas flow. Three forces: the capillary force, the gravitational force, and the gas-to-liquid frictional force are simultaneously acting on the liquid film. Since the triangular zone formed from adjacent pellets is always filled with the static liquid, the routine of liquid movement will be different from that of Ng (1986), in a manner like that of Sicardi et al. (1980, 1981).

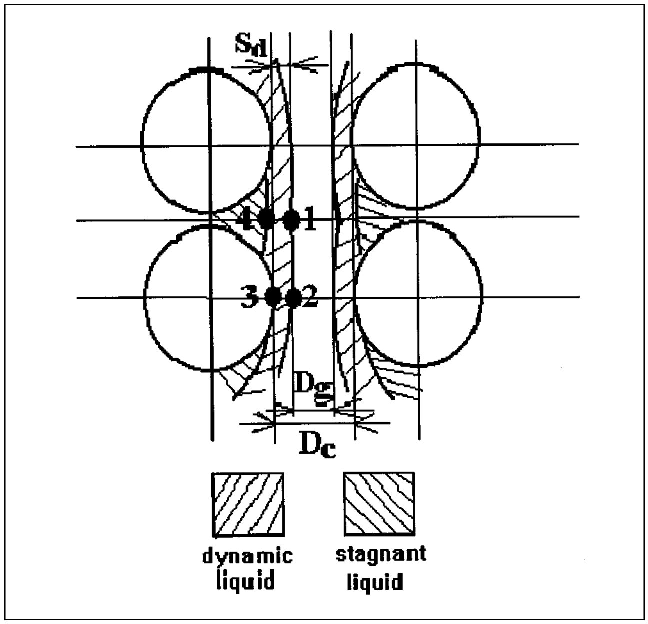

Figure 1. Physical model of the trickle bed.

In Figure 1, the channel is assumed to be straight with a constant diameter D_c (m). For the occurrence of transition from trickling to pulsing flow, the most probable position is at point 2, where the gas velocity is at the highest and the liquid film is most unstable. The mechanical energy conservation equation for the gas phase between positions 1 and 2 can be expressed as

$$\frac{1}{2}\rho_g u_{g1}^2 + p_1 = \frac{1}{2}\rho_g u_{g2}^2 + p_2 + 4 f_i \frac{L}{D_g}\rho_g \frac{\left(\bar{u}_g - \bar{u}_l\right)^2}{2} \quad (1)$$

where L is the distance (m) between positions 1 and 2 and is equal to $D_p/2$. u_{g1} and u_{g2} are interstitial gas velocities at these two points. Since the flow channel is almost straight with a constant diameter D_c, the following approximation can be made

$$u_g \doteq u_{g1} \doteq u_{g2} \doteq \bar{u}_g \quad (2)$$

In Eq. 1, f_i is the gas-liquid interfacial friction factor. Since there has been no correlation of this parameter proposed until now, one turns to the viewpoint of Taitel and Dukler (1976), who believed that the interfacial friction factor f_i (dimensionless) could be set equal to f_g (dimensionless), regardless of whether the interface is wavy or not. The friction factor for the gas phase refers to the work of Lockhart and Martinelli (1949), which is expressed in the generalized Blasius form

$$f_g = \frac{C_g}{Re_g^m} = \frac{C_g}{\left(\dfrac{D_g u_g \rho_g}{\mu_g}\right)^m} \tag{3}$$

In Eq. 3, the following coefficients are used: $C_g = 0.046$, and $m = 0.2$ for turbulent gas flow at $Re_g > 2{,}000$ and $C_g = 16$; and $m = 1.0$ for laminar gas flow at $Re_g < 1{,}000$.

