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Effect of Operating Pressure on the Extent of Hysteresis in a Trickle Bed Reactor

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The dependence of hydrodynamic parameters, such as pressure drop, on the flow history of the bed is called hysteresis. This phenomenon is most commonly associated with changes of flow distribution and flow patterns with the flow history. Many studies have shown that increased operating pressure will affect flow distribution and wetting efficiency; however, there seems to be no study of the effect of the elevated operating pressure on the extent of hysteresis. In this study, an experimental investigation of the hysteresis in a high-pressure trickle bed has been performed. A hysteresis factor has been introduced to quantify the extent of hysteresis and was found to be a strong function of gas and liquid operating flow rates as well as the operating pressure. In the region of lower liquid velocities, hysteresis is present regardless of the operating pressure or gas velocity. In the region of higher liquid velocities, increases in both pressure and gas velocity will lower the extent of hysteresis. For the range of conditions considered in this study, the extent of hysteresis was uniquely determined by the pressure drop in the Levec mode, regardless of the operating pressure or velocities. The results are interpreted in terms of the phenomenological analysis of Al-Dahhan and Dudukovic (Al-Dahhan, M. H.; Dudukovic, M. P. *Chem. Eng. Sci.* **1994**, *49*, 5681–98), which relates operating pressure and gas velocity to the flow distribution and wetting efficiency in a trickle bed reactor.

Introduction

Trickle bed reactors (TBRs) are widely utilized multiphase reactors in which gas and liquid phases flow cocurrently down a fixed bed of catalyst. These reactors are most commonly used in petroleum and petrochemical processes such as hydrodesulfurization and hydrogenation. They also find applications in wastewater treatment for oxidation of organic pollutants, volatile organic compound abatement in air pollution control, and bioprocesses (Dudukovic et al.;[2] Sie and Krishna[3]).

Among the basic design and operating parameters for trickle beds are pressure drop and liquid-phase holdup. These parameters are not only very dependent on the operating conditions, such as flow rates and bed characteristics, but also exhibit dependence on the flow history of the bed. This is termed hysteresis or the multiplicity of hydrodynamic states in trickle beds. In the landmark study by Kan and Greenfield,[4] the gas flow rate, for a fixed liquid velocity, was looped between zero and the prescribed maximum value. Pressure drop was higher for the branch formed by increasing gas velocity (the upper branch) than the corresponding values in the branch formed by decreasing gas velocity (the lower branch). Based on this and similar studies (Christensen et al.,[5] Levec et al.,[6] Lutran et al.,[7] Ravindra et al.,[8] Gunjal et al.,[9] and Maiti et al.;[10] see also Maiti et al.[11] and references therein), it is evident that the extent of hysteresis depends on the packing size, particle characteristics (e.g., whether the particles are porous), and fluid properties (such as surface tension). The existence of hysteresis has been attributed to the fact that predominant flow structures, for example, film flow or rivulet flow, are different for the upper and lower branches (Christensen et al.,[5] Lutran et al.[7]). The flow structure determines the extent of the interaction between the phases and thus each leads to distinct values of hydrodynamic parameters, such as the pressure drop and liquid holdup.

In the studies performed by Ellman et al.,[12] Wammes et al.,[13] Larachi et al.,[14] Al-Dahhan and Dudukovic,[1] Nemec et al.,[15] Highfill and Al-Dahhan,[16] and Iliuta et al.,[17] it was shown that increased operating pressure alters the phase interaction, hydrodynamic parameters, and flow regime transition. However, there seems to be no study in the literature performed to examine the effect of the operating pressure on the extent of hysteresis in TBRs, even though, typically, industrial trickle beds operate at elevated pressure.[18] As mentioned earlier, operating pressure affects the phase interactions and wetting efficiency and, hence, the flow pattern. Thus, it is expected that it will affect the extent of hysteresis as well. Therefore, the focus of this study is to experimentally examine the effect of elevated pressure on the extent of hysteresis in pressure drop in a TBR.

In this work, different flow histories were achieved in two ways: by looping the liquid velocity for a fixed gas velocity and operating pressure and by setting different initial states of the bed using four prewetting modes (van der Merwe and Nicol,[19] Loudon et al.[20]). In both procedures, the intention is to bring the system into the same operating conditions, with the only distinction being the flow history, and to quantify the resulting difference in the pressure drop.

In the studies by van der Merwe and Nicol[19] and Loudon et al.,[20] the variation in the pressure drop and liquid holdup was examined as a function of the applied prewetting mode for a nitrogen–water 3-mm glass beads system at atmospheric conditions. Five cases of prewetting were considered, namely: a non-prewetted bed, Levec, Kan-liquid, Kan-gas, and Super (for which we will use the name Nicol) prewetted bed. In the Levec mode, the bed is flooded and the liquid is then allowed

