LOADING AND DRAINING OF PERIODICALLY OPERATED TRICKLE BED REACTOR

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ABSTRACT

Periodic interruption of liquid flow in cocurrent trickle beds appears to be an attractive mode of operation. For modelling these intermittent flow reactors, loading and draining times must be known. Experiments were undertaken using beds of activated carbon with water and air as the fluid phases. Loading time was taken as water breaktrough. The gas flow was continuous while the time between the end of drainage and the start of filling was varied to simulate different periods. Drainage experiments followed the liquid flow leaving the bed as a function of time. Liquid hold ups were determined after the filling and draining measurements. Variable considered were particle size, gas and liquid velocities. Loading closely follows the plug flow model; drainage shows tailing but does not follow literature models. Static and dynamic hold ups at zero gas flow agree with literature correlations for the larger particle size used. A gas velocity effect on both static and dynamic hold up was observed.

INTRODUCTION

Haure et al.(1989) found that periodic interruption of liquid flow in cocurrent trickle beds increased the rate of an exothermic catalytic reaction in which both reactant enter the reactor in the gas phase. Because the increase is about 40 to 50%, this type of periodic operation merits more detailed study. Periodic flow interruption was considered for the scrubbing of SO_2 from stack gas (Haure et al., 1989) because sulfur can be recovered as a moderately concentrated sulfuric acid. A model for flow interruption has been proposed that neglects the time needed to fill the bed when liquid is readmitted and the time to drain the bed when the flow is closed off (Haure et al., 1990; Stegasov et al., 1992). It is evident that filling and draining limit how short a cycle can be in operations with flow interruption. A further question that arose out of model development was how large should the hold up be when (i) only gas flows through the bed and (ii) both gas and

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liquid flow through it. In other words, whether periodic operation influence the hold up was questioned.

A literature survey encompassing the last 20 years failed to disclose any research on the filling time required to establish hydrodynamically steady flow through the column. Also, the character of the flow in the bed during the filling phase seems not to have been investigated. On the other hand, there is a rich literature on gravity drainage from packed beds. Unfortunately, this literature deals primary with drainage from filter cake or centrifuge solids, the particle size of which is often more than two orders of magnitude smaller than trickle bed packing and thus capillary forces prevail. However, gas flow was either absent or countercurrent to the liquid flow. Cocurrent gas flow was treated by Wakeman (1979, 1982). Therefore, it seemed to be useful to study the drainage behaviour experimentally. The literature was helpful for interprating the data obtained. The question of hold up in periodic flow stoppage, of course, had to be answered experimentally.

Objectives of this study were thus 1) to measure the filling time, defined as the time after flow stars to when liquid begins to fall from the bottom of the trickle bed, 2) to determine the flow character during filling, 3) to measure the rate of drainage as a function of time, and 4) to measure the liquid hold up when both gas and liquid flow cocurrently through the bed (referred to as static hold up). In addition, the influence of particle size at to levels, gas velocity at three levels and liquid velocity and water saturation of the air at two levels on the above group of measurements were examined.

Experimental Equipment

The schematic diagram is shown in Figure 1. Because the experiments were carried out in support of experimental and simulation studies of periodic interruption, trickle bed dimensions, packing materials used and particle size, and gas and liquid velocities were chosen to mirror earlier periodic operation studies. Filling and draining behaviour were observed using a 3.6 cm cylindrical trickle bed. The trickle bed wall was lucite, so that the filling and the draining flows could be observed. Granular active carbon with 0.8 and 3.5 mm particle sizes was used as packing. The bed was 40.4 cm high and supported by a fine stainless steel screen. The gas phase was saturated air, which wets activated carbon.

Each activated carbon charge was dried and weighed. A stopwatch with a resolution of 0.1 s was used. Filling experiments were begun by passing air through the bed; then at time zero, liquid was fed to the bed. Air flow was continuous. The time was recorded when water first fell from the bottom of the bed. Flow thereafter was collected in a narrow graduated cylinder and the volume change with time determined.

