The Hydrodynamics of Trickling Flow in Packed Beds

Part II: Experimental Observations

The liquid holdup and the pressure drop for two-phase trickling flow in a packed bed were measured experimentally. Different values of those parameters were obtained as the liquid flow rate was increased and then decreased, indicating a multiplicity in hydrodynamic states. This behavior was observed even when the gas flow rate was zero in the bed. It was determined that the hysteresis exhibited by the process is due to imperfect wetting of the packing and to the difference between advancing and receding contact angles at the gas-liquid-solid contact lines. The reduced wetting conditions are also the cause of the increase in liquid holdup observed when the surface tension of the liquid is decreased. In this case, the amount of liquid retained in the bed is controlled by the extension of the wetted regions due to the more favorable contact angle as the surface tension decreases. The transition from the trickling to the pulsing flow regime was also dependent on the history of the process.

A new correlation for predicting liquid holdups and pressure drops for trickling flow in packed beds is proposed for the decreasing liquid flow rate operating mode. This correlation is based on the experimental determination of the liquid phase relative permeability as a function of the liquid phase reduced saturation and the determination of the gas phase relative permeability as a function of the gas phase saturation and the gas phase Reynolds number. The new data are analyzed in the light of the theory developed in the first part of this paper.

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SCOPE

Extensive experimental studies of the hydrodynamics of trickle-bed reactors have been conducted in recent years. These studies are aimed primarily at the development of empirical correlations as the way to predict the hydrodynamic performance of such systems. Despite the large amount of work done in this area, there are still uncertainties associated with the prediction of pressure drops and holdups for these systems. The lack of understanding of this complex hydrodynamic behavior has resulted in correlations that are valid only over limited ranges of experimental conditions.

The main objective of this work is to present a detailed experimental study of the hydrodynamics of trickling flow in packed beds. This study deals with the experimental evaluation of liquid holdups and pressure drops for a wide range of operating conditions, giving special attention to the dependence of these parameters on the history of the process, i.e., to the presence of hydrodynamic hysteresis.

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The traditional concepts of relative permeabilities are used to analyze the experimental results and to compare trickle bed hydrodynamics to the predictions of conduit models developed in Part I (Sáez et al., 1986).

CONCLUSIONS AND SIGNIFICANCE

An experimental study of the hydrodynamics of trickling flow in packed beds was conducted. This study provided evidence of the existence of a multiplicity of hydrodynamic states, not only under gas-liquid flow operation but also when the gas phase is stagnant in the column. The hydrodynamic performance of the system depends on the history of the process. Specifically, hydrodynamic parameters such as pressure drop and liquid holdup are functions not only of the operating conditions but also of the operating mode; i.e., different results are obtained when the operating conditions are reached by increasing the liquid flow rate or by decreasing it to the desired value. This phenomenon is explained by the fact that imperfect wetting conditions exist in the bed. When there are partially dry zones, the hysteresis of the contact angle at the gas-liquid-solid contact line leads to different configurations in the distribution of the liquid according to the operating mode and, therefore, to different holdups and pressure drops. The hysteresis of liquid holdups and pressure drops in two-phase flow is appreciable for small packing particles (spheres of 0.3 cm dia.) for which differences of almost one order of magnitude were observed in the pressure drop between the two modes of operation. This fact is of great importance to the design of trickle-bed reactors.

The multiplicity of hydrodynamic states also affects the transition from trickling to pulsing flow regime. It was observed that the transition to pulsing flow at a given gas phase Reynolds number occurred at a larger liquid flow rate in the increasing liquid flow rate mode of operation than in the decreasing flow rate mode.

The effect of surface tension on the liquid holdup was studied by performing experiments with pure water and aqueous solutions of a surfactant as working fluids. It was observed that under the same operating conditions, a decrease in the surface tension of the liquid resulted in an increase in the amount of liquid retained in the medium. This behavior is the opposite of that followed by conduit models in which perfect wetting conditions are assumed. The presence of imperfect wetting provides an explanation of the experimental trends. A reduction in surface tension causes a reduction in the contact angle and a subsequent increase in the amount of liquid retained. This effect is more dramatic in the increasing flow rate mode of operation, in which the wetting efficiency is expected to be smaller.

The experimental data obtained were analyzed by using the concepts of relative permeabilities. The liquid phase relative permeabilities calculated from the data exhibited hysteresis, but within any of the operating modes they were insensitive to surface tension effects and packing characteristics. The experimental liquid relative permeabilities compared well to those predicted by the conduit models in the absence of surface tension effects.

The experimentally obtained gas phase relative permeabilities followed the same qualitative trends predicted by the conduit models. However, quantitative comparisons were not accurate.

The liquid phase relative permeability corresponding to the decreasing liquid flow rate mode of operation was correlated with the reduced liquid saturation. The gas phase relative permeability was correlated with the gas phase saturation and Reynolds number. This allowed the derivation of a correlation for predicting liquid holdups and pressure drops that gave mean relative deviations with respect to experimental values of 3.5% for the liquid holdups and 13% for the pressure drops. This correlation represents a significant improvement over previous correlations and it provides a better physical interpretation of the process.

Introduction

A large number of previous studies of the hydrodynamics of trickling flow in packed beds have resulted in the accumulation of an extensive array of experimental observations. The interpretation of the physics of the process has traditionally relied on phenomenological analogies aimed at the development of empirical correlations to predict pressure drops and liquid holdups in the bed. Of these two parameters, the study of pressure drops has received more attention owing to the direct relation that it has to the design of trickle-bed reactors. Four different approaches have been used to predict pressure drops in gas-liquid cocurrent flow through packed beds:

1. The two-phase flow pressure drop has been expressed as a function of the pressure drops of gas and liquid in one-phase flow through the packed bed. The correlation is of the same form as those used to predict pressure drops in two-phase flow through tubes. This approach was developed by Larkins (1959) and it has subsequently been used in most of the investigations performed in the field (Weekman, 1963; Reiss, 1967; Charpentier et al., 1968; and Midoux et al., 1976).
2. The pressure drop has been correlated to the liquid and gas

phase Reynolds numbers by means of a friction factor type of relation. This method was proposed by Turpin and Huntington (1967) and later used by Specchia and Baldi (1977) to correlate pressure drops in the pulsing regime. This method was also used recently by Kan and Greenfield (1979) in correlating their pressure drop information.

3. An Ergun-type equation with adjustable parameters has been used to calculate two-phase pressure drops by Hutton and Leung (1974) and Specchia and Baldi (1977).

4. Sáez and Carbonell (1985) analyzed the gas-liquid hydrodynamics of trickling flow by applying the traditional concepts of relative permeabilities.

Some of the correlations proposed in the works mentioned above have been compared by Sáez and Carbonell (1985) and Tosun (1984).

The prediction of liquid holdup is sometimes treated as a problem that is separate from the prediction of pressure drops. Of all the approaches presented above, only the investigations following the techniques outlined in methods 3 and 4 have tried to relate these two parameters. In the other approaches the liquid holdup has been directly correlated with dimensionless numbers representative of the characteristics of the flow (Reynolds and Galileo numbers) in the manner presented by Otake and Okada (1953), or it has been related to the single-phase pressure drops (Midoux et al., 1976). In this work we analyze a new set of experimental data for liquid holdups and pressure drops using the relative permeability approach originally proposed by Sáez and Carbonell (1985). The data will then be compared to the predictions of the conduit models developed in Part I of this paper (Sáez et al., 1986).

