Properties of Pulsing Flow in a Trickle Bed

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An experimental study of pulsing flow in trickle beds is presented, based on data from two measuring techniques. New evidence on pulse arrangement and propagation in the bed, and data on basic pulse characteristics (frequency, celerity, length, duration) as well as liquid holdup and pressure drop measurements are included. Some of these data, such as the length of the liquid-rich zone of pulses, are not currently available. Two flow regions exhibit different trends of pulse characteristics, "mild" and "wild" pulsing for relatively small and large liquid flow rates, respectively. New findings are compared with previously available data and correlations. An effort to develop new or modify existing generalized correlations is made for the aforementioned quantities.

Introduction

Packed beds in which a liquid and a gas flow concurrently downwards are usually referred to as *trickle beds*. This type of equipment is very frequently selected in chemical reactor design (mainly for oil refining) as outlined by Dudukovic and Mills (1986). *Pulsing flow* is observed in trickle beds under relatively high gas and liquid mass-flow rates. This particular flow regime is identified by the alternating passage of liquid-rich and gas-rich two-phase flow down the packed column, and it is associated with high mass-and heat-transfer rates. Due to the intensive interaction between the phases, it is considered suitable for fast reactions (Rao and Drinkenburg, 1985). Thus, there is considerable interest in predicting pulsing flow characteristics. In addition to time-averaged quantities, such as pressure drop and total liquid holdup, it is important to know various pulse properties such as frequency, celerity, and duration/length of liquid-rich and gas-rich zones. These quantities are essential for modeling pulsing flow (Rao and Drinkenburg, 1985; Dankworth et al., 1990; Dimenstein and Ng, 1986). They can be also very helpful in developing rules for design and scale-up of commercial units (Koros, 1986).

Several review articles on trickle beds in general are available in the literature: Satterfield (1975), Charpentier (1976), Hofmann (1977), Gianetto et al. (1978), Herskowitz and Smith (1983), Koros (1986), Zhukova et al. (1990). The recent review by Gianetto and Specchia (1992) places emphasis on the operation at elevated pressures. Most of the reported work on pulsing flow is devoted to average properties; that is, pressure drop, liquid holdup, and mass-transfer rates, for which several correlations are available. However, the influence of gas and liquid flow rates and of other system variables on pulse characteristics is not adequately dealt with in the literature. Some data are reported in the early studies by Weekman and Myers (1964), Beimesch and Kessler (1971), and Sato et al. (1973). More recently, Drinkenburg and his collaborators (Blok and Drinkenburg, 1982; Blok et al., 1983; Rao and Drinkenburg, 1983) obtained the most complete set of data on pulse frequency and celerity, using a conductance technique. Each probe consisted of a pair of parallel screens, placed 1 cm apart, covering the entire column cross section. Two such probes were placed in a packed section, 5 cm apart, at the end of the bed. It will be pointed out, however, that the possible influence of those closely spaced screens on the measured pulse characteristics has not been assessed. A limited amount of data, based on local pressure measurements, were presented by Dimenstein and Ng (1986) to support their modeling efforts.

Some information on pulsing flow characteristics was obtained by Christensen et al. (1986) in a "two-dimensional" packed column of rectangular cross section, in their study of the effect of column cross-sectional area. A novel microwave probe used in that study appeared to be sensitive only within a limited holdup range between 0.3 and 0.7.

To explain the macroscopic behavior observed in packed beds, Melli et al. (1990) employed a two-dimensional test sec-

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tion consisting of O-rings fixed between parallel transparent walls in a regular pattern in order to mimic the void space of packed beds. The observed macroscale flow regimes were attributed to various combinations of microscale patterns, which were the outcome of local competition between liquid and gas in the packing interstices. Using the same model "packed bed," Kolb et al. (1990) tried to identify the flow regimes from the power spectra of the sound detected at the outlet of the column. They found useful acoustic signatures of certain flow regimes as well as the transitions between them. The two-dimensional test section adopted in these articles has the advantage of easy flow visualization, but represents a porosity structure very different from that of an actual trickle bed. Since there are no contact points among the particles, the walls effects are very significant and the whole structure is uniform.

The main purpose of this work is to improve our understanding of the mechanics of pulsing flow by collecting a complete set of sufficiently detailed and accurate data. In previous work (Tsochatzidis and Karabelas, 1994b), in the same experimental setup employed here, evidence was obtained suggesting that complete and radially fairly uniform wetting prevails during pulsing flow. Thus, a novel nonintrusive conductance technique (Tsochatzidis et al., 1992) providing accurate, cross-sectionally averaged, instantaneous records is quite suitable for this study. By monitoring on-line or collecting time records of conductance, all the essential information is obtained, including the pulse propagation, pulse characteristics (frequency, celerity, length, duration), holdup profiles, as well as the respective time-averaged quantities. Measurements of pressure drop are also made. In the following sections the experimental procedures are described first, and the new data on pulse properties are outlined next. Preliminary experimental results of this work have been reported in a conference (Tsochatzidis and Karabelas, 1991).

Experimental Setup and Procedures

Equipment

Experiments are carried out in a cylindrical column made of Plexiglas. The column inner diameter is 14 cm and the column length 124 cm. The experimental system is shown in Figure 1. The experimental setup is also described in Tsochatzidis and Karabelas (1994a). Dry filtered air is saturated with water before entering the column to avoid temperature gradients due to evaporation. To minimize air flow fluctuations, a dampener is used. The experiments are carried out at room temperature (about 20°C). Water is sprayed uniformly onto the top of the packing through a perforated distributor and air is introduced by means of another perforated tubular section.

The packing material is fairly uniform unpolished glass spheres of 6 mm diameter. The column is packed layer by layer to make sure that the porosity of the bed is uniform. The void fraction of the bed was determined to be $\epsilon = 0.36$, and the bed specific surface area $S = 640\ m^{-1}$. The packing material is supported, at the bottom of the column, by a rigid stainless steel screen.

A newly developed conductance technique was employed that was capable of providing accurate instantaneous measurements of pulsing flow characteristics. Two identical con-

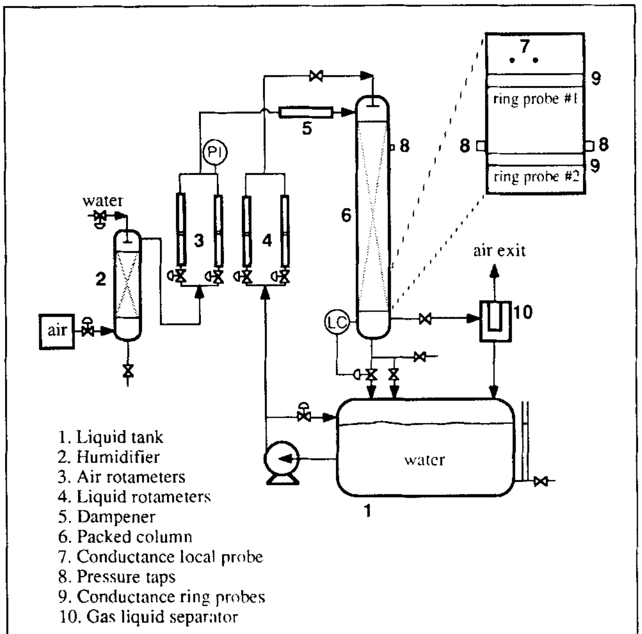

Figure 1. Experimental setup.

ductance probes were installed in the lower half of the packed column. Each probe consisted of two ring electrodes flush mounted onto the column wall to avoid disturbing the local porosity of the bed. The performance of these probes measuring conductance of gas-liquid mixtures in pipes and packed beds was studied experimentally and theoretically by Tsochatzidis et al. (1992). The ring electrodes had a width of 3 mm. A distance of 3 cm between the rings was chosen to achieve satisfactory spatial resolution, that is, ring spacing shorter than the length of the disturbance (pulses) being investigated. Criteria for selecting this ring electrode spacing were discussed by Tsochatzidis et al. (1992), where it was also shown that a distance of 7 cm from the bottom of the bed (where the lower ring electrode was placed) was more than adequate to avoid end effects.