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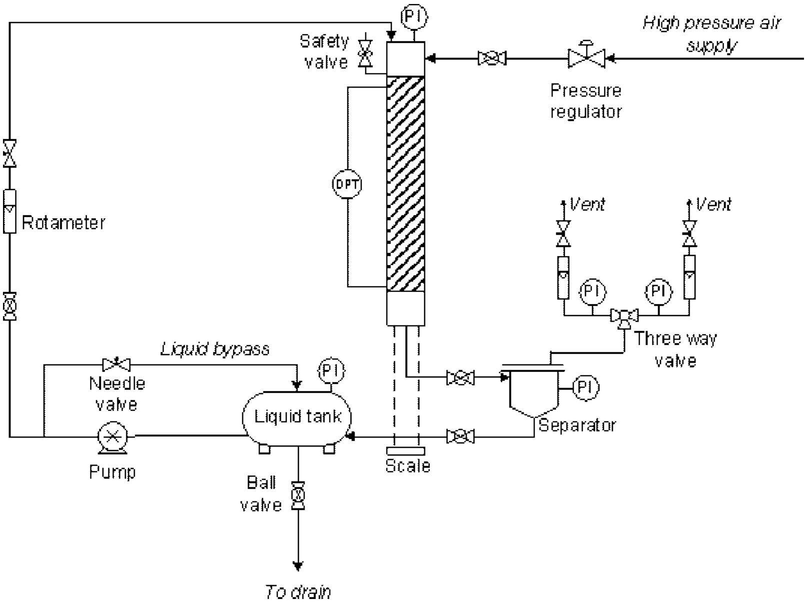

Figure 1. High-pressure trickle bed reactor: experimental setup.

to drain under gravity. After that, gas and liquid flows are initiated. In the Kan-liquid mode, the gas velocity is kept at the operating value while the liquid velocity is increased in order to reach the pulsing regime and is then reduced to the operating value. In the Kan-gas mode, the liquid velocity is kept at the operating value and the gas velocity is varied as before. The Nicol prewetted bed is achieved by first flooding the bed and then draining it without interruption of liquid flow. During this process, the liquid velocity is kept at the operating value, and gas flow is initiated after drainage is complete. Significant differences in the values of pressure drop and liquid holdup were found among the cases described above. A nonprewetted bed invariably exhibited the lowest pressure drop and liquid holdup. The Nicol and Kan-liquid modes had the highest values of pressure drop, while the Kan-gas mode had the highest values of liquid holdup. Among the prewetted beds, the Levec mode always had the lowest pressure drop and liquid holdup. The authors attributed the higher values of pressure drop and liquid holdup to film flow and the lower values to rivulet flow. A very interesting result obtained in these studies is that the Kan-gas mode had intermediate values of pressure drop while at the same time exhibited the highest corresponding liquid holdup. The authors attributed this to the fact that the flow textures are a combination of film and rivulet flow and that their interplay with the effect of tortuosity of the path is such that pressure drop is reduced without affecting the resulting liquid holdup.

The four cases of prewetting mentioned above will be examined in this study at varying operating pressures up to 8 barg. To avoid repacking the bed, which changes the bed structure, the nonprewetted mode was not studied here.

The effect of the increased pressure on the extent of hysteresis can be described using the phenomenological analysis proposed by Al-Dahhan and Dudukovic.[1] These authors introduced five limiting cases that are determined by the values of operating pressure and gas velocity. Their effect on the hydrodynamic parameters, such as pressure drop, liquid holdup, catalyst wetting efficiency, and gas−liquid interfacial area, was examined. Limiting cases are interpreted in terms of the dimensionless pressure drop defined as $(\Delta P/L)/\rho_L g$. The analysis of Al-Dahhan and Dudukovic[1] is summarized next.

Case 1: No Gas Flow, All Pressures. The dimensionless pressure gradient $(\Delta P/L)/\rho_L g$ is zero and the liquid flow is driven by gravity only. The system is characterized by the largest value of liquid holdup for a given liquid velocity. Due to poor spreading of the liquid, it also exhibits the smallest values of catalyst wetting efficiency and gas−liquid interfacial area at given liquid velocity.

Case 2: Low Pressure and Low Gas Superficial Velocity. The dimensionless pressure drop is small, and its change with gas velocity can be neglected. Hence, the liquid flow is gravity driven and to a large extent gas independent; i.e., it does not depend on the operating pressure and gas flow rate.

Case 3: Low Pressure and High Superficial Gas Velocity. In this case, for a fixed gravitational force, i.e., given liquid density, the pressure gradient increases with increased gas velocity, and as a consequence, the dimensionless pressure gradient increases as well. This results in a decrease in liquid holdup and an increase in the catalyst wetting efficiency and gas−liquid interfacial area at given liquid velocity. This is the result of the increased spreading of liquid on both the macrolevel (across the reactor) and microlevel (over the external area of the packing particle) due to higher gas flow rate.

Case 4: High Pressure and Low Gas Superficial Velocity. The pressure drop and dimensionless pressure gradient increase due to the higher gas density. The resulting effect is similar to case 3, but less pronounced since the pressure gradient is more sensitive to velocity changes than to gas density changes.

Case 5: High Pressure and High Gas Superficial Velocity. In this case, gas−liquid interactions are the most pronounced. Shear stress on the gas−liquid interface increases significantly, and due to this, liquid holdup decreases while catalyst wetting efficiency and gas−liquid interfacial area increase. This effect is more pronounced at higher liquid flow rates than at low flow rates.

This analysis will be extended to interpret the effect of elevated pressure on the extent of hysteresis.

