Origins of Pulsing Regime in Cocurrent
Packed-Bed Flows

B.A. Wilhite\textsuperscript{+}, B. Blackwell\textsuperscript{++}, J. Kacmar\textsuperscript{++}, A. Varma\textsuperscript{#} and M. J. McCready\textsuperscript{*}

Department of Chemical Engineering
University of Notre Dame
Notre Dame, IN 46556

*Correspondence regarding this article should be addressed to M. J. McCready

\textsuperscript{+} Current Address: Department of Chemical Engineering, Massachusetts Institute of Technology, Cambridge MA.

\textsuperscript{++} Current Address: Department of Chemical Engineering, University of Minnesota, Minneapolis, MN.

\textsuperscript{#} Current Address: School of Chemical Engineering, Purdue University, West Lafayette, IN.

March 2004
Abstract

The mechanism of the formation for cocurrent downflow pulse flow was studied experimentally in a packed-bed of inert spheres of 3, 6, and 8 mm using an air-water flow. By measuring the flow distance until pulses are observed, the spatial growth rate of convective disturbances within the pulsing-flow regime were determined. Observations indicate that pulses form from trickling-flow as the result of a global convective instability. Further, experiments indicate that an analogous transition exists for the formation of pulses from the dispersed bubble flow regime, except that pulses form as the flowrates are adjusted to become less severe. Existing global instability models based on averaged (dispersed flow) momentum equations were modified to explain experimental results. A key uncertainty in modeling pulse formation from trickle flow is the regularization (i.e., stabilization) force. Reexamination of this issue suggests some mechanistic inconsistencies with surface tension which had been used in previous studies. Consistent with the present experiments, it is proposed that gravity may be the primary restoring force. Incorporating gravity stabilization into the dispersed flow equations provides predictions that are at least as good as the previous models. A similar dispersed flow model is used to explain the bubbly flow to pulse transition. While predictions agree with experimental data for part of the range, model accuracy is limited by the accuracy of constitutive expressions for interaction forces between phases.

Keywords: Packed-bed, trickling-flow, pulsing-flow, dispersed-bubbling flow, convective instability.
Introduction

Multiphase reactions, in which gas and liquid reactants are selectively converted into desired products using solid catalysts, provide the basis for a large number of chemical, petrochemical, biochemical and polymer processes (Mills et al. 1992, Dudukovic et al. 1999). A common configuration for carrying out such reactions is a three-phase packed-bed reactor, in which gaseous and liquid reactants flow in cocurrent manner over a stationary bed of catalyst. Multiple flow regimes may occur within this system, depending upon bed parameters, fluid properties and throughputs. The trickling-flow regime exists at low gas and liquid flows and is characterized by rivulet and/or film flow of the liquid phase over the packing, with the gas phase passing continuously through the remaining void. At increased gas and liquid throughputs, liquid-rich waves, or 'pulses', form and traverse the column length at almost regular time intervals, resulting in local fluctuations in liquid holdup, pressure drop, heat and mass transfer rates. It should be noted that the majority of industrial processes operate at or near the transition from trickling- to pulsing-flow (Dudukovic et al., 1999). Additionally, pulsing flow enhances overall heat and mass transport while reducing axial dispersion, making it a potentially attractive mode of operation. Recent experimental work has demonstrated that, holding all other parameters constant, reactor operation in the presence of pulses results in up to 30% increase in reaction rate (Wilhite et al., 2001). These findings clearly indicate that an accurate understanding of this regime, including mechanism of pulse formation and necessary conditions for onset of pulsing flow, are crucial for optimum reactor design.
Macroscopic models for two-phase flow in vertical packed-beds, based on spatially averaged mass and momentum equations and employing constitutive equations for drag and capillary forces, have been developed to predict transition from trickling to pulsing-flow. Perturbation analysis of the steady state corresponding to trickling-flow is used to develop criteria for the onset of bed instability, or pulsing-flow. Calculations by Grosser et al. (1988) compared favorably with experimental data obtained in an air-water system. An improved model by Dankworth et al., (1990) qualitatively predicted the hysteresis behavior of the trickling to pulsing-flow transition. The most recent model developed by Attou and Ferschneider (1999), including modified terms for the fluid drag and bed capillary forces, compared well with experimental data for a range of fluid and bed properties. However, close agreement between observed pulse formation within a particular bed depth and flow conditions predicted from a stability model does not prove that the model is correct. Hydrodynamic instabilities in flowing systems are usually "convective instabilities" (Huerre and Monkewitz, 1985) meaning that the disturbance grows as it travels in the flow direction. As discussed by Bruno and McCready (1988) for waves in a channel and Krieg et al. (1995) for pulsing flow, if the disturbance grows with distance, at the neutral stability prediction of a model, the growth rate is zero and thus infinite distance would be required for the disturbance to reach a macroscopic size. If the model is correct, disturbances would be observed in a finite column length only at conditions beyond those predicted for neutral stability, and the location of observed disturbances would approach the inlet as the conditions are adjusted to increase the growth rate. It is this idea that Wilhite et al. (2001) exploited to allow reaction studies to be performed both with and without pulsing at constant flow conditions. Because this
criterion for transition has not been applied, it is not clear that the models discussed above correctly characterize pulse formation.

Melli and coworkers (1990) described bed behavior in terms of single-pore flow regimes based upon visual observations using high-speed video imaging of an idealized two-dimensional column. It was noted that while bed-scale pulses first appeared near the column exit at the onset of pulsing-flow, their origin was found in smaller scale disturbances that originated near the column inlet and grew downstream. Within the pulsing flow regime, individual pores were observed to be in trickling-flow between pulses, and as the dispersed-bubbling regime within the pulses. At the transition from pulsing to dispersed-bubbling flow, the gas-rich (trickling-flow) regions reduced in size until becoming trapped as localized pockets as the remainder of the bed operated under dispersed-bubbling flow. Visual investigations in a three-dimensional laboratory-scale column reported by Tsochatzidis and Karabelas (1994) also confirmed localized disturbances analogous to those reported for two-dimensional systems, and suggested a mechanism for pulse formation based on the growth of such disturbances as they traverse the column until spanning the entire column cross section. These reports of local events growing into pulses are inconsistent with the models of Grosser et al. (1988) and Attou and Ferschneider (1999). Because these models are based on the dispersed flow equations, the "instability" is manifested as a disturbance in void fraction that grows with distance. Owing to the averaging employed in the formulation, the void fraction is not defined in a local region, but instead only in regions of many particles. Further, the
formulation is one-dimensional (in the axial direction) such that pulse formation occurring by transverse growth is also inconsistent with these models.

