Cocurrent Downflow of Air and Water in a Two-Dimensional Packed Column

Multiple hydrodynamic states were observed during cocurrent downflow of air and water under trickling flow conditions in a packed bed of rectangular cross section. Although the multiplicity was exhibited by both pressure gradient and liquid holdup, the pressure gradient showed the largest variations at identical conditions. The multiplicity is interpreted as being due to the liquid flowing in two different modes, namely, film flow and rivulet flow.

The characteristics of pulsing flow in a packed bed of rectangular cross section were found to be appreciably different from those reported in the literature for flow in small-diameter cylindrical columns. The most significant observation in the packed bed of rectangular cross section was that the pulses did not always span the column cross section, unlike the case in small-diameter columns. The lower pressure drop and pulse velocity in the packed bed of rectangular cross section are believed to result from the bypassing of gas around the edge of the pulses. The location of the pulses was found to depend on the quality of gas and liquid distribution at the top of the column, and the shape of the top surface of the packing. A distributor configuration in which the gas was injected directly into the bed was found to be the most desirable, and is recommended in industrial practice.

G. Christensen, S. J. McGovern, S. Sundaresan
Department of Chemical Engineering
Princeton University
Princeton, NJ 08544

Introduction

A trickle-bed reactor is one in which a gas and a liquid flow cocurrently downward over a solid packing. The use of such reactors is widespread in the petroleum and chemical industries. The many advantages and disadvantages of trickle-bed reactors have been reviewed by Satterfield (1975) and Shah (1979). A variety of flow regimes have been observed in trickle beds. At low liquid and gas flow rates, the trickling regime, in which the liquid trickles down over the packing and the gas phase is continuous, is observed. At high gas and liquid flow rates, a complex and fascinating flow pattern commonly referred to as pulsing is observed. The origin of this pulsing flow pattern and its characteristics have been a subject of perennial charm, and several studies on pulsing flow behavior can be found in the literature. A number of industrial reactors operate near the trickling-to-pulsing transition (Satterfield, 1975). The other predominant flow patterns are spray and bubble flow. Spray flow occurs at high gas and low liquid flow rates, while bubble flow occurs at low gas and high liquid flow rates. The gas-liquid systems used in the study of trickle bed hydrodynamics can be classified in two categories, foaming and nonfoaming systems. The differences between foaming and nonfoaming systems have been highlighted by Charpentier and Favier (1975). In what follows, we restrict our attention to nonfoaming systems. A number of flow regime maps have been published for nonfoaming systems in trickle-bed reactors (Charpentier et al., 1971; Charpentier and Favier, 1975; Chou et al., 1977; Gianetto et al., 1978; Midoux et al., 1976; Sato et al., 1973b; Specchia and Baldi, 1977; Talmor, 1977; Tosun, 1984a; Turpin and Huntington, 1967; Weekman and Myers, 1964). The most widely used flow regime map is that proposed by Charpentier and Favier (1975).

An important consideration in the trickling regime is the fashion in which the liquid is distributed in the column. For example, a fraction of the liquid entering the packed bed flows through it in the form of rivulets while the remainder of the liquid flows in the form of films over the solid packing. Rivulet flow is, in general, undesirable as it leads to poor contacting between the gas, the liquid, and the catalyst, and hence to a decrease in

Correspondence concerning this paper should be addressed to S. Sundaresan.
The present address of S. J. McGovern is Mobil R & D Corp., Paulsboro, NJ 08066.

the effective rate constants for the conversion of the liquid phase reactants. A recent review by Koros (1981) summarizes the effect of rivulet flow on important design variables such as contacting efficiency and catalyst utilization. A good understanding of the liquid distribution pattern in the bed is clearly of significant practical interest. Kan and Greenfield (1978, 1979) observed multiple hydrodynamic states in the trickling flow through a column packed with small particles (<1.8 mm). Such multiplicity has also been observed by Levec et al. (1984, 1986). The pressure drop and holdup for specified gas and liquid flow rates have been found to depend on the manner in which the flows were established. This multiplicity is intimately connected with the liquid distribution in the bed. Hence a systematic study of the multiple hydrodynamic states should further our understanding of liquid distribution in the trickling regime; this is one of the objectives of the present study.

Pulsing flow is characterized by large fluctuations in pressure drop and liquid holdup. Most of the experimental work on pulsing reported in the literature has been performed in small-diameter (<0.1 m) cylindrical columns. These studies have led to a visualization of pulsing flow as the alternating passage of liquid-rich and gas-rich regions down the column. Such a visualization has been used by Dimenstein and Ng (1986) to develop a mathematical model for pulsing flow. Weekman and Myers (1964) visualized the liquid-rich region in the pulsing flow as a wavelike torus with a sharp leading edge and a trailing wake. Experimental evidence supporting the above visualization has been obtained in several studies (Beimesch and Kessler, 1971; Blok and Drinkenberg, 1982; Blok et al., 1983; Rao and Drinkenberg, 1983).

There exists, however, a fundamental difficulty in extrapolating the visualization of pulsing from the laboratory columns to large-diameter industrial columns. In every laboratory study, the torus-shaped liquid-rich region in the pulsing flow apparently filled most of the column cross section, suggesting that the size and shape of the liquid-rich region in the pulsing flow were limited by the column diameter. It is difficult to believe that this would happen in large-diameter industrial columns. Under conditions where the size and shape of the liquid-rich region in the pulsing flow are no longer restricted by the column diameter, the nature of pulsing may be very different from what has been deduced from studies in small diameter columns. The second objective of our study is to develop a conceptual model for the pulsing flow in packed columns with large cross-sectional area.

In order to achieve these objectives, we have studied the cocurrent downflow of air and water in a packed column of rectangular cross section (to obtain an effectively two-dimensional behavior). The choice of the bed geometry was also motivated by the use of a microwave probe for holdup measurement, as described in the next section.

Experimental

The column used in our study was rectangular in cross section, being constructed of aluminum channels with Plexiglas walls. It had internal dimensions of 0.051 × 0.457 × 1.83 m. Glass beads of 3 mm nominal diam. were used as packing material. The void fraction of the packed column was determined to be 0.368. The liquid feed to the column (tap water) was distributed in two stages. The primary distribution was effected through a pipe with holes in its sides, positioned across the column. The subsequent secondary distribution of the liquid took place through 75 4.8 mm copper tubes inserted in a distributor plate located horizontally at the top of the packing. The air was cooled and filtered before passing through the rotameters and the column. As the feed air was essentially saturated with water vapor, humidification of the air prior to entering the column was deemed unnecessary. The air entering the column was distributed through five 11 mm tubes inserted in the distributor plate. The distributor plate was located about 0.1–0.15 m above the top of the packing. The 4.8 mm tubes in the distributor plate were not long enough to reach into the packing. Thus the liquid "rained" downward a certain distance before entering the packing. The five 11 mm tubes in the distributor plate could be inserted into the packing, if desired. At the base of the column, the air and water were separated in a disengaging section. From here the air was vented to the atmosphere while the water was recirculated.

The pressure drop was measured through differential pressure transducers mounted at a number of locations in the column. The liquid holdup at any desired location in the column was estimated with the help of a microwave probe. Briefly, a beam of microwave radiation (0.075 × 0.038 m) was directed through the column and its absorption by water was measured. A schematic diagram of the circuitry used is shown in Figure 1. The output from the unit was a DC voltage. The microwave probe was calibrated in a test cell filled with beads. The test cell

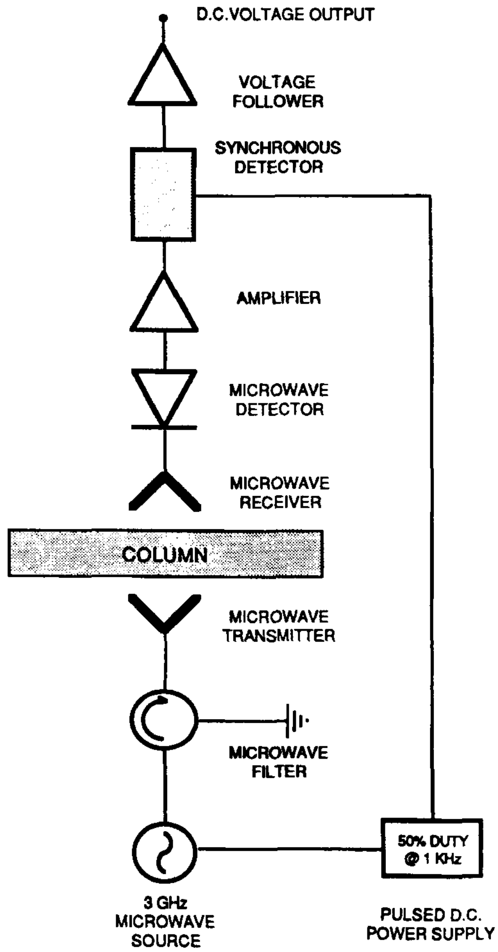

Figure 1. Circuit diagram for microwave unit.

was partially filled with water. Air was bubbled through at different rates and the voltages recorded. The liquid holdup was estimated from the water levels with and without air flow. The microwave calibration curve prepared in this manner is shown in Figure 2. The limitation of the microwave probe is quite evident. It is hardly sensitive outside the region $0.3 < \beta < 0.7$. Thus it is an unsatisfactory probe for developing holdup correlations. The primary attraction of the microwave probe is that it could be moved easily from one location to another in the bed and therefore gross maldistribution of liquid, if present, could readily be detected.