Experimental Setup and Operating Conditions

Figure 1 is a schematic of the high-pressure trickle bed reactor setup. It is designed to operate at pressures up to 10 barg (∼115 psig). It consists of a reactor column, gas and liquid delivery system, and gas−liquid separator, and it enables simultaneous measurement of liquid holdup and pressure drop. The reactor

Table 1. Experimental Setup and Operating Conditions

column diameter, m	0.163
bed height, m	0.68
fluids used	water and air
packing	glass beads, 3-mm diameter
bed porosity	0.41
system pressure, barg	1–8
gas velocity, mm/s	27–94
liquid velocity, mm/s	1.9–9.5
prewetting modes considered	Levec, Kan-gas, Kan-liquid, Nicol

is a stainless steel column with a 16.3-cm inner diameter. Liquid recirculates in a closed loop: it is stored in a pressurized tank and supplied via a high-pressure pump to the distributor at the top of the column. After the gas−liquid separation, the liquid is directed back to the tank. The system is pressurized using a high-pressure air supply. The top distributor dispenses liquid uniformly across the bed. Once the system is pressurized by the gas supply, the gas flow is regulated using the valves located after the gas−liquid separator. Gas, introduced at the top of the column, flows through tubes that are spaced uniformly in the axial direction through the distributor. The liquid flow rate is controlled using the needle valve and measured using the flowmeter placed in the inlet line. The gas flow rate is measured with two flowmeters (high and low range) located after the gas−liquid separator. Two Validyne differential pressure transducers, (low range 1.25 psid and high range 5 psid), with pressure taps spaced 50 cm apart, were used for the pressure drop measurements. The experimental work consisted of two parts: (1) setting the different initial state of the bed using the four prewetting modes, and (2) looping the liquid velocity (i.e., increasing gradually the liquid velocity from a preset minimum to a maximum value, measuring pressure drop at each steady state, and then reducing the liquid velocity over the same range of values) for a fixed gas velocity and operating pressure. Both procedures enabled us to investigate the effect of flow history on the pressure drop and the extent of its hysteresis.

Table 1 summarizes the experimental conditions. The bed was packed with 3-mm glass beads to a height of 0.68 m. The resulting average porosity of the bed was 0.41. The fluids used were water and air. The experiments covered five operating pressures (1, 2, 4, 6, and 8 barg). The gas (air) density at these pressures is in the range 1−8 kg/m^3 and therefore corresponds to typical hydrogen gas densities at industrial conditions used in commercial hydrogenation units where pressures and temperatures are typically in the range 1−20 MPa and 300−400 °C, respectively.[21] As shown in Al-Dahhan et al.,[18] the hydrodynamics of high-pressure hydrogen can be simulated by lower pressure provided that the gas densities are matched. The present results are therefore applicable to industrially relevant cases. Since the case of the nonprewetted bed was not considered in this study, it was possible to examine all of the above conditions without any need for repacking the column. Thus, the results obtained are not affected by a change in the overall porosity or the structure of the bed. The experimental procedure was as follows. In the Nicol prewetting mode, the bed was first prewetted and drained until only residual holdup remained. The pressure was increased to the operating value. Then, liquid flow was set to the appropriate value for the current experiment, and the bed was flooded by closing the outlet valve. Once the bed was flooded, the outlet valve was opened, allowing liquid to drain while keeping the liquid flow on. After the bed drained fully, the gas flow was initiated. Pressure drop was recorded once the steady state was reached. In the Levec prewetting mode, the bed was flooded, liquid flow was shut off, and the bed was drained. Then, the gas flow was initiated. Ten minutes later,

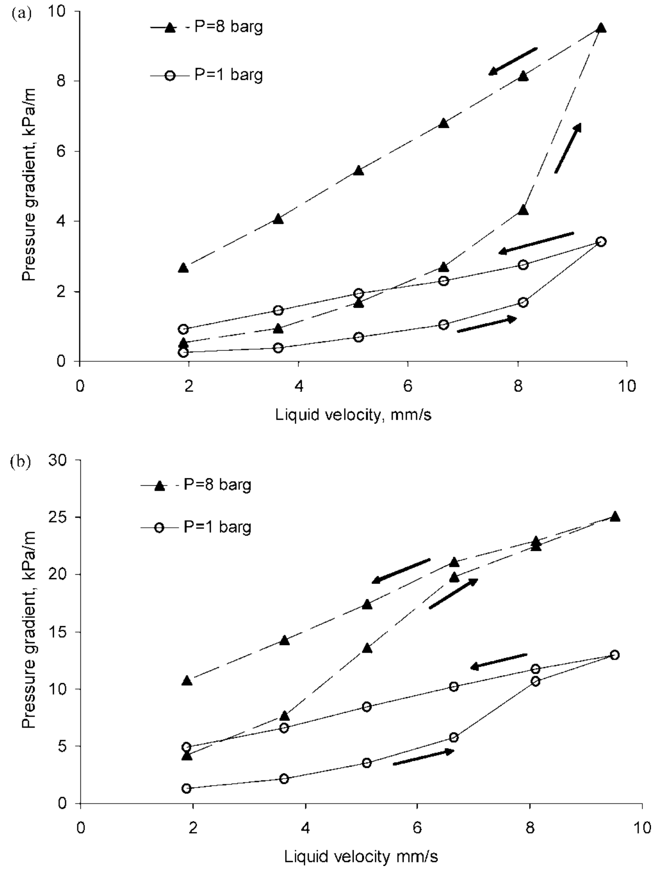

Figure 2. Hysteresis loops: (a) U_G = 27 mm/s; (b) U_G = 90 mm/s.

liquid flow was set to the operating value, and after steady state was reached, measurements were taken. The Kan-liquid and Kan-gas procedures were obtained by reaching the pulsing regime by increasing the liquid- and gas-phase flow rates, respectively, and then setting them to the operating values.