The aim of the present work is to resolve existing questions regarding the mechanism of pulse formation and to determine the best framework for a theoretical model. The location of pulse appearance was observed for different conditions, and results indicate that pulses form from trickling-flow as the result of a global convective instability. Further, it appears that a similar transition exists for the formation of pulses from dispersed-bubble flow regime. This transition is also a convective instability, in which pulses form as the liquid flowrate is decreased, thus allowing regions of higher void fraction to appear. In light of these observations, models based on the dispersed flow equations are presented for both transitions. For the trickling- to pulsing-flow transition, the primary difference from previous models is reformulation of the pressure difference between the gas and liquid phases to be determined by gravity. This term allows stabilization by gravity, which acts to smooth disturbances in the void fraction. In previous models, surface tension was argued to be the primary stabilizing term. However as will be discussed below, this proves difficult to justify mechanistically because surface tension does not appear to be able to spread out regions of higher liquid concentration, which are the precursors of pulses, for both wetting and non wetting liquids.

**Experimental**

While qualitative trends regarding the location of pulses within the column at the transition from trickling- to pulsing-flow have been reported in numerous sources, no
quantitative information exists regarding pulse appearance throughout the pulsing-flow regime. It was therefore of interest to determine the column height at which individual pulses appear as a function of gas and liquid flow for multiple packing geometries, to assess the validity of describing the pulsing-flow regime in terms of a convective disturbance.

**Apparatus**

Experiments were performed using an air-water system employing multiple packing sizes over a range of gas and liquid flows. A schematic of the apparatus utilized is shown in Figure 1. The column used for this study was 1.3 m length, 1.27 cm in wall thickness, 7.62 cm ID acrylic tube, packed with non-porous glass spheres to a height of 1.25 m. A highly porous section of foam followed by a fine screen served as a precursor to the packing to ensure even distribution of liquid. Uniform introduction of gas and liquid to the column was provided by a 10 cm high monolith "honeycomb" inlet section, with roughly 1/3 of the passages (0.35 cm diameter) used for gas flow and the remainder liquid, following the design used by Herskowitz and Smith (1978). The column emptied into a six-gallon separation tank, which in turn drained back to the reservoir. House air was supplied at 0.965 MPa in a 1/2-in line equipped with particulate filter and oil traps. Supply pressure to the apparatus was reduced using a Speedair model R72G-2AT-RMG single-stage regulator to 0.201 MPa. The gas flow rate from the regulator to the column inlet was measured using a Omega model FL-1502A flow meter. Water was supplied from a 6 gallon reservoir using a Eastern model U-34D centrifugal pump. The liquid flow
rate was measured using an Omega FL-1501A flow meter for low flows and F&P model 10A4555X for increased rates.

**Experimental Technique**

Prior to each experiment, water was passed through the column at a sufficient rate to ensure complete wetting of the bed, in order to achieve reproducible observations regarding transition behavior. Gas and liquid rates were then set to the desired values and the column was allowed to equilibrate for a minimum of 15 minutes before observations were made. The point at which a specific pulse was first visually identified was marked. Measurements were based upon visual identification to avoid restricting observations to a fixed location, as would be the case with pressure transducers or conductance probes. The pulse onset distance was then measured from the column inlet to the axial position at which the individual pulse was first visually identified. Preliminary experiments were performed to determine the experimental error associated with this technique. As shown in Figure 2, standard deviation converged to a relatively stable value within 20 measurements for all three packing diameters. Average values for pulse onset distance were therefore calculated from groupings of 20 measurements at each set of operating conditions. Averaged onset distances obtained using 3,6 and 8 mm glass spheres over the range of gas and liquid flow rates encompassing the pulsing-flow regime are presented in Figure 3.
Results and Discussion

The observations demonstrate that at a constant gas flow, pulses first appear near the column exit as the liquid flow rate is increased. Further increases in liquid rate result in a rapid decrease in onset location, until the majority of the bed operates in the presence of pulses. At further increments of liquid flow, pulses retreat to the column outlet, marking the transition from pulsing-to-dispersed-bubbling flow. In all experiments, there always existed a finite portion of the bed near the column inlet in which pulses did not occur, as indicated by the presence of non-zero values for the pulse onset distance. At the trickling to pulsing transition, this inlet region appeared to behave as trickling-flow, while at the pulsing-to-dispersed bubbling transition, the inlet region appeared similar to the dispersed-bubbling regime.

In each limiting case, pulses appeared to form from traveling disturbances, observed as waves with higher liquid fraction, originating within the inlet region. Results presented for all three packing sizes indicate two trends regarding the interplay of the disturbance growth rate and velocity. First, near the onset of pulsing flow an increase in liquid feed resulted in an increase in disturbance growth rate relative to the disturbance velocity. Second, near the transition from pulsing- to dispersed-bubbling flow, an increment in the liquid throughput resulted in an increase of disturbance velocity relative to the growth rate. These trends then suggest that the growth rate increases from zero at the onset of pulsing-flow to a maximum value within the fully developed regime, before decreasing to zero at the onset of dispersed-bubbling flow.
The experimental results illustrate the need for accurate understanding of the spatial growth rate in order to predict the transition from trickling- to pulsing-flow, as reflected in the slope of the data near this transition (Figure 3). It is seen that if the column length is doubled, then the minimum liquid Reynolds number for pulse appearance decreases by \(\sim 10-20\%\). The uncertainty could be larger for the dispersed-bubble to pulsing-flow transition, owing to milder slopes in pulse onset distance with liquid Reynolds number approaching that limit. It should be noted that this dependence upon column length is not accounted for by available hydrodynamic models or regime maps.

**Theory**

Experiments indicate that for both the trickling to pulse transition and the bubbly to pulse transition, pulses appear to form as the result of a convective instability manifested as void waves traveling through the entire bed, not a specific local feature of the fluid dynamics. Equivalent phenomena have been observed in horizontal gas-liquid packed-bed flows (Krieg, et al., 1995), as well as for other wave behavior in non-packed conduits (Jurman et al., 1992; Lahey, 1991). It is therefore reasonable to attempt to model the transitions to pulsing flow using linear stability analysis of spatially (or ensemble) averaged mass and momentum equations, an idea that dates to Jackson (1963). These equations work best when there is no particular structure to the base state flow, such as a dilute gas-solid flow or a dispersed bubbly gas-liquid flow. While radial inhomogeneities exist in the trickling-flow regime, most commonly in the form of preferentially gas- or
liquid-filled channels or local stagnant zones, the length scales of such inhomogeneities are small compared to that of the most important fluid dynamic phenomenon (i.e. void waves). Thus, ensemble-averaged equations independent of flow structure and utilizing closure expressions that capture the small-scale behavior are applicable. Here we use these equations and discuss their validity for the trickling to pulsing and bubbly to pulsing-flow transitions.