The hydrodynamics of trickling flow through packed beds exhibits an interesting phenomenon that has been mostly overlooked in the classical literature on the subject. We are referring to a multiplicity in hydrodynamic states, first reported by Kan and Greenfield (1978). These investigators observed that the pressure drop and liquid holdup depended not only on the operating conditions but also on the history of the process, i.e., the operating mode. Specifically, when the process was operated by fixing a liquid flow rate and then adjusting the gas flow rate to the desired value, they observed that the hydrodynamic performance of the system was strongly dependent on the maximum gas flow rate to which the process was subjected after fixing the liquid flow rate. Kan and Greenfield speculated that this hysteresis was due to an irreversible breakup of liquid bridges transverse to the direction of the flow that occurs as the gas flow rate is increased. They concluded that the maximum gas flow rate was enough to characterize the history of the process and went on to propose a correlation for calculating pressure drops and holdups (Kan and Greenfield, 1979) that included the gas phase Reynolds number given by the maximum gas flow rate as an additional parameter. The experiments conducted by these investigators were performed with small particles (spheres of less than 2 mm dia.) and the air-water system, indicating that the strong surface tension forces might have been the source of the phenomenon. However, when the surface tension of the water was decreased by adding a wetting agent, the same behavior was observed. The multiplicity of hydrodynamic states has not been reported in other works dealing with small particles, such as those of Tosun (1984), who carried out experiments with the air-water system and particle diameters less than 2 mm, and Morsi et al. (1984), who used aqueous solutions as working liquids and a particle diameter of 1.16 mm. In the present work we provide additional evidence of the existence of hysteresis in trickling flow through packed beds and we present an experimental study of this phenomenon. As will be shown, the maximum gas flow rate does not seem to be the most important factor determining the hysteresis behavior. We found hysteresis even when the gas flow rate is zero in the column. We also present results on the influence of particle diameter and liquid surface tension on the hysteresis and pulsing behavior.

Experimental

Apparatus

The experimental system used in this investigation is shown in Figure 1. The column was made of Plexiglas and had a 17.2 cm ID and 130 cm total packing length. The packing section consisted of six removable segments 20 cm long and one 10 cm segment. The column was equipped with seven pressure taps located axially at nearly equal separation and connected to a water manometer, alcohol manometer, or Lambrecht manometer, depending on the magnitude of the measured pressure drop.

The packing materials used were 0.3 and 0.6 cm dia. glass spheres. The spheres were supported in the packing section of the column by a stainless steel screen which was mounted on a bundle of 1.5 cm OD PVC tubes that had been glued together and served as an outlet distributor.

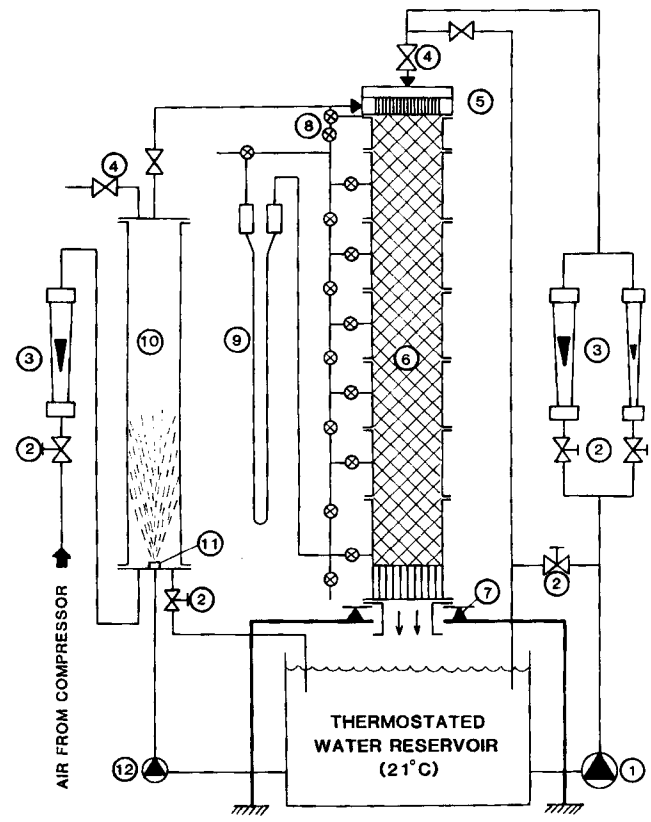

Figure 1. Experimental apparatus.

1. Pump
2. Regulating valves
3. Rotameters
4. Ball valves
5. Gas-liquid distributor
6. Column packing
7. Weighing device
8. Stopcocks
9. Manometers
10. Air humidifier
11. Nozzle
12. High-pressure pump

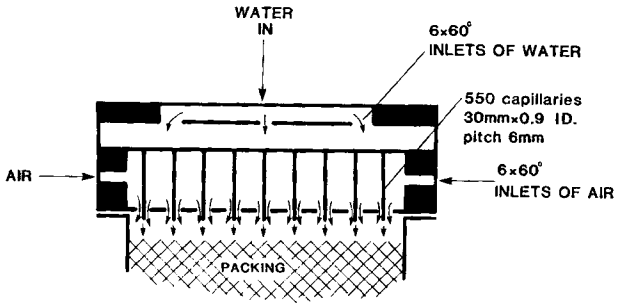

Figure 2. Inlet distributor.

The flow rates of air and water were measured by means of calibrated rotameters. The water temperature was kept constant at 21°C during the experimental runs. Atmospheric air coming from a compressor was saturated with water at 21°C in a spray column before entering the packed bed.

The column was equipped with a flow distributor, Figure 2, consisting of 550 capillaries at 0.6 cm pitch with 0.09 cm ID and 3.0 cm length through which water was pumped into the column. The capillaries were placed between two Pertinax plates. The bottom plate had circular holes around the capillaries that had a diameter slightly larger than the outer diameter of the tubes. The air was introduced to the chamber formed between the plates and it exited the distributor through these holes. The top of packed section of the column rested about 0.5 cm below the distributor.

The dynamic liquid holdup was measured by weighing the column during the operation by means of a strain gauge (N/SL from BLH Electronics, USA) with a sensitivity of 360 μV per kg. This high sensitivity allowed the measurement of weight differences of about 5 g without appreciable error from a dry weight of column of 80 kg.

The structural properties of the column, as well as the range of operating conditions used in this work, are summarized in Table 1.

Procedure

Two types of experiments were performed: one consisting of only liquid flowing through the bed in the presence of a stagnant gas phase, and another with gas and liquid flowing downward through the bed.

In the stagnant gas operation, the column was preflooded and then allowed to drain for about 20 min; at this point a residual amount of liquid (static holdup) remained in the bed. The column was then kept at atmospheric pressure and the liquid flow rate was gradually increased from zero to the value at which it was observed that the column was completely flooded. The liquid flow rate was then gradually decreased to zero. During both operating modes (increasing and decreasing flow rate) the dynamic liquid holdup was measured as a function of the liquid flow rate.

