EXPERIMENTAL VERIFICATION OF HYDRODYNAMIC MULTIPLICITY IN AN INDUSTRIAL TRICKLE BED REACTOR

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Trickle bed hydrodynamics has undergone intensive investigation in the last 5 decades. In this context, hydrodynamics refer to the measurement and correlation of parameters like two-phase pressure drop, liquid holdup, wetting (or contacting) efficiency, gas-liquid and liquid-solid mass transfer coefficients and areas, flow regimes and axial dispersion. These parameters directly impact the design (size, catalyst loading), operation (operating cost, thermal stability, catalyst utilization and catalyst life) and optimization of trickle bed reactors (TBR).

The greatest drawback of data and correlations available in the open literature is the fact that the hydrodynamic parameters were determined almost exclusively on laboratory and pilot scale apparatus. In addition, the systems used in the studies were generally far removed from those encountered in industrial practice. As such, low pressure air with water as liquid phase together with glass beads as packing are often employed. More recently, considerable attention has been given to the effect of high pressures on the hydrodynamics and some industrial packings and fluids have come into consideration (see the review by Al-Dahhan et al., 1997).

In this study, we focus on the phenomenon of hysteresis in the hydrodynamic parameters, first reported by Kan & Greenfield (1978, 1979) and subsequently studied experimentally by Levec et al. (1986), Christensen et al. (1986), Lazzaroni et al. (1988, 1989), Lutran et al. (1991), Wang et al. (1995), Ravindra et al. (1997), Sederman & Gladden (2001) and Van der Merwe & Nicol (2005). All of these studies were laboratory scale, operated at low pressures and employed air, glass and water (sometimes with added surfactants to decrease the surface tension). It was observed that pressure drop varied by as much as 700 % depending on the exact history of gas and liquid flows, as well as the prewetting procedure employed.

There are in fact two broadly classified pressure drop regions: an upper branch of the hystersis loop for beds that were subjected to high gas and/or liquid flow rates in their past, and a lower branch of the hysteresis loop for beds that either had not been prewetted or had drained (as a result of liquid flow interruption). The upper region is dominated by uniform flow (films spread evenly over all of the available surface). The lower region is dominated by non-uniform flow (localized rivulet or channelling flow). They are denoted as the Kan mode (high fluid rates in the past) and the Levec mode (post-draining operation) respectively.

There are several strong arguments supporting the notion that hydrodynamic multiplicity is unlikely to exist in industrial reactors:

1. The surface tension of petrochemical liquids at the temperatures encountered in industry (in excess of 250ºC) is typically in the order of 5-10 mN/m, i.e. seven times lower than that of water (73 mN/m at 25ºC). The low surface tension is expected to allow the liquid to spread over the packing – resulting in uniform film flow regardless of the flow history.
2. The contact angle between glass and water is 31.7º, indicating non-perfect wettability. Catalysts, however, are porous. This implies that the particles become liquid filled (due to capillary action) – resulting in a zero contact angle (i.e. perfect wettability). Levec et al. (1986) in fact attributed their hysteresis observations to the difference between advancing and receding contact angles – a difference that does not exist if the contact angle is zero. A (effectively) perfectly wettable solid is expected to again ensure uniform film flow.
3. Industrial applications are operated at high pressure (tens to hundreds of bar). It is well established (Al-Dahhan et al., 1997) that increased gas

density (due to pressure) causes the gas to smear the liquid over the solid, thereby increasing the wetting efficiency and the flow uniformity.

4. Intermittent shut-downs (trips) cause the bed to be properly wetted eventually since the liquid is believed to take a different path each time it is reintroduced.
5. The existence of wandering rivulets (i.e. that the rivulets change path stochastically over long periods of time) will cause the bed to be properly wetted and the liquid to be uniformly distributed after several days of operation.

In this work, an industrial trickle bed reactor is investigated during regular operation in a large petrochemical refinery. It is perfectly suited to an investigation into industrial hydrodynamic multiplicity:

- It is a typical industrial reactor according to points 1 to 4 listed above. Details are given in Table 1.
- It is operated at low gas and liquid mass fluxes. This ensures operation in the trickle flow regime (as apposed to the transition, pulse, spray or bubble flow regimes where hysteresis does not exist).
- There is a single bed. There are no intermediate liquid or gas feeds.
- The reactor operates nearly isothermally (absolute temperature difference of 2 % between inlet and outlet).
- The catalyst cokes very slowly. Periods of stable gas and liquid feed rates of up to 30 days were found during which the pressure drop did not increase.
- The pressure drop is measured across the entrance and exit lines. An electronic log of historic data going back several years is available for liquid and gas flow rates and pressure drop.