*Formulation of the model*

The basic averaged equations for two-phase interpenetrating flows are given by Drew and Passman (2000):

\[
\frac{\partial \sum_k \phi_k}{\partial t} + \nabla \cdot \sum_k \phi_k \vec{v}_k = 0 \quad (1)
\]

\[
\frac{\partial \sum_k \phi_k \vec{v}_k}{\partial t} + \nabla \cdot \sum_k \phi_k \vec{v}_k \vec{v}_k = \nabla \cdot \sum_k \left( \tau_k + T_k^{Re} \right) + \sum_k b_k \quad (2)
\]

where \( \phi_k \) represents the fraction of the bed occupied by phase \( k \), \( \vec{v}_k \) are the interstitial velocities of phase \( k \) and \( \bar{\rho}_k \) are the mass-weighted average density of the \( k^{th} \) phase, \( \bar{T}_k \) is the total viscous stresses, \( T_k^{Re} \) is the Reynolds stress and \( b_k \) represents body forces upon the fluid. It is important to point out that these equations describe flow of interspersed fluid phases, and with proper closures can give accurate description of flow in complex multiphase systems. The most extensive use of such equations has been for predicting onset of instabilities in the local void fraction for solid-gas and gas-liquid flows (Lahey, 1991). Equations 1 and 2 are functionally equivalent to the set of equations employed by Anderson and Jackson (1967), who give closures that we will adapt for the bubbly-pulse
transition, and by Grosser et al (1988), which we will use and modify for the trickle to pulse transition. As discussed by Anderson and Jackson (1968), the two momentum and mass balance equations can be converted into a single wave equation of the general form

$$C_2 \frac{\partial^2 \phi}{\partial t^2} + C_1 \frac{\partial \phi}{\partial t} + C_0 \phi = 0$$

(3)

with spatial derivatives contained in terms $C_2$, $C_1$ and $C_0$. Substitution of a small perturbation in the volume fraction term in the form of

$$\phi = \phi \exp(st + jwz)$$

(4)

where $s$ is a complex frequency and $w$ is a real wavenumber in the flow direction $z$ and $j = \sqrt{-1}$, yields the neutral stability condition

$$s^2 \cdot W_1 + s \cdot W_2 + 2sjw \cdot W_3 \phi \cdot W_4 + jw \cdot W_5 = 0$$

(5)

Details from Grosser et al. (1988) are given in Appendix I. Equations of this type have been used extensively in hydrodynamic analysis, including falling films (Chang and Demekhin 2002, Balakotaiah et al. 1995) and sheared liquid layers (Fukano, 1986; Jurman and McCready, 1989), as well as classical stability analysis (Whitham, 1974). Using this methodology, the analysis of Grosser et al. (1988) yielded the following expressions for $W_i$ coefficients corresponding to the trickling-flow regime:

$$W_1 = \frac{\partial g}{\partial \rho} + \frac{\partial \rho}{\partial g}$$

(6)

$$W_2 = \phi \frac{\partial \rho}{\partial \rho} \frac{\partial g}{\partial \rho}$$

(7)

$$W_3 = \frac{\partial u^0}{\partial g} + \frac{\partial u^0}{\partial \rho}$$

(8)
Analysis of the above expressions indicates that $W_1$ contributes to destabilization as do the first two terms of $W_4$. Term $W_3$ provides stabilization, but not enough to overcome the other terms. Thus the surface tension, the last term of $W_4$, is the key to creating regions of stability so that a transition to instability can be computed. More discussion of the mechanism will be given below, but it is useful to note that if surface tension is stabilization the void fraction “waves”, it must be acting to cause drainage of liquid from higher fractions (lower void) to lower fraction (higher void).

Grosser et al. (1988) introduced surface tension as a stabilizing force into their model expressions by employing a capillary pressure model adapted from flow in porous media, the Leverett drainage function to relate the gas- and liquid-phase pressures,

$$\rho_g \bigg[ \rho_l \bigg] = \frac{1}{k} \bigg[ \frac{\mu_g}{\mu_l} \bigg]^{1/2} \frac{\partial \ln (\theta)}{\partial (\frac{z}{R})} \bigg).$$

(11)

It should be noted that this closure expression dictates that the gas-phase pressure is greater than that of the liquid phase. Grosser et al. (1988) don’t explain the mechanism for how surface tension provided stabilization so this will be attempted here. For the case of a wetting (i.e., contact angle $> \theta/2$) liquid in a partially filled void, then the pressure within the trapped liquid will be less than that of the gas, as illustrated in Figure 4 by the concave shapes to the liquid interfaces which become less concave left to right.
(indicative of lower to high liquid fraction). As the liquid fraction inside a given pore increases, the interfacial curvature decreases, corresponding to an increase in the liquid pressure (and reduction in the pressure difference). Thus, if liquid trapped in higher liquid fraction regions is somehow connected to liquid-poor regions, the lower pressure in the liquid poor regions would draw liquid from the liquid-rich regions, resulting in a net redistribution of local liquid fraction that acts to stabilize the bed voidage. The rate at which this could occur would presumably be determined by the value of the surface tension and the viscosity of the liquid. Because of the importance of the interface shape, it is not obvious how to extend this stabilization mechanism to non-wetting systems. However, studies have been published for both wetting and non-wetting systems and the basic instability mechanisms appear to be similar. This suggests that it is possible that a mechanism independent of capillary action, acts to stabilize voidage waves in the trickling-flow regime.