Table 1. Properties of Column and Packing and Operating Conditions

Column dia.	17.2 cm
Total packing Length	130 cm
Sphere dia. (glass)	0.30 and 0.60 cm
Bed porosity (ϵ)	0.375–0.385
Static holdup (both sphere sizes) (ϵ_β^o)	0.022
Surface tension range	0.0388–0.0725 N/m
Mass flow rate ranges:	
Air	0–0.37 kg/m^{2}s
Water	0.06–25.1 kg/m^{2}s

The two-phase flow experiments were performed in the following manner. Before starting every set of runs, pulsing flow was induced by operating at very high gas and liquid flow rates. This assured that all sections of the packing were completely wetted and provided excellent reproducibility. The gas and liquid flows were shut down simultaneously and the column was allowed to drain for 20 min. Afterward, the gas flow rate was set at a fixed value and the liquid flow rate was gradually increased until the pulsing flow regime was reached. The liquid flow rate was then decreased. Pressure drops and dynamic holdups were measured during both modes of operation (increasing and decreasing liquid flow rates). The column was packed segment by segment to make sure that the porosity of the bed was uniform. Each segment was packed by carefully filling it with particles. The second segment was assembled on top of the first; this procedure was repeated from the bottom to the top of the bed. Differences in global porosities between two consecutive packing procedures were always less than 2%. A reproducibility of 12% was found in the measurement of static holdup. The dynamic holdup showed a 15% reproducibility for low values ($\epsilon_\beta^D < 0.03$) and a 5% reproducibility for high values ($\epsilon_\beta^D > 0.2$) within the same mode of operation. The reproducibility of the pressure drop measurements was estimated to be 5% at low values of the pressure drop (below 200 mm of water) and up to 20% near the transition to the pulsing flow regime.

All the experiments were carried out with the air-water system. The only physical property varied was the surface tension, which was reduced by adding sodium octadecil sulphate to the water in very small concentrations (below 0.5 g/L). The range of surface tensions used is shown in Table 1. The surface tensions were measured by the pendant drop method.

The Reynolds, Galileo and Eötvös numbers were calculated using the formulas consistent with Part I of this paper

$$Re_\alpha^* = \frac{\rho_\alpha u_\alpha d_p \epsilon}{\mu_\alpha (1-\epsilon)} \qquad \alpha = \beta, \gamma$$

$$Ga_\alpha^* = \frac{\rho_\alpha^2 g d_p^3}{\mu_\alpha^2} \frac{\epsilon^3}{(1-\epsilon)^3}$$

$$E\ddot{o}^* = \frac{\rho_\beta g d_p \epsilon}{\sigma(1-\epsilon)}$$

The interstitial velocities for the gas and liquid were calculated directly from the volumetric flow rate of each phase, the column area A, and the porosity of the bed

$$u_\alpha = \frac{Q_\alpha}{A\epsilon} \qquad \alpha = \beta, \gamma$$

The measurements of dynamic holdup ϵ_β under no-gas flow conditions were used to compute k_β, the relative permeability of the liquid phase according to Eq. 1 of Part I with the gas phase Reynolds number set equal to zero

$$k_\beta = A \frac{Re_\beta^*}{Ga_\beta^*} + B \frac{Re_\beta^{*2}}{Ga_\beta^*}$$

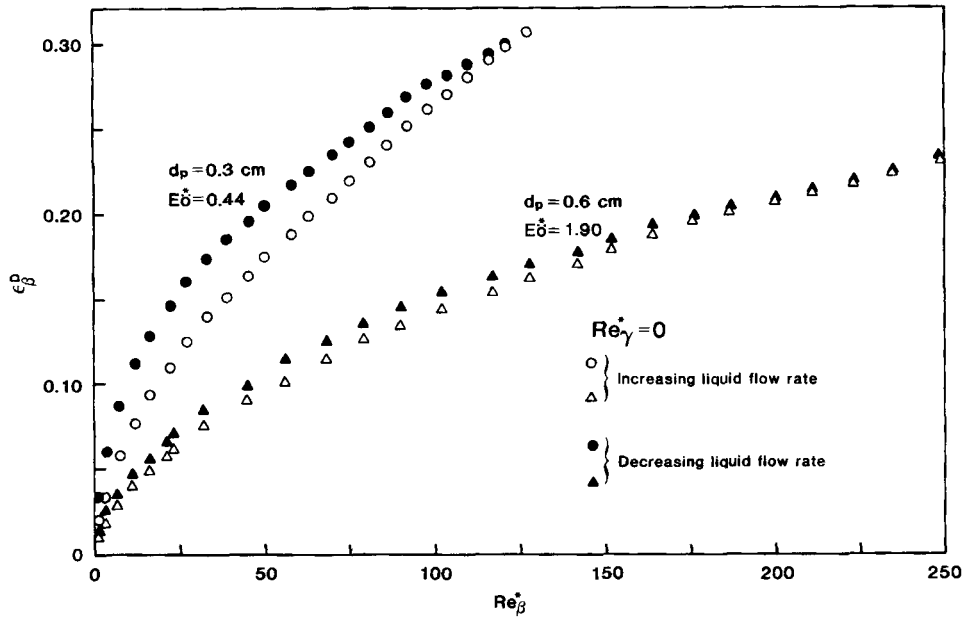

Figure 3. Experimental dynamic holdup for stagnant gas phase operations, air-water system.

Values of $A = 180$ and $B = 1.8$ were used (Macdonald et al., 1979). For each value of Re_β^* there is a different k_β value at a different reduced saturation δ_β. From measured dimensionless pressure drops in the gas phase under two-phase flow conditions one can calculate k_γ, the relative permeability for the gas phase, according to Eq. 4 of Part I

$$k_\gamma = \frac{1}{\psi_\gamma}\left[A\frac{Re_\gamma^*}{Ga_\gamma^*} + B\frac{Re_\gamma^{*2}}{Ga_\gamma^*}\right]$$

For each different pair of gas and liquid flow rates there is one measured dynamic holdup and a measured pressure drop. Since k_γ is calculated from the equation above, a plot of the calculated k_γ as a function of the measured gas phase saturation S_γ is easily generated.

Results and Discussion

Stagnant gas phase

Experimental results obtained for the case of the stagnant gas phase operation are presented in Figures 3 and 4. The dynamic holdup is shown as a function of the Reynolds and Galileo numbers of the liquid phase, for two different surface tensions. A multiplicity of hydrodynamic states is evident from these results. This hysteresis is more noticeable for the case of small particles ($d_p = 0.3$ cm). For a given particle diameter and a

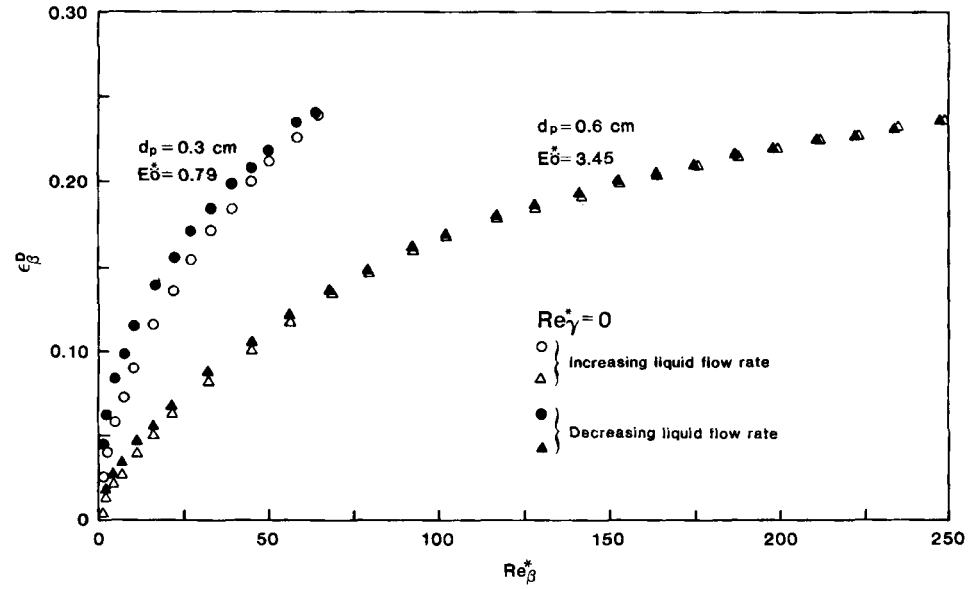

Figure 4. Experimental dynamic holdup for stagnant gas phase operations, air-water-surfactant system.

given surface tension, the dynamic holdup increases as the liquid flow rate is increased (lower curve of the loops). When the liquid flow rate is decreased (upper curve), the dynamic holdup takes larger values. If the mode of operation is changed at an intermediate liquid flow rate, the decreasing liquid flow rate curve would take values within the loop. Hence, the curves shown represent an envelope of all the possible hydrodynamic states. The hysteresis pattern under stagnant gas phase conditions has not been reported before and it leads to very interesting implications. First of all, hysteresis in gas-liquid flow operations has been explained (Kan and Greenfield, 1978) as being the result of the distortion induced in the gas-liquid interface shapes by the flow of gas. The fact that the phenomenon is present under stagnant gas conditions implies that the mechanism producing the hysteresis pattern is not completely explained by that hypothesis.