An AC carrier voltage of 25 kHz frequency is applied across each probe in order to eliminate capacitive impedance. Electronic signal analyzers convert the response to analog output signals (Tsochatzidis et al., 1992). The analog signal from a probe is uniquely related to the conductance of the medium, which is in turn affected by the liquid fraction. Each probe is connected to a separate analyzer to permit simultaneous measurements. The signals from the analyzers are fed to an A/D converter and recorded to a computer. A sampling rate of 100 Hz is sufficient and data are collected over a time period of 20.48 s.

Signal analysis

An example of two simultaneously recorded signals, for superficial liquid mass-flow rate $L = 20.509$ kg/m^2·s and superficial gas mass-flow rate $G = 0.257$ kg/m^2·s is shown in Figure 2 (traces a,b). The voltage difference between the base and the peak of the disturbance in traces a, b of Figure 2 is in the range of 0.5 to 2 V, which is indicative of the satisfactory resolution of the technique. It is evident that each peak cor-

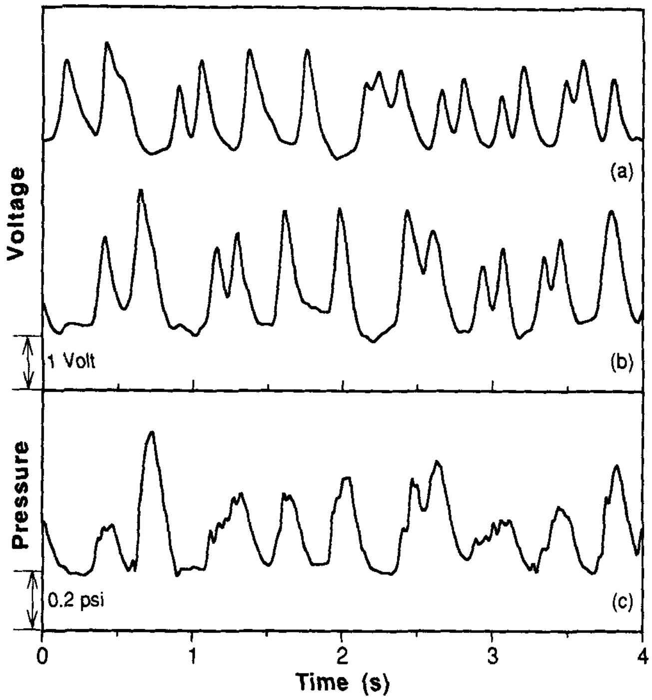

Figure 2. Typical simultaneous traces from conductance ring probes (a, b) and pressure transducer (c).

Flow rates: $L = 20.509$ kg/m^2·s; $G = 0.257$ kg/m^2·s.

responds to the passage of the liquid-rich zone of a pulse, followed by the low conductance (gas-rich) zone. The two traces are nearly identical except for a time-lag, corresponding to the time required by each pulse to cover the distance between the probes (20 cm). A steep increase is observed in the signal (corresponding to the liquid holdup) associated with the front of the liquid-rich zone, followed by a more gradual decrease in the gas-rich zone, which makes difficult the precise determination of boundaries between the liquid- and the gas-rich zones. Pulses are formed by blocking of some flow channels distributed throughout a cross section (Sicardi and Hofmann, 1980; Tsochatzidis and Karabelas, 1994a). This stochastic process can occasionally produce double or (seldom) triple pulses, as shown in Figure 2. As the gas and liquid flow rates increase, the generation of double or triple pulses tends to increase. In these twin pulses usually the second one exhibits higher voltage in its liquid-rich zone.

The shape of the peaks in Figure 2 bears some resemblance to that of pulses presented by Blok and Drinkenburg (1982; their Figure 3). However, the latter display a nearly constant holdup in the gas-rich zone (a flat "base" as they call it) that is not observed in any of the traces of this study. The extent to which the screens, placed in the packing by these authors, are responsible for those flat "bases" is unknown.

Statistical analysis of time records from the conductance probes was performed to determine the main pulse characteristics. Using time series analysis techniques (Bendat and Piersol, 1986), the power spectral density function was computed, from which the characteristic pulse frequency was obtained. To reduce the error and to increase the resolution of the computed spectra, the spectral density functions were obtained by dividing the samples into segments and by computing the power spectrum of each segment (Bartlett method). This operation was followed by averaging the spectra and frequency smoothing. The pulse celerity was obtained via the cross-correlation function of two simultaneously recorded signals. To determine the pulse celerity, the distance separating the two conductance ring probes was divided by the time delay of the maximum of the cross-correlograms. The correlation coefficient, corresponding to the primary maximum in the cross-correlation plots, was always quite high (above 0.70), indicating a clear detection of the time delay and of the respective celerity of pulse propagation.

Holdup measurements

To calibrate the probes (or to measure the temporal variation of liquid holdup) only one probe was used at a time, while the other was disconnected to avoid electronic interference. Using a technique based on electrically operated quick-closing valves, the dynamic liquid holdup, h_d, was independently determined at various gas and liquid flow rates, while the signal from the upper ring probe was recorded simultaneously. The time-average value from the recorded signal was used in this calibration, which gave very reproducible results. The calibration was performed in the trickling flow regime in which a nearly uniform liquid distribution is considered to prevail (Specchia et al., 1974). In order to achieve high values of liquid holdup, some runs were carried out close to the transition region between trickling and bubbling flow. As shown by Tsochatzidis and Karabelas (1991), the data used to prepare the calibration curve were in very good agreement with trickling flow data from the literature.

Tsochatzidis et al. (1992) show that the conductance probes are capable of providing fairly accurate (cross-sectionally av-

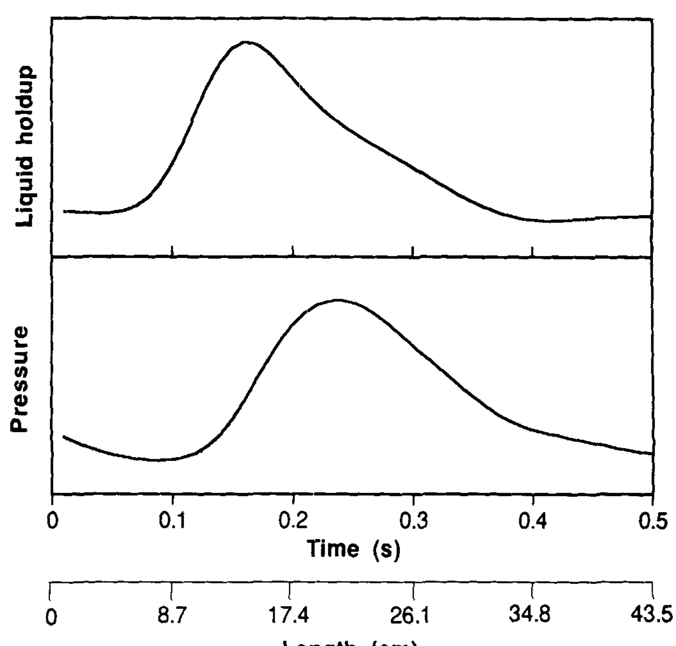

Figure 3. Simultaneous traces of liquid holdup and pressure variation in one pulse unit.

Flow rates: $L = 20.509$ kg/m^2·s; $G = 0.257$ kg/m^2·s.

eraged) holdup data. There is already evidence (Herskowitz and Smith, 1983; Chou et al., 1979; Tsochatzidis and Karabelas, 1994b) that in the pulsing flow regime the two-phase mixture is fairly evenly distributed in the radial direction. Therefore, the calibration for the dynamic liquid holdup made in the trickling flow regime can be used with confidence in the pulse flow regime as well.