The gas-phase pressures p_1 and p_2 are related through the following equations

$$p_3 = p_2 + \frac{2\sigma}{r} \tag{4}$$

$$p_3 = p_4 + \rho_1 g \frac{D_p}{2} \tag{5}$$

and

$$p_1 = p_4 \tag{6}$$

thus

$$p_1 = p_2 + \frac{2\sigma}{r} - \rho_1 g \frac{D_p}{2} \tag{7}$$

In Eq. 4, r is the radius of the liquid film surrounding the packing at position 2, and is expressed as

$$r = \frac{D_p}{2} + S_d \tag{8}$$

where S_d is the film thickness (m) as shown in Figure 1, which can be obtained as the ratio of the dynamic liquid holdup to the area wetted by the flowing film. The value of S_d is found in the order of 0.05 to 0.1 mm for the 1 mm pellets (Sicardi et al., 1981), 0.118 to 0.2 mm for the 8.25 mm pellets, and 0.01 to 0.1 mm for the Shell hydrodesulfurization process under different flow conditions (Satterfield et al., 1969). Since the packing size in a trickle bed is normally between 1 to 3 mm, the liquid film radius r (m) can be roughly approximated by $D_p/2$.

From discussions about the model parameters through Eqs. 2 to 8, the final Bernoulli equation is derived

$$f_g \rho_g \frac{D_p}{D_g} u_g^2 = \frac{2\sigma}{D_p/2 + S_d} - \rho_l g \frac{D_p}{2} \tag{9}$$

The gas-flow channel diameter D_g (m) is related to the two-phase flow channel diameter D_c (m) and the film thickness S_d (m) through

$$D_g = D_c - 2S_d \tag{10}$$

where D_c is different from the bed equivalent diameter since the flow channel model introduced here is not simplified to such a high degree that all the porous media characteristics are lost. In this consideration, the bed average channel diameter proposed by Ng (1986) is adopted in this work

$$D_c = \sqrt{\frac{4\epsilon}{\pi N_C}} \tag{11}$$

with

$$N_C = \frac{6(1-\epsilon)}{\pi D_p^2} \tag{12}$$

The bed porosity ϵ (dimensionless) in Eqs. 11 and 12 can be obtained via experiments or via correlation such as (Mueller, 1991)

$$\epsilon = 0.379 + \frac{0.078}{D_t/D_p - 1.80} \tag{13}$$

Given the value of ϵ as 0.38, the channel diameter D_c is evaluated to be $0.64 D_p$. In comparison to Eq. 8, the inaccuracy generated from neglect of the film thickness S_d is almost doubled in Eq. 10. Relations in estimation of S_d have been developed by Satterfield et al. (1969) and Sicardi et al. (1981). Satterfield's method is limited to liquid single-phase flow and is based on volumetric liquid flow rate, and this is not suitable for this work. Instead, Sicardi's method is used

$$S_d = \frac{V_d}{(\alpha_w - \alpha_{ws} + \alpha'_{ws}) a_s} = \frac{h_d \cdot D_p}{(\alpha_w - \alpha_{ws} + \alpha'_{ws}) \cdot 6(1-\epsilon)} \tag{14}$$

where

$$\alpha_w = 1, \quad \alpha_{ws} = 0.42 \quad \text{and} \quad \alpha'_{ws} = 0.21 \tag{15}$$

To avoid the tedious trial-and-error process to evaluate the dynamic liquid holdup, which is conventionally correlated by the Lockhart-Martinelli parameter, a distinct relationship developed by Rao et al. (1983) is recommended

$$h_d = 0.33 \left(\frac{\overline{Re}_l}{\overline{Re}_g}\right)^{1/3} a_s^{1/3} \tag{16}$$

where $\overline{Re}_l$ and $\overline{Re}_g$ are Reynolds numbers (dimensionless) based on superficial gas and liquid velocities, and a_s is the specific surface area of the bed (m^{-1})

$$a_s = \frac{6(1-\epsilon)}{D_p} \tag{17}$$

Through Eqs. 9 to 17, the interstitial gas velocity u_g (m/s) can be obtained, and based on this, the superficial linear velocity v_g (m/s) for pulsing flow to occur is obtained through