Attou and Ferschneider (2000) also attempt to introduce capillary pressure as a stabilizing force, by employing the following closure expression to relate the gas and liquid-phase pressures:

\[ p_g - p_l = \frac{1}{d_1} + \frac{1}{d_2} \]  \hspace{1cm} (12)

where \( d_1 \) and \( d_2 \) are the two principle radii of curvature of the gas-liquid interface at the pore scale for the liquid films. In their figure 3, which shows liquid covering a particle creating a convex surface, these two radii are just those necessary to give the curvature of the liquid on the particle which is necessarily convex. For this geometry, the pressure in the liquid is thus higher than the gas (see our figure 5) and thus eq. 12 has the wrong sign.
However if equation 12 is used as is, stabilization will occur, consistent with equation 11, because the gas pressure is higher than the liquid and this difference decreases as the liquid fraction increases. Thus the error in the sign fixes Attou’s otherwise inconsistent argument. This is shown in figure 5 where the left sphere with less liquid (signifying lower liquid fraction) has a higher pressure (smaller radius) than the one on the right with more liquid (larger r). If a connection (e.g., a fluidic bridge) is made between them then the higher pressure liquid will flow (to the right) into the lower pressure region. This is the opposite of what is needed for stabilization!

The conceptual and mathematical errors of Attou and Ferschneider (2000) aside, there is still some question about how surface tension can act as the stabilizing force in trickling-flow. If surface tension were the dominant stabilizing force, then for the case of a local region of higher than average liquid fraction introduced into a packed column, the excess liquid would be expected to conduct upward as well as downward. However, in experiments where a dye solution was injected into the column midsection in the presence of cocurrent gas and liquid downflow, the excess liquid flowed only in the direction of gravity. Identical results were obtained in the absence of gas and liquid flow. While this does not prove that surface tension is not the correct regularizing force, it suggests that gravity could be important and should be considered in the stability analysis of trickling-flow.

The importance of gravity in preventing pulse flow is supported by recent studies by Motil et al. (2003) who have shown that the stable trickle-flow regime does not occur
under microgravity conditions, indicating that gravity is necessary for trickle flow. Gravity is responsible for reducing the static liquid holdup and, in the absence of gravity, higher liquid holdup would facilitate pulsing.

The question that arises when exploring how gravity can act to stabilize the trickle regime, is what term is missing that would be responsible for gravity stabilization? A possible answer is that the closure for the pressure difference between the phases is not given by a surface tension term but has an effect of the hydrostatic pressure in the discontinuous phases. Previous models discussed above have assumed $P_g - P_l$ to be purely a function of surface tension. However, consider the situation shown in figure 6a where the phases are not truly evenly dispersed but there is an uneven radial distribution of the liquid fraction, which commonly occurs in packed-bed systems under trickling-flow conditions. At a given axial position, the averaged liquid-phase pressure would be greater than that of the gas phase, owing to the hydrostatic head associated with local regions of elevated liquid holdup as seen in figure 6b. The magnitude of the difference would depend on the length-scale and local liquid fraction corresponding to these regions. Visual evidence suggests that these ‘clusters’ are typically multiple (~4-10) particles in size, which is here termed $\bar{d}$ with $d_p$ being the particle diameter. For an average cluster height of $\bar{d}d_p$, liquid density $\bar{\rho}_l$, total void fraction $\bar{e}_{\text{bed}}$, and liquid fraction $\bar{e}(z)$, hydrostatic arguments suggest a new closure expression relating the gas and liquid pressures:

$$P_l(z)\bar{d}P_g(z) = \frac{1}{2} \bar{\rho}_l g \bar{d}D_p \bar{e}(z)/\bar{e}$$  \hspace{1cm} (13)
where the 1/2 comes from vertical averaging. Substituting eqn (13) into the stability model of Grosser et al. (1988) yields an identical solution except for the $W_4$ term, which becomes:

$$W_4 = \frac{(u_{gz})^2}{\bar{d}_g} + \frac{(u_{lz})^2}{\bar{d}_l} \frac{1}{2} \frac{\rho \cdot g \cdot \bar{d} \cdot \bar{d}}{\bar{d}_{bed}}$$

(14)

The predicted trickling-to-pulsing transition, using the above modifications for the case of $\bar{d} = 2$ and $\bar{d} = 9$, are compared with predictions from Grosser et al. (1988) and experimental data of Sato et al. (1973) in Figure 7 using physical parameters summarized in Table 1. It is seen that if the pertinent length scale for a region of higher liquid fraction is 9 particle diameters, then predictions are similar to the Grosser model and roughly consistent with the experimental data. The improved model provides good accuracy and is consistent with observations made under both normal and zero gravity conditions.

**Modeling the Pulsing-to-Dispersed Bubbling Flow Transition**

Visual observations in figure 3 under pulsing-flow conditions suggest that the transition from dispersed-bubble to pulsing-flow, with decreasing liquid flow for constant gas flow, also results from a convective instability. At this upper boundary of the pulsing-flow regime, the base state flow pattern is now dispersed-bubble flow, as opposed to trickling-flow, which is the base state flow pattern at the lower boundary. Thus, the pulsing-flow behavior is a convective disturbance existing between two *distinct* base states. As mentioned above, a model to describe the dispersed-bubble regime and the transition to
pulsing can be based on the same basic equations as the trickle to pulse transition. The model differs from those employed to predict the trickling-to-pulsing transition in the closure expressions and by the inclusion of significant interaction terms between the two fluid phases.

In contrast to the analysis for trickling flow instability where both phases are continuous and there is no particular structure to the phases, for a bubbly flow the liquid is continuous but the gas exists as entrained bubbles. Mathematical formulation for this situation is analogous to a fluidized bed (Anderson and Jackson, 1968) except that the particles are bubbles and the drag relations are thus altered as is the direction of the flow. Drag from the solid phase is also ignored. Stability analysis was applied to a traveling frame of reference moving at a velocity equal to that of the gas phase; thus the average relative velocities \( u_0 \) and \( v_0 \) of the liquid and gas phases were defined as:

\[
u_0 = 0 \tag{16}\]

\[
\begin{align*}
u_0 &= \frac{Q_l}{Q_g (1 - e_l) \text{ bed} \cdot a_{bed}} \frac{1}{(1 - e_l)} \end{align*}\tag{15}
\]

for liquid and gas volumetric flow rates \( Q_l \) and \( Q_g \) and a bed cross-section area \( a_{bed} \).

Under dispersed-bubbling conditions, Rode et al. (1994) observed that the ratio of gas to liquid velocity ranged from 1.3 - 2.4; thus the relative liquid velocity within the frame of reference opposes gravity.

Interfacial drag was obtained using the expressions of Ishii and Zuber (1979) for distorted particles, thus providing the solution to the state of uniform fluidization within the traveling frame of reference corresponding to stable dispersed-bubbling flow,
\[ (\mathcal{B}(Q_0)) \cdot u_0 \cdot (1 - \mathcal{B}(Q_0)) \cdot (r_l - r_g) \cdot g = 0 \]  

(17)

\[ \mathcal{B}(Q_0) = 2 \mathcal{B}_r \cdot u_0 \cdot (1 - \mathcal{B}(Q_0)) \cdot \frac{g \cdot (Q_l - Q_g) \cdot u_0}{Q_{r,0} \cdot \mathcal{B}} \]  

(18)

where \( \mathcal{B}(Q_0) \) is the base state, liquid-gas drag coefficient.