The static liquid holdup, h_s, is determined using a section of the column. Starting with a dry packing, the section is flooded with a known amount of liquid and then is allowed to drain. As usual (Rao and Drinkenburg, 1985) from the difference of the initial amount and the weight of the drained liquid, the static holdup is determined to be 0.02. Levec et al. (1986) report $h_s = 0.022$ for 3- and 6-mm-dia. glass spheres. Van Swaaij et al. (1969) report that for $h_d/h_s > 8$ axial dispersion in trickle beds is very small and comparable to single-phase flow. This empirical condition is satisfied in most of our experiments in the pulsing flow regime.

Pressure measurements

For measuring two-phase pressure drop through the bed and recording of pressure fluctuations due to pulses, taps were drilled in the column wall and sensitive pressure transducers (R. D. P. Electronics Ltd., Model TJE true gauge) were mounted on special supports. The hole connecting the column to the transducer was drilled at an angle to allow gas purging and to ensure that it was always filled with liquid, thus avoiding damping of pressure fluctuations. One transducer was placed at the top section and another at the lower half of the column. The upper transducer could detect pressures in the range 0–10 psig, while the lower one is in the range 0–5 psig. Both transducers had accuracy of $\pm 0.1\%$ F.S. and frequency response of 1 kHz. The output signals of the transducers were fed to an A/D converter and recorded to a computer, with a sampling frequency of 100 Hz. The pressure drop through the packed bed was determined by subtracting the average values of the two pressure signals.

An example of the pressure fluctuations, taken from the downstream transducer is shown in Figure 2 (trace c). This trace is simultaneously recorded with the conductance traces. The downstream transducer was mounted just above the lower conductance probe. Thus, traces b and c of Figure 2 are comparable. It appears that the maximum pressure difference over a pulse is 0.2–0.4 psi (1,400–2,800 N/m^2), which is in general agreement with data from previous investigations (Rao and Drinkenburg, 1983, 1985). The sharp peaks of the conductance trace, corresponding to the passage of the liquid-rich zone of pulses, are smoother in the pressure trace due to the different averaging (in the longitudinal direction) of the two measuring techniques. The gas-rich zone of a pulse, corresponding to the low pressure and low conductance regions of the traces, seems to be better depicted in the conductance trace. Moreover, some of the twin pulses are apparently misrepresented in the pressure trace.

Results and Discussion

Pulse shape and propagation

By comparing the simultaneously recorded traces b and c of Figure 2 (from the lower conductance ring probe and the pressure transducer) it is apparent that there is a time lag between the peaks of the two quantities. The pressure peaks *lag* behind the liquid holdup maxima. This phenomenon becomes more pronounced if a correction is made on the pressure trace to account for the small difference in the axial location of the two probes (3.5 cm). Cross-correlations of simultaneously recorded holdup and pressure time series show that the corresponding mean distance between peaks is 4.0–7.5 cm throughout the flow rate range of the tests. It is noted that the pulse celerity (required in these calculations) is determined here for each set of flow conditions. The present data show that greater delays are associated with lower gas flow rates, for a constant liquid flow rate. These results are at variance with reported observations by Sato et al. (1973) that in low frequency pulsing flow no time lag was detected between the sharp increase in the pressure and the passage of the pulse front (the latter determined by motion pictures), while there was some evidence of time lag in high frequency pulsing flow.

In Figure 3 traces of one pulse unit are depicted, corrected for probe location. The flow rates are the same as in Figure 2. The pressure peak corresponds to the end of the liquid-rich zone (or the front of the following gas-rich zone), while the minimum of the pressure is just ahead of the front of the liquid-rich zone. This *moving pressure difference* obviously provides the force required to push the liquid-rich zone downstream. A similar phenomenon is observed in the slug flow regime for horizontal two-phase flow in a tube (Dukler and Hubbard, 1975). In general the shape of the pulse does not change dramatically with the flow rates. In mild pulsing (low flow rates) a rather symmetrical liquid-rich zone is formed. With increasing flow rates a more abrupt rise is associated with the front of the liquid-rich zone, especially in holdup traces, while a more gradual holdup reduction is shown in its tail.

To obtain information about the pulse arrangement over a column cross section, a third pressure transducer was mounted at the lower half of the column, on the diametrically opposite side of the original downstream transducer. Cross-correlations of the two simultaneously recorded pressure traces from these transducers (not presented here due to space limitations) show that the two signals are almost identical. It is evident that well-developed pulses are *radially symmetrical* and they apparently span the entire column cross section. Therefore, valid pressure measurements can be made anywhere in the circumference of a column section. Pulses not spanning the cross section, as reported by Christensen et al. (1986) for their two-dimensional packed column, are not observed in these tests and might characterize columns of larger diameters. In a parallel experimental study for the onset of pulsing (Tsochatzidis and Karabelas, 1994a), some slightly asymmetric pulses are observed only very close to the trickling-to-pulsing transition boundary. These "protopulses" (as they are called by Melli et al., 1990) are rectified with a small increase of flow rates giving rise to well-developed pulsing flow.

To further examine the pulse shape and position, a separate "local" conductance probe is employed. It is composed of a pair of nickel spheres having the same diameter as the packing material. One sphere is placed at the center of the bed cross section and 4.5 cm above the upper ring probe while

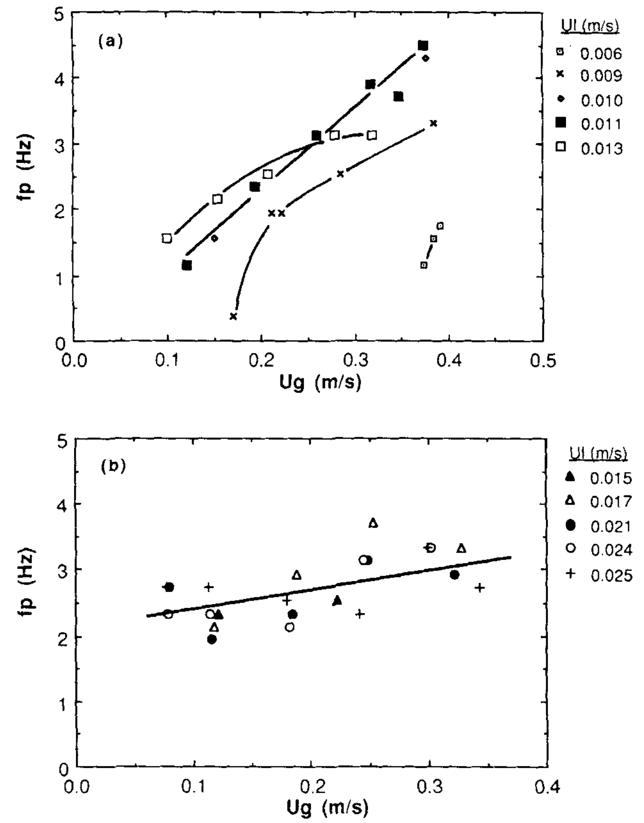

Figure 4. Variation of pulse frequency with gas superficial velocity for small (a) and large (b) liquid superficial velocities.

another is located at the same level and at a radial distance of 3.5 cm (half the bed radius). This arrangement of the "local" probe gives useful information on the conditions in the core of the bed in the pulsing flow regime. Traces of simultaneous measurements with the local probe and with the lower ring probe are very similar (Tsochatzidis, 1994).

From the traces obtained with three different sensors (ring probes, "local" probes, and pressure transducers), it is concluded that developed pulses cover the entire column cross section, and that radial nonuniformities are apparently insignificant at the scale investigated. The front of the liquid-rich zone appears to be geometrically fairly flat and horizontal as reflected in the steep front of practically all the traces. Visual observations by Tsochatzidis and Karabelas (1994a) support these remarks.