The onset of pulsing was determined in three different ways, by visual observation of pulses, by fluctuations in pressure drop, and by fluctuations in holdup (i.e., microwave output). Not surprisingly, all three methods agreed very well. When pulsing was first observed, it was detected at the bottom of the column. The location in the column where pulsing initiates moved upstream as the flow rates were increased. This is consistent with the observations of previous investigators.

The pulse velocity was also measured in three ways. The first, and easiest, was by estimating the time required for the pulse to travel a certain distance in the column by visual observation and stopwatch. This was inaccurate at high pulse velocities. The increase in local pressure corresponding to a pulse was used in the other two methods. Two pressure taps 0.076 m apart vertically were connected across a differential pressure transducer. A sample output obtained from this transducer is shown in Figure 3. At time *A*, the pulse passes the upper pressure tap; it arrives at the lower tap at time *B*. From this time interval the velocity can be calculated. The microwave unit was placed 0.22 m above the upper pressure tap. Both the microwave and the pressure transducer outputs were recorded simultaneously. As seen in Figure 3, the time *C-D* is the time taken for the pulse to travel from the microwave unit to the upper pressure tap. In general, the agreement between the three methods was found to be satisfactory, except at very high pulse velocities. The majority of the pulse velocity data was collected by using just the pressure transducer.

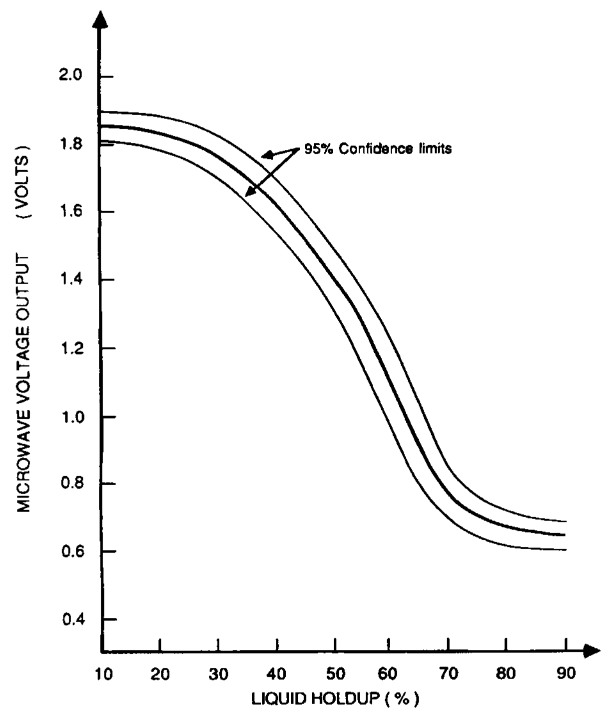

Figure 2. Calibration curve for microwave unit.

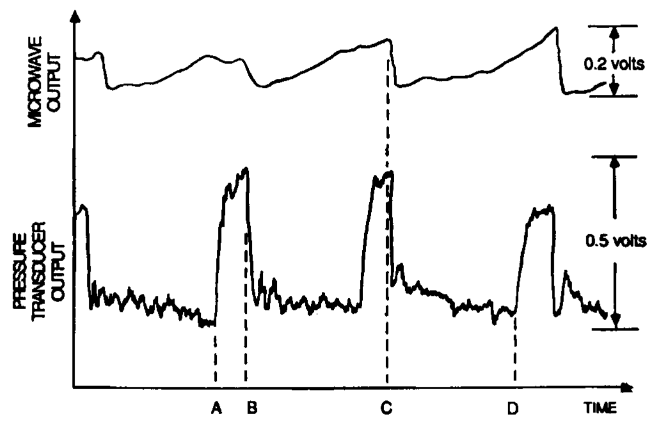

Figure 3. Typical outputs of differential transducer and microwave unit in pulsing flow.

The pulse frequency was calculated by measuring the time taken for a given number of pulses to pass through the column. This was done by visual observation of pulses, by pressure transducer output, and by microwave output. All three methods were in good agreement, as one would expect.

Results and Discussion

Multiple hydrodynamic states in the trickling regime

The bed was completely wetted at first and then allowed to drain before any series of experimental runs was started. Two types of experiments were carried out. In the first series, henceforth referred to as constant gas flow rate experiments, the desired gas flow rate was established; then the liquid flow rate was varied in different ways to study the multiplicity of hydrodynamic states. In the second series of experiments, starting from a prewetted bed, the desired liquid flow rate was first established and subsequently the gas flow rate was varied as desired; these are referred to as constant liquid flow rate experiments.

Pressure drop

Multiplicity of hydrodynamic states was observed over virtually the entire trickling regime. The multiplicity in pressure gradient observed in a constant gas flow rate experiment for the air-water system is shown in Figure 4. Also shown is the effect of adding 1 wt. % ethanol to the water (which leads to a decrease in the surface tension). In both cases, starting from zero liquid flow rate, as the liquid flow rate is increased the lower curves are obtained. Starting from the liquid flow rate corresponding to the onset of pulsing (shown by * in these curves), if the liquid flow rate is decreased to zero, the upper curves are obtained. We shall discuss the curves obtained for the air-1 wt. % ethanol in water system in a later section on the effect of surface tension; for the moment we will restrict our attention to the results obtained for the air-water system. Only the two end points are common for the two curves. Provided that one started from one of these points (zero liquid flow rate or pulsing), both curves could be reproduced to within 10%. Proceeding in a large number of

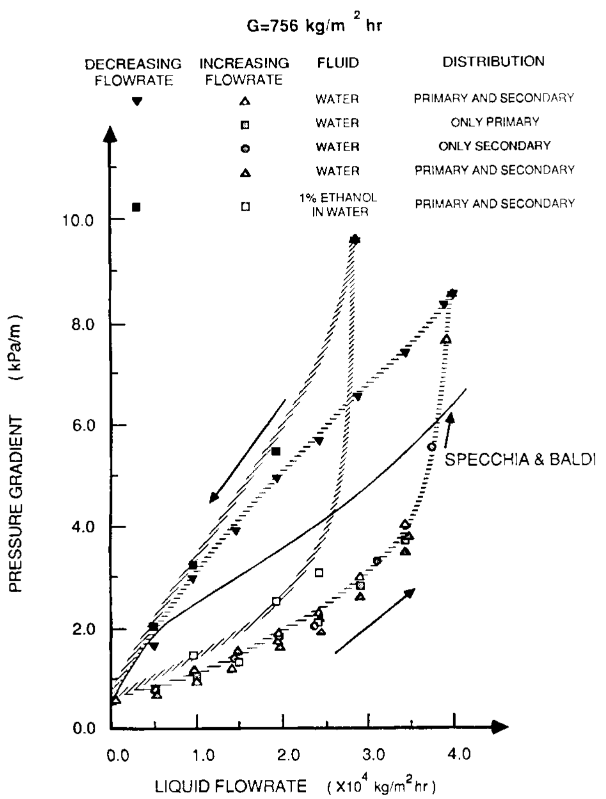

Figure 4. Pressure gradient vs. liquid flow rate at a constant gas flow rate.

Also displayed, effect of liquid distribution on pressure gradient corresponding to lower limiting curve for air-water system.

small steps or a small number of large steps produced the same steady states. As can be seen from Figure 4, the pressure gradients for the upper curve at intermediate liquid flow rates are as much as 100% larger than those for the lower curve. It was found that the fractional difference in pressure gradient between the upper and lower curves becomes smaller as the gas flow rate increases.

To test the importance of gas and liquid distribution at the top of the column, three sets of experiments were carried out at a single gas flow rate. In each set, the liquid flow rate was increased in small steps from zero until the onset of pulsing. In the *first set of experiments* the distributor plate was removed so that only the primary distributor described earlier was present. In the second set the distributor plate was in place while the primary distributor for liquid was removed. The third set contained both primary and secondary distributors. The measured pressure gradients are shown in Figure 4. In the interior of the trickling regime, hardly any difference between the three sets of experiments could be seen. Thus it appears that in the trickling regime a modest attempt to obtain uniform distribution is quite adequate. The same conclusion has been reached by previous investigators (e.g., Herskowitz and Smith, 1978). The distribution, however, may affect the trickling-to-pulsing transition (i.e., conditions under which pulsing was first detected) and the details of the flow in the pulsing regime itself. These will be described later. In all the experiments described below, both distributors were used.