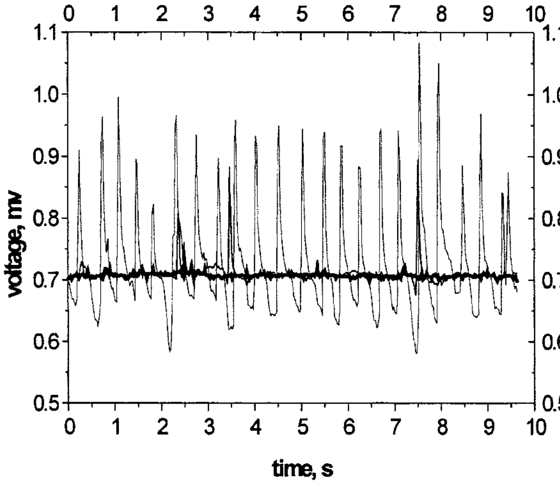

Figure 2. Measurements of the gas-phase pressure fluctuations.

Air-water system with ceramic spheres of $D_p = 3$ mm at constant gas flow rate of $v_g = 105$ cm/s, pressure transducer position: 10 cm from the bottom. —— trickling flow, $v_1 = 0.48$ cm/s; —— transitional flow, $v_1 = 0.53$ cm/s; —— pulsing flow, $v_1 = 0.65$ cm/s.

correction with the liquid average saturation β according to Ng (1986)

$$v_g = \epsilon u_g (1 - \alpha)$$

where

$$\alpha = 4\left[\sqrt{1-\beta} - (1-\beta)\right] \tag{18}$$

and

$$\beta = \left[\left(\frac{200}{Re_l} + 1.75\right)\frac{v_l^2}{gD_p}\frac{1-\epsilon}{\epsilon^3}\right]^{1/4} \tag{19}$$

Results and Discussions

The pulsing transition point is usually defined as the condition under which the stable two-phase flow terminates. It is observed in the present work that at the transition points, the bed cross section is only partially blocked by weak and short liquid slugs, and these points are in good agreement with those observed by Charpentier and Favier (1975) and Chou et al. (1977) as will be shown in Figure 6. It is also reasonable to assume that at pulsing transition points the gas phase is in the well-developed turbulent flow regime. Such a consideration is derived from visual observations and from the gas-phase pressure fluctuations depicted in Figure 2. It has been further identified from simulations that the gas Reynolds number is of the order of 2,000 to 8,000. Under turbulent flow conditions, a preliminary evaluation of the validity of the model is possible.

To make a more general analysis, the value of Re_g is set from 1,000 to 10,000. In this range, it is found that $Re_g^{0.2}$ only varies from 3.98 to 6.31, and the coefficient f_g is found in Eq. 3 from 0.0073 to 0.012. The magnitude of other variables

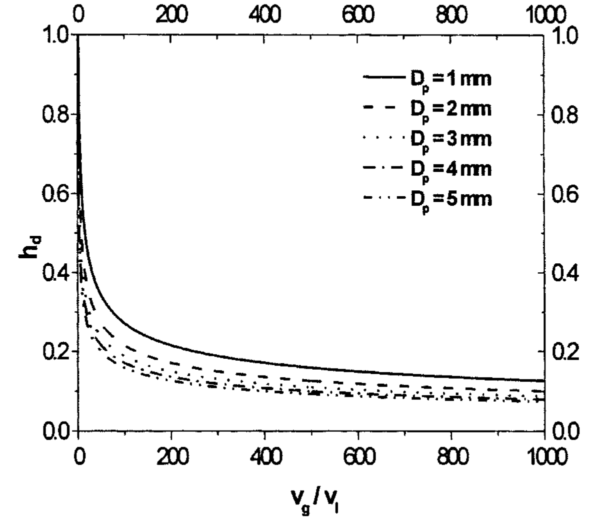

Figure 3. Prediction of dynamic liquid holdup for the air-water system.

such as D_p/D_g and S_d in Eq. 9 can be obtained through Eq. 10 and Eqs. 15, 16, and 17. Figures 3 and 4 are the results of the above calculations. In Figure 4, the film thickness S_d is found to be between $0.04\,D_p$ and $0.08\,D_p$ for different pellet diameters, and is in agreement with previous reports of Satterfield et al. (1969) and Sicardi et al. (1981). Since $D_c = 0.64\,D_p$, the value D_p/D_g is averaged at 2.0. Moreover, with the influence of S_d neglected in the expression of the film radius r, Eq. 9 reduces to

$$(0.015 \text{ to } 0.024)\cdot \rho_g u_g^2 = \frac{4\sigma}{D_p} - \rho_l g \frac{D_p}{2} \tag{20}$$