Table 1. Experimental detail

Liquid flux range	0.74 – 1.22 kg/m^2s
Gas flux range	0.004 - 0.011 kg/m^2s
Liquid phase	Petrochemical
Gas phase	Hydrogen
Liquid surface tension	10 mN/m (approx.)
Operating pressure	4 MPa
Catalyst	Commercial 1 x 3 mm extrudate (porous)

Figure 1 is an excerpt of such historic data. It shows liquid flow rate and pressure drop for constant gas flow rate before and after a liquid interruption. Note that the liquid flow rate is very stable but there are periodic interruptions and rate variations. The gas flow rate exhibits considerable noise, but whenever the liquid rate is constant, the average gas flow rate usually remains relatively constant as well. The pressure drop also has a noise component that is attributable to the noise in the gas flow rate.

In figure 1, the pressure drop directly *after* the reintroduction of liquid is *lower* than directly *before* the interruption. The liquid rate, gas rate, coking condition and other system parameters were identical for these two points. The only difference is that the bed had been subjected to a high liquid flow rate some time before the interruption (marked L_{max} on figure 1), after which the liquid flow rate was reduced to the lower value that is encountered at the points at which we measure the pressure drop (marked L_1). This analysis was repeated for several instances where the bed had fortuitously been subjected to a high liquid rate ($L=L_{max}$), followed by a reduction to a lower rate ($L=L_1$), followed by an interruption ($L=0$) and then a reintroduction at the previous rate ($L=L_1$). Figure 2 shows that after a liquid feed interruption - reintroduction event the pressure drop is lower. This is understood easily in terms of the laboratory scale experiments. Upon liquid interruption, the liquid drains under gravity. When the liquid is re-introduced, the reactor is operated in the Levec mode. An increase in the liquid rate improves liquid spreading. Upon a subsequent decrease, the pressure drop is higher because the bed had moved toward more uniform operation. Qualitatively, the pressure drop loop is shown in figure 3.