For each of the experimental conditions, the order in which the modes were used was invariably as follows: Nicol, Levec, Kan-liquid, and Kan-gas. It was verified that the order in which the modes are applied does not affect the resulting pressure drop gradient or the extent of hysteresis. This was confirmed by varying the sequence for a couple of sets of experimental conditions. Thus, all the results are to be attributed purely to the modes used and not to the sequence in which they are applied.

In each of the looping experiments, the bed was first flooded and then drained until only static holdup remained. Thus, Levec prewetting mode was used. Then, for a fixed operating pressure (1 or 8 barg) and gas velocity (27 or 94 mm/s), liquid velocity was looped from 1.9 to 9.5 mm/s (which is short of the pulsing velocity) and back. The pressure drop was recorded for each of the liquid velocities.

Results and Discussion

The results for pressure drop, based on the looping liquid velocity experiments, are given in Figure 2. The data points group into two distinct branches: one formed by increasing liquid velocity ("lower branch") and one formed by decreasing liquid velocity ("upper branch"). The pressure drop is always higher for the case of decreasing liquid velocity and is also always higher for the case of higher operating pressure. Hysteresis is present irrespective of the operating pressure or gas velocity. In the lower branch, liquid spreads across a progressively higher

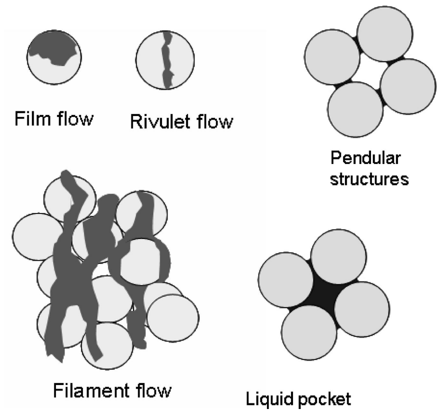

Figure 3. Trickle flow patterns (adapted from Lutran et al.[7]).

area of the column. Some of the liquid films that were developed in this process remained irrigated in the upper branch.[10] The enhanced spreading led to a higher pressure drop. The generalization of this interpretation is that the hysteresis is a phenomenon caused by a change in the predominant flow pattern during the hysteresis loop.[22] In the lower branch, liquid flows predominantly in the form of filaments (Figure 3). With an increase of liquid velocity, the filaments enlarged and merged, creating predominantly film flow. Once the liquid flow rate was decreased, predominant flow patterns were again filaments, but with the larger contribution coming from film flow. Similar analysis was used by Wang et al.,[22] to develop a model capable of conceptually quantifying the extents of film and rivulet flow. In their model, fitting parameters are used to assign areas of reactors associated with film and filament flow in the lower branch.

In Figures 4–6, the pressure drop is given as a function of the applied prewetting mode. The data indicate existence of hysteresis for the high-pressure system under investigation. Values of pressure drop can differ up to 300% just due to the applied prewetting mode. Pressure drop is the highest for the Nicol and Kan-liquid prewetting modes, while the Levec mode tends to exhibit lower values.

The analysis of Loudon et al.,[20] discussed above, can be extended as follows. The initial state of the bed depends on the applied prewetting procedure and, in terms of flow structures, will consist of the patterns shown in Figure 3. Draining the bed, i.e., applying the Levec mode, will yield an initial state with predominantly pendular structures with small contributions from liquid pockets. Among others, this was demonstrated in the experiments performed by Kramer,[23] in which the bed was flooded and drained, and the resulting structures were photographed. Pendular structures are located between two touching spheres, and liquid is held by the capillary forces (Figure 3). The Nicol mode apparently yields an initial state that has a bigger contribution of liquid pockets due to irrigation of the bed during draining. Due to very high liquid flow rate, the Kan-liquid mode yields a similar initial state of the bed, while the Kan-gas mode gives an intermediate state between the Levec and Kan-liqud. The initial state of the bed determines the resulting flow distribution, just as in the cases of prewetted and nonprewetted beds,[7] and hence, the variability of such state is the cause of the observed hysteresis. The better initial irrigation of the bed present in the Kan-liquid and Nicol modes yields flow distribution and patterns with corresponding higher pressure drops. One indication that this corresponds to the actual flow patterns was given by van Houwelingen et al.[24] They examined the distribution of wetting efficiency for the case of the Kan-