The multiplicity pattern of the hydrodynamic states is more

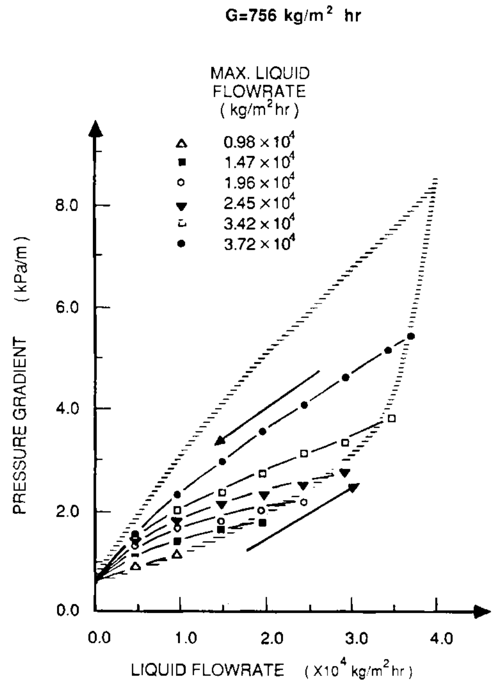

Figure 5. Effect of decreasing liquid flow rate from various maximum values.

complicated than shown in Figure 4. Instead of just two curves as shown in this figure, a family of curves could in fact be generated. Figures 5 and 6 illustrate two families of curves at the same gas flow rate. In Figure 5, starting from zero liquid flow rate, the liquid flow rate was increased to various maximum values (as described in the figure) and then decreased back to zero. The lower limiting curve was followed every time as the liquid

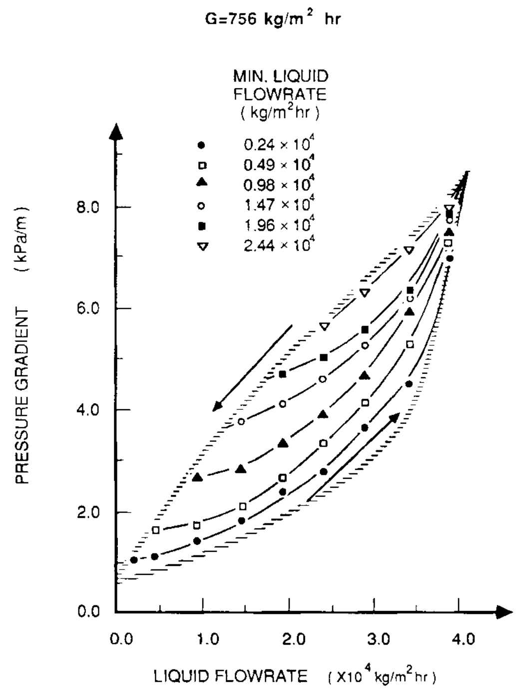

Figure 6. Effect of increasing liquid flow rate from various minimum values.

flow rate was increased. However, the return path is dependent on the maximum liquid flow rate attained. The higher the maximum liquid flow rate for a given curve, the closer the return path is to the upper limiting curve. In Figure 6, starting from the liquid flow rate corresponding to pulsing inception (L_p), the liquid flow rate was decreased to various minimum values (as described in the figure) and then increased back to L_p. The upper limiting curve was followed every time as the liquid flow rate was decreased while the return path is dependent on the minimum liquid flow rate attained. The lower the minimum liquid flow rate, the closer the return path is to the lower limiting curve. The inner points (between the two limiting curves) shown in Figures 5 and 6 were found to be quite stable. For a few points the flow rates were maintained in excess of 2 h and little variation in pressure gradient was observed. It is now clear that every point inside the region bounded by the two limiting curves can be realized through an appropriate sequence of experimental conditions.

The effect of repeatedly increasing and decreasing the liquid flow rate between two fixed limits was also investigated. If one of the fixed limits is zero liquid flow rate, then, for the cyclic variation, operating loops of the type shown in Figure 5 determined by the maximum liquid flow rate are obtained. Similarly, if one of the fixed limits is a liquid flow rate corresponding to pulsing inception, operating loops of the type shown in Figure 6 determined by the minimum liquid flow rate are obtained. The important result to note is that there is a unique loop determined by the two fixed limits (zero liquid flow and pulsing). However, if both the fixed limits lie in the open interval between zero and pulsing, such uniqueness is not realized. This is illustrated in Figure 7, for liquid flow rate cycling between 9,780 and 37,100 kg/m^2 · h, where the asymptotic operating loops (along with their evolution) are shown for two different initial conditions (zero liquid flow and pulsing). This figure highlights the strong history dependence of the observed pressure gradient.

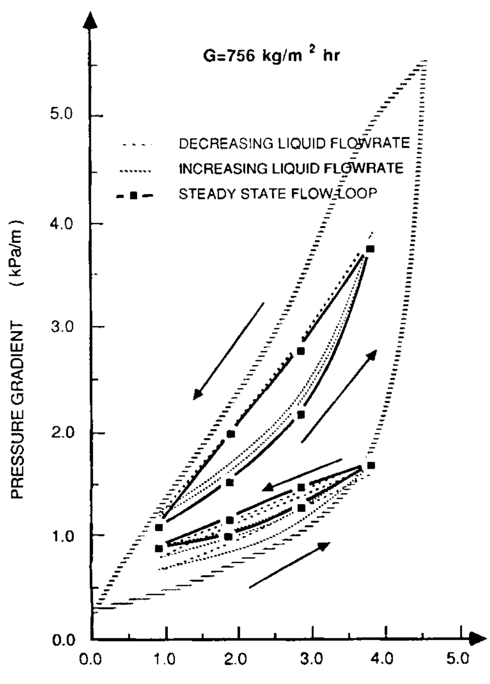

Figure 7. Asymptotic inner multiplicity loops for liquid flows cycling between 9,780 and 39,100 kg/m^2 · h.

Results for two different initial conditions: zero liquid flow (increasing liquid flow rate) and pulsing (decreasing liquid flow rate).

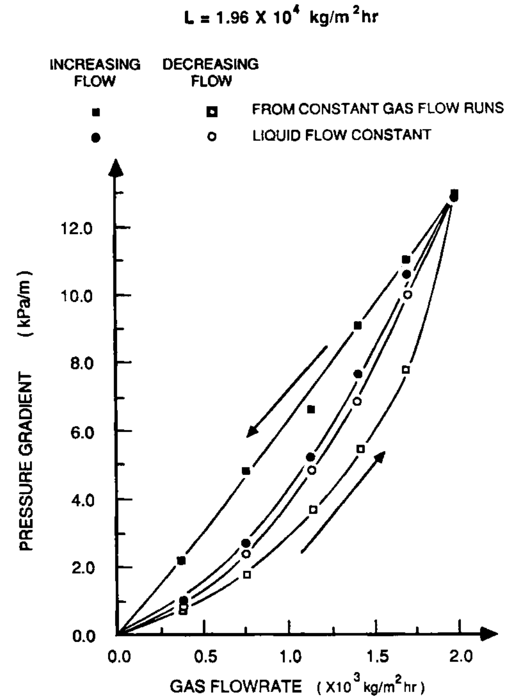

Figure 8. Pressure gradients obtained during constant liquid flow rate and constant gas flow rate runs at similar conditions.

The results discussed thus far were obtained from constant gas flow rate experiments. The pressure gradients obtained at various gas flow rates for a constant liquid flow rate are presented in Figure 8. Superimposed are the pressure gradient results under similar conditions corresponding to the upper and lower limiting curves of constant gas flow rate experiments (extracted from figures such as no. 4). It is seen that the hysteresis is more pronounced for the constant gas flow rate experiments.

Liquid holdup

As discussed in the Experimental section, the liquid holdup could not be measured accurately using the microwave probe. The only attractive feature of this probe was that it could easily be moved from one location to another in the bed. The best accuracy that can be expected is ± 0.03 around a liquid holdup of 0.50, with the error increasing as the holdup decreases. At a holdup of 0.20, the uncertainty has increased to a value of ± 0.10. The size of this uncertainty associated with the microwave probe should be kept in mind when one analyzes the liquid holdup data presented below. Error bars are not included in the results described below for the sake of clarity.

A constant gas flow rate experiment in the trickling regime, in which the liquid flow rate was increased from zero to pulsing in small steps, was repeated several times. The pressure gradient measurements of various runs at identical liquid flow rates did not reveal any appreciable run-to-run variations. The microwave absorption for the various liquid flow rates in the different runs was also measured at a fixed location in the column. The run-to-run variations in the holdup values determined from the microwave calibration were found to be within the uncertainty limit associated with the microwave probe.

The effects of the distributor on the trickling flow characteristics were investigated using three different distributor configurations, as already described in the previous section on pressure drop, and see Figure 4. The differences in the liquid holdup measured at a fixed position in the column for the three different distributors were within the uncertainty limit associated with the microwave probe.

For the poorest gas and liquid distribution (i.e., only using the primary distributor), the holdup data measured at five different locations are presented in Figure 9, along with the average value and the 95% confidence limits. It appears that the distributor has no great effect on the steady state liquid distribution in the bed. This is consistent with our earlier conclusion (Figure 4) that a modest effort to distribute the gas and liquid at the top of the column is quite adequate in the trickling regime.