Pulse frequency

The pulse frequency is found to increase with both gas and liquid superficial velocities only for rather small values of U_l as shown in Figure 4a. For values of U_l higher than 0.015 m/s the pulse frequency seems to depend only on U_g, as depicted in Figure 4b. For obtaining rough estimates of pulse frequency at relatively high liquid flow rates (Figure 4b), the following linear expression may be used (for $U_l > 0.015$ m/s and 0.07 m/s $< U_g <$ 0.33 m/s):

$$f_p = 2.13 + 2.87\,U_g. \tag{1}$$

Weekman and Myers (1964) report that the pulse frequency tends to increase with the liquid rate up to approximately 20 $kg/m^2 \cdot s$; and that at higher liquid rates the frequency actually declines owing to increasing coalescence of the pulses. These observations are generally consistent with the present results.

It will be pointed out that some uncertainty is associated with the determination of the pulse characteristic frequency to which one may attribute the noticeable scattering of some of the present data (Figure 4b), also observed in data from the literature. First, the pulse frequency seems to be dependent, to a limited extent, on the probe axial location, possibly due to the coalescence of pulses. The new data reported here correspond to a probe placed 0.27 m above the bottom of the column. However, it is observed visually that at high liquid flow rates, the pulse frequency is slightly higher at the point where the pulses are formed (approximately 10 cm below the column entrance). The same observations are made by Dimenstein and Ng (1986), and are attributed to the change in pressure that causes a considerable expansion of the gas. Second, in certain cases the usual determination of the characteristic frequency from the main peak of the power spectrum involves some uncertainty. At low liquid flow rates (approximately $U_l < 0.015$ m/s) a steep peak characterizes the power spectra, for all gas flow rates, indicative of a single frequency. However, at higher liquid flow rates and intermediate gas rates, a second smaller peak appears in the spectrum at frequencies 1.5–2 Hz higher than the main peak, which corresponds to the double pulses. At higher gas flow rates, the second peak in the power spectrum vanishes and a broad peak is obtained due to coalescence of the twin pulses (Tsochatzidis, 1994).

Blok and Drinkenburg (1982) report that the pulse frequency is proportional to the difference between the real liquid velocity, u_l, and the real liquid velocity at the trickling-to-pulsing transition, u_{lt}. They propose that, with increasing liquid flow rate (at a given gas flow rate) the excess liquid represented by the difference $(u_l - u_{lt})$ is transported through the bed by an increasing number of pulses. They claim that for a given packing, u_{lt} appears to be almost constant. Therefore, pulse frequency, f_p, may be linearly related with u_l. Our pulse frequency data lend some support to these arguments, displaying a roughly linear u_l dependence, only for small U_l values. In fact, even though the measured u_{lt} values are not constant (Tsochatzidis and Karabelas, 1994a), their range of variation is small.

Christensen et al. (1986) found, in their *two-dimensional* column, the pulse frequency to increase sharply with increasing liquid flow rate, while only a weak dependence on the gas flow rate was noted. This may be explained by recalling that they observed pulses not spanning the entire column cross section and that the gas bypassed those liquid-rich zones. Hence, in their setup, the gas may not have significantly affected the pulses. Perhaps for the same reason they report values of pulse celerity lower than those of Rao and Drinkenburg (1983) for an identical packing material. Qualitatively the results of Kolb et al. (1990) concerning pulse frequency are in accord with the present data, but their values of f_p are much higher, possibly due to the quite different apparatus employed.

In attempting to develop a generalized correlation, the

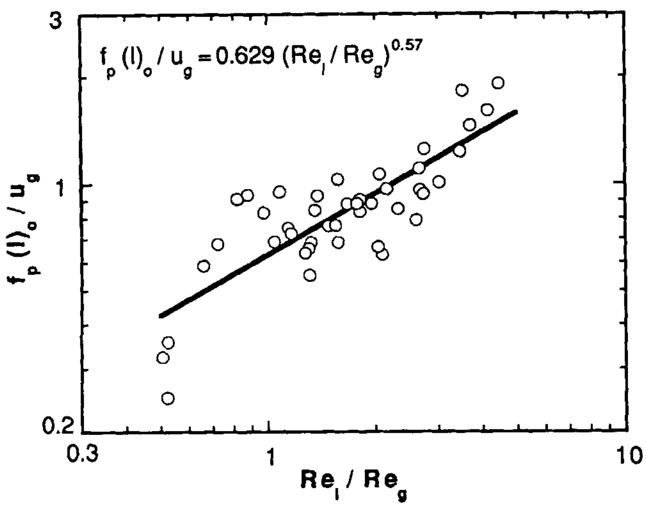

Figure 5. Correlation of dimensionless pulse frequency with liquid and gas Reynolds numbers (Eq. 2).

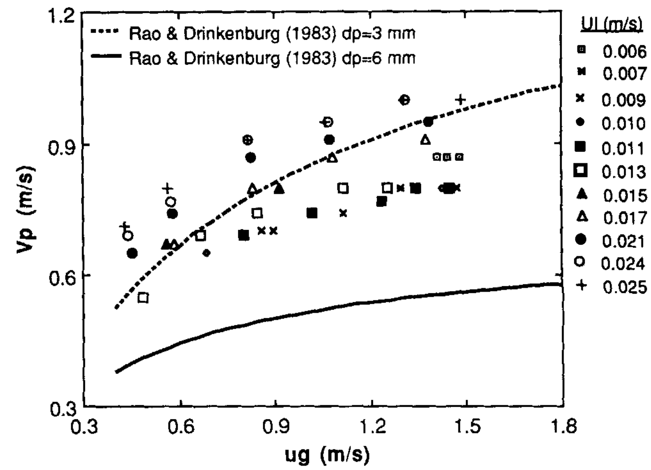

Figure 6. Influence of gas and liquid flow rates on pulse celerity.

pulse frequency should be rendered dimensionless using an appropriate time scale based on hydrodynamic quantities. The ratio of a typical pulse length over the real gas velocity, l/u_g, provides such a time scale. As reported in a subsequent section, the average pulse length tends to become almost constant at high liquid flow rates. Thus for simplicity the value $(l)_0 = 0.3$ m is selected here as the characteristic length. In Figure 5 the dimensionless frequency is plotted vs. the ratio of liquid and gas Reynolds numbers, which are based on real phase velocities and on particle radius (Tsochatzidis and Karabelas, 1994a). The correlation

$$\frac{f_p(l)_0}{u_g} = 0.629\left(\frac{Re_l}{Re_g}\right)^{0.57} \tag{2}$$

is clearly tentative. The real gas and liquid velocities are defined as $u_g = U_g/(\epsilon - h_t)$ and $u_l = U_l/h_t$ where h_t is the total liquid holdup that is treated in a subsequent section.

Pulse celerity

The effect of measured real gas velocity, u_g, and of the superficial liquid velocity on pulse celerity, V_p, is shown in Figure 6. Sato et al. (1973) report that the celerity for low frequency pulses is almost independent of flow rates of both gas and liquid. A number of studies (Blok and Drinkenburg, 1982; Rao and Drinkenburg, 1983; Christensen et al., 1986) suggest that the liquid flow rate has a very weak influence or no influence at all on the pulse celerity, which is dependent only on gas flow. The present data agree with this observation only for small liquid flow rates. For higher liquid rates, the influence of U_l on V_p is not insignificant, as also found by Weekman and Myers (1964). Dependence of pulse celerity on liquid flow rate is also reported by Kolb et al. (1990). The effect of gas velocity on V_p is somewhat stronger than that of U_l. However, at high real gas velocities, the pulse celerity data tend to approach an asymptotic value, as is also pointed out by Rao and Drinkenburg (1983).

Data corresponding to $U_l = 0.006$ m/s (the smallest liquid velocity) seem to be associated with rather high V_p values. These data points correspond to conditions very close to the trickling-to-pulsing transition boundary. The pulses formed at these not well-developed pulsing flow conditions may be influenced by small fluctuations of the flow rates (mainly of the gas flow) which may tend to increase the pulse celerity.