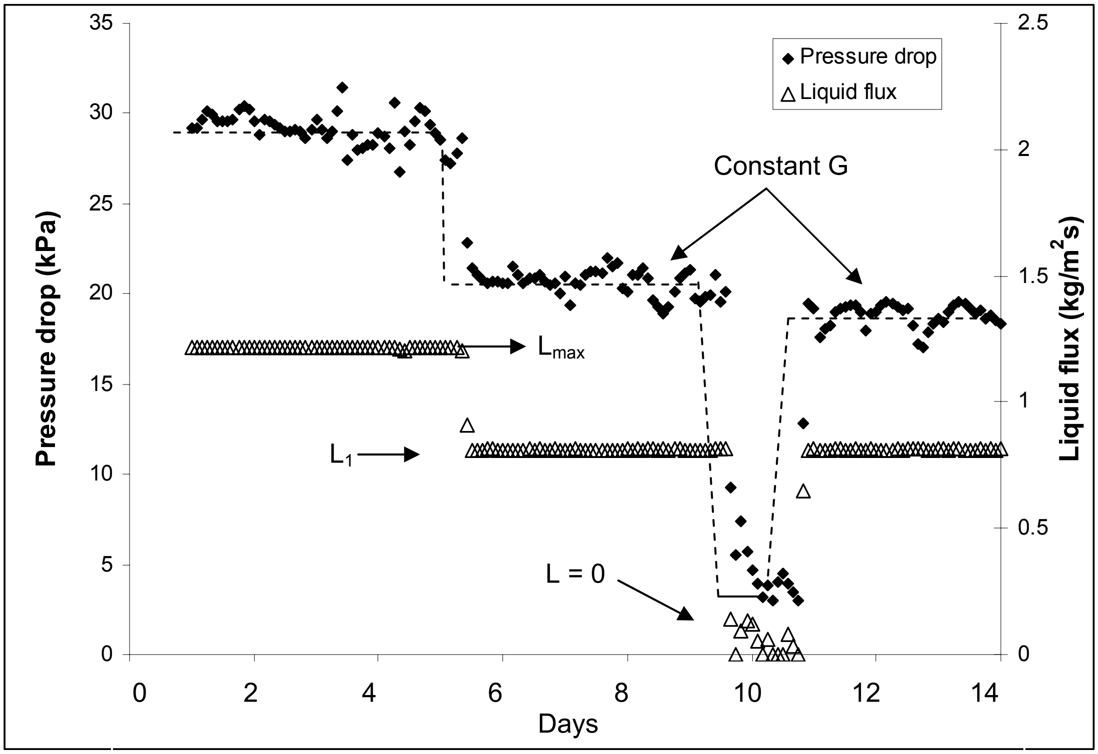

Figure 1. Hysteresis analysis strategy

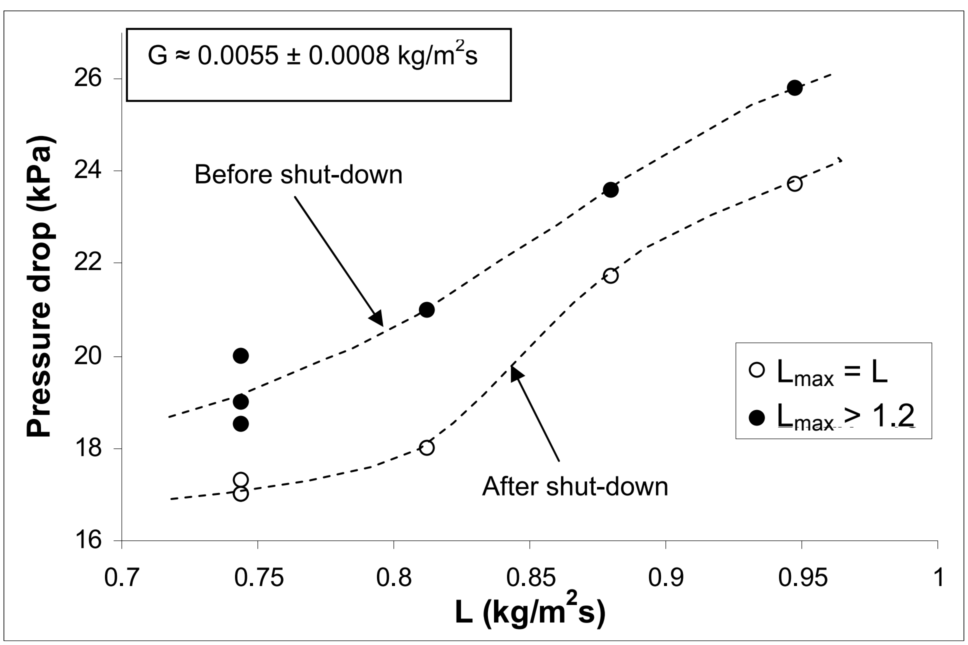

Figure 2. Experimental hysteresis behaviour at different liquid flow rates

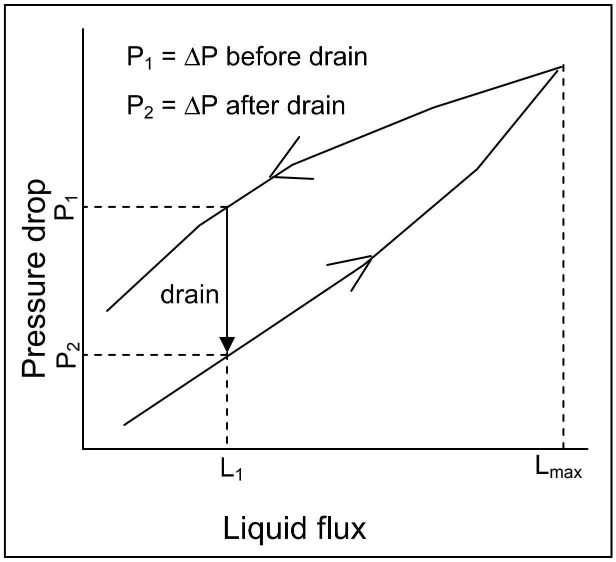

Figure 3. Qualitative hysteresis behaviour with liquid flow
(after Levec et al., 1986)

A severe increase in L is required for transition to the pulsing flow regime. The small increases observed here suffice only to increase the flow uniformity slightly and the reactor is essentially operating slightly above the lower hysteresis curve (i.e. in the Levec mode).

This study constitutes the first experimental evidence of hydrodynamic hysteresis in an industrial trickle bed reactor during regular operation. Despite low liquid surface tension, high-pressure operation, perfectly wettable particles and intermittent liquid feed interruptions the hysteresis exists. Since nearly all pressure drop correlations are based upon Kan mode data, there is a need to investigate the implications that multiple hydrodynamic states have on TBR performance.

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