Corresponding to the multiple hydrodynamic states observed in pressure gradient, a difference was seen in the liquid holdup between increasing and decreasing liquid flow rates. The liquid holdup for increasing liquid flow rate was slightly larger than that for decreasing flow rate. However, the difference in holdup between increasing and decreasing liquid flow rates was less than the uncertainty associated with the measurement technique; but this trend was observed with nearly every set of data obtained. In the analysis presented below we assume that the holdup is approximately independent of the history.

The trend we observed seems to be different from the results reported by Levec et al. (1984, 1986). While they observed lower pressure gradients with increasing liquid flow rates than with decreasing liquid flow rates (in agreement with our results, as can be seen, for example, in Figure 4), they found the liquid holdup for increasing liquid flow rate to be somewhat smaller than that for decreasing liquid flow rate.

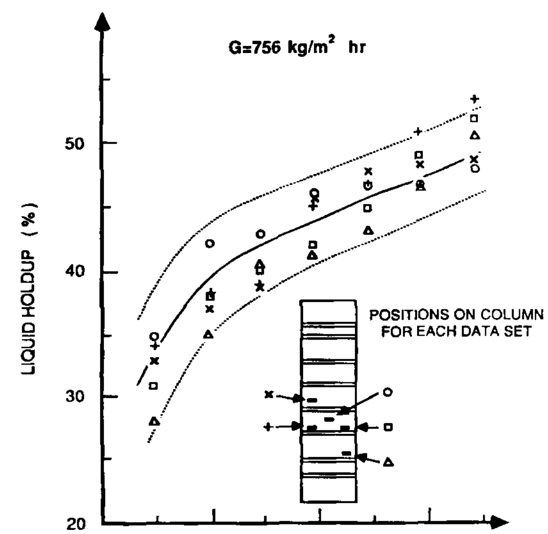

Figure 9. Variation of liquid holdup with position in the column.
— average liquid holdup ···· 95% confidence limits.

Kan and Greenfield (1978, 1979) always observed higher pressure gradients and lower holdups with increasing gas flow rates than with decreasing gas flow rates. This was not found to be the case with our measurements.

Liquid distribution in the column

Visual observation of the column during a constant gas flow rate experiment in the trickling regime suggests the following picture. Starting from zero liquid flow rate, as the liquid flow was commenced the liquid was seen to move down the column in a few large rivulets. These appeared to be approximately 0.01 m in width and were spaced horizontally about every 0.04 m. These rivulets meandered down the packing, coalescing and splitting as they went. The following liquid traveled down these rivulets and did not appear to spread any more. The rivulets were apparently very stable; some were watched for over 2 h with no changes in their shapes being observed. On increasing the liquid flow rate, the rivulets would grow in size, but seldom in number or flow path. Just prior to pulsing, rippling was observed on the rivulets, indicative of increased gas-liquid interaction. At this point the rivulets began to split, forming several smaller rivulets. When pulsing was reached, the liquid was

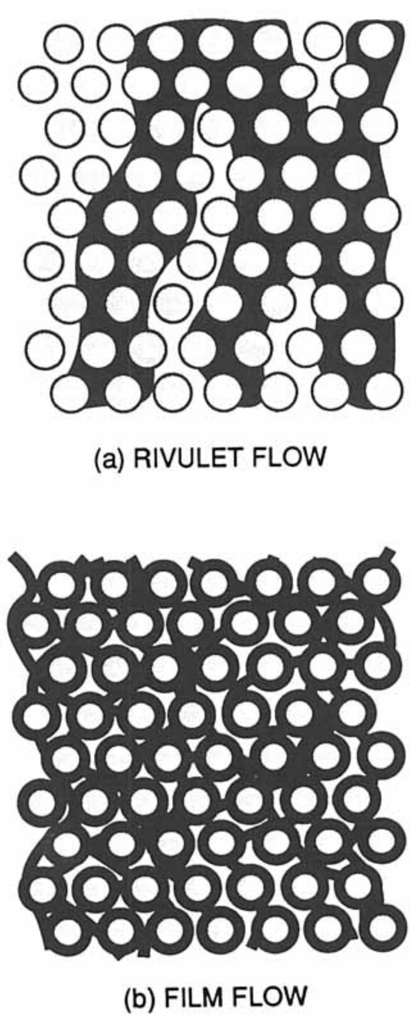

Figure 10. Rivulet and film flow models.

spread evenly over the packing. On reducing the liquid flow rate, this evenly spread distribution was apparently maintained and no coalescence of these thin films into rivulets could be observed. Eventually a state was reached where the flow rate had been reduced so much that the liquid supply was not enough to maintain all of the thin films and dryer sections could be observed. The two liquid distributions are shown schematically in Figure 10.

The multiple hydrodynamic states observed can readily be rationalized in terms of these different liquid distributions. In trickle flow, the pressure gradient is primarily due to the gas flow. If one considers the packing as a large number of capillaries, the two different liquid distributions affect the gas flow area in different ways. For the large rivulets produced when the liquid flow is increased, whole groups of capillaries are blocked while the remainder are only slightly affected. For the evenly distributed thin film flow obtained when the liquid flow is decreased from pulsing, a fraction of the gas flow area in essentially every capillary is taken away by the liquid. If one assumes that the capillaries are parallel and noninteracting, it is straightforward to show that the pressure gradient would be smaller in rivulet flow than in evenly distributed film flow for the same liquid holdup.

As the total drag force exerted on the liquid by the gas is much greater in evenly distributed film flow than in rivulet flow, a somewhat larger average liquid velocity in evenly distributed film flow than in rivulet flow appears likely. It then follows that a somewhat lower liquid holdup in film flow than in rivulet flow observed in our study for similar gas and liquid flow rates is not unrealistic.

The families of curves shown in Figures 5 and 6 are also explained by the above model. Observation of the column while obtaining the results in Figure 5 showed that while the rivulets grew in size with increasing liquid flow rate, hardly any change in the rivulet size was detectable when the liquid flow rate was decreased. Thus, when the flow rate is decreased, the rivulets become porous, in effect turning into a region of thin liquid films trickling over the packing with gas flowing in the remaining void. As discussed earlier, the pressure gradient is greater for film flow than for rivulet flow. Thus the results of Figure 5 are hardly surprising. The results of Figure 6 also can be rationalized in a similar fashion. In summary, in the trickling regime an increase in the liquid flow rate results in the growth of the rivulets, while a decrease in the liquid flow rate results in a transformation of the rivulets into thin films of liquid trickling over the packing.

One would expect the spatial variation in the liquid holdup to be substantial in rivulet flow. Yet the microwave probe did not reveal large spatial nonuniformities in holdup (Figure 8). Thus it appears that the region sampled by the microwave probe (at least $0.075 \times 0.038 \times 0.051$ m) is large compared with the characteristic length scale of spatial nonuniformities in rivulet flow.

It is instructive to analyze the correlations in the literature at this stage. Implicit in every correlation is the assumption that the liquid is evenly distributed. This corresponds to film flow, i.e., the upper limiting curve obtained in constant gas flow rate experiments. Thus it is unrealistic to expect the correlations to predict anything but the upper limiting curve. It is also clear, in light of the complex hydrodynamic behavior in trickle flow, that experimental data intended for correlation development should be gathered under constant gas flow conditions, with the liquid flow rate varying only in the decreasing direction starting from pulsing flow conditions.

The correlations based on the Lockhardt-Martinelli parameter (Larkins et al., 1961; Midoux et al., 1976; Sato et al., 1973a; Tosun, 1984b) in general were unsatisfactory. This is not surprising as these correlations were derived largely from experimental results in the pulsing regime. The correlations of Specchia and Baldi (1977) and Saéz and Carbonell (1985), which are valid only in the trickling regime, yield reasonable estimates of pressure drop in film flow and the liquid holdup (provided good estimates of model parameters such as static holdup and modified Ergun constants are available). A sample comparison of the predictions of the Specchia and Baldi correlation with our data is shown in Figure 4. At low liquid flow rates, the correlation of Specchia and Baldi predicts the upper limiting curve shown in Figure 4 reasonably accurately, but its predictions drop off at higher flow rates. This should be expected of this type of correlation since liquid-gas interactions become increasingly important at these higher flow rates. The only way the liquid affects this correlation is through a decrease in the area for gas flow resulting from the presence of liquid in the column.