The stability analysis of Anderson and Jackson (1968) can then be applied to this solution to determine the growth rate and velocity of voidage disturbances within the traveling frame of reference.

The details of this analysis are provided in Appendix II. Table 2 summarizes values for all parameters used for this study. The predicted pulse onset distance near the transition to dispersed-bubbling flow regime is presented in Figure 8a as a function of gas and liquid Reynolds number. As demonstrated in Figure 8b, at low gas flows, the minimum liquid flows for stable fluidization predicted by this model agree reasonably well with experimental data for the pulsing-to-dispersed-bubble transition as determined in the present work and also by Sato and coworkers (1973). This agreement is less satisfactory for high gas flows, but is expected to improve with the addition of solid-liquid drag and refining the liquid-gas drag expressions.

**Concluding Remarks**
Experiments were performed to measure the dependence of spatial onset for pulses on the gas and liquid flows, spanning the pulsing-flow regime. Results indicate that the distance from the column inlet at which pulses first appear approaches the column length at the transition from trickling to pulsing flow and at the upper limit of pulsing to dispersed-bubble flow. This onset distance approaches a minimum value, corresponding to a majority of the bed operating in the presence of pulses, between two boundaries of the pulsing-flow regime. This is the first time that pulse formation from a bubbly flow basestate has been reported. An improved version of the model of Grosser et al. (1988) was proposed, based upon findings indicating that gravity, not surface tension, is the restoring force in the trickling-flow regime. While the present study cannot confirm that surface tension is not regularizing the small amplitude void fraction waves, the question of how it can act similarly for both wetting and non-wetting situations and the need to justify it is indeed stronger than gravity as important issues are clarified here. In addition, a model based on the stability of a fluidized-bed of distorted particles was developed and shown capable of describing trends in pulse onset distance and conditions for the start of the dispersed-bubbling flow regime, thus demonstrating that this transition also follows a convective instability mechanism. While models presented in the current work agree qualitatively with reported trends, additional information regarding the interaction terms in their corresponding base flows is required to improve the quantitative agreement with experimental data.

Acknowledgements
We gratefully acknowledge the National Science Foundation (grant EEC-9700537) and the Arthur J. Schmitt Chair Fund at the University of Notre Dame for support of this research. B. A. Wilhite gratefully acknowledges the award of a Schmitt Fellowship. We are also indebted to James Smith for his assistance with construction of experimental apparatus.
**Notation**

\[ a = \text{cross-sectional area, m}^2 \]
\[ A = \text{disturbance amplitude, m} \]
\[ C = \text{virtual mass coefficient, dimensionless} \]
\[ d = \text{diameter, m} \]
\[ g = \text{gravitational acceleration, m.sec}^{-1} \]
\[ h = \text{channel height, m} \]
\[ i = \text{unit vector in direction of fluid flow within mobil frame of reference} \]
\[ k = \text{perturbation wave vector} \]
\[ n = \text{local mean value of number of fluidized particles per unit volume} \]
\[ p = \text{pressure, N.m}^2 \]
\[ p' = \text{local mean value of particle pressure} \]
\[ Q = \text{volumetric feed rate, m}^3\text{.sec}^{-1} \]
\[ \text{Re} = \text{Reynolds number, dimensionless} \]
\[ t = \text{time, sec} \]
\[ u = \text{superficial fluid velocity within mobile frame of reference, cm.sec}^{-1} \]
\[ \bar{u} = \text{superficial fluid velocity within mobile frame of reference, cm.sec}^{-1 \]
\[ v = \text{bubble velocity within mobile frame of reference, cm.sec}^{-1 \]
\[ x = \text{axial position, m} \]
Greek Letters

\( \boldsymbol{e} \) = fluid-particle drag coefficient, kg.cm\(^{-3}\).sec\(^{-1}\)

\( \epsilon \) = void fraction, or voidage

\( \bar{\mu} \) = fluid phase effective bulk viscosity, kg.m\(^{-1}\).sec\(^{-1}\)

\( \tilde{\mu} \) = particle phase effective bulk viscosity in state of uniform fluidization, kg.m\(^{-1}\).sec\(^{-1}\)

\( \tilde{\mu}' \) = particle phase effective shear viscosity in state of uniform fluidization, kg.m\(^{-1}\).sec\(^{-1}\)

\( \bar{\eta} \) = viscosity of phase \( i \), kg.m\(^{-1}\).sec\(^{-1}\)

\( \rho \) = density, kg.m\(^{-3}\)

\( \sigma \) = interfacial surface tension, kg.sec\(^{-2}\)

\( \gamma \) = arbitrary scaling term for proposed interphase pressure expression, dimensionless

\( \lambda \) = spatial growth rate, m\(^{-1}\)

Subscripts

0 = in state of uniform fluidization

1 = first-order perturbation term

bed = of packed-bed

g = gas phase, within idealized frame of reference

G = gas phase, overall

i = initial value, corresponding to nominal disturbance

k = of \( k^{th} \) fluid phase
l = liquid phase, within idealized frame of reference
L = liquid phase, overall
P = of pulse, corresponding to fully formed disturbance
p = packing
x, y, z = cartesian coordinates

**Literature Cited**


Appendix I: Stability Analysis for Trickling-Flow, from Grosser et al. (1988)

If it is assumed that the gas and liquid are evenly distributed within the bed such that lateral variations in the dependant variables do not exist, then the following expressions describe the trickle-bed system:

\[
\square(z) + \square_{l}(z) = \square \tag{II.1}
\]

\[
\frac{d}{dz} \{l_{uz}\} = 0 \tag{II.2}
\]

\[
u_{ux} = u_{uy} = 0 \tag{II.3}
\]

\[
\square \square u_{z} \frac{d u_{z}}{dz} = \square \square \frac{d p_{l}}{dz} + F_{z} + \square \square g \tag{II.4}
\]

\[
p_{g}(z) \square p_{l}(z) = \left[ \square J[\square(z)] \right]^{1/2} \tag{II.5}
\]

where x and y denote cartesian coordinate directions perpendicular to gravity, and z axis is parallel to gravity. \(F_{iz}, \ i = l, g\) denote the drag forces acting in the z direction, taken from Saez and Carbonell (1985). Solutions to the uniform steady state can then be obtained from the above set of expressions. Linear stability analysis is performed by applying a one-dimensional perturbation around the uniform steady state.