In comparison, Ng's (1986) model is written as

$$\frac{1}{2}\rho_g u_g^2 = \frac{4\sigma}{D_p} - \rho_l g \frac{D_p}{2} \tag{21}$$

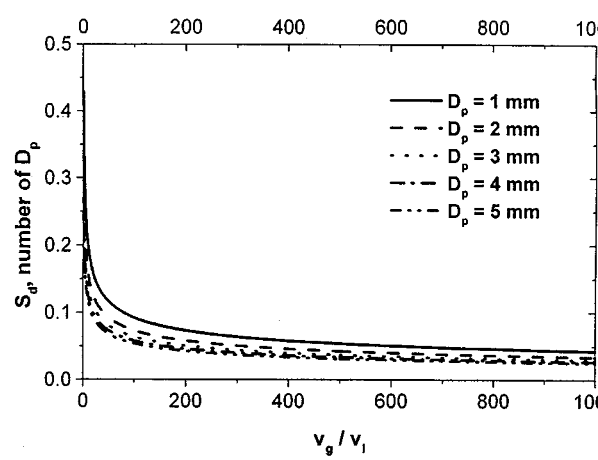

Figure 4. Prediction of liquid film thickness for the air-water system.

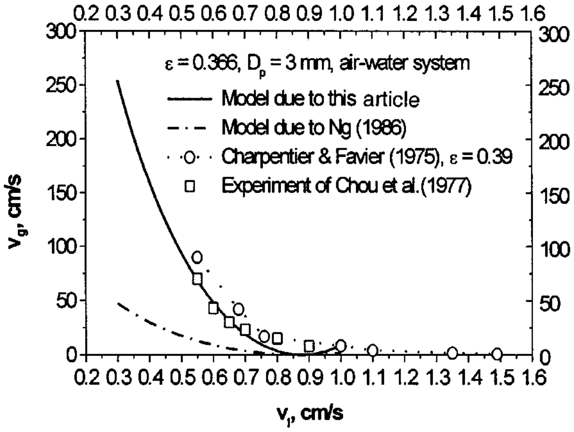

Figure 5. Verification of model prediction with experiment: dense packing case.

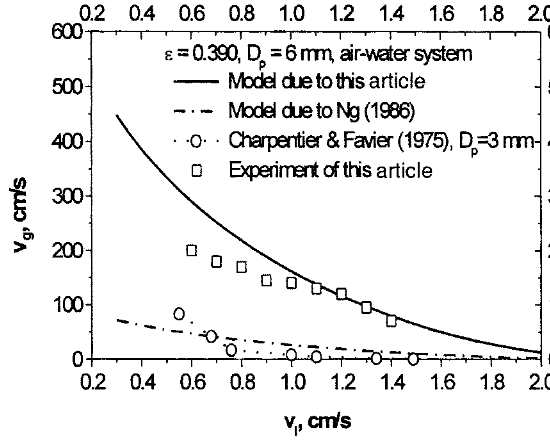

Figure 7. Comparison of model prediction with experiment at an increased packing size.

The only difference between these two equations is the coefficient ahead of the term $\rho_g u_g^2$ on the lefthand side. It is speculated under identical conditions that the interstitial gas velocity u_g evaluated from Eq. 20 would be 4.65 to 5.77 times that of Eq. 21. Since Ng's prediction for the gas velocity is usually 1/4 to 1/5 of the experimental value, the result given by Eq. 20 appears very promising.