The trends observed in this investigation lead to the conclusion that the multiplicity of hydrodynamic states is a consequence of imperfect wetting of the packing. When the bed is preflooded and then drained, dry regions are formed locally and the liquid remaining in the bed (static holdup) is retained by means of a counterbalance of forces that includes gravitational forces and interfacial tension forces at the gas-liquid interface and at the three-phase contact lines. As the liquid flow rate is increased from this static situation, a liquid flow pattern is established in which dry regions still remain. The increase in the liquid flow rate induces a larger accumulation of liquid in the wet regions, which results in a larger force exerted by the liquid at the three-phase contact line. Eventually, the wetted area starts to increase until perfect wetting conditions are reached. When the liquid flow rate is decreased, the process described above is reversed only in a qualitative manner. The difference between both modes of operation lies in the fact that in the increasing flow rate mode the three-phase contact line is advancing over a dry surface, whereas in the decreasing flow rate mode the contact line is receding from a wet surface. It is a well-known fact (Adamson, 1976, Ch. VII) that the contact angle of a three-phase system is larger when the contact line advances than when it recedes. A larger contact angle means that a larger force has to be exerted in order to cause a change in the location of the three-phase contact line. In terms of the process under consideration, this implies that a larger fraction of the bed will remain dry in the increasing flow rate mode than in the decreasing flow rate mode at the same operating conditions. Hence, the decreasing flow rate mode provides more efficient conditions in terms of liquid distribution and therefore it yields larger holdups, which is precisely the trend observed in Figures 3 and 4.

Another interesting observation gathered from the results presented in Figures 3 and 4 is that for a given particle size and a given liquid phase Reynolds number, the dynamic holdup increases as the surface tension is decreased. This trend was also observed by Jesser and Elgin (1943) in experiments with water and surfactants, and by Charpentier and Favier (1975) in experiments involving water and organic liquids. This experimental trend is completely opposite to the predictions of the periodically constricted tube model developed in Part I of this work. It can be observed in Figure 13 of Part I that, at a given value of Re_β^*/Ga_β^*, a decrease in surface tension (increase in Eötvös number) causes a decrease in the liquid holdup. This fact provides another indication that the wetting of the packing has a very important effect on the amount of liquid retained in the bed. When the surface tension decreases, the contact angle at the three-phase contact line decreases (Adamson, 1976) causing more wetting in the bed and, therefore, an increase in the liquid holdup. The effect of surface tension on the holdup is stronger in the increasing liquid flow rate mode. When the surface tension is increased (see Figures 3 and 4), the holdups corresponding to decreasing flow rate mode increase slightly, whereas those corresponding to increasing flow rate mode increase appreciably, to the point that the hysteresis disappears for the 0.6 cm particles. This effect is consistent with the hypothesis made before that the degree of wetting controls to a large extent the amount of liquid retained. The increasing flow rate mode exhibits more extended dry regions and thus it is more sensitive to changes in the surface tension.

As discussed in Part I, the prediction of liquid holdups depends on knowledge of the liquid phase relative permeability, k_β, as a function of the reduced saturation of the liquid phase, δ_β. Under no-gas flow conditions, Eq. 1 of Part I reduces to

$$k_\beta = A\frac{Re_\beta^*}{Ga_\beta^*} + B\frac{Re_\beta^{*2}}{Ga_\beta^*} \tag{1}$$

The liquid phase relative permeabilities corresponding to the experiments performed in this investigation are presented in Figure 5 for the increasing liquid flow rate mode, and in Figure 6 for the decreasing liquid flow rate mode. Values for the Ergun constants of $A = 180$ and $B = 1.8$ were used, following the recommendations of Macdonald et al. (1979). The static holdup, required to calculate δ_β (see Eq. 5, Part I), was measured experimentally and a single value of 0.022 was obtained (Table 1), independent of the Eötvös number. In the range of Eötvös numbers considered (see Table 3) the correlation developed by Sáez and Carbonell (1985a) predicts values greater than 0.04, which is 80% larger than the experimental observation.

The increasing flow rate mode gives relative permeabilities that change somewhat with the Eötvös number without showing a definite trend. The variations of k_β are within 0.1 unit for a

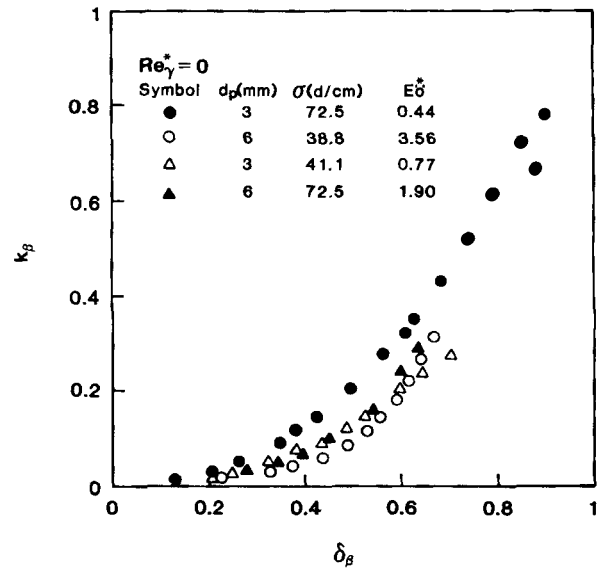

Figure 5. Liquid phase relative permeabilities, increasing liquid flow rate mode.

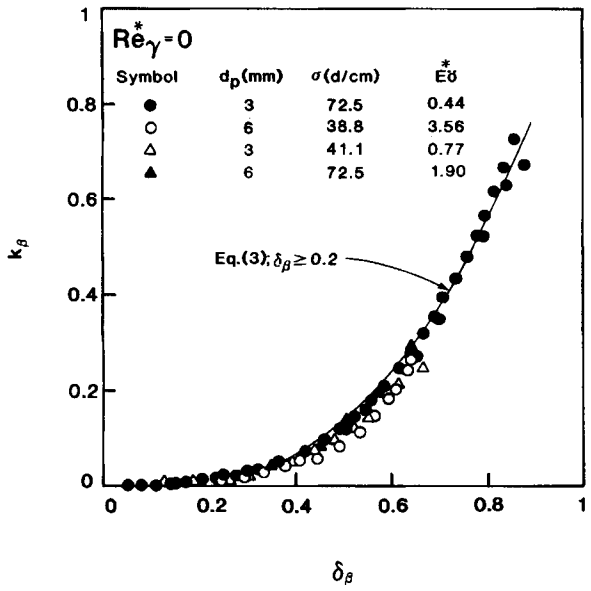

Figure 6. Liquid phase relative permeabilities, decreasing liquid flow rate mode.

change of almost an order of magnitude in the Eötvös number, so that they are not highly sensitive to this parameter for practical purposes. The relative permeabilities corresponding to the decreasing flow rate mode show a complete insensitivity to the Eötvös and Galileo numbers, Figures 6 and 7.