\[
\square(z,t) = \square^{0} + \square_{i}(z,t) \tag{II.6}
\]

\[
u_{z}(z,t) = u_{z}^{0} + u_{ni}(z,t) \tag{II.6}
\]

\[
p_{l}(z,t) = p_{l}^{0}(z) + p_{ni}(z,t) \tag{II.6}
\]
By introducing (II-6) into equations (II-1-5) and linearizing, the following expressions are obtained in terms of the first-order perturbations and uniform stability solutions,

\[
\frac{\partial \bar{\Theta}_1}{\partial t} + \bar{u}_l \frac{\partial \bar{\Theta}_1}{\partial z} + u^0_i \frac{\partial \bar{\Theta}_1}{\partial z} = 0
\]  
(II.7)

\[
\bar{\Theta}_1 + \bar{\Theta}_4 = 0
\]  
(II.8)

\[
\bar{\Theta}_i \frac{\partial \bar{u}_l}{\partial t} + u^0_i \frac{\partial \bar{u}_l}{\partial z} = \bar{\Theta}_i \frac{\partial p^0_i}{\partial z} \frac{\partial \bar{\Theta}_1}{\partial z} + \bar{\Theta}_1 \frac{\partial g}{\partial \bar{\Theta}_1} + \bar{\Theta}_1 \frac{\partial u_i}{\partial \bar{\Theta}_1} + \bar{\Theta}_4
\]  
(II.9)

\[
p_{g_1} \bar{p}_{l_1} = \left( \frac{k}{\bar{J} \bar{\Theta}_1^2} \right)^{1/2} \bar{J} \bar{\Theta}_1^2 \bar{\Theta}_1
\]  
(II.10)

where

\[
\bar{J} \bar{\Theta}_1^2 = \left( \frac{dJ}{d\bar{\Theta}_1} \right)^0
\]  
(II.11)

\[
\bar{\Theta}_i = \frac{\partial F^0_i}{\partial \bar{u}_l}; \bar{\Theta}_i = \frac{\partial F^0_i}{\partial \bar{\Theta}_1}
\]  
(II.12)

The usual procedure for linear stability analysis is followed by assuming

\[
u_{l_i} = \hat{u}_i, \exp\{s t + j \bar{\Theta}_1\}
\]

\[
\bar{\Theta}_i = \bar{\Theta}_i, \exp\{s t + j \bar{\Theta}_1\}
\]  
(II.13)

\[
p_{l_i} = \hat{p}_i, \exp\{s t + j \bar{\Theta}_1\}
\]

where \( j = \sqrt{-1} \). Introducing equations (II-13) into eqns (II-7-12) and simplifying yields:

\[
W_1 s^2 + s(W_2 + 2 j \bar{\Theta}_1) \bar{\Theta}_1 \bar{\Theta}_4 \bar{\Theta}_4 j \bar{\Theta}_5 = 0
\]  
(II.14)

where

\[
W_1 = \frac{\partial \bar{\Theta}_1}{\partial \bar{\Theta}_4} + \frac{\partial \bar{\Theta}_1}{\partial \bar{\Theta}_4}
\]  
(II.15)
\[ W_2 = \frac{\partial j}{\partial \theta} \frac{\partial l}{\partial \theta} \]  \quad (II.16)

\[ W_3 = \frac{\partial g}{\partial g} u_{ge}^0 + \frac{\partial g}{\partial g} u_{le}^0 \]  \quad (II.17)

\[ W_4 = \frac{\partial g}{\partial g} \left( u_{ge}^0 \right) + \frac{\partial l}{\partial l} \left( u_{le}^0 \right) + \frac{\partial g}{\partial g} u_{le}^0 \]  \quad (II.18)

\[ W_5 = \frac{F_{le}^0}{\theta} + \frac{F_{ge}^0}{\theta} \frac{\partial g}{\partial g} \frac{\partial l}{\partial l} + \frac{\partial g}{\partial g} u_{le}^0 + \frac{\partial g}{\partial g} u_{ge}^0 \]  \quad (II.19)

Given that the criteria for stability dictates that the real part of s be less than or equal to zero, the condition for the transition from trickling to pulsing flow becomes:

\[ W_1 W_5^2 + 2W_2 W_3 W_5 + W_2^2 W_4 = 0 \]  \quad (II.20)
Appendix II. Derivation of Stability Expressions for Fluidized-Bed System

The basic equations describing the motion of a fluidized system, as developed by Anderson and Jackson (1967), consist of two continuity equations, one for the fluid and one for the fluidized gas phase:

\[
\frac{\partial \phi}{\partial t} + \frac{\partial}{\partial x_k} (\phi \cdot u_k) = 0 \quad (I.1)
\]

and

\[
\frac{\partial}{\partial t} \left( 1 \phi \right) + \frac{\partial}{\partial x_k} \left[ (1 \phi) v_k \right] = 0 \quad (I.2)
\]

combined with the two vector equations of motion,

\[
\frac{\partial}{\partial t} \left[ \begin{bmatrix} \phi u_i \\ \frac{\partial \phi}{\partial x_k} \\ \phi u_k \end{bmatrix} \right] + u_k \frac{\partial u_k}{\partial x_k} = \frac{\partial \phi}{\partial x_k} \begin{bmatrix} n 
\end{bmatrix} + \phi g_i \quad (I.3)
\]

\[
\frac{\partial}{\partial t} \left[ \begin{bmatrix} \phi v_i \\ \frac{\partial \phi}{\partial v_k} \\ \phi v_k \end{bmatrix} \right] + v_k \frac{\partial v_k}{\partial x_k} = \frac{\partial \phi}{\partial x_k} \begin{bmatrix} n 
\end{bmatrix} + (1 \phi) (\phi, \phi) g_i + \frac{\partial \phi}{\partial x_k} \quad (I.4)
\]

where

\[
n_i = \phi (\phi) \cdot (u_i \phi v_i) + (1 \phi) \cdot C \phi \frac{d}{dt} (u_i \phi v_i) \quad (I.5)
\]

\[
\phi_k = \phi p \phi_k + \phi (\phi) \frac{\partial u_m}{\partial x_m} \phi_k + \phi (\phi) \frac{\partial u_k}{\partial x_k} \phi_k + \frac{2}{3} \phi_k \frac{\partial u_m}{\partial x_m} \quad (I.6)
\]

and

\[
\phi_k = \phi p \phi_k + \phi (\phi) \frac{\partial v_m}{\partial x_m} \phi_k + \phi (\phi) \frac{\partial v_k}{\partial x_k} \phi_k + \frac{2}{3} \phi_k \frac{\partial v_m}{\partial x_m} \quad (I.7)
\]
Subsequent derivations from the above equations employed the following expression for the relative acceleration in the virtual mass term:

\[
\frac{d}{dt}(\mathbf{u}_i \cdot \mathbf{v}_j) = \sum \frac{\partial \mathbf{u}_i}{\partial t} + \mathbf{u}_k \cdot \frac{\partial \mathbf{u}_i}{\partial x_k} + \sum \frac{\partial \mathbf{v}_j}{\partial t} + \mathbf{v}_k \cdot \frac{\partial \mathbf{v}_j}{\partial x_k}
\]