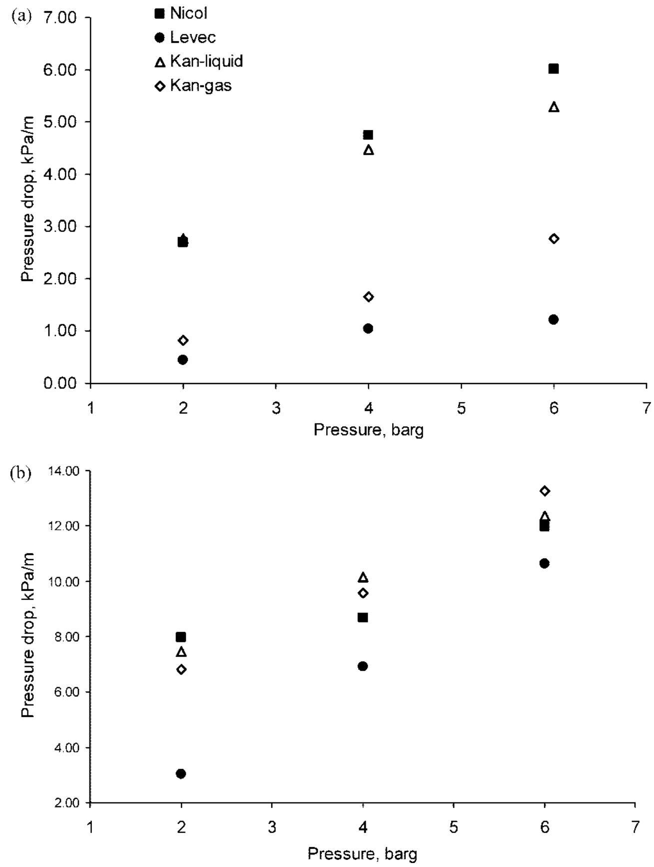

Figure 4. Dependence of the pressure gradient on the applied prewetting mode. Low gas velocity case ($U_G = 36$ mm/s). (a) Low liquid velocity ($U_L = 3.6$ mm/s); (b) high liquid velocity ($U_L = 9.52$ mm/s).

liquid and Levec prewetting modes. The Levec mode had a bimodal distribution of wetting efficiency, while the Kan-liquid mode exhibited more homogeneously distributed wetting efficiency with a higher average value.

Effect of Operating Conditions on the Extent of Hysteresis. Figure 2 and Figures 4–6 indicate that the difference in the pressure drop due to the flow history of the bed (i.e., the extent of hysteresis) is a strong function of the operating flow rates. For the lower flow rates (Figures 4a and 5a), the system exhibits behavior similar to the low-pressure data of Loudon et al.[20] Three distinct regions can be seen: the Levec mode with the lowest, the Kan-liquid and Nicol with the highest, and the Kan-gas with intermediate values of pressure drop. These three regions are also present for intermediate (Figure 5a) and high (Figure 6a) gas velocity cases, provided that liquid velocity is in the low range. Increasing the liquid velocity (Figures 4b, 5b, and 6b) will diminish the difference in the pressure drop data. In a study by Wang et al.,[22] the same operating conditions were reached by looping first the liquid and then the gas velocity. The difference between the pressure drops of the upper and lower branches was higher when liquid flow was varied. The present study leads to a similar conclusion—the liquid velocity has a more pronounced effect on the extent of hysteresis than the gas velocity. The reason is that the initial state of the bed, dependent on the applied prewetting procedure as discussed earlier, will have a less pronounced effect on the flow distribution in the case of a higher liquid velocity.

In order to quantify the effect of operating flow rates and pressure on the extent of hysteresis, a hysteresis factor was introduced:

$$f_H = 1 - \frac{(\Delta P/L)_{\text{lower branch}}}{(\Delta P/L)_{\text{upper branch}}} \quad (1)$$

For the prewetting modes investigation, the lower branch is Levec mode, while the upper branch is Kan-liquid mode. The no-hysteresis case is defined as $f_H = 0$, while increased values of the hysteresis factor indicate progressively higher extents of hysteresis. According to the phenomenological analysis discussed above, increased gas flow rate will enhance the liquid spreading and wetting efficiency (case 3). Operating pressure will have a similar, but less pronounced effect (case 4). The effects of both gas velocity and operating pressure increase in the case of a higher liquid velocity. As a result, it is expected that the hysteresis factor, for a fixed liquid velocity, will be a decreasing function of both operating pressure and gas velocity, since enhanced spreading will diminish the influence of the initial state of the bed on the resulting pressure drop. Figure 7 gives the hysteresis factor based on the looping liquid velocity data. Figure 8a gives the hysteresis factor based on the prewetting modes investigation. It can be seen that f_H shows strong dependence on the liquid flow rate and that two regions of values can be defined (Figure 8a): a high hysteresis region for the low liquid velocity and low hysteresis region for the higher liquid velocity. In the low liquid velocity region, the hysteresis factor maintains a high value irrespective of pressure or gas velocity. This implies that in this region the improvement in liquid spreading due to higher gas−liquid interactions (achieved through either higher gas density, i.e., operating