Precise predictions of the pulsing flow conditions are given in Figures 5 to 10. Figures 5 and 6 are comparisons of predictions by different models with the correlation of Charpentier and Favier (1975), as well as experimental results of Chou et al. (1977) and those of the authors. The result is found to be satisfactory, both in trend and magnitude. This finding confirms, to a large degree, the validity of the model of the present work. There is also a comparison made for a larger packing with $D_p = 6$ mm (Figure 7), and the result is also promising. Influence of particle size is shown in Figure 8, where it is found that within the abscissa range, the profiles corresponding to $D_p = 3$ mm and $D_p = 1.9$ mm both exhibit two branches. It can be speculated that the left branch is the boundary between trickling flow and pulsing flow, while the right one is the boundary between dispersed bubbling flow and pulsing flow.

Effects of liquid viscosity and surface tension are separately investigated, as shown in Figures 9 and 10. It should be noted that in these two figures, no experimental results are given for comparison despite the correlation of Charpentier and Favier (1975). The liquid is not real, since the viscosity cannot actually be changed while simultaneously keeping the surface tension constant, and vice versa. More importantly, foaming cannot be prevented, although the purpose is only to study surface tension's influence. From Figures 8, 9 and 10, the predictive trends are found in agreement with the well accepted observations and correlation of Charpentier and Favier (1975), that is, pulsing flow tends to occur under conditions with small packing size, high liquid viscosity, and low surface tension. These facts are all ascribed to the occlusion mechanism as suggested by Hofmann (1978) and Sicardi et al. (1979). Theoretically, they can be explained in context of this article. Decreasing the packing size makes the channel diameter D_c decrease (Eqs. 11 and 12) and the film thickness

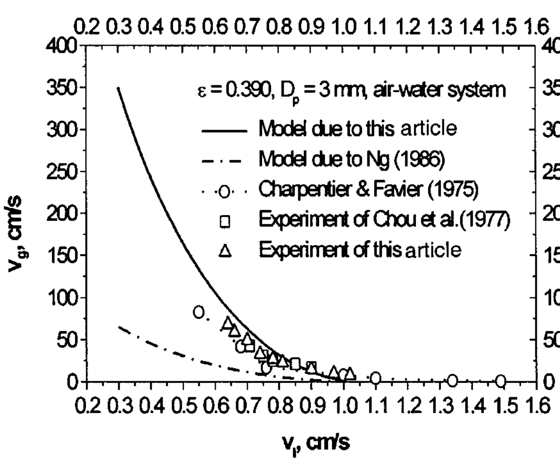

Figure 6. Verification of model prediction with experiment: normal packing case.

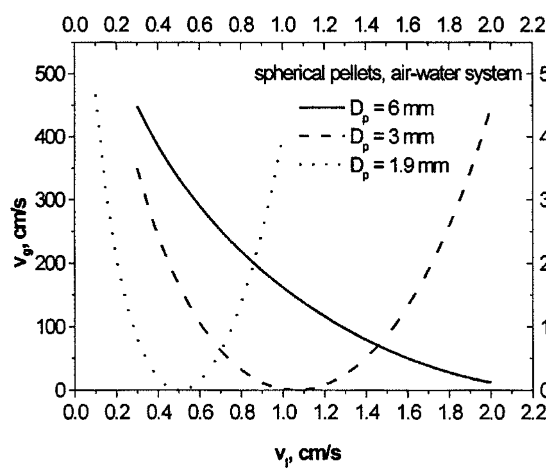

Figure 8. Prediction of the influence of packing size on pulsing flow inception.

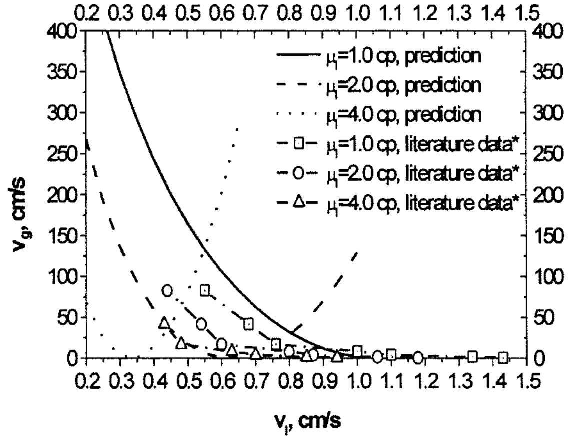

Figure 9. Influence of liquid viscosity on pulsing flow inception (Charpentier and Favier, 1975).