A common observation in the literature, confirmed by the new data, is that at high gas flow rates, the pulse celerity is *smaller* than the real gas velocity. Figure 6 shows that V_p is smaller than u_g, for u_g values greater than ~ 0.8 m/s, irrespective of the liquid flow rate. In these cases, gas must "penetrate" the pulse and gas bubbles or fingers must be formed in the liquid-rich zone of the pulse as suggested by Blok and Drinkenburg (1982).

A comparison of data from this work with a correlation proposed by Rao and Drinkenburg (1983) (to the authors' knowledge the only one available in the literature) shows that the latter significantly underpredicts the measured pulse celerities for a 6-mm packing. However, good agreement is obtained between the new data and the directly measured celerity by Rao and Drinkenburg (1983) for 3-mm particles (dashed line in Figure 6). This probably suggests that the pulse celerity may not significantly depend on particle size (for the same shape of packing), contrary to what the Rao and Drinkenburg (1983) correlation indicates. Moreover, that correlation neglects the influence of U_l. By nonlinear regression analysis of the present data an empirical correlation is obtained, as shown in Figure 7a. The pulse celerity is computed from the real gas velocity and the superficial liquid velocity as

$$V_p = \frac{1.03 u_g^{0.79}}{1 + 0.76 u_g^{1.27}}(1 + 25.4\, U_l) \tag{3}$$

for $0.4\ \text{m/s} < u_g < 1.5\ \text{m/s}$ and $0.011\ \text{m/s} < U_l < 0.025\ \text{m/s}$. For smaller values of U_l the influence of this quantity on pulse celerity can be neglected.

The pulse celerity can be expressed in dimensionless form with respect to u_g, since gas supplies the necessary force for

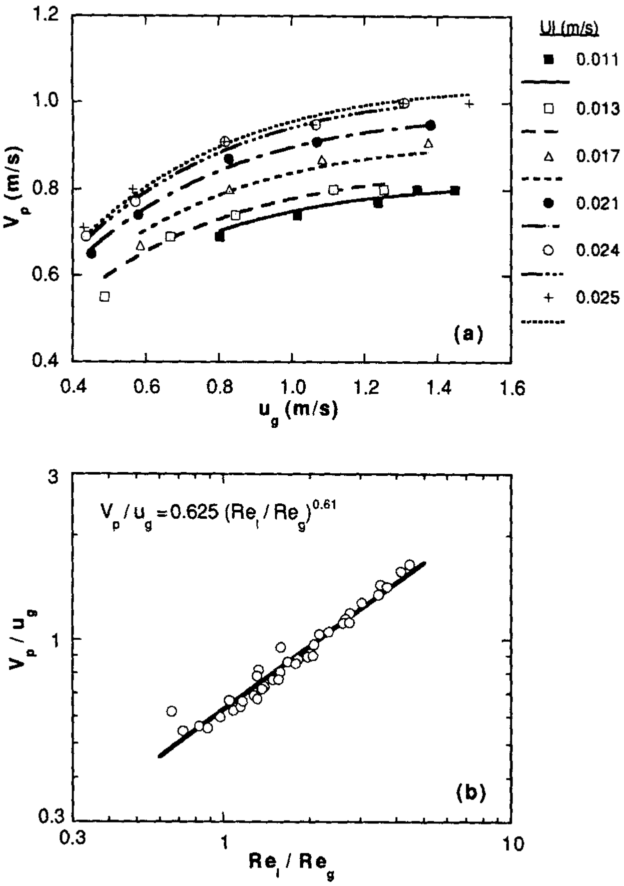

Figure 7. Correlation for: (a) pulse celerity (Eq. 3) vs. experimental data; (b) dimensionless pulse celerity with Reynolds numbers (Eq. 4).

the movement of the pulse as has been discussed earlier. For $V_p/u_g < 1$ the real gas velocity is greater than the pulse celerity and gas "penetrates" the liquid-rich zone. The dimensionless pulse celerity is correlated very well with the ratio of liquid over gas Reynolds number, as depicted in Figure 7b:

$$\frac{V_p}{u_g} = 0.625 \left(\frac{Re_l}{Re_g} \right)^{0.61}. \tag{4}$$

The ratio Re_l/Re_g effectively eliminates the dependence on packing diameter, which is in accord with previously made observations (Figure 6). This matter, however, deserves further study. Equation 3 is a better correlation for modeling and calculations, but Eq. 4 shows more clearly the dependence of pulse celerity on basic variables such as u_l and u_g.

Pulse length

The pulse length (or height) is obtained by dividing the pulse celerity by the pulse frequency ($l_p = V_p/f_p$). In that way the pulse length includes both liquid-rich and gas-rich zones of a pulse. This definition is valid only for well-developed flow, where the whole bed is occupied by alternating liquid-rich and gas-rich zones; that is, excluding some data very close to the transition boundary. It is observed that l_p is large near the trickling-to-pulsing transition boundary and tends to decrease with increasing gas velocity. This observation is in accord with the data of Weekman and Myers (1964), but it differs from the results of Blok and Drinkenburg (1982) and Rao and Drinkenburg (1983), probably due to the different (and somewhat arbitrary) definition of the pulse height they use. For high liquid flow rates, the pulse length seems to be almost constant and roughly equal to 0.3 m. A similar observation is reported by Rao and Drinkenburg (1983). More details are provided by Tsochatzidis (1994).

In addition to the preceding calculation of the mean pulse length, estimates are obtained of the length of consecutive liquid-rich zones, l_{l-r}, from the time series of the conductance ring probes. The average value of each time series is employed here as a criterion for the determination of the liquid-rich zone; that is, pulse segments above the average are considered to represent liquid-rich zones. In that way the duration of each liquid-rich zone is determined, from which (with the known pulse celerity) the liquid-rich length is calculated. Visual observations (Tsochatzidis and Karabelas, 1994a) are in line with the liquid-rich lengths determined by this procedure. Figure 8 shows probability density distributions of the liquid-rich zones for various flow rates. Each histogram corresponds to a specific liquid superficial velocity for several gas velocities. All distributions are asymmetric with a tail to the right (skewed to the right). The maximum value is in the range of 5–10 cm (with the exception of some data from the first histogram of $U_l = 0.006$ m/s and some from the last histogram of very high flow rates). At $U_l = 0.009$–0.011

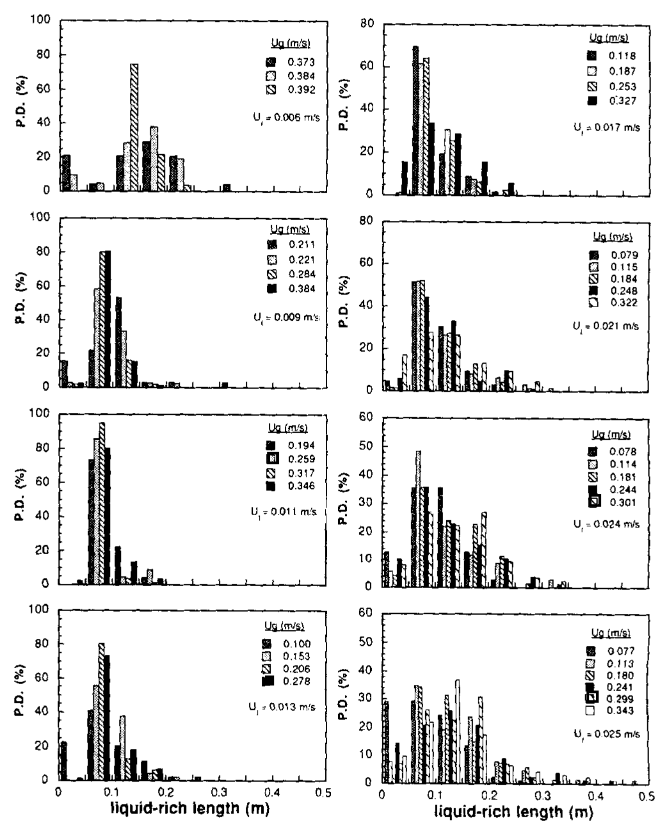

Figure 8. Probability density distributions of the liquid-rich lengths.