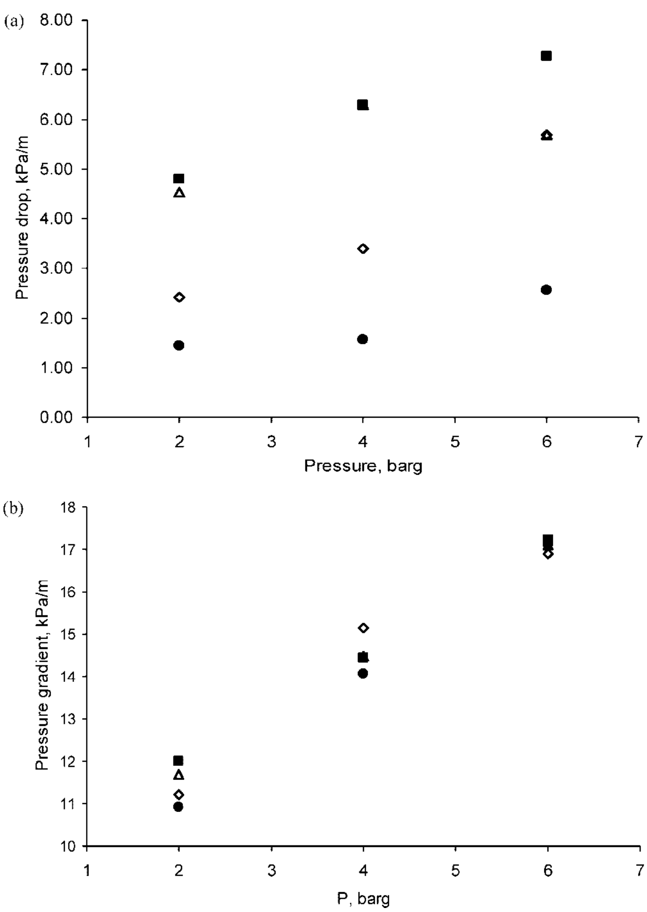

Figure 5. Dependence of the pressure gradient on the applied prewetting mode (for symbols see, Figure 4). Intermediate gas velocity case ($U_G = 58$ mm/s). (a) Low liquid velocity ($U_L = 3.6$ mm/s); (b) high liquid velocity ($U_L = 9.52$ mm/s).

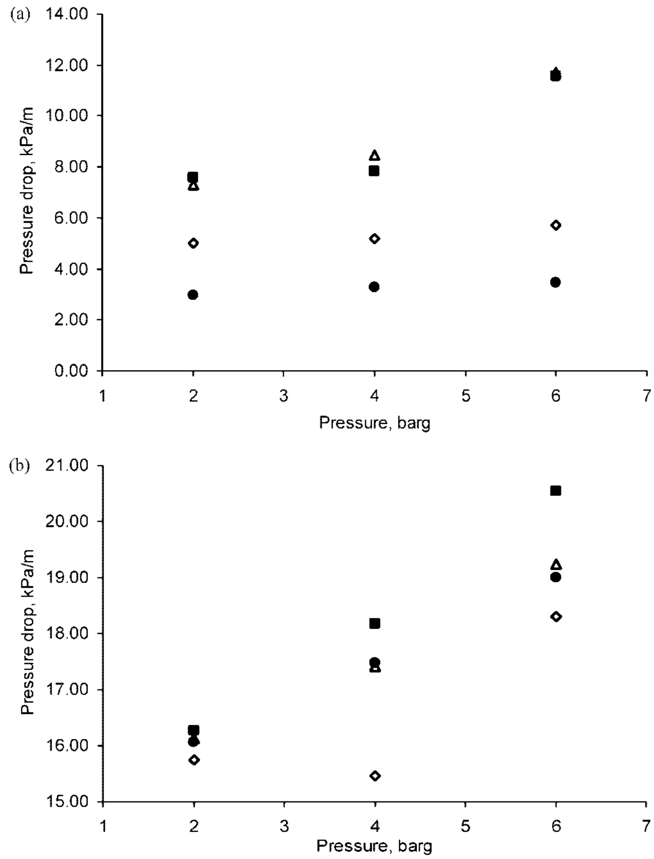

Figure 6. Dependence of the pressure gradient on the applied prewetting mode (for symbols, see Figure 4). High gas velocity case ($U_G = 90$ mm/s). (a) Low liquid velocity ($U_L = 3.6$ mm/s); (b) high liquid velocity ($U_L = 9.52$ mm/s).

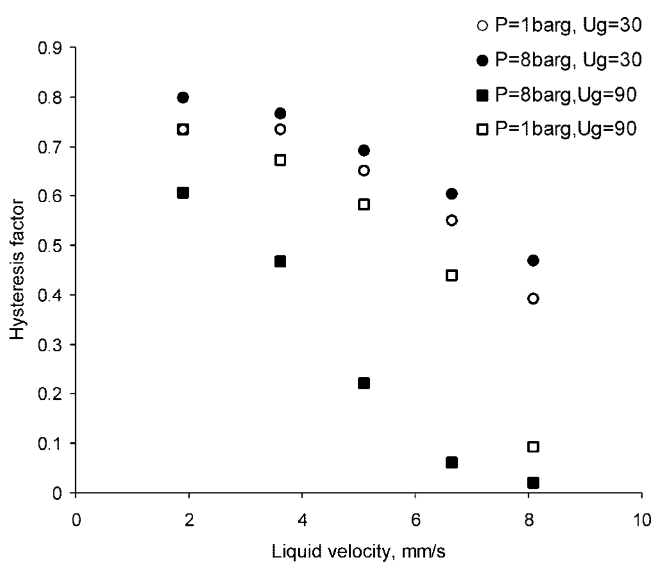

Figure 7. Hysteresis factor (eq 1) based on looping liquid velocity data.