The appropriate solution of the above system of equations is the steady state of uniform fluidization, in which the local mean particle velocity is zero throughout the system,

\[
\mathbf{u} = \mathbf{u}_0 = i \mathbf{u}_0; \quad \mathbf{v} = \mathbf{v}_0 = 0; \quad \mathbf{g} = \mathbf{g}_0
\]  

which satisfy the continuity expressions, and reduce the equations of motion to:

\[
\text{grad} p_0 \cdot i \mathbf{g}(\mathbf{u}_0 \cdot \mathbf{u}_0) + i \mathbf{g} = 0
\]  

\[
\mathbf{g}(\mathbf{u}_0 \cdot \mathbf{u}_0)(1 \cdot \mathbf{g}_0 \cdot (\mathbf{g}_0 \cdot \mathbf{g}_0)) = 0
\]

where the drag coefficient, \( \mathbf{g} \), is related to the voidage by the relationship of Ishii and Zuber (1979) for distorted particles, as follows:

\[
\mathbf{g}(\mathbf{u}_0) = 2 \mathbf{u}_0 (1 \cdot \mathbf{g}_0) \sqrt{\frac{g(\mathbf{g}_0 \cdot \mathbf{g}_0)}{\mathbf{g}_0}}
\]

Combining this with the equations (I.9-I.10) of motion provides a solution for the state of uniform fluidization, so long as equation (I.10) can be satisfied by a value of \( \mathbf{g} \) greater than the minimum fluidization voidage. The uniform fluidization solution is then augmented by a small perturbation, following Anderson and Jackson (1968):

\[
\begin{align*}
\mathbf{u} &= \mathbf{u}_0 + \mathbf{u}_1 = i \mathbf{u}_0 + \mathbf{u}_1 \\
\mathbf{v} &= \mathbf{v}_0 + \mathbf{v}_1 = \mathbf{v}_1 \\
\dot{\mathbf{a}} &= \dot{\mathbf{a}}_0 + \dot{\mathbf{a}}_1 \\
p &= p_0 + p_1
\end{align*}
\]
all terms of degree greater than one in small quantities are neglected, leading to the following linear equations obtained from the continuity expressions and equations of motion:

\[
\frac{\partial \mathcal{Q}}{\partial t} + u_0 \frac{\partial \mathcal{Q}}{\partial x} + \mathcal{Q}_0 \text{ div } u_i = 0 \quad (1.1')
\]

\[
\mathcal{Q}_0 \frac{\partial \mathcal{Q}}{\partial t} + (\mathcal{Q}_0 + \frac{1}{3} \mathcal{Q}_0) \text{ div } v_i = 0 \quad (1.2')
\]

\[
\mathcal{Q}_g (\mathcal{Q}_0) + \mathcal{Q}_0 \cdot \mathcal{Q}_0 \cdot v_i + C_0 \frac{\partial v_i}{\partial t} + u_0 \frac{\partial u_i}{\partial x} = 0 \quad (1.3')
\]

\[
i \cdot (\mathcal{Q}_g \mathcal{Q}_0) + \mathcal{Q}_0 \cdot (u_i \cdot \mathcal{Q}_0) \cdot u_0 \frac{\partial \mathcal{Q}_0}{\partial t} + \mathcal{Q}_0 \frac{\partial p_0}{\partial t} \text{ div } \mathcal{Q} + (\mathcal{Q}_0 + \frac{1}{3} \mathcal{Q}_0) \text{ div } v_i + \mathcal{Q}_0 \frac{\partial v_i}{\partial t} = 0 \quad (1.4')
\]

Where

\[
\mathcal{Q}_0 = \frac{d \mathcal{Q}}{dn} \Big|_{n=\mathcal{Q}_0} = \frac{\partial \mathcal{Q}}{\partial t} \Big|_{n=\mathcal{Q}_0} \quad (1.13)
\]

\[
p_0 = \frac{d p}{dn} \Big|_{n=\mathcal{Q}_0} = \frac{\partial p}{\partial t} \Big|_{n=\mathcal{Q}_0} \quad (1.14)
\]

A solution to the above equations is then sought in the form of plane waves:

\[
u_i = \hat{u}_i \exp(st) \exp(ik \cdot x)
\]

\[
v_i = \hat{v}_i \exp(st) \exp(ik \cdot x)
\]

\[
\mathcal{Q} = \mathcal{Q}_i \exp(st) \exp(ik \cdot x)
\]

\[
p_i = \hat{p}_i \exp(st) \exp(ik \cdot x)
\]

(1.15)
where \( \hat{u}_i, \hat{v}_i, \hat{u}_v, \hat{v}_v \) are the amplitudes of the perturbations in fluid velocity, particle velocity, voidage, and pressure, respectively. \( k \), which has real components, is the wave number vector of the disturbance, whose wavelength \( \sigma \) is therefore given by

\[
\sigma = 2\sigma / |k|
\]

(1.16)

in general, \( s \) is a complex number which may be written

\[
s = \sigma \sigma i\sigma
\]

(1.17)

The imaginary part then determines the velocity of wave propagation within the model system,

\[
V_p = \sigma / |k|
\]

(1.18)

while the real part determines the rate of growth or decay of the waves with time.