A rough estimate of the fraction of liquid flowing in film may be obtained from the liquid flow rate (at the same gas flow rate) required to give the same pressure drop for purely film flow (assuming that the upper limiting curve corresponds to purely film flow). For the operating condition under investigation, the fraction of liquid in film flow is taken to be equal to this liquid flow rate for purely film flow divided by the actual liquid flow rate. The fraction of liquid in film flow for the lower limiting curves for several gas flow rates is shown in Figure 11. In general, the

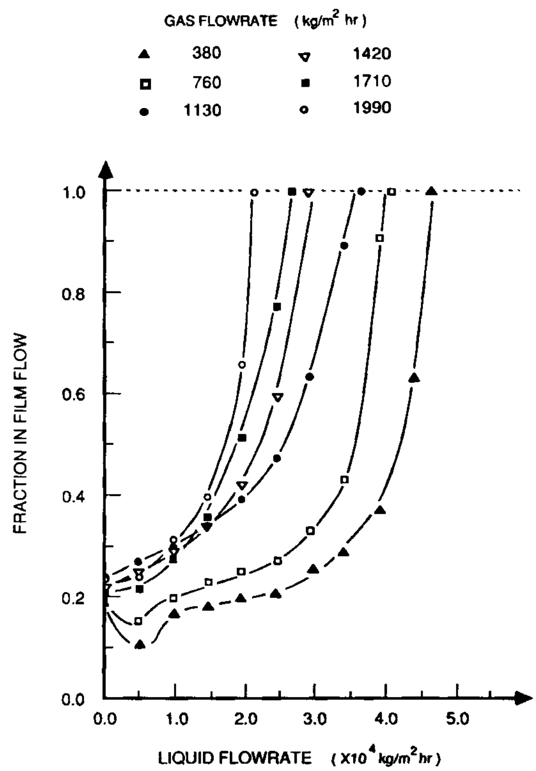

Figure 11. Variation of fraction of liquid flowing as films with liquid flow rate, as estimated from the lower limiting curves.

fraction of liquid in film flow increases sharply shortly before pulsing. For moderately high liquid flow rates (above 1×10^4 kg/m$^2 \cdot$ h), the higher the gas flow rate, the larger is the fraction of liquid in film flow—indicative of larger gas-liquid interactions. At low liquid flow rates, the curves cross each other and the trend is more complex. This is quite likely to be an artifact of the estimation procedure and hence we do not associate any significance to the features observed at low liquid flow rates.

The presence of multiple hydrodynamic states in trickle flow may clearly be of significance to the start-up and operation of industrial trickle bed reactors in trickle regime. Two common start-up practices are:

1. Establish the liquid flow rate and then introduce the gas.
2. Establish the desired gas flow rate and then introduce the liquid.

The present study reveals that the first strategy is better than the second one. For the best distribution of liquid in the reactor, the liquid flow should be increased to its highest possible value or until pulsing inception, whichever is lower, and subsequently reduced to the desired operating point.

Since the pressure drop for specified gas and liquid flow rates increases as the fraction of liquid in film flow increases, a decrease in the pressure drop over a period of time (caused, for example, by the inevitable fluctuation in throughputs) is indicative of an increase in the fraction of liquid in rivulet flow and hence a decline in the catalyst utilization. The strategy required to reestablish a more uniform liquid distribution is also clear from the present study.

Effect of surface tension

In order to understand the effect of surface tension on hydrodynamic multiplicity in the trickling regime, experiments were carried out with 1 wt. % ethanol in water as the circulating liquid. The upper and lower limiting curves for a constant gas flow rate experiment for the ethanol-water mixture are compared with those for pure water in Figure 4. The liquid flow rate corresponding to pulsing inception is lowered by the decrease in surface tension caused by the addition of ethanol, consistent with the findings of other researchers (Chou et al., 1977). The lower limiting curve for the ethanol-water mixture is located above that for pure water. This is not surprising, as rivulets would spread and split more easily if the surface tension were lower. The upper limiting curve for the ethanol-water mixture is located above that for pure water. The liquid holdups for the upper limiting curve of the ethanol-water mixture were found to be slightly lower than those corresponding to pure water under the same conditions (not shown). It therefore follows that the higher pressure gradients for ethanol-water mixture should be due to increased gas-liquid interaction (rippling) arising from a decrease in surface tension.

Trickling-to-pulsing transition

Although it was found that the manner in which the flows were established can have a significant effect on the resulting liquid distribution and pressure gradient in the trickling regime, no hydrodynamic multiplicity (other than the inherent fluctuations characteristic of pulsing) was detected either in the trickling-to-pulsing transition or the pulsing characteristics. Hence, in what follows we do not make any reference to the flow history in the bed.

The onset of pulsing was determined when only the primary distributor was used (term poor distribution) and when both distributors were used (termed good distribution). It was found that for a given gas flow rate, pulsing began at a lower liquid flow rate for poor distribution, Figure 12. Thus the quality of distribution at the top of the bed can affect the conditions at which pulsing is first observed. This differs from the measurements in trickling regime, Figure 4, where it was found that a modest effort to distribute the liquid at the top was adequate to establish an equilibrium liquid distribution in the column. We will discuss the role of gas-liquid distribution at the top in greater detail later.

One of the most accepted flow regime maps is that of Charpentier and Favier (1975). In general, close agreement between this correlation and our data for both air-water and air-1 wt. % ethanol in water systems was obtained. Noting that the correlation was developed from data in small, cylindrical columns while our data were obtained in a bed of very different geometry, one may conclude that the trickling-to-pulsing transition is essentially independent of bed geometry. The onset of pulsing is believed to be related to the occlusion of flow channels in the packed column by the ripples on the surface of the liquid film (Sicardi et al., 1979; Sicardi and Hofmann, 1980; Ng, 1986). Thus it is not surprising that the onset of pulsing is essentially independent of the bed geometry.

The results described below were obtained with both the primary and secondary distributors in place, unless mentioned otherwise.

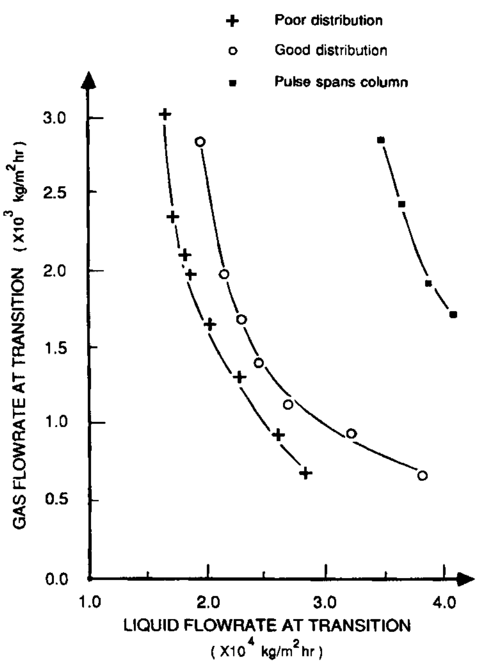

Figure 12. Effect of liquid distribution on the trickling-to-pulsing transition.

Onset of pulsing, + when only primary distributor for the liquid is used, O when both primary and secondary distributors are used. ■, conditions at which pulses first become large enough to span column cross section.

Pulsing flow

It was found that in the pulsing regime, the gas and liquid distribution at the top of the bed had little effect on pulse properties such as velocity, frequency, average pressure drop, and holdup. This differs from our observation regarding the onset of pulsing.

Pressure gradient

The pulsing regime pressure gradients were measured over a wide range of gas and liquid flow rates. The effect of liquid flow rate on the pressure gradient at a constant gas flow rate is shown in Figure 13. Shown are the average pressure gradient along with the maximum and minimum observed during the fluctuations. The pressure gradient correlations in the literature are compared with our data in Figure 13. These correlations were developed from experimental data on two-phase flow in small-diameter columns. It is generally believed that the correlations of Sato et al. (1973a) and Midoux et al. (1976) are reasonably accurate, with that of Tosun (1984b) representing a recent refinement.

The pressure gradients measured in our column are generally lower than the predictions of these correlations. The difference is pronounced at low gas and liquid flow rates, and it decreases as the flow rates are increased. An explanation for this result will be presented in the later section on pulse growth and shape.

Liquid holdup

As with pressure drop, the liquid holdup is characterized by fluctuations during pulse flow. Blok and Drinkenburg (1982) have presented the following picture for holdup variation in a pulse unit comprising a liquid-rich region followed by a gas-rich region. The liquid holdup rises sharply at the front of the liquid-rich region and then drops off slowly to the value corresponding to the gas-rich region. The drop in the liquid holdup at the tail of the liquid-rich region, however, occurs relatively quickly when compared with the length of the entire pulse unit.

The variation of liquid holdup in a pulse unit measured in our experimental system is shown in Figure 14. A sharp rise in liquid holdup is seen at the front of a pulse unit, in agreement with the results of Blok and Drinkenburg (1982). However, the decrease in the holdup behind the front seems to take place rather slowly, in fact almost linearly all the way until the next pulse unit.

The average liquid holdup corresponding to the conditions in Figure 13 is presented in Figure 15. Very good agreement with the correlations of Sato et al. (1973a) and Midoux et al. (1976) is evident.

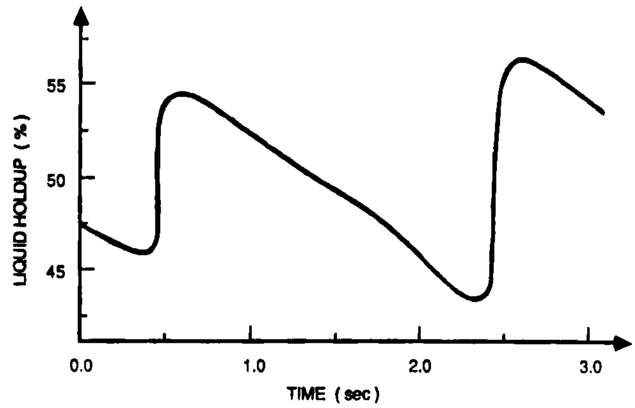

Figure 14. Variation of liquid holdup at a fixed location with time.