The drainage rate was monitored by accumulating the liquid leaving the bed in graduated cylinder and noting the increase in volume with time until liquid flow from the bed ceased. The volum collected during the drainage experiment measured the dynamic hold up, defined as the fraction of the interparticle void volume filled with liquid during liquid flow through the bed less the static hold up.

Static hold up was determined by weighing the bed after drainage was complete and substracting the original weight of dry bed. Void volum can be calculated from the weight of carbon and the porosity of the BPL activated carbon.

Filling Behaviour

Figure 2 shows the filling time per meter of activated carbon vs. the superficial liquid velocity, i.e., the volumetric rate of water feed divided by the bed cross sectional area. Other parameters in the figure are the carbon mean diameter and the superficial air velocity through the bed. The filling time is the time for the first liquid to drop from the bottom of the bed after water flow to the bed is started. The figure shows that this filling time is independent of the carbon particle size and the gas velocity.

Water leaving the bed was collected and measured as a function of time. Figure 3 shows that flow leaving the bed is a linear function of time and is identical to the water feed rate. This means that for the bed dimensions and flow rate used, liquid moved through the bed in plug flow. Within the limits of measurement accuracy, fingering does not occur. The plug flow character was observed in all experiments including those at higher gas velocities and experiments in which liquid was retained in the bed from previous filling and draining steps.

Figure 4 compares filling times for three different filling conditions. The bottom group of curves represents filling times when water is retained in the bed. These measurements were intended to simulate the technique of periodic flow interruption investigated by Haure et al. (1989, 1990). The gas flow is continuous in this operation so that after the liquid flow is interrupted, leaving residual water in the porous carbon particles and at particle interstices, evaporation commences. Evaporation removes this hold up and thus the filing times increases. The difference in filling times is discernible at low liquid velocities (i.e., between 5 min of flow interruption and 60 min), but at velocities 6 to 8 mm/s or greater, it cannot be measured. Data were obtained for only the 3.6 cm diameter particles.

The uppermost line in Figure 4 and the accompanying data points provide filling times for a dry bed of carbon. The line and the data points for the 3.6 cm particles were taken from Figure 2. It is evident from a comparison of the dashed line and the data points for periodic flow interruption (lower cluster of lines) that even after 60 min of flow interruption, the carbon bed contains almost all of the liquid retained from the previous dousing.

The solid line in Figure 3 above the dashed line for filling of the dry bed is the calculated line, assuming liquid plug flow and no gas flow in the bed. It represents the maximum filling time. The difference between the two lines is the gas hold up in the bed.

Filling times for any bed depth can be estimated from Figure 4. They can also be obtained by deviding the volumetric liquid flow rate into the product of the difference between the total hold up under liquid flow and the total hold up with just gas flow through the bed times the bed void volume. In terms of dynamic and static hold up:

$$t_f = (\beta_d - \beta_s)\varepsilon V_b/Q_l \qquad (1)$$

Equation (1) is simply the plug flow model for bed filling.

Draining Behaviour

All the experimental data collected are shown in Figure 4 as the liquid collected vs. time after the water flow was halted. Draining is a slower and much more complex process. Figure 5a shows the draining behaviour when the gas flow has also been terminated. This is typical of gravity flow data in the literature (Wakeman and Vince, 1986) and shows a particle size effect that would be expected for laminar flow in the deraining bed. In contrast to published data, there is an effect of the water flow rate just prior to flow interruption. This and the effect of the gas superficial velocity are examined in Figure 5b and 5c for the 0.8 mm and 3.6 mm particle respectively. Compa- rison of the curves for V_l = 5.1 mm/s and 13 mm/s at V_g = 0 illustrated the water flow- rate effect. This effect arises from dynamic hold up that is a function of the liquid loading. The effect of gas velocity can be seen by comparing the curves for V_g = 0 and V_g = 80 mm/s for V_l = 13 mm/s in Figure 5b. Virtually the same behaviour can be seen in Figure 5c for the 3.6 mm particles using the curves for V_g = 0.80 mm/s and V_l = 8.4 and 13 mm/s.