Substitution of equation (15) into equations (1'-4') yields a set of eight homogeneous linear algebraic equations. The resulting simplified determinantal equation corresponding to this system can be obtained as:

\[
\text{(I.19)}
\]

Substitution of the plane wave equation for voidage into the above equation yields:

\[
\text{(I.19)}
\]
\[
\Box \left[ A s^2 + (B + cD + i2bF)s + (eD \Box b^2 F + ibBE) \right] = 0
\]  

(1.20)

where

\[
A = 1 + \frac{\Box_g \Box_b}{\Box_g 1 \Box_b} + \frac{C_0}{\Box_g (1 \Box_b)}
\]  

(1.21)

\[
B = \frac{\Box_g \Box_b}{\Box_g 1 \Box_b} \frac{\Box_b}{\Box_b} \frac{g}{\bar{u}_0}
\]  

(1.22)

\[
E = 1 \Box 2 \Box_b + \Box_b \frac{n_0 \Box_b}{\Box_b}
\]  

(1.23)

\[
D = \frac{\Box_g \Box_b}{\Box_g 1 \Box_b}
\]  

(1.24)

\[
F = 1 + \frac{C_0}{\Box_b}
\]  

(1.25)

are properties of the unperturbed system, and

\[
b = \frac{\bar{u}_0 k_x}{\Box_b}
\]  

(1.26)

\[
c = \frac{\Box_b + \frac{4}{3} \Box_b}{\Box_b (1 \Box_b)} |k|^2
\]  

(1.27)

\[
e = \frac{n_0 \rho_0}{\Box_b (1 \Box_b)} |k|^2
\]  

(1.28)

which is in turn satisfied by the non-trivial solution

\[
A s^2 + (B + cD + i2bF)s + (eD \Box b^2 F + ibBE) = 0
\]  

(1.29)

whose roots are given by
\[
\begin{align*}
\dot{q} &= \frac{B}{2A} \pm \frac{Dc}{B} \pm \sqrt{\frac{(1 + w)^2 + q^2}{2} + (1 + w)} \\
\dot{q} &= \frac{B}{2A} \pm \frac{2Fb}{B} \pm \sqrt{\frac{(1 + w)^2 + q^2}{2} (1 + w)} \\
\end{align*}
\]  
(I.30) 

where

\[
\begin{align*}
w &= 2\left(\frac{Dc}{B} + \frac{Dc}{B} + 4\frac{b}{B} \frac{F(A \parallel F)}{b^2} \frac{ADe}{b^2}\right) \\
q &= 4\left(\frac{b}{B} \frac{AE \parallel F}{Fb} \frac{Fb}{b^2} \frac{B}{b^2}\right) \\
\end{align*}
\]  
(I.32) 

and

\[
\begin{align*}
\frac{Q_G}{(1 \parallel b) \cdot A_{bed} \cdot Q_{bed} \cdot k_x} \\
\end{align*}
\]  
(I.34)
Table 1: Simulation Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$G_r$</td>
<td>1.2</td>
</tr>
<tr>
<td>$L_r$</td>
<td>1000</td>
</tr>
<tr>
<td>$G_m$</td>
<td>1.8x10^-5</td>
</tr>
<tr>
<td>$L_m$</td>
<td>1.2x10^-3</td>
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<tr>
<td>$p_d$</td>
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<td>$d_{bed}$</td>
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<tr>
<td>$\theta$</td>
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</tr>
<tr>
<td>$g$</td>
<td>9.8</td>
</tr>
<tr>
<td>$A$</td>
<td>1.8</td>
</tr>
<tr>
<td>$B$</td>
<td>180</td>
</tr>
</tbody>
</table>
Table 2: Simulation Parameters for Fluidized Bed Model Employing Expressions of Anderson and Jackson (1968).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Function</th>
<th>Expression</th>
</tr>
</thead>
<tbody>
<tr>
<td>( g )</td>
<td>1.2</td>
<td>( g )</td>
<td>( 2 \cdot f_f \cdot u_0 \cdot (1 - f_b) \cdot \frac{g}{f_b} )</td>
</tr>
<tr>
<td>( l )</td>
<td>1000</td>
<td>( l )</td>
<td>( (\text{from Ishii and Zuber, 1979}) )</td>
</tr>
<tr>
<td>( g )</td>
<td>9.8</td>
<td>( g )</td>
<td>( 9.8 )</td>
</tr>
<tr>
<td>( C_o )</td>
<td>0</td>
<td>( C_o )</td>
<td>( 0 )</td>
</tr>
<tr>
<td>( \frac{g}{l} + \frac{4}{3} l^3 )</td>
<td>2</td>
<td>( \frac{n_0 g l}{l} )</td>
<td>( 0.5 \cdot \left( 1 + l^3 \right) )</td>
</tr>
<tr>
<td>( l )</td>
<td>0.074</td>
<td>( l )</td>
<td>( 0.074 )</td>
</tr>
<tr>
<td>( k_x )</td>
<td>4</td>
<td>( k_x )</td>
<td>( 4 )</td>
</tr>
<tr>
<td>( k_r )</td>
<td>0</td>
<td>( k_r )</td>
<td>( 0 )</td>
</tr>
</tbody>
</table>
Captions for Figures

Figure 1: Schematic of Experimental Apparatus. 1. House air supply; 2. flowmeter; 3. bourdon gauge; 4. inlet / distribution section; 5. packed column; 6. gas / liquid separation tank; 7. liquid reservoir; 8. centrifugal pump; 9. drain.

Figure 2: Standard deviation in pulse onset distance vs. number of measurements.

Figure 3: Pulse onset distance vs. liquid Reynolds number in column packed with (a) 3 mm, (b) 6 mm, and (c) 8 mm spheres.

Figure 4: Illustration of liquid-filled pores of increasing liquid fraction, from Grosser et al. (1988).

Figure 5: Illustration of two adjacent, completely wetted packing elements, from Attou and Ferschneider (2000).

Figure 6: Illustration of column cross-section at an arbitrary radial position, showing local regions of elevated liquid holdup.

Figure 7: Results of modified trickling-to-pulsing model and comparison with experiments; (□) 2.59 mm packing, from Sato et al. (1973); (□) 5.61 mm packing, from Sato et al. (1973); (□) 3 mm packing; (□) 6 mm packing (□) 8 mm, (--) Grosser et al. (1988); (-) Current Model.

Figure 8: Results of pulsing to dispersed-bubble flow transition model; (a) Predicted pulse onset distance vs. Re_L, (b) Comparison with experimental data: (□) 2.59 mm packing, from Sato et al. (1973); (□) 5.61 mm packing, from Sato et al. (1973); (□) 3 mm packing; (□) 6 mm packing (□) 8 mm; (--) model.
Figure 2
Figure 3
Figure 4

Higher curvature when less liquid

Lower curvature when more liquid
Figure 7
Figure 8