S_d increase (Figure 4). Increasing the liquid viscosity increases the liquid saturation β (Eq. 19) and, therefore, the flow area α (Eq. 18). Decreasing the liquid surface tension will make the liquid film deform, even at a low gas velocity (Eqs. 9 or 20).

Conclusions

Beginning with the fundamental equations of fluid dynamics, from the understanding of the manner of the existence of gas and liquid in a trickle bed, and from the principles of two-phase pipe flow, the authors were able to develop a pulsing inception predictive model. The validity of this model was confirmed with respect to the air-water system by experimental data from different sources. Extensions to systems other than air and water, using the same basic principles, are expected to be promising, although more fundamental work will be required.

Regarding the relation of the theory in the mechanism of pulsing flow inception, Ng's (1986) attempt may be considered as the sufficient condition, while that of the present approach may be considered as the necessary one. That is to say, Ng's model emphasizes the kinematic energy of the gas, and should be considered as the preliminary and sufficient condition. In comparison, the present model demands much higher gas flux to provide sufficient gas-liquid frictional force in overcoming the stabilizing capillary force, and should be considered as the essential and necessary condition.

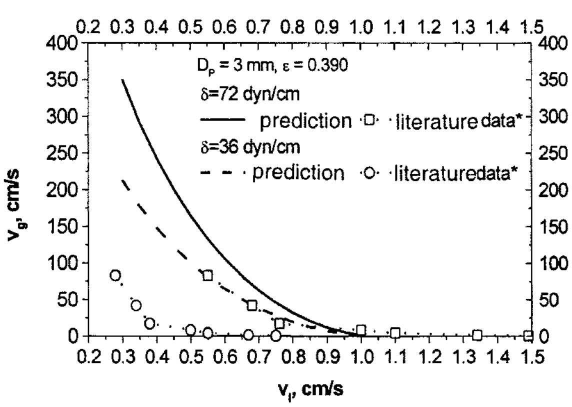

Figure 10. Influence of liquid surface tension on pulsing flow inception (Charpentier and Favier, 1975).

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Notation

C_g = parameter in Eq. 3, dimensionless
D_p = packing diameter, m
D_t = packed bed diameter, m
f_g = friction factor in the gas phase, dimensionless
h_d = dynamic liquid holdup, dimensionless
m = parameter in Eq. 3, dimensionless
N_C = number of channels per unit sectional area, dimensionless
p = pressure, N/m^2
V_d = volume of dynamic liquid, m^3
α = fraction of flow channels that is occupied by the liquid, dimensionless
α_w = fraction of total wetted area, dimensionless
α_{ws} = fraction wetted by the static liquid, dimensionless
α'_{ws} = fraction of meniscus of static liquid, dimensionless
β = average liquid saturation in the bed, dimensionless
μ = gas or liquid viscosity, kg/m·s
ρ = gas or liquid density, kg/m^3
σ = gas-liquid surface tension, N/m

Subscripts

1, 2, 3, 4 = locations in Figure 1
g = gas phase
l = liquid phase

Literature Cited

Baker, O., "Simultaneous Flow of Oil and Gas," *Oil Gas J.*, **53**, 185 (1954).

Charpentier, J. C., C. Prost, W. P. M. Van Swaaij, and P. Le Goff, "Etude de la Retention de Liquid dans une Colonne a Garnissage arrosee a Contre-courant et à Cocourant de Gaz et de Liquide: Representation de sa Texture par un Modele à Films, Filets et Gouettes," *Chimie et Industrie, Genie Chimique*, **99**, 803 (1968a).

Charpentier, J. C., C. Prost, and P. Le Goff, "Ecoulement Ruisselant de Liquide dans une Colonne a garnissage. Determination des vitesses et des Debits Relatifs des Films, des Filets et des Gouttes," *Chimie et Industrie, Genie Chimique*, **100**, 53 (1968b).