Although drainage from beds in the presence of gas flow has been both measured and analyzed (Wakeman, 1979, 1982), conditions in this study differ because gas flow occurred prior to drainage and continues at the same velocity during drainage. In Wakeman's studies, gas flow begins with drainage and pressure drop rather than velocity is maintained constant.

Despite system differences, one would expect the same physical influences to be important. In Wakeman's studies, the knee of the drainage curve results from two processes: the dropping liquid head in the bed as drainage proceeds followed by a slower process of film or interstitial drainage from the particles (Wakeman and Vince, 1986). A sophisticated analysis of the V_g = 0 drainage is given by Wakeman and Vince, who allow for capillary pressure and for the effect of liquid retention in the bed on the Darcy's Law permeability. This leads to liquid retention or static hold up that varies with bed depth. The hold up is very small at the upper surface of the bed and increases slowly with depth before rising rapidly to almost the pre drainage hold up at the bottom of the bed. The Wakeman analysis assumes β = 1 at t = 0 and that only gravity acts on the liquid. Although the analysis could be modified for β < 1 and gas phase drag (following Wakeman, 1982) a numerical solution is necessary; thus, the analysis leads to a drainage chart specific to bed and liquid properties.

For purposes of modelling periodic liquid flow interruption, or determining the shortest practicle cycle periods, the detail provided by the Wakeman and Vince treatment is unnecessary and generalized drainage charts are inconvenient. Thus, for the analysis of the data in Figure 5, an analysis presented by Nenniger and Storrow (1958) has been adapted. Only liquid external to the porous carbon particles is considered. This is justified by a mean pore diameter in the carbon of about 2 nm. Water wets carbon

preferentially so that the particle interior is non draining. The hold up is assumed to be uniform through the bed after the drainage is complete. Following Nenniger and Storrow, this static hold up is associated with capillary pressure that can be expressed as liquid height, h_c. If h_l is the momentary liquid height in the bed, Darcy's law for liquid drainage through the bed of solids can be written as

$$-dQ/dt = \{(h_l - h_c)/h_l\}(\rho_l k A g/\mu_l) \qquad (2)$$

The dynamic-plus static hold up in the presence of liquid flow is the hold up at t = 0 and is assumed independent of axial position. Thus, at any time t > 0

$$Q(t) = (h\beta_s + h_l\beta_d)A\varepsilon \qquad (3)$$

where Q(t) is the external liquid remaining in the bed. Eliminating h_l from Eq. (2), integrating this expression from Q_o=$h(\beta_s+\beta_d)A\varepsilon$ at t =0 to Q(t) at t, substituting in the final liquid volum Q_∞=$(h-h_l)\beta_s A\varepsilon + h_l\beta_d A\varepsilon$ at t = ∞ and Q_l=$h_c\beta_d A\varepsilon$, and letting Q_d=volume of liquid draineded from the bed yields

$$\rho_l k A g\, t/\mu_l = Q_d + \beta_d Q_c \ln\{(Q_d)_\infty/((Q_d)_\infty - Q_d)\} \qquad (4)$$

Equations (4) have two parameters that must be evaluated from experimental data:k and Q_c. In Eq. (4), Q_d and $(Q_d)_\infty$ can be read from the drainage curve so both k and Q_c should be calculable. Indeed, if h>>h_c, then k can be calculated from the slope of the drainage curves at t = 0.

When this model was tested against the data shown in Figure 4, Q_c assumed a negative value. Therefore, it is concluded that the analysis of Wakeman and the simpler analysis of Nenniger and Storrow are not applicable to this situation. These authors deal with a bed of solids supported on a screen. For this reason, the capillary pressure expressed as a liquid height must be negligible. Indeed, the knee shown in Figure 5 is probably due to drop formation beneath the screen supporting the packed bed as the liquid drains from the bed.