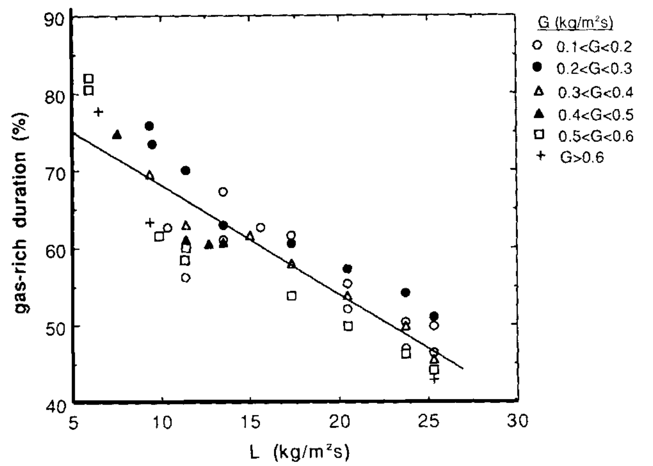

Figure 9. Influence of gas and liquid flow rates on pulse duration.

The line is represented by: % gas-rich duration = 80 − 1.4 L (kg/m^2·s).

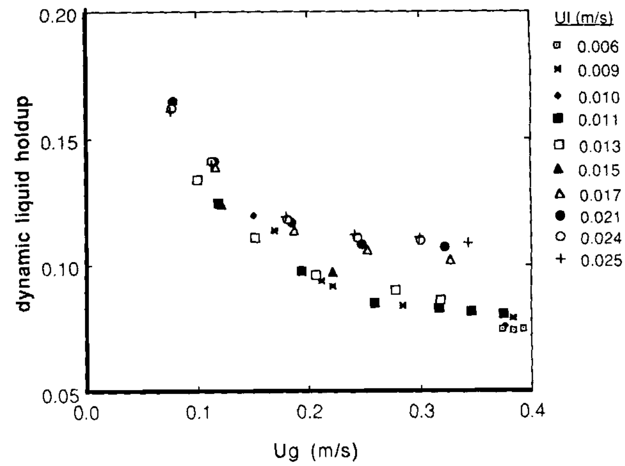

Figure 10. Influence of gas and liquid flow rates on the dynamic liquid holdup in the pulsing flow regime.

m/s a well-organized and repeatable flow is observed with more than 80% of the liquid-rich zones having the same length. An increase of the liquid velocity tends to spread the liquid-rich lengths. At low liquid velocities, an increase of gas velocity tends to make the distribution narrow and the flow more regular. The opposite trend is observed at high liquid flow rates where gas velocities tend to spread the data, but their influence is weaker in comparison with that of the liquid flow rates. The spreading with increased gas flow rate is also observed in histograms of the distribution of time intervals between the pulses, reported by Helwick et al. (1992). In data of the average liquid-rich length (not presented here due to space limitations) different trends are evident for mild and wild pulsing.

Pulse duration

The duration of the liquid- or the gas-rich zone of pulses (otherwise termed intermittence) is expected to influence the bed pressure drop and the overall mass- and heat-transfer rates. From this point of view, knowledge of this quantity is important for determination of the bed performance and for pulsing flow modeling. Previous studies dealing with the pulsing flow characteristics in trickle beds suggest that the gas-rich zone is normally larger than the respective liquid-rich zone of a pulse.

As before, the average value of the conductance signal is employed as a criterion to determine the gas-rich duration of a pulse. The percentage of time below the average is considered to represent the gas-rich duration for specific flow rates. Figure 9 shows that the gas-rich (or equivalently the liquid-rich) duration depends only on the liquid and not on the gas flow rate. This may imply that the liquid flow is mainly responsible for the pulse geometry. The reproducibility of measurements in Figure 9 (from different runs) is better than 3%, which is surprising if someone recalls that pulse frequency or celerity are not characterized by such a good reproducibility. A linear expression fits the data well. Furthermore, it is noted that some data in Figure 9, for very high liquid flow rates, are below 50%, meaning that at these conditions the gas-rich zone is somewhat smaller than the respective liquid-rich zone. This is an indication that the bubbling flow boundaries are approached.

Liquid holdup

This quantity is defined as the ratio of the liquid volume to the total volume of the bed. For a nonporous packing material, the total liquid holdup is comprised of the dynamic and the static component as follows:

$$h_t = h_d + h_s.$$

The dynamic liquid holdup, h_d, is measured in this work and corresponds to the liquid drained from the bed over a specified time period after the flow is stopped. The static component is determined as explained in the section on Experimental Setup and Procedures. Figure 10 shows that the mean dynamic liquid holdup tends to decrease with increasing gas flow rate. An almost insignificant increase of the liquid holdup is observed by increasing the liquid flow rate.

The pulsing flow regime is characterized by a significant temporal variation of the liquid holdup. This is evident in Figure 11 where typical holdup traces are presented. It is clear that peak values approximately 40% over the mean holdup are associated with the liquid-rich pulses, and that gas-rich zones (of relatively longer duration) do not exhibit a similar holdup reduction. Figure 11 also indicates that using the mean holdup value as a criterion for the determination of the liquid-rich zone, double or triple pulses are usually counted as one pulse unit.

The holdup in the liquid-rich and in the gas-rich zones of a pulse are quantities essential for the prediction of bed performance and the development of macroscopic models, such as those of Dimenstein and Ng (1986) and of Dankworth et al. (1990). Data for holdup in the liquid-rich and gas-rich zones ($h_{d,l\text{-}r}$ and $h_{d,g\text{-}r}$, respectively) are extracted in the following way. A computer program is employed to determine the peak and trough values of each pulse in the traces (like those of Figure 11). By averaging separately the peak and the

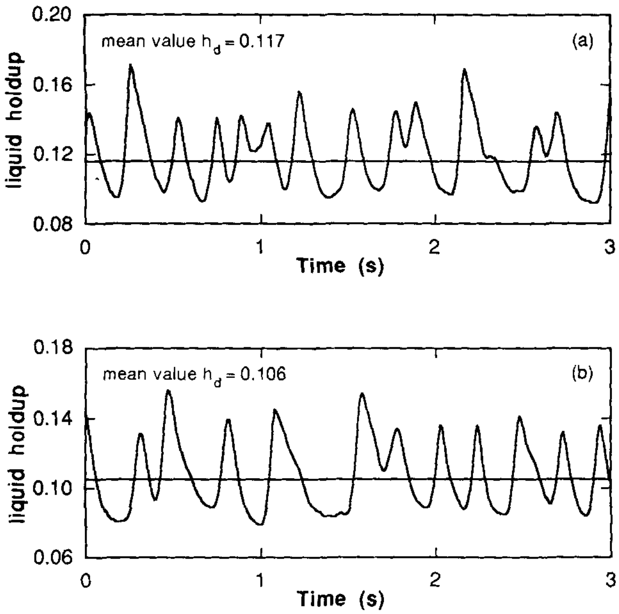

Figure 11. Holdup variation in the pulsing flow regime: (a) $L = 20.509$ kg/m²·s and $G = 0.257$ kg/m²·s; (b) $L = 17.291$ kg/m²·s and $G = 0.366$ kg/m²·s.

trough values, the quantities $h_{d,l\text{-}r}$ and the $h_{d,g\text{-}r}$ are calculated. The standard deviation of the peak or of the trough values of each trace is in the range 2–17% of their respective mean values. In relatively small gas and liquid flow rates (mild pulsing) the standard deviation of the peak values is higher than that of the trough values. This means that more repeatable gas-rich zones (concerning liquid holdup) are formed under these conditions. At higher gas and liquid flow rates (wild pulsing) this tendency is reversed. In general, the influence of gas and liquid flow rates on $h_{d,l\text{-}r}$ and $h_{d,g\text{-}r}$ is similar to that observed for the mean liquid holdup (Tsochatzidis, 1994).