pressure or higher gas velocity) is not significant. Note that these results are in agreement with X-radiography imaging of flow structures of the Levec mode, performed by van der Merwe et al.,[25] that showed no significant improvement of liquid spreading due to increased gas velocity. The data points corresponding to higher liquid velocity are grouped in terms of both operating pressure and gas flow rate. The lowest values of hysteresis factor (Figure 8a, condition: $P = 6$ barg, $U_G = 90$ mm/s) correspond to the highest pressure−highest gas velocity conditions, i.e., case 5 of the phenomenological

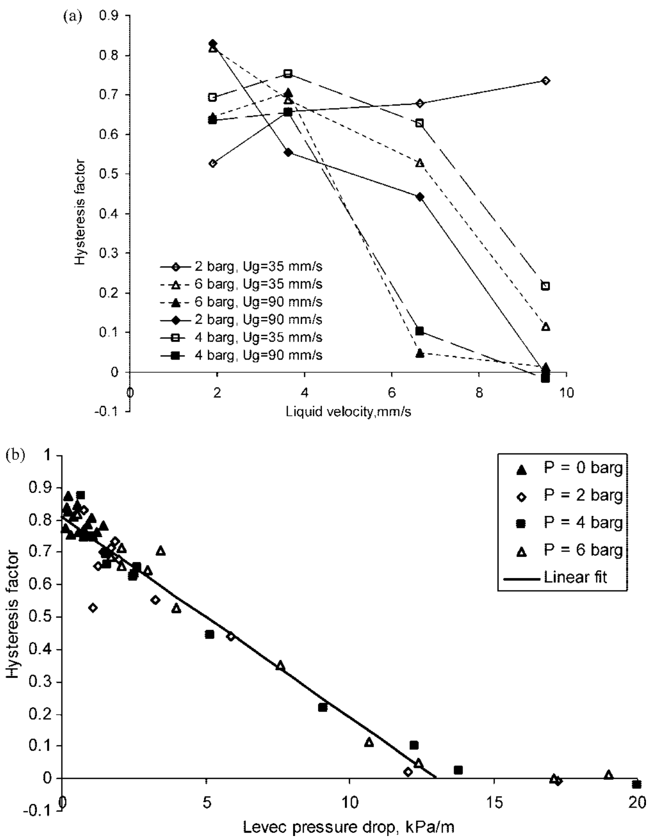

Figure 8. (a) Hysteresis factor based on the prewetting modes investigation, (b) Hysteresis factor as a function of pressure drop in the Levec mode. $P = 0$ barg data adapted from Loudon et al.[20] (water−nitrogen−3-mm glass beads system, $U_L = 3-9$ mm/s, $U_G = 20-90$ mm/s).

analysis. Gas velocity has a bigger effect on the extent of hysteresis, as seen in the three bottom curves in Figure 8a, which are all for high gas velocity ($U_G = 90$ mm/s) conditions. This is in accordance to case 3 of the phenomenological analysis. The top three curves are for the low gas velocity ($U_G = 35$ mm/s) condition and, corresponding to the case 4 of the analysis, are decreasing functions of operating pressure.

Figure 8b shows the hysteresis factor as a function of the pressure drop in the Levec mode. Note that the data all fall on one line despite the fact that they correspond to vastly different operating conditions (pressure, gas, and liquid velocity). This implies that only the Levec mode pressure drop uniquely determines the extent of hysteresis, as the other prewetting modes' pressure drop data do not show this trend. The linear fit of data with the nonzero value of hysteresis factor, i.e., for the pressure drops of up to 13 kPa/m, has a slope of about −0.06 (Figure 8b). It would be instructive to examine whether the slope of this line is a function of other system parameters, such as the size of packing, surface tension, and density of the liquid phase. Such an enlarged data set would allow development of the correlation for the prediction of the hysteresis factor.

Concluding Remarks

The experimental results based on both the looping liquid velocity and the prewetting modes investigations indicate the presence of hysteresis in a high-pressure trickle bed. The resulting difference in pressure drop can reach up to 300−400%. A hysteresis factor has been introduced to quantify the extent of hysteresis and was found to be a strong function of the operating flow rates and pressure. For the lower liquid flow rates, hysteresis seems to persist regardless of the pressure or gas flow rate. However, in the region of higher liquid flow rates, increases in both pressure and gas flow rates will decrease the extent of hysteresis.

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Nomenclature

f_H = hysteresis factor, defined in eq 1
g = gravitational acceleration, m/s
P = pressure, barg
U_G = gas superficial velocity, mm/s
U_L = liquid superficial velocity, mm/s
$\Delta P/L$ = pressure drop gradient, kPa/m
ρ_L = liquid-phase density, kg/m^3

Literature Cited

(1) Al-Dahhan, M. H.; Dudukovic, M. P. Pressure drop and liquid holdup in high pressure trickle-bed reactors. *Chem. Eng. Sci.* **1994**, *49* (24B), 5681–5698.

(2) Dudukovic, M. P.; Larachi, F.; Mills, P. L. Multiphase reactors - revisited. *Chem. Eng. Sci.* **1999**, *54* (13−14), 1975–1995.

(3) Sie, S. T.; Krishna, R. Process development and scale up: III. Scale-up and scale-down of trickle bed processes. *Rev. Chem. Eng.* **1998**, *14* (3), 203–252.