Because the amount of liquid drained after the falling head approaches the screen is quite small, this amount is neglected and it is assumed that the draining time can be approximated by the time it takes for the liquid head to movethrough the bed. This neglects the logarithmic term on the right hand side. The draining behaviour can be described by means of the Darcy coefficient k, now. This was calculated from the first two measurements of the drainage collected within one minute of interruption of liquid flow.

Figure 6 shows values of the Darcy coefficient k as a function of the superficial gas velocity for the 0.8 mm and 3.6 mm carbon particles.Individual data points are labelled with the liquid velocity. It is apparent that particle size has just a small effect on the Darcy coefficient at gas velocities above 22mm/s. However, the coefficient increases with the gas velocity as expected, up to 22 mm/s, indicating that the higher drag of the gas flow forces the greater volume of liquid through the packed bed. The Darcy coefficients of Figure 5 permit an estimate of the draining time for any bed depth for the particle sizes and velocity ranges used in the experiments.

Draining experiments were carried out using dry air and water saturated air, because it was thought possible that draining might require along enough time for evaporation to influence the drainage behaviour. Air saturation, however, did not effect drainage and in Figures 4 and 5 data for flows of saturated and dry air have been averaged. Points for different liquid velocities prior to flow interruption are shown in Figure 5. V_l is important when there is no air flow through the bed. The Darcy coefficient, k, increases with increasing V_l. As V_g increases, the effect of V_l on k decreases. Liquid velocity changes β_d so the influence suggest that $(\beta_s + \beta_d)\rho_l$ should be used in Eq. (2) in place of ρ.

Hold Up

Static hold up is the liquid held in the interstices of the trickle bed packing and retained in the bed by capillary forces when liquid flow goes to zero. It generally correlates with the Eötvös number, the ratio of gravitational and surface tension forces.

Figure 7 plots the static hold up as a function of bed volume vs. the Eötvös number. Because the Eötvös number is independent of liquid and gas velocities, results in this study are taken at Eö = 0.17 and 3.4. Most published data, including points from other studies shown in the figure, are at $V_g = 0$. Experimental values in Figure 7 at $V_g = 0$ scatter at Eö = 3.4. If the mean our data at each Eotvos number is used, the static hold up displays a higher dependence on this parameter than do published data.

With gas flowing through the bed, $\varepsilon\beta_s$ is about 50% lower at $d_p = 0.8$ mm. Experimental data at $d_p = 3.6$ mm are close to zero at $V_g = 80$ mm/s and fall off the plot. With larger particles, V_g has a large influence on B_s and the relationship given by the solid line in Figure 6 significantly over estimates the static hold up.

Dynamic hold up at zero gas velocity is plotted against the particle Reynold number based on liquid properties. The data for the 3.6 mm particles agrees closely with data presented by Wammes et al. (1991), whose data were collected for a particle size of 3±0.5 mm. The measurements for the smaller particles are about twice the dynamic hold up predicted by Wammes et al. (1991). This suggests that the correlation against particle Reynolds number is inadequate. Dynamic hold up was also measured as a function of gas velocity. For the larger particles, the influence of the gas was negligible, but with the 0.8 mm particles, an increase of about 10% in dynamic hold up with gas velocity was observed. An explanation would be that with no gas flow, liquid channelling occurs in the bed, the preferred path being that which offer the least resistance. Liquid flows by gravity without pressure drop. With cocurrent gas flow, the gas competes with the liquid for the preferred paths, causing redistribution of the liquid through less preferred paths. Pressure drop that now occurs provides the energy for this redistribution. Our data suggest that this phenomenon depends on bed porosity.

Wammes et al. (1991) also correlated their dynamic hold up data as the product of dynamic hold up and the Galileo number to the exponent 0.42. These experimental

results compared with the data of these authors in Figure 8. Now data of at zero and finite gas velocities are shown. Introduction of the Galileo number has just a small effect on the differences between the hold up for 3.6 and 0.8 mm particles. Data for zero gas flow are also shown and the ~10% effect mentioned above remains evident.