The liquid phase relative permeability for the increasing flow rate mode and water as working fluid can be expressed as a function of the reduced saturation by

$$k_\beta = \delta_\beta^{2.0} \tag{2}$$

For the decreasing flow rate mode, k_β is accurately represented by

$$k_\beta = \begin{cases} \delta_\beta^{2.9}, & \delta_\beta \geq 0.2 \\ 0.25\,\delta_\beta^{2.0}, & \delta_\beta < 0.2 \end{cases} \tag{3}$$

Equation 3 holds for all the experiments carried out in the present work in the decreasing flow rate mode.

These representations for k_β are shown in Figure 7, along with experimental data obtained in this investigation. Two additional experimental points, obtained by Wijffels et al. (1974) in the decreasing flow rate operating mode, are also shown. Note that Eq. 3 is a good representation of data spanning three orders of magnitude of the Galileo and Eötvös numbers.

In a previous work, Sáez and Carbonell (1985) obtained a relation between k_β and δ_β based on data taken from the literature without discriminating between operating modes (which were often not reported). The result (Eq. 7, Part I) was a power fit with an exponent of 2.43. Note that this value coincides with the average value of the exponents of Eqs. 2 and 3, indicating that the data used by Sáez and Carbonell probably corresponded to both modes of operation.

The recognition of the hysteresis phenomenon implies that one should use different correlations for different modes of operation. The decreasing flow rate mode is the more significant of the operating modes since it yields larger liquid retentions and

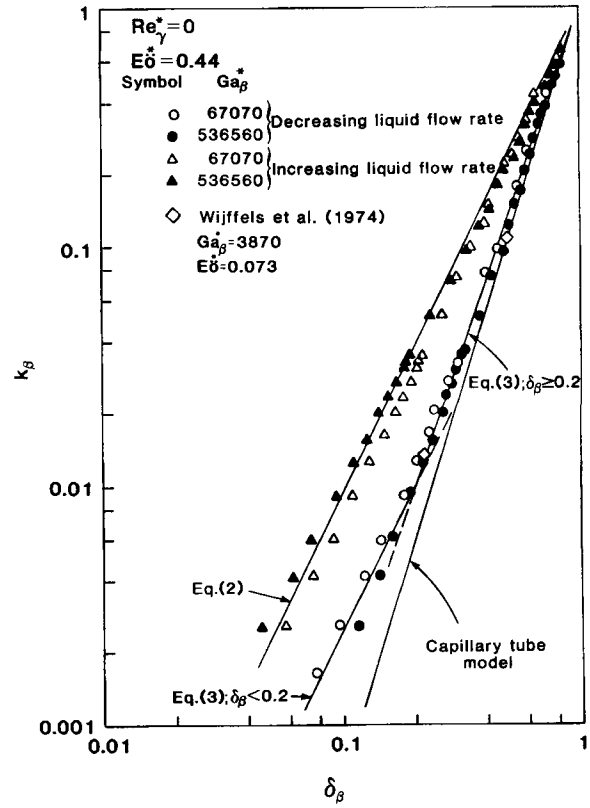

Figure 7. Liquid phase relative permeabilities, comparison between operating modes.

better wetting characteristics, which are desirable features for practical applications (Hofmann, 1977). By combining Eqs. 1 and 3 we find a correlation for predicting reduced saturations in this operating mode,

$$\delta_\beta = \begin{cases} \left(A\dfrac{Re^*_\beta}{Ga^*_\beta} + B\dfrac{Re^{*2}_\beta}{Ga^*_\beta}\right)^{0.35}, & \delta_\beta \geq 0.2 \\ 2\left(A\dfrac{Re^*_\beta}{Ga^*_\beta} + B\dfrac{Re^{*2}_\beta}{Ga^*_\beta}\right)^{0.50}, & \delta_\beta < 0.2 \end{cases} \tag{4}$$

This correlation is compared to the experimental data obtained in the present work in Figure 8. The prediction of reduced saturation by means of Eq. 4 yields a mean relative deviation of 3% when compared to 95 data points.

The periodically constricted tube model developed in Part I does not provide a good representation of the trends followed by the experimental data. The results of the model indicated that the relative permeability curves were sensitive to the Eötvös number (Figure 12, Part I), a fact that is not observed experimentally. One of the reasons for this discrepancy might be that the conduit models assumed perfect wetting of the packing surfaces, which is now always true in the real system. In Figure 7 the results of the straight capillary tube model are compared to experimental data. The capillary tube curve is very close to that of the periodically constricted tube when surface tension effects are absent ($E\ddot{o} \rightarrow \infty$). Notice that the model represents fairly well the data corresponding to the decreasing liquid flow rate operating mode. This is due to the fact that this mode of opera-

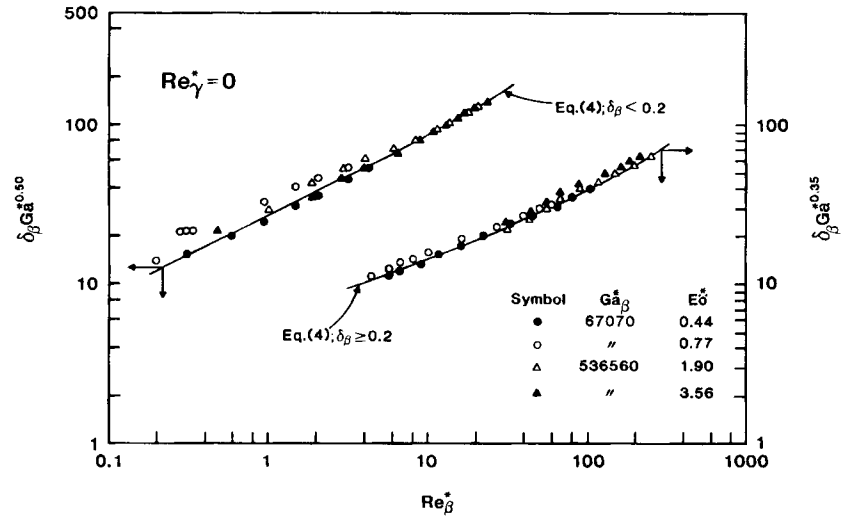

Figure 8. Prediction of liquid holdup under stagnant gas conditions, decreasing liquid flow rate mode.

tion yields the maximum wetting efficiency and the capillary tube model assumes perfect wetting. The differences between the model and the experimental data become more appreciable in the region of low liquid holdups. This can be ascribed to the absence of static holdup in the capillary tube model.

Gas-liquid flow

Representative results obtained in the present work for dynamic holdup and pressure drop in gas-liquid cocurrent flow are shown in Figures 9 and 10. The hysteresis phenomenon is also evident in these results. Differences of up to 30% were observed in the dynamic holdup depending on the mode of operation. The most dramatic effect of hysteresis is on pressure drops, Figure 10, which in some cases exhibit differences of almost an order of magnitude when the increasing and decreasing liquid flow rate modes are compared.

The decreasing liquid flow rate mode exhibits larger holdups, as was the case in the stagnant gas operation, Figure 9. Larger holdups mean more obstruction to the flow of gas, and this results in larger pressure drops, Figure 10.