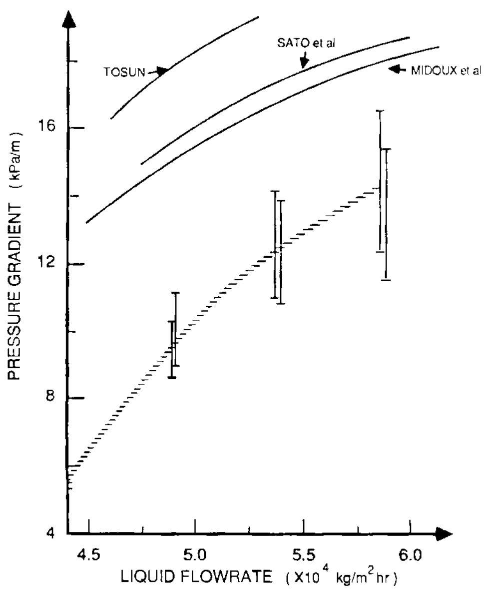

Figure 13. Pressure gradient vs. liquid flow rate at a constant gas flow rate.

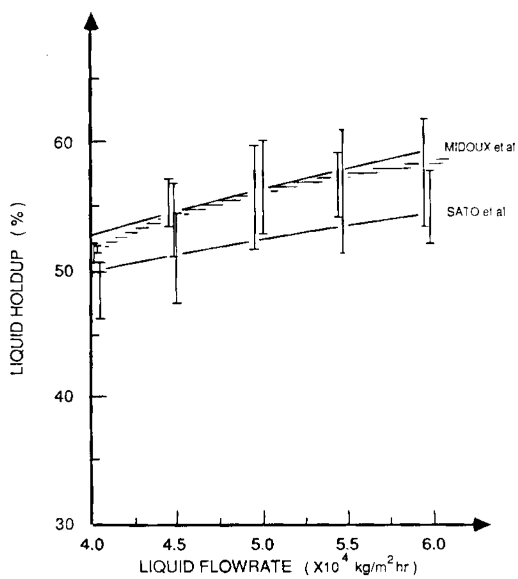

Figure 15. Liquid holdup vs. liquid flow rate at a constant gas flow rate.

Pulse velocity

It was originally thought that the velocity of the pulse was equal to the actual velocity of gas in the column. It has been now established that the pulse velocity is usually smaller than the actual gas velocity (Blok and Drinkenburg, 1982; Blok et al., 1983). Beimesch and Kessler (1971) found the liquid flow rate to have a strong effect on the pulse velocity, while more recent studies (Blok and Drinkenburg, 1982; Blok et al., 1983; Rao and Drinkenburg, 1983) suggest that liquid flow rate has little effect on the pulse velocity. We found only a weak dependence of the pulse velocity on liquid flow rate. Our experimental results are compared with the correlation of Rao and Drinkenburg for pulse velocity in Figure 16. The pulse velocities measured in our column are generally lower than those predicted by Rao and Drinkenburg.

Pulse frequency

In our experiments, the pulse frequency was found to increase sharply with increasing liquid flow rate, while only a weak dependence on the gas flow rate was observed. This is opposite of what was observed with liquid holdup and pulse velocity. The trend observed in our column is consistent with the results of Blok and Drinkenburg (1982), Blok et al. (1983), and Rao and Drinkenburg (1983). Blok and Drinkenburg have proposed that at a given gas flow rate, the liquid in excess of the liquid flow rate corresponding to the trickling-to-pulsing transition is transported through the bed in the liquid-rich region of the pulse units. Their experimental observations that the pulse velocity, the holdup in the liquid-rich region of a pulse unit, and the size of this liquid-rich region do not vary appreciably with the liquid flow rate, when combined with their proposition, implies that the pulse frequency should increase with liquid flow rate and in fact be proportional to the difference between the actual liquid velocity (V_ℓ) and the actual liquid velocity at the trickling-to-pulsing transition ($V_{\ell t}$). The pulse frequencies measured in our system over a wide range of gas and liquid flow rates are plotted against this excess liquid velocity ($V_\ell - V_{\ell t}$) in Figure 17. A reasonably straight line is indeed obtained. Also shown are the predictions of Rao and Drinkenburg's (1983) correlation for an identical packing material. The pulse frequencies measured in our system are somewhat lower than the predictions.

The overall height of a pulse unit can be obtained by dividing the pulse velocity by the pulse frequency. The overall pulse height is large near the trickling-to-pulsing transition. An increase in the excess liquid velocity results in a decrease in the overall height of the pulse unit, while an increase in the gas flow rate tends to make the pulse unit longer.

Pulse growth and shape

As mentioned in the Introduction, the primary objective of our study is to develop a conceptual model for pulsing in packed columns with large cross-sectional area. Visual observation of pulses (more precisely, pulse fronts) in our column revealed several interesting features. Recall that our column is rectangular (0.051 × 0.457 m) in cross section. When pulsing is first observed (i.e., at trickling-to-pulsing transition), the pulses apparently spanned the 0.051 m thickness of the column, as it could be seen from either side of the column. This was also found to be the case in the interior of the pulsing regime as well. Thus the flow patterns observed in our experiments appear to be essentially two-dimensional. In the discussion below, we assume this two-dimensional flow behavior and present our observations on pulsing as viewed from either of the two 0.457 × 1.83 m sides of the column.

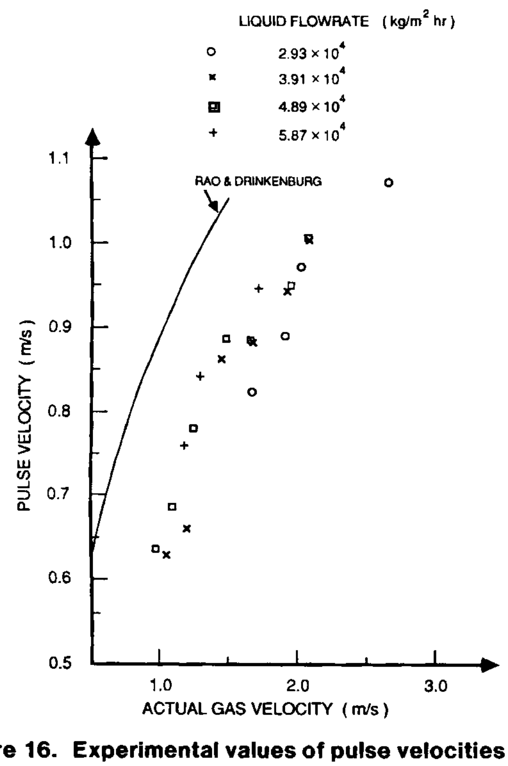

Figure 16. Experimental values of pulse velocities vs. actual gas velocity.

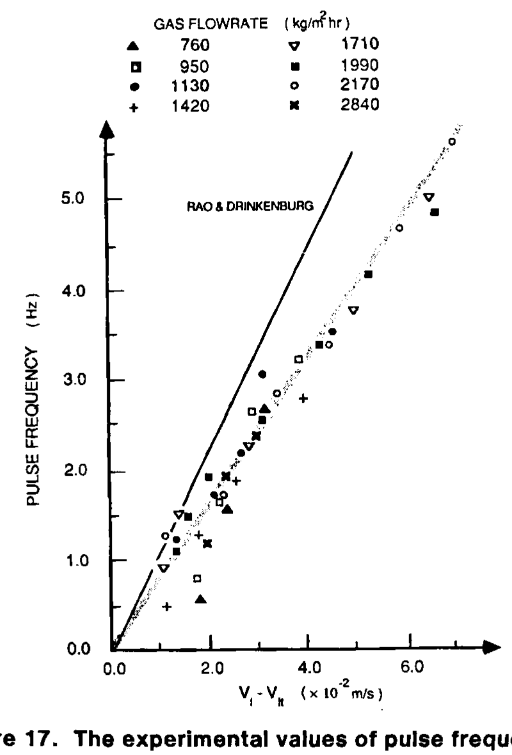

Figure 17. The experimental values of pulse frequencies vs. $V_\ell - V_{\ell t}$ (see Notation).

The first and very striking observation is that, unlike previous studies in small cylindrical columns, the pulses did not always span the column cross section. The location of the pulses was affected by the way in which the gas and liquid were distributed at the top of the column. This will be discussed in the next section. The pulses, although not always spanning the column cross section, were always observed to move vertically downward.

At the transition from trickling to pulsing, the pulses were only about 0.08 m wide, which is much smaller than the 0.457 m width of the column. As the flow rates were increased, the width of the pulse increased. It turned out that the pulse width could indeed become large enough to span the 0.457 m column width. For various gas flow rates, the liquid flow rate at which the pulses were first observed to span the entire 0.457 m column width was determined and is shown in Figure 12. Note that the flow rates at which the pulses first span the column cross section are considerably higher than those corresponding to the onset of pulsing. It appears reasonable to conclude the following. For small-diameter cylindrical columns, the conditions at which the pulses first span the column cross section are very close to those corresponding to the onset of pulsing. As the column diameter increases, the conditions at which the pulses first span the column cross section shift further and further into the pulsing regime. For very large diameter columns, the pulses may never span the column cross section at realistic flow rates.