Although these experimental results show particle size and gas velocity effects that have not been reported by Wammes et al. (1991), the agreement of the data suggests that cycling does not affect the static and dynamic hold up measured in these three phase reactor.

Conclusions

1. Loading or filling follows the plug flow model.
2. Draining does not follow literature model.
3. Static and dynamic hold ups at zero gas flow agree with literature correlations for the larger particle size used.
4. Static and dynamic hold ups are influenced by gas velocity.

Acknowledgments

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Notation

A	bed cross sectional area, m^2
d_p	particle diameter, m
Eö	Eötvös Number
g	gravitational acceleration, m/s^2
h	bed height, m
h_c	liquid height due to capillary pressure in bed, m
h_l	momentary liquid height in bed, m
k	Darcy coefficient for packing, m^2
Q(t)	momentary liquid retained by bed, m^3
Q_l	liquid volume fed to bed, m^3
Q_d	volume of liquid drained from bed, m^3/s
Q_0	initial liquid volume retained by bed, m^3
Q_∞	ultimate liquid volum retained by bed after draining, m^3
t	time, s
t_f	filling time for the bed, s
V_b	volume of bed, m^3
V_g	volume of bed occupied by gas, m^3
V_l	volume of bed occupied by liquid, m^3

Greek Letters

β_d	fraction in the bed space filled by liquid under flow conditions
β_d	fraction in the bed space filled by liquid under static conditions
ε	overall porosity of bed
μ_l	dynamic liquid viscosity, kg m/s
ρ_l	liquid density, kg/m^3