The hysteresis exhibited by the process also affects the transition from the trickling to the pulsing flow regime. It was

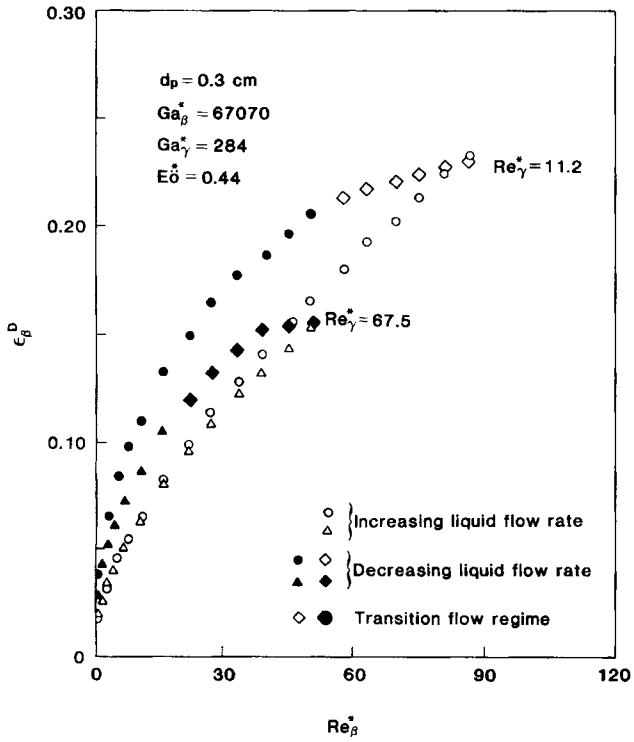

Figure 9. Experimental dynamic holdup for gas-liquid operation.

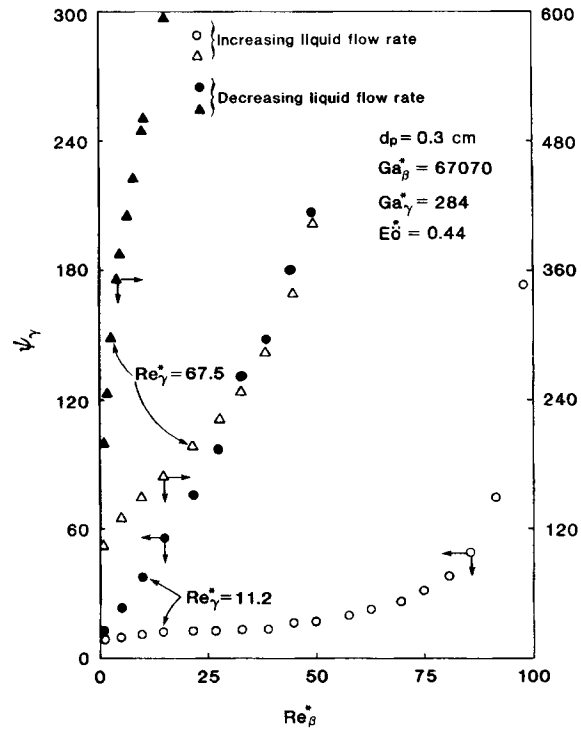

Figure 10. Experimental pressure drop for gas-liquid operation.

observed experimentally that the transition to pulsing flow at a given gas flow rate occurred at a larger liquid flow rate in the increasing flow rate mode, Figure 9. In other words, when pulsing flow is reached by increasing the liquid flow rate, the unstable pattern of flow persists as the liquid flow rate is decreased. This observation indicates that the presence of pulsing flow is dependent on the amount of liquid retained in the bed. Under the same operating conditions (gas and liquid flow rates), a larger liquid holdup increases the possibilities of the presence of the pulsing flow regime. Under pulsing flow conditions, the packing is perfectly wetted due to the motion through the bed of regions of high liquid content (pulses), and therefore hysteresis in the liquid holdup disappears, as observed in Figure 9 (the curves corresponding to the two operating modes coincide as the pulsing regime is reached). The pressure drop curves shown in Figure 10 do not form a closed loop because this parameter was not measured after transition to pulsing was observed.

The gas phase relative permeabilities can be calculated from the experimental data gathered on two-phase flow by means of the following equation (Part I, Eq. 4),

$$k_\gamma = \frac{1}{\psi_\gamma}\left\{A\frac{Re_\gamma^*}{Ga_\gamma^*} + B\frac{Re_\gamma^{*2}}{Ga_\gamma^*}\right\} \tag{5}$$

The results are shown in Figure 11 for the increasing liquid flow rate mode and in Figure 12 for the decreasing liquid flow rate mode. Only experimental data corresponding to particles of 0.3 cm dia. are shown in those plots. The results for particles of 0.6 cm dia. show similar trends. The multiplicity of hydrodynamic states affects the gas phase relative permeabilities too; the values of this parameter under similar conditions are appreciably larger in the increasing liquid flow rate mode.

The values of A and B, 180 and 1.8, respectively, were chosen according to the recommendations of Macdonald et al. (1979). These values are an average over a wide variety of porous media and, depending on the structure of the porous matrix, they can vary appreciably. For example, for beds packed with spheres A

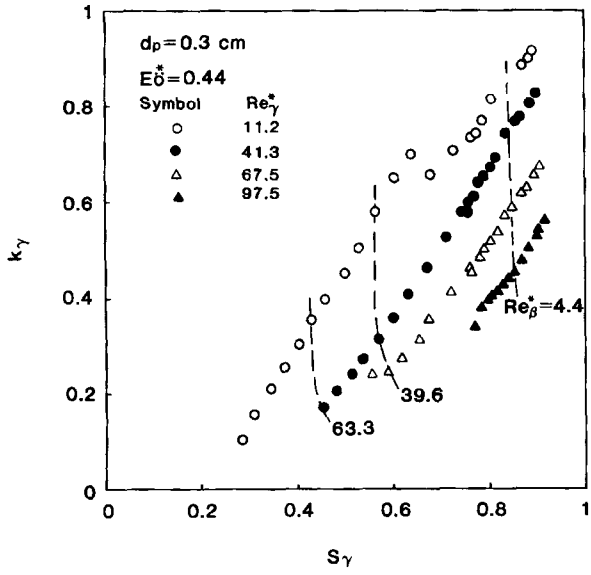

Figure 11. Gas phase relative permeabilities, increasing liquid flow rate mode.

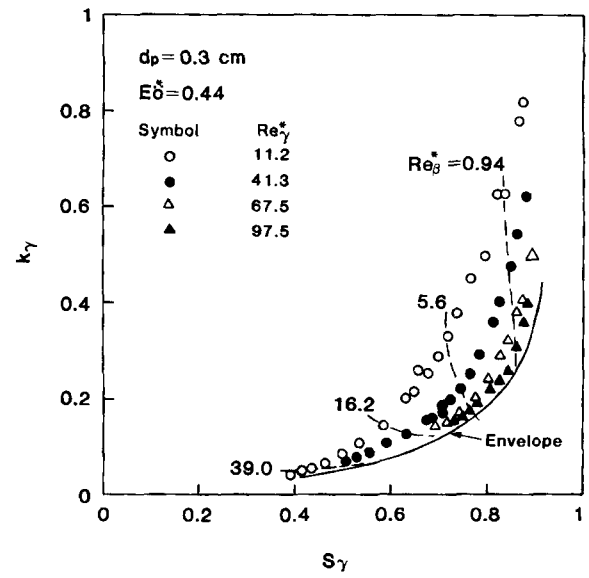

Figure 12. Gas phase relative permeabilities, decreasing liquid flow rate mode.

and B are reported (Macdonald et al.) to be in the neighborhood of 160 and 1.5, respectively, which would result in changes of about 10% in the calculated values of k_γ by using Eq. 5.