Charpentier, J. C., "Liquid Texture in Films, Rivulets and Drops, and Residence Time Distribution in Gas-Liquid Absorption Packed Columns," *3rd CHISA Congress, Section G, Marianske Lazne*, Czechoslovakia (Sept. 12–20, 1969).

Charpentier, J. C., and M. Favier, "Some Liquid Holdup Experimental Data in Trickle-Bed Reactors for Foaming and Nonfoaming Hydrocarbons," *AIChE J.*, **21**, 1213 (1975).

Cheng, Z. M., and W. K. Yuan, "Estimating Radial Velocity of Packed Beds with Low Tube-to-Particle Diameter Ratios," *AIChE J.*, **43**, 1319 (1997).

Chou, T. S., F. L. Worley, Jr., and D. Luss, "Transition to Pulsed Flow in Mixed-Phase Cocurrent Downflow through a Fixed Bed," *Ind. Eng. Chem. Proc. Des. Dev.*, **16**, 424 (1977).

Ergun, S., "Fluid Flow through Packed Columns," *Chem. Eng. Prog.*, **48**, 89 (1952).

Hofmann, H., "Multiphase Catalytic Packed-Bed Reactors," *Catal. Rev. Sci. Eng.*, **17**, 71 (1978).

Holub, R. A., M. P. Dudukovic, and P. A. Ramachandran, "Pressure

Drop, Liquid Holdup, and Flow Regime Transition in Trickle Flow,'' *AIChE J.*, **39**, 302 (1993).
Larkins, R. P., R. R. White, and D. W. Jeffrey, ''Two-Phase Cocurrent Flow in Packed Beds,'' *AIChE J.*, **7**, 231 (1961).
Lockhart, R. W., and R. C. Martinelli, ''Proposed Correlation of Data for Isothermal Two-Phase, Two-Component Flow in Pipes,'' *Chem. Eng. Prog.*, **45**, 39 (1949).
Mueller, G. E., ''Prediction of Radial Porosity Distributions in Randomly Packed Fixed Beds of Uniformly Sized Spheres in Cylindrical Containers,'' *Chem. Eng. Sci.*, **46**, 706 (1991).
Ng, K. M., ''A Model for Flow Regime Transitions in Cocurrent Down-Flow Trickle-Bed Reactors,'' *AIChE J.*, **32**, 115 (1986).
Rao, V. G., M. S. Ananth, and Y. B. G. Varma, ''Hydrodynamics of Two-Phase Cocurrent Downflow through Packed Beds,'' *AIChE J.*, **29**, 467 (1983).
Satterfield, C. N., A. A. Pelossof, and T. K. Sherwood, ''Mass Transfer Limitations in a Trickle-Bed Reactor,'' *AIChE J.*, **15**, 226 (1969).
Sicardi, S., H. Gerhard, and H. Hofmann, ''Flow Regime Transition in Trickle-Bed Reactors,'' *Chem. Eng. J.*, **18**, 173 (1979).
Sicardi, S., and H. Hofmann, ''Influence of Gas Velocity and Packing Geometry on Pulsing Inception in Trickle-Bed Reactors,'' *Chem. Eng. J.*, **20**, 251 (1980).
Sicardi, S., G. Baldi, A. Gianetto, and V. Specchia, ''Catalyst Areas Wetted by Flowing and Semistagnant Liquid in Trickle-Bed Reactors,'' *Chem. Eng. Sci.*, **35**, 67 (1980).
Sicardi, S., G. Baldi, V. Specchia, I. Mazzarino, and A. Gianetto, ''Packing Wetting in Trickle Bed Reactors: Influence of the Gas Flow Rate,'' *Chem. Eng. Sci.*, **36**, 226 (1981).
Taitel, Y., and A. E. Dukler, ''A Model for Predicting Flow Regime Transitions in Horizontal and Near-Horizontal Gas-Liquid Flow,'' *AIChE J.*, **22**, 47 (1976).
Talmor, E., ''Two-Phase Downflow Through Catalyst Beds,'' *AIChE J.*, **23**, 868 (1977).

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