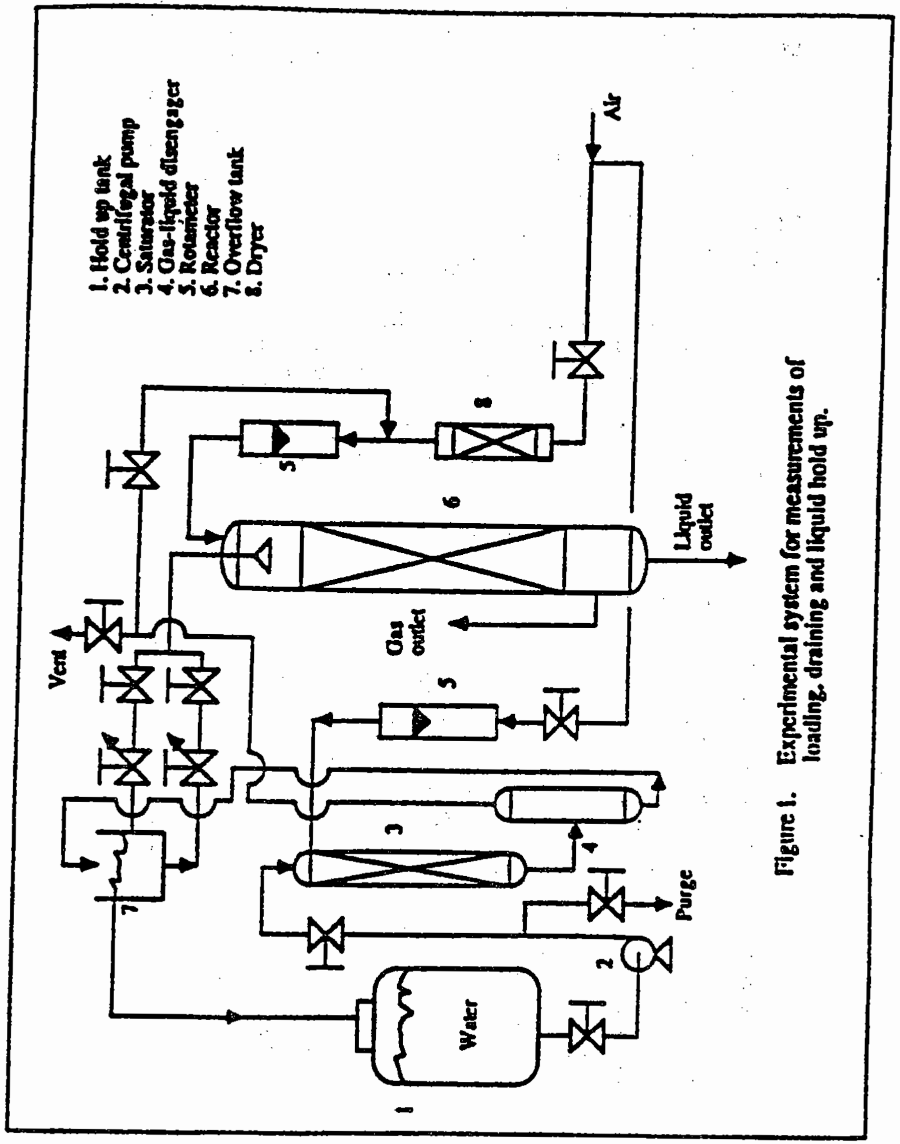


Figure 1. Experimental system for measurements of loading, draining and liquid hold up.