The experimental gas phase relative permeabilities show the same trends as those followed by the predictions of the straight and constricted tube models developed in Part I (see Figures 4 and 14, Part I) although the quantitative behavior is appreciably different. The curves at constant liquid phase Reynolds numbers are almost vertical lines for low values of the gas phase Reynolds number. The curves at constant gas phase Reynolds numbers approach a unique curve as the value of that parameter becomes large. This unique curve represents the envelope of the gas phase relative permeability curves. For the case of the decreasing liquid flow rate mode, the envelope was found by extrapolation to infinite gas Reynolds numbers. The envelope curve is well represented by the equation

$$k_\gamma^{(ENV)} = 0.045 + 0.67\,S_\gamma^7, \quad 0.4 < S_\gamma < 0.9 \tag{6}$$

This curve, represented as a solid line in Figure 12, is considerably lower than the envelopes predicted by the conduit models developed in Part I. It is interesting to point out that even though in this work we could not obtain data exactly on the envelope curve, Eq. 6 represents well the extrapolated envelope of the data corresponding to particles of both 0.3 cm and 0.6 cm dia., indicating that this envelope is independent of the Galileo numbers of both phases, as it was in the conduit models.

The theoretical models and the experimental data indicate that a single curve is not a very adequate representation of the gas phase relative permeability relations. For each particle size used, and hence for each Galileo and Eötvös number, k_γ was correlated to the gas phase saturation by means of an equation of the form

$$k_\gamma = S_\gamma^n \tag{7}$$

where the exponent n is a function of the gas phase Reynolds number. The values of n that minimize the mean relative devia-

Table 2. Exponent of Gas Phase Relative Permeability Relations, Decreasing Liquid Flow Rate Mode

$d_p = 0.3$ cm		$d_p = 0.6$ cm	
Re_γ^*	n	Re_γ^*	n
11.3	3.6	22.9	5.3
41.3	4.9	83.9	6.2
67.5	6.0	137.3	6.9
97.5	6.8	198.4	7.5

tion between the experimental and calculated values of k_γ are presented in Table 2. If a fit similar to Eq. 7 were performed to the results obtained with the conduit models, then n would be a function only of Re_γ^*/Ga_γ^* (Part I). However, in the real case, n seems to correlate better with the parameter Re_γ^*, as shown in Figure 13. This observation is not conclusive since just two values of the gas phase Galileo number were investigated, but it provides a good representation if Ga_γ^* is in the range used in the present work.

The knowledge of n as a function of Re_γ^* allows the development of a correlation for calculating pressure drops and liquid holdups under two-phase flow conditions. The liquid holdup is calculated by means of Eq. 3, Part I,

$$\frac{1}{k_\beta}\left\{A\frac{Re_\beta^*}{Ga_\beta^*}+B\frac{Re_\beta^{*2}}{Ga_\beta^*}\right\}-\frac{1}{k_\gamma}\left\{A\frac{Re_\gamma^*}{Ga_\gamma^*}+B\frac{Re_\gamma^{*2}}{Ga_\gamma^*}\right\}\frac{\rho_\gamma}{\rho_\beta}=1 \quad (8)$$

where k_β is given by Eq. 3 and k_γ is obtained from Eq. 7 with the appropriate value of n, which can be estimated from Figure 13 or Table 2. After the liquid holdup and hence δ_β and S_γ have been calculated, the dimensionless pressure drop is estimated by

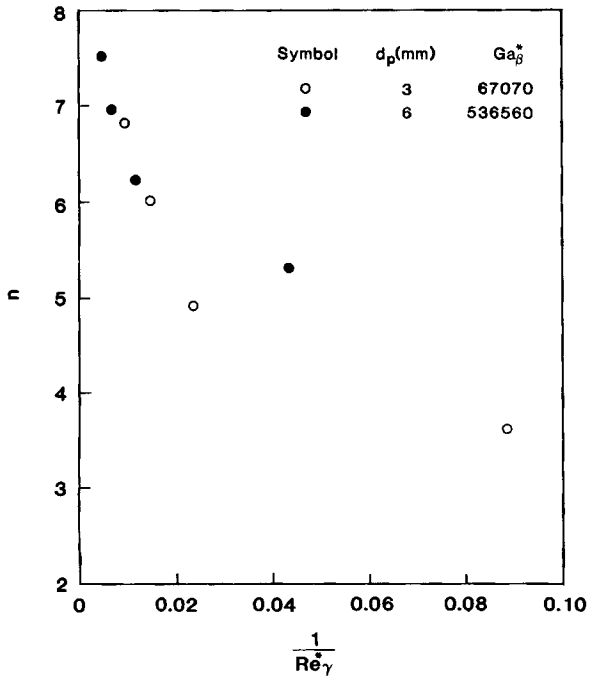

Figure 13. Gas phase relative permeability exponent, decreasing liquid flow rate mode.

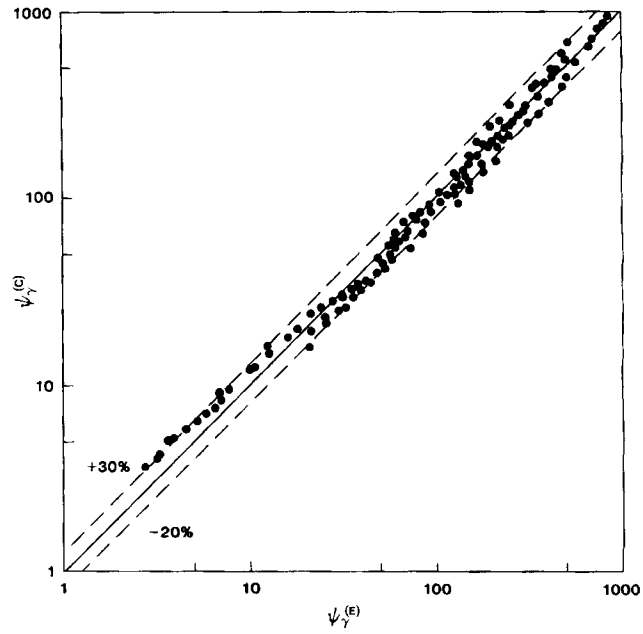

Figure 14. Comparison of predicted and experimental pressure drops.

means of Eq. 4, Part I,

$$\psi_\gamma=\frac{1}{k_\gamma}\left\{A\frac{Re_\gamma^*}{Ga_\gamma^*}+B\frac{Re_\gamma^{*2}}{Ga_\gamma^*}\right\} \quad (9)$$

The calculated values of the pressure drop are compared in Figure 14 to the experimental values. The experimental data used in this analysis were obtained in the ranges reported in Table 3. The correlation gives a mean relative deviation of 13% in the prediction of pressure drops for 137 experimental data points. The predictions of liquid saturations for the same set of experimental data are shown in Figure 15. The mean relative deviation was 3.5%. The correlation was not compared to experimental data in the literature due to the fact that the operating mode is not usually reported in previous works. Further experimental analyses are required in order to establish the dependence of the gas phase relative permeability curves on the independent variables. In any case, the correlation proposed in this work represents an improvement over previous studies.