Previous descriptions of pulses report an essentially flat front for the liquid-rich region. Visual observation of pulse fronts in our column revealed that the pulse fronts were approximately flat only when the pulses spanned the column cross section. Pulses not spanning the column cross section were curved concavely downward; this is shown schematically in Figure 18. An attempt was made to measure this curvature using pressure transducers at a number of positions at the same height. This failed as the response of the transducers proved to be too slow to provide the necessary resolution. The curvature of the pulses apparently depends on the gas and liquid flow rates. It was found that the curvature decreased when either flow rate was increased. When a step increase in either flow rate was made, it was observed that the width of the pulses (fronts) would increase instantaneously. This was accompanied by a decrease in the curvature. Subsequently, the pulses would slowly decrease in width and increase in curvature until an equilibrium curvature was achieved.

Our observations suggest the following picture for pulsing. Let us assume that an amount of liquid has collected in one place and a pulse has been formed. This liquid-rich region is pushed down the column by the gas behind it. The velocity at which this liquid-rich region moves down the column is lower than the actual velocity of the gas in the bed, Figure 16. Thus the gas behind this liquid-rich region is in effect able to move past it. One can think of two ways for the gas to move past the *liquid-rich* region:

1. The gas passes through the liquid-rich region.
2. The gas travels around the edge of the liquid-rich region, in effect bypassing it, as shown schematically in Figure 18.

For small pulses, one can expect the resistance experienced by the gas for going around the edge of the liquid-rich region to be smaller than that for passing through it and hence the preferred path for the gas behind the liquid-rich region will be to travel around it. If the gas is indeed going around the liquid-rich region, the pressure at the edge of the liquid-rich region will be lower than that at the center, thereby causing the pulses to be curved. As the pulses get wider, the pressure gradient in the lateral direction is less and hence the curvature of the pulse decreases. When the liquid-rich region spans the column cross section, all the gas must pass through it and hence there is little lateral pressure gradient. Therefore the pulse is essentially flat.

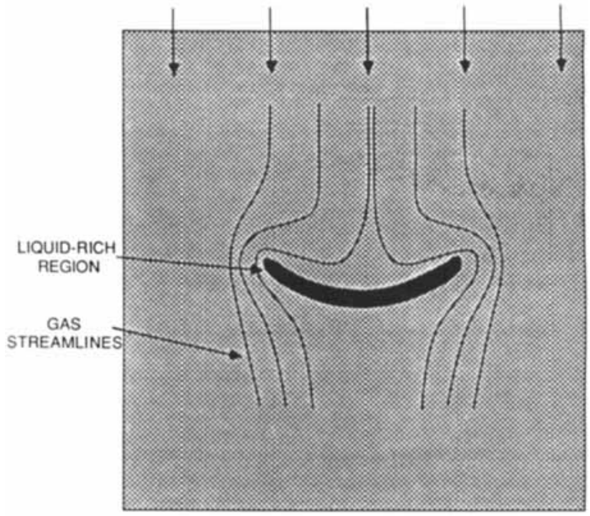

Figure 18. Representation of gas bypassing the liquid-rich region when the region does not span the column cross section.

The fact that the pulses do not always span the column cross section in our system helps to explain why the pulse velocity, the pulse frequency, and the pressure gradient measured in our column are in general lower than those obtained by previous researchers in small-diameter columns.

In the small-diameter columns used by previous researchers, the pulses spanned the column cross section for virtually every gas and liquid flow rate in the entire pulsing regime. Thus the gas behind the liquid-rich region has to pass through this liquid-rich region. On the other hand a less resistive path, one passing around the edge of the liquid-rich region, is available for gas flow in our column. Thus it is hardly surprising that the pressure gradient at specified flow rates is lower in our column when compared to that in the small-diameter columns. At high gas and liquid flow rates when the pulses were large enough to span our column, the measured pressure gradients were indeed found to be closer to the predictions of the correlations. Thus one can conclude that the literature correlations are valid only when the pulses span the column cross section. The applicability of these correlations to large-diameter columns used commercially is therefore rather questionable.

In a similar way, one can argue that the pulse velocities measured in our column are lower than those predicted from the work of Rao and Drinkenburg on small-diameter columns because of the gas bypassing and the concomitant lower pressure gradients in our column.

In small-diameter columns, the size of the liquid-rich region in a pulse unit does not change appreciably with liquid flow rate (Blok and Drinkenburg, 1982; Blok et al., 1983; Rao and Drinkenburg, 1983). This is not necessarily true in our system. The width of the pulse was found to increase with increased liquid flow rate (at a constant gas flow rate), indicating that the amount of liquid transported by a liquid-rich region increased as the liquid flow rate was increased. Thus the frequency of pulses in our system would not be expected to increase as rapidly with liquid flow rate as seen in smaller diameter columns; see Figure 17.

Pulse location

The distribution of gas and liquid at the top of the column, as well as the shape of the top surface of the packing, was found to have a marked effect on the location where the pulses were observed, and this is described below. Since the pulses were always observed to span the 0.051 m thickness of the column and move vertically downward, for the purpose of the following discussion we shall be concerned only about the location of the center of a pulse front with respect to either of the two 0.051 × 1.83 m sides (as viewed from either of the two 0.457 × 1.83 m sides).

For a very poor liquid distribution, the pulses were observed to form and traverse the column directly below the region where the highest liquid irrigation rate occurred. For example, when all but a few of the holes (intended for liquid flow) in the distributor plate were blocked so that the liquid rained over a small region as shown in Figure 19a, the pulses were observed to form directly below this region. This is not surprising as the formation of the pulses, being intimately related to the occlusion of channels for gas flow by the liquid, should indeed be favored in a region where the liquid holdup is the highest. As the flow rates increased, the pulses were observed to widen and ultimately span the column cross section even in the case of the poor liquid distribution shown in Figure 19a.

Even when the liquid was distributed uniformly (i.e., all the holes in the distributor plate intended for liquid flow were open),

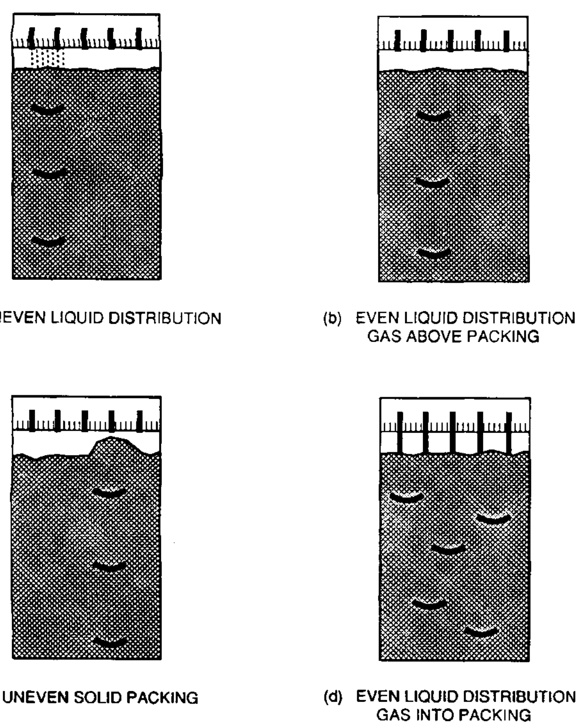

Figure 19. Effect of different distributions at top of column on position of pulses.

the pulses were observed to form at a preferential location, as shown in Figure 19b. In this set of experiments, the five tubes in the distributor plate intended for gas flow were not inserted into the packing, Figure 19b, so that there was an unpacked region below the distributor plate where the gas had to flow before entering the packing. (In most of the laboratory-scale as well as large-scale commercial columns, this type of gas distribution in which the gas flows through a certain length of unpacked column is indeed employed.) Visual inspection of the top surface did not reveal any unevenness. The column was repacked with the top surface essentially flat and the distributor still as shown in Figure 19b, and the experiments were repeated. Pulses were still observed to form at a preferred location, although this preferred location was not the same as that in the previous set of experiments. Thus it appears that the inherent nonuniformities in the packing had an effect on the location at which the pulses formed.

In an effort to understand the role of packing nonuniformities, two-phase cocurrent downflow experiments were carried out in a cylindrical column (0.076 m ID × 1.22 m) packed with Plexiglas beads of nominal diameter (3 mm). Dibutyl phthalate, which has the same refractive index as the Plexiglas packing, was the liquid used in this set of experiments. As a result of this refractive index matching, the movement of the gas bubbles in the column could be followed visually (provided the density of the gas bubbles was not too high). One of the observations from this study was the preferred entry of the gas into the packing at specific places, when a gas-liquid distribution of the type shown in Figure 19b was employed. One of the factors determining this preferred entry point was the smoothness of the top of the packing. It was observed that the gas preferred to enter the packing at its highest point. To check this hypothesis, the glass beads at the top of the packing in the rectangular column were arranged into a pile, as shown schematically in Figure 19c. The formation of the pulses was observed to take place directly beneath this pile. The pile was moved to several different locations and in every instance the pulses were observed directly beneath the pile. A logical explanation as to why the gas prefers to enter the packing at the highest point is elusive at the present time. However, given that the gas enters the packing preferentially at this location, it is easy to understand why the pulses are formed here. The occlusion of the gas flow channel by the liquid occurs more easily in this region as a result of increased gas-liquid interaction arising from a higher gas flow rate.