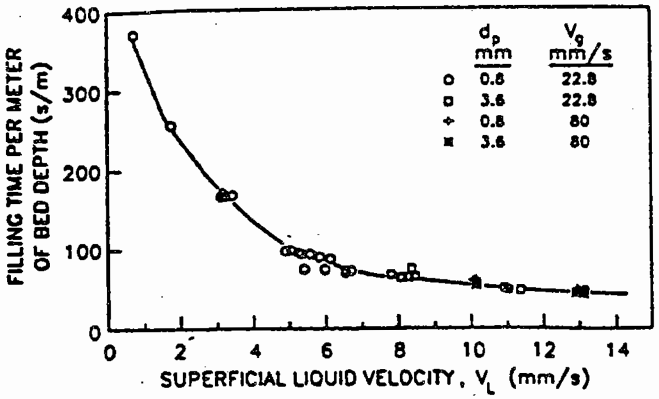


Figure 2. Filling time at various superficial liquid velocities

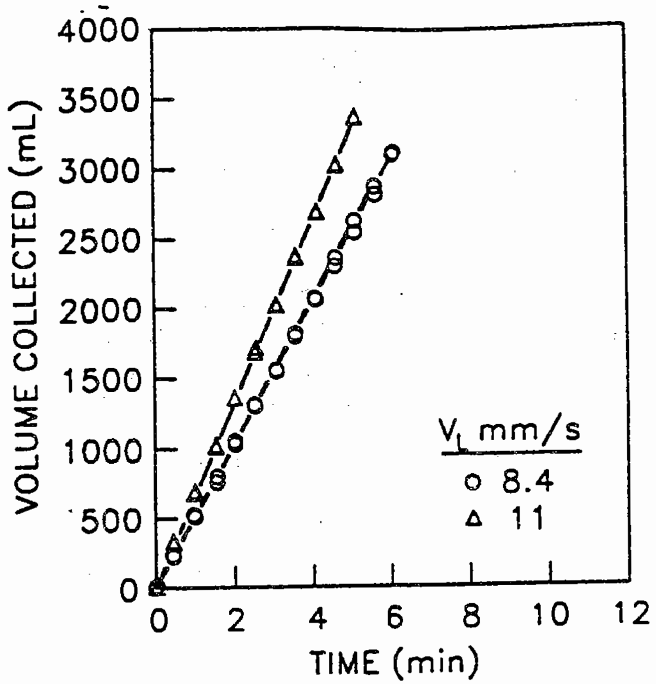


Figure 3. Filling volume vs. time $d_p = 3.6$ mm and $V_g = 22.8$

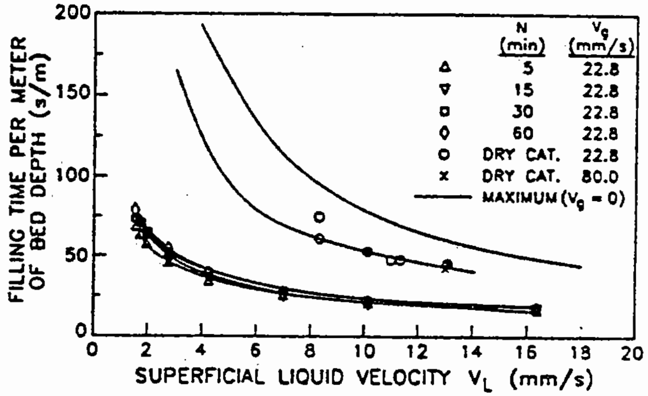
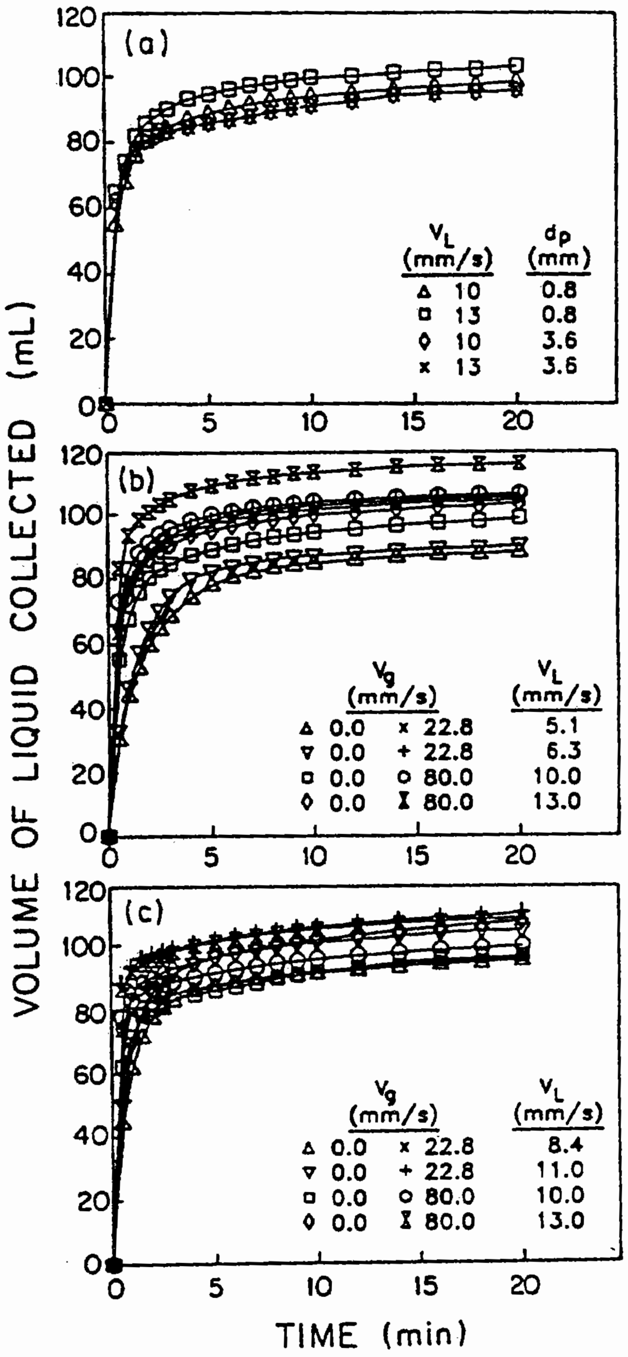


Figure 4. Filling time at various liquid velocities and bed water content for $d_p = 3.6$ mm. Upper two lines are calculated from Equation (1). N is the internal between the interruption of continuous liquid flow through the column and the beginning of the filling experiment.