The highest measured total holdup in the liquid-rich zone is 0.22 as compared to a maximum of 0.36 in perfect flooding. This leads to the conclusion that the liquid-rich zone of a pulse contains a large number of bubbles or gas clusters. Visual observations confirm this conclusion (Tsochatzidis and Karabelas, 1994a). Beimesch and Kessler (1971) used a conductance probe to measure the volumetric liquid fraction at a point in the bed. By integration of the local values, they determined that the average liquid fraction (based on the void space in the bed) in the primary liquid portion of the pulse (liquid-rich zone) is 0.40, while that of the gas portion (gas-rich zone) is 0.08. The proposed value for the liquid-rich zone is close to our data, while the value of the gas-rich zone is very low.

By noting the similarity of data for the mean holdup and for that in the liquid- and gas-rich zones, Blok and Drinkenburg (1982) present two correlations for the total "pulse" and "base" holdup, as they call, $h_{t,l\text{-}r}$ and $h_{t,g\text{-}r}$, respectively. They correlate $h_{t,l\text{-}r}/\epsilon$ and $h_{t,g\text{-}r}/\epsilon$ with the quantity (U_g/S), where S is the specific bed surface area. A correlation of the same form is proposed by Blok et al. (1983) for the mean total liquid holdup, h_t. A comparison is presented in Figure 12 between data of this work and the preceding correlations. The x-axis of Figure 12 represents a modified gas Reynolds number, Re_g^*, which is defined as

$$Re_g^* = \frac{U_g \rho_g}{\mu_g S}$$

and contains the ratio U_g/S proposed by Drinkenburg and his collaborators. Holdup data of this work are somewhat overpredicted even though they follow the trend of the correlations given earlier. The deviation is larger in Figure 12a, while it is almost insignificant in Figure 12c. To explain this discrepancy it will be recalled that Blok and Drinkenburg (1982) used a tracer technique to calibrate their conductivity cells, while the draining method was employed in this work. Kushalkar and Pangarkar (1990) report that the two different procedures may result in deviating holdup values, with the

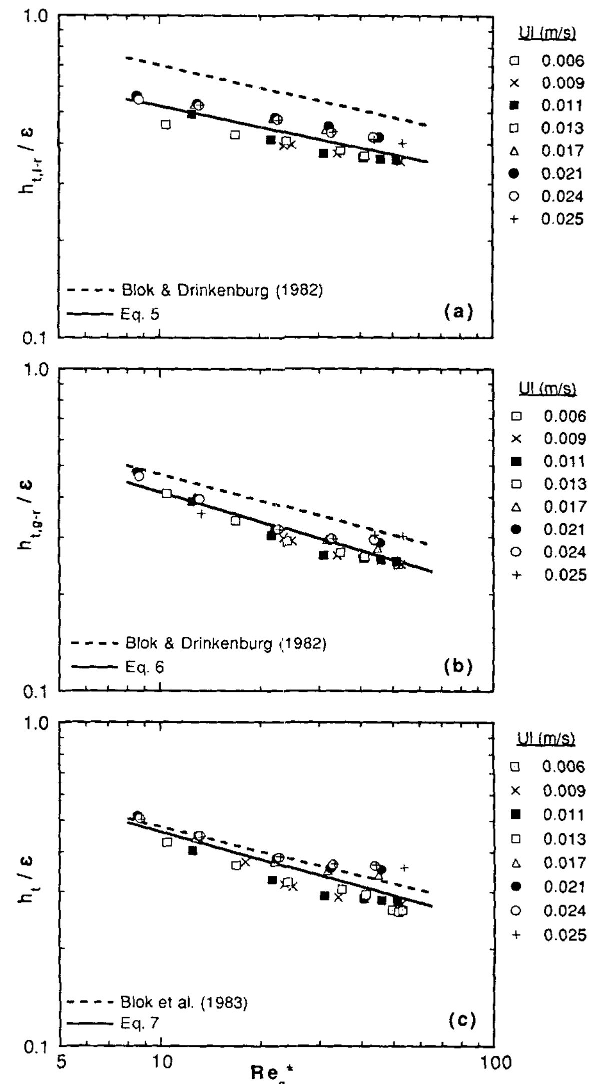

Figure 12. Comparison between data of liquid holdup and correlations.

(a) Total liquid holdup in the liquid-rich zone of a pulse; (b) total liquid holdup in the gas-rich zone of a pulse; (c) total mean liquid holdup in the pulsing flow regime.

tracer technique leading to higher values. Another reason is that, compared to the flush-mounted probes used in this work, a very different probe arrangement (screens) was employed by Blok and Drinkenburg (1982). Moreover, as mentioned earlier, the nearly flat gas-rich zones in their trace (used to determine $h_{t,g\text{-}r}$) are not observed in any of the traces obtained in this study.

Using the modified gas Reynolds number, the following liquid holdup correlations are proposed, from the best fit of the new data (Figure 12):

$$h_{t,l\text{-}r}/\epsilon = 0.856(Re_g^*)^{-0.215} \qquad (5)$$

$$h_{t,g\text{-}r}/\epsilon = 0.822(Re_g^*)^{-0.299} \qquad (6)$$

$$h_t/\epsilon = 0.866(Re_g^*)^{-0.278}. \qquad (7)$$

It will be noted that the ratio $h_{t,l\text{-}r}/h_{t,g\text{-}r}$ is almost constant ranging from 1.2 to 1.5 for the flow rates examined here. A somewhat larger range (1.5 to 2) is observed by Blok and Drinkenburg (1982). For high flow rates this ratio could be taken as constant and equal to 1.5.

Using a large database, Ellman et al. (1990) developed recently a generalized liquid holdup correlation. The latter systematically overestimates holdup (compared to the new data) but the data fall within the $\pm 50\%$ error band given by Ellman et al. (1990). More details of such comparisons are given by Tsochatzidis (1994).

Pressure drop

The pressure drop is related to the liquid holdup in trickle beds. As the liquid holdup increases, the resistance to gas flow also increases. Figure 13 shows that the two-phase pressure drop per unit length of the bed, $\Delta P/\Delta L$, increases with both gas and liquid mass-flow rates. This variation is almost linear in the pulsing flow regime. Such trends are in agreement with the observations made by other investigators (Rao et al., 1983; Rao and Drinkenburg, 1983). Dimenstein and Ng (1986) report a nonlinear dependence of pressure gradients on the liquid mass-flow rate, with an exponent somewhat greater than one. In Figure 13 pressure drop measurements from (single-phase) gas flow through dry packing are also included.

The pressure drop data are satisfactorily correlated with the sum of the liquid and gas Reynolds numbers as depicted in Figure 14:

$$\frac{\Delta P/\Delta L}{\Delta \rho g} = 6.1 \quad 10^{-6}(Re_l + Re_g)^{1.885}. \qquad (8)$$

In this correlation the pressure drop is made dimensionless with the static pressure of the bed (de Santos, 1991). It must be pointed out that the exponent value is not unusual for turbulent flow in simpler systems.

The present data agree fairly well with a correlation by Ellman et al. (1988) falling within the reported $\pm 60\%$ error limits. Of interest is also a pressure drop correlation by Larachi et al. (1991), derived from high-pressure experimental data. This correlation slightly underpredicts the new data

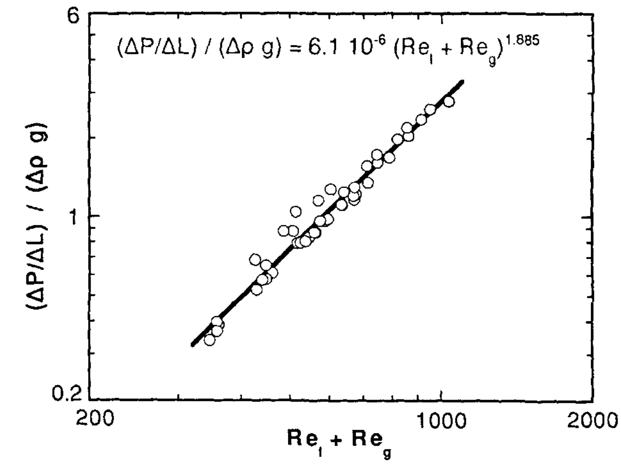

Figure 14. Correlation of dimensionless pressure drop with gas and liquid Reynolds numbers (Eq. 8).