In the first attempt to apply the relative permeability analysis to trickling flow in packed beds, Sáez and Carbonell (1985) represented the gas phase relative permeability by means of a unique curve, equivalent to Eq. 7 with an exponent $n = 4.8$. That work did not discriminate between different modes of operation nor did it consider possible changes in k_γ with the gas phase

Table 3. Range of Independent Dimensionless Variables in Present Study

$d_p = 0.3$ cm	$d_p = 0.6$ cm
$Ga_\beta^* = 67{,}070$	$Ga_\beta^* = 2{,}272$
$Ga_\gamma^* = 284$	$Ga_\gamma^* = 536{,}560$
$Re_\beta^* = 0.3$–50	$Re_\beta^* = 0.5$–130
$Re_\gamma^* = 10$–100	$Re_\gamma^* = 20$–200
$Eö^* = 0.44$–0.77	$Eö^* = 1.90$–3.56

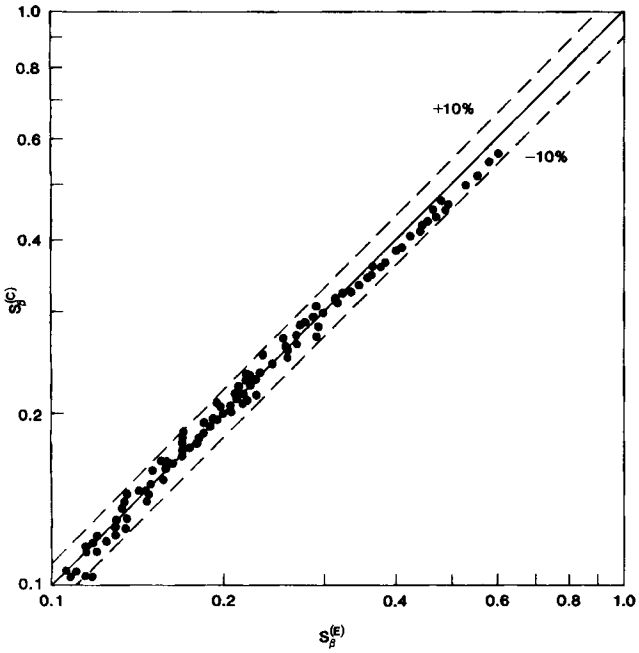

Figure 15. Comparison of predicted and experimental liquid saturations.

Reynolds number. That first correlation gave mean relative deviations of 13% in the prediction of liquid holdups and 22% in the prediction of pressure drops, values that are higher than those obtained with the new correlation. We must point out, however, that the new correlation has only been tested against the data used to develop it and this causes an obvious bias in the comparison. If the primitive correlation proposed by Sáez and Carbonell is used to predict the pressure drops and holdups obtained in this experimental study, mean relative deviations of 13% for the liquid holdup and 32% for the pressure drop are obtained. These values are higher than those given by the new correlation, as expected, but they are consistent with the errors obtained previously. These results confirm that the first correlation was statistically significant, so that it can be used with the confidence predicted by the errors reported. For practical purposes, the new correlation represents a quantitative improvement and a better representation of the physics of the process.

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Notation

A = area of the bed
A = constant in the viscous term of the Ergun equation
B = constant in the inertial term of the Ergun equation
d'_h = hydraulic diameter, $= d_p\,\epsilon/(1-\epsilon)$
d_p = sphere diameter
$E\ddot{o}^*$ = Eötvös number based on the hydraulic diameter, $\rho_\beta g d'^2_h/\sigma$
Ga^*_α = Galileo number of the α phase based on the hydraulic diameter, $\rho^2_\alpha g\, d^3_p\, \epsilon^3/\mu^2_\alpha (1-\epsilon)^3$
n = exponent in the gas phase relative permeability relation, Eq. 7
k_α = relative permeability of the α phase
Re^*_α = Reynolds number of the α phase based on the hydraulic diameter, $\rho_\alpha u_\alpha d_p \epsilon/\mu_\alpha(1-\epsilon)$
S_α = saturation of the α phase, ϵ_α/ϵ
u_α = interstitial velocity of the α phase, $Q_\alpha/A\epsilon$

Greek letters

δ_β = reduced saturation of the liquid phase, $S_\beta - \epsilon^0_\beta/\epsilon$
ϵ = bed porosity
ϵ_β = liquid holdup per unit volume of bed
ϵ^0_β = static liquid holdup
ϵ^D_β = dynamic liquid holdup, $= \epsilon_\beta - \epsilon^0_\beta$
ρ = density
σ = surface tension
ψ_α = dimensionless pressure drop of the α phase per unit length, $1 - (\Delta \bar{P}_\alpha/\rho_\alpha g \ell)$

Subscripts and superscripts

(C) = calculated
(E) = experimental
(ENV) = envelope
β = liquid phase
γ = gas phase

Literature Cited

Adamson, A. W., *Physical Chemistry of Surfaces,* Wiley, New York, 3rd ed., Ch. VII (1976).

Charpentier, J. C., et al., "Study of Liquid Retention in Cocurrent and Countercurrent Gas-Liquid Flow in a Packed Bed," *Chimie et Ind. Génie Chim.*, **99**, 803 (1968).

Charpentier, J. C., and M. Favier, "Some Liquid Holdup Experimental Data in Trickle-Bed Reactors for Foaming and Non-Foaming Hydrocarbons," *AIChE J.*, **21**, 1,213 (1975).

Hofmann, H., "Hydrodynamics, Transport Phenomena, and Mathematical Models in Trickle-Bed Reactors," *Int. Chem. Eng.*, **17**, 19 (1977).

Hutton, B. E. T., and L. S. Leung, "Cocurrent Gas-Liquid Flow in Packed Columns," *Chem. Eng. Sci.*, **29**, 1,681 (1974).

Jesser, B. W., and J. C. Elgin, "Studies of Liquid Holdup in Packed Towers," *Trans. Inst. Chem. Engrs.*, **39**, 277 (1943).

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

———, "Pressure Drop and Holdup in Two-Phase Cocurrent Trickle Flows through Beds of Small Packings," *Ind. Eng. Chem. Proc. Des. Dev.*, **18**, 740 (1979).

Larkins, R. P., "Two-Phase Cocurrent Flow in Packed Beds," Ph.D. Thesis, Univ. of Michigan (1959).

McDonald, I. F., et al., "Flow through Porous Media—The Ergun Equation Deviated," *Ind. Eng. Chem. Fund.*, **18**, 199 (1979).

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).

Morsi, B. I., et al., "Hydrodynamics and Gas-Liquid-Solid Interfacial Parameters of Cocurrent Downward Two-Phase Flow in Trickle-Bed Reactors," *Chem. Eng. Commun.*, **25**, 267 (1984).

Otake, T., and K. Okada, "Liquid Holdup in Packed Towers—Operating Holdup without Gas Flow," *Kagaku Kogaku*, **17**, 176 (1953).

Reiss, L. P., "Cocurrent Gas-Liquid Contacting in Packed Columns," *Ind. Eng. Chem. Proc. Des. Dev.*, **6**, 486 (1967).

Sáez, A. E., and R. G. Carbonell, "Hydrodynamic Parameters for Gas-Liquid Cocurrent Flow in Packed Beds," *AIChE J.*, **31**(1), 52 (1985).

Sáez, A. E., R. G. Carbonell, and J. Levec, "The Hydrodynamics of Trickling Flow in Packed Beds. I: Conduit Models," *AIChE J.*, **32**(3), 353 (1986).

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

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

Turpin, J. L., and R. L. Huntington, "Prediction of Pressure Drop for Two-Phase, Two-Component Concurrent Flow in Packed Beds," *AIChE J.*, **13**, 1,196 (1967).
Weekman, V. W., "Heat Transfer and Fluid Flow for Concurrent, Gas-Liquid Flow in Packed Beds," Ph.D. Thesis, Purdue Univ. (1963).
Wijffels, J. B., J. Verloop, and F. J. Zuiderweg, "Wetting of Catalyst Particles under Trickle Flow Conditions," *Chem. Reaction Eng.* **II**, *ACS Monograph Ser.* **133**, 151 (1974).

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