(a) $V_g = 0$ mm/s; (b) $d_p = 0.8$ mm; (c) $d_p = 3.6$ mm

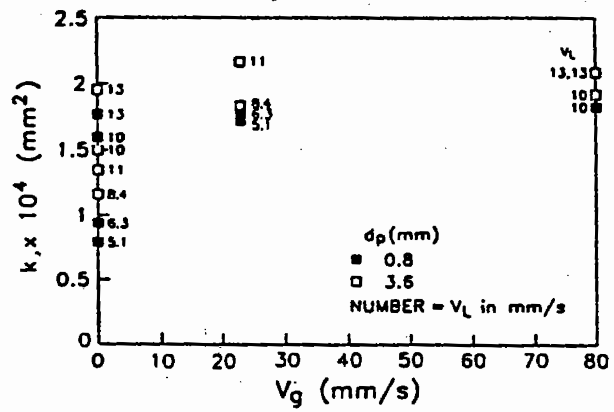


Figure 6. Darcy coefficient k and the height of the undrained bed as a function of the superficial gas velocity for the 0.8- and 3.6-mm carbon particles

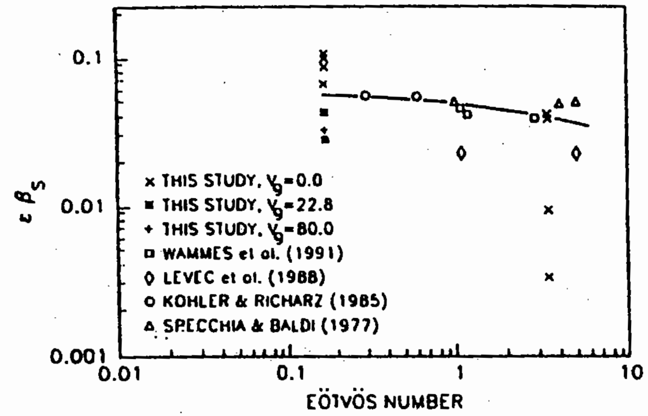


Figure 7. Static liquid hold-up as a function of the Eötvös number

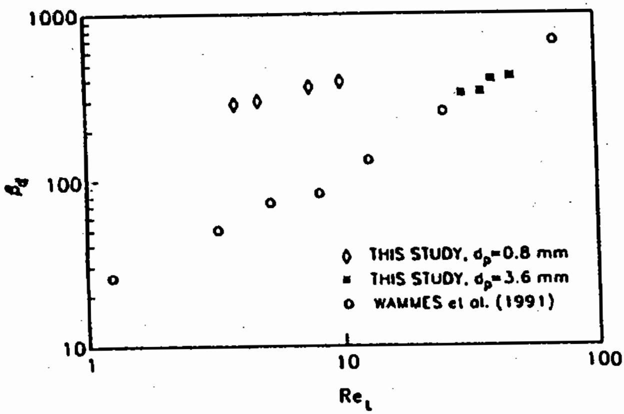


Figure 8. Dynamic liquid hold-up as a function of the liquid Reynolds number

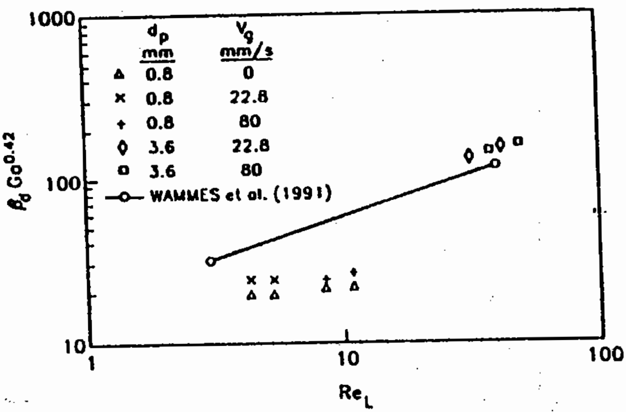


Figure 9. Dynamic liquid hold-up function vs. liquid Reynolds number