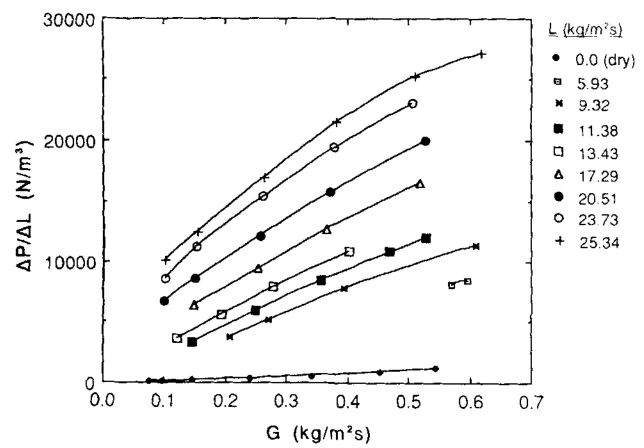

Figure 13. Influence of gas and liquid flow rates on the pressure drop in the pulsing flow regime; also shown are pressure drop data of gas flow through dry packing.

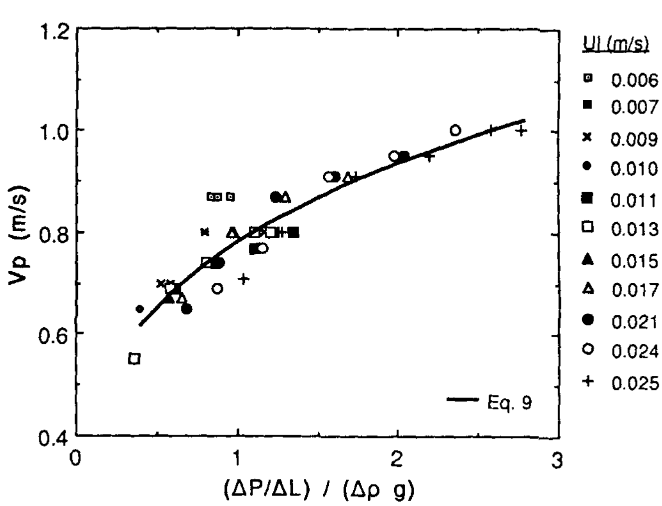

Figure 15. Correlation of pulse celerity with pressure drop.

that fall well within the error limit of $\pm 50\%$ proposed by the authors.

Finally, an interesting correlation is found between pressure drop in pulsing flow and pulse celerity data shown in Figure 15:

$$V_p = 0.78\left(\frac{\Delta P/\Delta L}{\Delta \rho g}\right)^{0.26}. \quad (9)$$

With increasing pressure drop (driving force) the pulse celerity tends to increase almost linearly at small pressure drop values. At higher $\Delta P/\Delta L$, pulse celerity data tend to approach an asymptotic value. From Eq. 9 the pulse celerity can be estimated if only the pressure drop is known. It is noted that Rao and Drinkenburg (1985) report a variation of pressure drop (due to the geometric and dynamic interaction) in the pulsing flow regime, with pulse frequency.

Concluding Remarks

Most of the data reported in this article are obtained with a conductance technique (Tsochatzidis et al., 1992) that allows accurate measurements of instantaneous, cross-sectionally averaged, holdup. These data are complemented with pressure measurements. Cross-correlation of simultaneously recorded liquid holdup and pressure signals clearly show that the pressure peaks lag behind the holdup maxima (liquid-rich zone of a pulse). Such traveling pressure differences provide the necessary driving force for moving the pulses. Evidence is also obtained that well-developed pulses are radially symmetric and that they span the entire column cross section in the system studied. However, in some industrial packed equipment of diameter much larger than the present one this symmetry may not exist and multiple pulses may appear, perhaps similar to those reported by Christensen et al. (1986).

A complete set of data is obtained on basic pulse characteristics such as frequency, celerity, length, and duration. Generally, two groups of data are discerned within the pulsing flow regime, exhibiting different trends. They are identified as "mild" and "wild" pulsing and correspond to relatively small and large liquid flow rates, respectively. Pulse frequencies tend to fluctuate somewhat, and in wild pulsing they seem to depend only on gas-flow rate. The pulse celerity depends on both gas and liquid flow rates, with a tendency to approach an asymptotic value at high real gas velocities. For real gas velocity, u_g, greater than ~ 0.8 m/s the pulse celerity is smaller than u_g, implying that gas must "penetrate" the pulse. This phenomenon appears to be responsible for an intensive transfer of momentum between phases and consequently for increased heat-and mass-transfer rates. Dimensionless pulse frequency and celerity are correlated with the ratio of liquid and gas Reynolds numbers.

Data on the length of the liquid-rich zones of pulses are reported for the first time here. Probability density distributions of such data indicate flow conditions under which very well-organized and repeatable pulsing flow prevails. The average pulse duration is found to depend essentially only on liquid flow rate. As regards the geometrical features of pulses, the results show that they are mainly affected by the liquid flow rate. However, the dynamic liquid holdup in the liquid-rich and gas-rich zones depends mainly on gas flow. New pressure drop data in pulsing flow are compared with existing correlations and found to be within their reported error limits. Useful correlations for most of the aforementioned quantities are proposed and/or modified. The data show an interesting correlation between measured pressure drop and pulse celerity.

The experiments reported here cover phenomena that manifest themselves in a length *macroscale* roughly of order 10 cm (considering the bed radius or a pulse characteristic length). Pulsing flow in the *microscale* (characteristic length of packing interstices or particle radius) was also studied in the course of this work using other appropriate techniques (Tsochatzidis and Karabelas 1994a,b). The latter provide information complementing the present data. Thus, there are sufficient reliable data to test and/or develop models of pulsing flow hydrodynamics, a prerequisite to simulating other physical or chemical processes.

The validity and applicability of the new results, on pulsing flow outside the range of conditions where they were obtained, is a matter of concern difficult to address at present. It is worth summarizing a few relevant observations, though. The effect of high operating pressure is of major importance in industrial trickle beds, and it is examined in recent studies. Wammes et al (1990) report that, when the reactor pressure is increased the trickling-to-pulsing transition boundary shifts toward higher liquid flow rates, while the dynamic liquid holdup is slightly decreased. Larachi et al. (1991) report that for values of the dimensionless group $(G/L)\sqrt{\rho_l/\rho_g} \geq 0.1$ and nonfoaming systems, the pressure drop at any high pressure can be predicted by using only measurements made at atmospheric pressure. Moreover, at low gas velocities ($U_g \leq 0.01$ m/s), the operating pressure appears to have no influence on liquid saturation, and measurements at atmospheric pressure may be sufficient for predictions at higher pressures. Turning to our data, there are indications (Figure 6) that the pulse celerity may be independent of packing particle size in the particle diameter range 1 to 10 mm. It appears therefore that the present results may be also applicable to somewhat different conditions. Of course, more experimental evidence combined with realistic modeling is required to address this important issue of scale-up.

Acknowledgment

The authors are grateful to the Commission of the European Communities for financial support of this work under contract JOU2-CT92-0067. N. A. Tsochatzidis would also like to extend his gratitude to Bodossakis Foundation for a scholarship.

Notation

d_p = particle diameter, m
D = bed diameter, m
r_p = particle radius, m
μ = dynamic viscosity, kg/(m·s)
ρ = density, kg/m^3

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Manuscript received Mar. 30, 1994, and revision received Dec. 5, 1994.