It was observed in our refractive-index matched studies that the entry of the gas at a few preferred locations could not be eliminated just be leveling the top surface of the packing. Thus, in addition to the shape of the top surface of the packing, there are other factors (packing characteristics) that could not be identified in our study which influence the gas entry point.

One conclusion that could be drawn from our study is that in order to ensure a uniform distribution of gas in the column, the gas should be directly injected into the column. One way to achieve this would be to extend the tubes in the distributor plate intended for gas distribution directly into the packing, as shown schematically in Figure 19d. This forces the gas to enter the packing at several locations. When this distributor configuration was used in our column, the pulses were observed randomly at at least four different locations. As the flow rates were increased, the pulses tended to coalesce and form larger ones. This was believed to indicate better gas distribution and this distributor configuration was used to obtain most of the hydrodynamic measurements discussed earlier.

In current industrial practice, to the best of our knowledge, the gas is not directly injected into the packing. In large columns used commercially, the packing is bound to settle with time, creating a top surface that is no longer level. Intuitively, the settling is more likely to occur in the center so that, after the settling, the packing will be concave downward with the highest points near the wall. Our studies indicate that this would encourage maldistribution, with the gas flowing preferentially near the walls of the column, which in turn may force the liquid to flow preferentially in the center. Direct injection of the gas into the packing should minimize such maldistributions.

Acknowledgments

This work was made possible by a Junior Faculty Career Development Grant awarded to S. Sundaresan by the Shell Foundation. The microwave unit was designed and built by R. Paglione, M. Nowogrodski, and F. J. Wozniak of Microwave Technology Center, RCA Corp., Princeton, NJ.

Notation

G = gas flow rate, $kg/m^2 \cdot h$
L = liquid flow rate, $kg/m^2 \cdot h$
V_ℓ = actual, i.e., interstitial, velocity of liquid, m/s
$V_{\ell t}$ = actual liquid velocity at trickling-to-pulsing transition
β = total liquid holdup, defined as the fraction of void volume occupied by the liquid

Literature cited

Beimesch, W. E., and D. P. Kessler, "Liquid-Gas Distribution Measurements in the Pulsing Regime of Two-Phase Concurrent Flow in Packed Beds," *AIChE J.*, **17**, 1160 (1971).

Blok, J. R., and A. A. H. Drinkenburg, "Hydrodynamic Properties of Pulses in Two-Phase Downflow-Operated Packed Columns," *Chem. Eng. J.*, **25**, 89 (1982).

Blok, J. R., J. Varkevisser, and A. A. H. Drinkenburg, "Transition to Pulsing Flow, Holdup and Pressure Drop in Packed Columns with Cocurrent Gas-Liquid Downflow," *Chem. Eng. Sci.*, **38**, 687 (1983).

Charpentier, J. C., M. Bakos, and P. LeGoff, "Hydrodynamics of Two-Phase Concurrent Downflow in Packed-Bed Reactors: Gas-Liquid Flow Regimes, Liquid Axial Dispersion, and Dead Zones," *Proc. 2nd Conf. "Quelques Applications de la Chimie Physique,"* Vestprem, Hungary, **2**, 31 (1971).

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).

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

Dimenstein, D. M., and K. M. Ng, "A Model for Pulsing Flow in Cocurrent Downflow Trickle-Bed Reactors," *Chem. Eng. Commun.*, **41**, 215 (1986).

Gianetto, A., G. Baldi, V. Specchia, and S. Sicardi, "Hydrodynamics and Solid-Liquid Contacting Effectiveness in Trickle-Bed Reactors," *AIChE J.*, **24**, 1087 (1978).

Herskowitz, M., and J. M. Smith, "Liquid Distribution in Trickle-Bed Reactors. I: Flow Measurements," *AIChE. J.*, **24**, 439 (1978).

Kan, K. M., and P. F. Greenfield, "Multiple Hydrodynamic States in Cocurrent Two-Phase Downflow through Packed Beds," *Ind. Eng. Chem. Process Des. Dev.*, **17**, 482 (1978).

———, "Pressure Drop and Holdup in Two-Phase Cocurrent Trickle Flows through Beds of Small Particles," *Ind. Eng. Chem. Process Des. Dev.*, **18**, 760 (1979).

Koros, R. M., "Scale-up Considerations for Mixed Phase Catalytic Reactors," *Multiphase Chemical Reactors,* **II**: *Design Methods,* A. E. Rodrigues, J. M. Calo, and N. H. Sweed, eds., Sijthoff and Noordhoff (1981).

Larkins, R. P., R. R. White, and D. W. Jeffrey, "Two-Phase Concurent Flow in Packed Beds," *AIChE J.*, **7**, 231 (1961).

Levec, J., A. E. Saéz, and R. G. Carbonell, "Holdup and Pressure Drop in Trickle-Bed Reactors," *8th Int. Symp. Chem. Reac. Eng.*, Edinburgh (1984).

———, "The Hydrodynamics of Trickling Flow in Packed Beds. II: Experimental Observations," *AIChE J.*, **32**(3), 369 (1986).

Midoux, N., M. Favier, and J. C. Charpentier, "Flow Pattern, Pressure Loss, and Liquid Holdup Data in Gas-Liquid Downflow through Packed Beds with Foaming and Nonfoaming Hydrocarbons," *J. Chem. Eng. Japan*, **9**, 350 (1976).

Ng, K. M., "A Model for Flow Regime Transitions in Cocurrent Downflow Trickle-bed Reactor," *AIChE J.*, **32**, 115 (1986).

Rao, V. G., and A. A. H. Drinkenburg, "Pressure Drop and Hydrodynamic Properties of Pulses in Two-Phase Gas-Liquid Downflow through Packed Columns," *Can. Chem. Eng. J.*, **62**, 158 (1983).

Saéz, A. E., and R. G. Carbonell, "Hydrodynamic Parameters for Gas-Liquid Cocurrent Flow in Packed Beds," *AIChE J.*, **31**, 52 (1985).

Sato, Y., T. Hirose, F. Takahashi, and M. Toda, "Pressure Loss and Liquid Holdup in Packed-Bed Reactor with Cocurrent Gas-Liquid Downflow," *J. Chem. Eng. Japan*, **6**, 147 (1973a).

Sato, Y., T. Hirose, F. Takahashi, M. Toda, and Y. Hashiguchi, "Flow Pattern and Pulsation Properties of Cocurrent Gas-Liquid Downflow in Packed Beds," *J. Chem. Eng. Japan*, **6**, 315 (1973b).

Satterfield, C. N., "Trickle-Bed Reactors," *AIChE J.*, **21**, 209 (1975).

Shah, Y. T., *Gas-Liquid-Solid Reactor Design,* McGraw-Hill, New York (1979).

Sicardi, S., H. Gerhard, and H. Hoffmann, "Flow Regime Transition in Trickle-Bed Reactors," *Chem. Eng. J.*, **18**, 173 (1979).

Sicardi, S., and H. Hoffmann, "Influence of Gas Velocity and Packing Geometry on Pulsing Inception on Trickle-Bed Reactors," *Chem. Eng. J.*, **20**, 251 (1980).

Specchia, V., and G. Baldi, "Pressure Drop and Liquid Holdup for Two-Phase Concurrent Flow in Packed Beds," *Chem. Eng. Sci.*, **32**, 515 (1977).

Talmor, E., "Two-Phase Downflow through Catalyst Beds. I: Flow Maps," *AIChE J.*, **23**, 868 (1977).

Tosun, G., "A Study of Cocurrent Downflow for Nonfoaming Gas-Liquid Systems in a Packed Bed. 1. Flow Regimes: Search for a Generalized Flow Map," *Ind. Eng. Chem. Process Des. Dev.*, **23**, 29 (1984a).

———, "A Study of Cocurrent Downflow for Nonfoaming Gas-Liquid Systems in a Packed Bed. 2. Pressure Drop: Search for a Correlation," *Ind. Eng. Chem. Process Des. Dev.*, **23**, 35 (1984b).

Turpin, J. L., and R. L. Huntington, "Prediction of Pressure Drop for Two-Phase, Two-Component Concurrent Flow in Packed Beds," *AIChE J.*, **13**, 1196 (1967).

Weekman, V. W., Jr., and J. E. Myers, "Fluid Flow Characteristics of Concurrent Gas-Liquid Flow in Packed Beds," *AIChE J.*, **10**, 951 (1964).

Manuscript received Nov. 5, 1985, and revision received Jan. 28, 1986.