(4) Kan, K.-M.; Greenfield, P. F. Multiple hydrodynamic states in cocurrent two-phase downflow through packed beds. *Ind. Eng. Chem. Process Des. Dev.* **1978**, *17* (4), 482–485.

(5) Christensen, G.; McGovern, S. J.; Sundaresan, S. Cocurrent downflow of air and water in a two-dimensional packed column. *AIChE J.* **1986**, *32* (10), 1677–1689.

(6) Levec, J.; Grosser, K.; Carbonell, R. G. The hysteretic behavior of pressure drop and liquid holdup in trickle beds. *AIChE J.* **1988**, *34* (6), 1027–1030.

(7) Lutran, P. G.; Ng, K. M.; Delikat, E. P. Liquid distribution in trickle-beds. An experimental study using computer-assisted tomography. *Ind. Eng. Chem. Res.* **1991**, *30* (6), 1270–1280.

(8) Ravindra, P. V.; Rao, D. P.; Rao, M. S. Liquid Flow Texture in Trickle-Bed Reactors: An Experimental Study. *Ind. Eng. Chem. Res.* **1997**, *36* (12), 5133–5145.

(9) Gunjal, P. R.; Kashid, M. N.; Ranade, V. V.; Chaudhari, R. V. Hydrodynamics of Trickle-Bed Reactors: Experiments and CFD Modeling. *Ind. Eng. Chem. Res.* **2005**, *44* (16), 6278–6294.

(10) Maiti, R.; Khanna, R.; Nigam, K. D. P. Trickle-Bed Reactors: Porosity-Induced Hysteresis. *Ind. Eng. Chem. Res.* **2005**, *44* (16), 6406–6413.

(11) Maiti, R.; Khanna, R.; Nigam, K. D. P. Hysteresis in Trickle-Bed Reactors: A Review. *Ind. Eng. Chem. Res.* **2006**, *45* (15), 5185–5198.

(12) Ellman, M. J.; Midoux, N.; Laurent, A.; Charpentier, J. C. A new, improved pressure drop correlation for trickle-bed reactors. *Chem. Eng. Sci.* **1988**, *43* (8), 2201–2206.

(13) Wammes, W. J. A.; Mechielsen, S. J.; Westerterp, K. R. The transition between trickle flow and pulse flow in a cocurrent gas-liquid trickle-bed reactor at elevated pressures. *Chem. Eng. Sci.* **1990**, *45* (10), 3149–3158.

(14) Larachi, F.; Laurent, A.; Midoux, N.; Wild, G. Experimental study of a trickle-bed reactor operating at high pressure: two-phase pressure drop and liquid saturation. *Chem. Eng. Sci.* **1991**, *46* (5−6), 1233–1246.

(15) Nemec, D.; Bercic, G.; Levec, J. The hydrodynamics of trickling flow in packed beds operating at high pressures. The relative permeability concept. *Chem. Eng. Sci.* **2001**, *56* (21−22), 5955–5962.

(16) Highfill, W.; Al-Dahhan, M. Liquid-solid mass transfer coefficient in high pressure trickle bed reactors. *Chem. Eng. Res. Des.* **2001**, *79* (A6), 631–640.

(17) Iliuta, I.; Aydin, B.; Larachi, F. Onset of pulsing in trickle beds with non-Newtonian liquids at elevated temperature and pressure.—Modeling and experimental verification. *Chem. Eng. Sci.* **2005**, *61* (2), 526–537.

(18) Al-Dahhan, M. H.; Larachi, F.; Dudukovic, M. P.; Laurent, A. High-Pressure Trickle-Bed Reactors: A Review. *Ind. Eng. Chem. Res.* **1997**, *36* (8), 3292–3314.

(19) van der Merwe, W.; Nicol, W. Characterization of Multiple Flow Morphologies within the Trickle Flow Regime. *Ind. Eng. Chem. Res.* **2005**, *44* (25), 9446–9450.

(20) Loudon, D.; van der Merwe, W.; Nicol, W. Multiple hydrodynamic states in trickle flow: Quantifying the extent of pressure drop, liquid holdup and gas-liquid mass transfer variation. *Chem. Eng. Sci.* **2006**, *61* (22), 7551–7562.

(21) Kundu, A.; Nigam, K. D. P.; Duquenne, A. M.; Delmas, H. Recent developments on hydroprocessing reactors. *Rev. Chem. Eng.* **2003**, *19* (6), 531–605.

(22) Wang, R.; Mao, Z.-S.; Chen, J. Experimental and theoretical studies of pressure drop hysteresis in trickle bed reactors. *Chem. Eng. Sci.* **1995**, *50* (14), 2321–2328.

(23) Kramer, G. J. Static liquid hold-up and capillary rise in packed beds. *Chem. Eng. Sci.* **1998**, *53* (16), 2985–2892.

(24) van Houwelingen, A. J.; Sandrock, C.; Nicol, W. Particle wetting distribution in trickle-bed reactors. *AIChE J.* **2006**, *52* (10), 3532–3542.

(25) van der Merwe, W.; Nicol, W.; de Beer, F. Trickle flow distribution and stability by X-ray radiography. *Chem. Eng. J. (Amsterdam, Netherlands)* **2007**, *132* (1−3), 47–59.

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