Chemical Engineering Science 61 (2006) 7448–7462

Chemical Engineering Science

www.elsevier.com/locate/ces

Induced pulsing in trickle beds — Particle shape and size effects on pulse characteristics

M.E. Trivizadakis, D. Giakoumakis, A.J. Karabelas*

Department of Chemical Engineering, Aristotle University of Thessaloniki & Chemical Process Engineering Research Institute, P.O. Box 361,
6th km Harilaou-Thermi Road, GR 57001 Thermi/Thessaloniki, Greece

Received 1 December 2005; received in revised form 26 June 2006; accepted 13 August 2006
Available online 30 August 2006

Abstract

Periodic liquid feeding of the ON–OFF type is investigated — at sufficiently high frequencies to be classified as "fast" mode of induced pulsing — in the range of mean gas and liquid flow rates corresponding to the steady "trickling flow" regime. Two of the most common types of catalyst-support particles, i.e. porous *spherical* and *cylindrical* extrudates, are employed to study the imposed pulse characteristics. Detailed information is obtained, on the axial propagation and attenuation of pulses, from instantaneous, cross-sectionally averaged holdup measurements. Key fluid-mechanical parameters studied include, aside from dynamic holdup and pressure drop, pulse celerity and intensity, as a function of fluid feed rates (G, L) and liquid cyclic frequency. Similar published data, for 6 mm glass spheres, are employed for comparison; it is shown that, for the particles examined, particle size has a pronounced effect, but not as significant as that of particle shape. For particles of comparable size, the cylindrical shape is associated with much greater global dynamic holdup and pressure drop, and with increased pulse attenuation. Moreover, packed extrudates exhibit a significant increase of holdup in the axial direction, recently also observed in steady trickling flow. For spherical particles, both time-average holdup and pulse celerity are practically constant along the bed for fixed L, G. Pressure drop, global holdup and pulse celerity are not affected by cyclic liquid feeding frequency, for both spherical and cylindrical extrudate particles. Based on the pulse attenuation characteristics, for the three particle types examined, recommendations are made on preferred conditions for induced pulsing (from the fluid dynamics point of view) which would maximize benefits. Overall, it appears that spherical packings hold significant advantages over cylindrical extrudates of comparable size. Finally, in view of the observed significant decay of imposed pulses along the bed, care should be exercised to properly interpret data obtained in short laboratory reactors (where pulse attenuation is limited) for scale-up of the much longer industrial beds.

Keywords: Trickle beds; Induced pulsing; Particle shape; Size effects

1. Introduction

There is considerable research activity in recent years on *forced periodic operation* of *trickle bed reactors* (TBR) aimed at improving their performance. Usually periodic changes are imposed on either liquid flow rate or liquid feed composition. In the more common case of periodic liquid feeding, the liquid flow rate may vary between a minimum, usually called "base", and a maximum called "pulse" or "peak"; the base and pulse feeding periods, t_b and t_p, respectively, may be equal (symmetric pulse) or unequal. The cycle period, $T = t_b + t_p$, the ratio $s = t_p/T$ (called "split") and the base and peak flow rates, L_b and L_p, are employed to define periodic feeding. This study examines imposed symmetric pulses (i.e. $s = 0.5$) with zero "base" (ON–OFF type) feeding. Two general approaches are taken in dealing with periodic feeding, characterized by either *low*- or *high-frequency*. One of the original papers in this area by Haure et al. (1989) is typical of the former approach, which has led to positive results, in terms of improved selectivity and conversion (e.g. Dudukovic et al., 1999; Silveston and Hanika, 2002). These improvements, for exothermic gas-limiting reactions, are attributed to temperature increase during the gas-rich part of the cycle, which apparently facilitates vapor phase reactions, thus enhancing reaction rate. This type of cyclic liquid feeding, characterized by

---

* Corresponding author. Tel.: +30 310 996201; fax: +30 310 996209.
E-mail address: karabaj@cperi.certh.gr (A.J. Karabelas).

0009-2509/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2006.08.041

a rather long period T (e.g. of order 10 min or more) may be called "*slow*" mode. Research on the other type of imposed periodic operation (i.e. at relatively high frequency) commenced only fairly recently (e.g. Tsochatzidis et al., 1997; Boelhouwer et al., 1999) in efforts to "transfer", to some extent, the well-known benefits of natural pulsing flow (i.e. enhanced transport coefficients, improved liquid distribution) into the range of relatively low average liquid feed rates, i.e. into "trickling flow" regime.

Very few experimental studies have been published so far, examining possible benefits of operating with relatively high frequency cyclic feeding. Wilhite et al. (2003) reported data, obtained under gas-limiting, isothermal conditions, indicating better performance for steady trickling flow compared to induced pulsing flow with period $T = 30\,\mathrm{s}$, split $s = 0.33$, and various ratios of peak to base flow rates. These authors also reported results of simulations (using idealized square-wave functions, constant throughout the bed, for feeding rate, holdup, and liquid–solid mass transfer coefficient), which suggested that enhancement in reactor performance due to induced pulsing may be expected for liquid-limiting systems. Banchero et al. (2004) and Urseanu et al. (2004) employed the same reaction (hydrogenation of α-methyl styrene), the former under effectively isothermal and the latter under non-isothermal conditions. Banchero et al. (2004) used cyclic ON–OFF feeding of rather *short* periods; Urseanu et al. (2004) imposed periodic feeding with non-zero base flow and presented some data taken with relatively large periods. Both papers report improved performance of the TBR under induced pulsing flow compared to steady trickling; in the former study the improvement is attributed to mass transfer enhancement, but in the latter thermal effects appear to have played a major role as indicated by the measured periodic temperature variation. According to the definition of fast and slow mode of induced pulsing, subsequently provided in more detail, some of the experiments of Banchero et al. (2004) belong in the fast mode, whereas the rest as well as those of Wilhite et al. (2003) and Urseanu et al. (2004) are at the borderline between fast and slow mode. It will also be noted that in these studies no hydrodynamic parameters were measured and some values necessary for data interpretation or modeling (e.g. for holdup) were taken from the literature for *constant* feeding, due to lack of such data for induced pulsing.

To pursue this research activity further and to explore the potential for possible industrial applications, there is a need for systematic work to clarify first the fluid-mechanical characteristics of forced periodic liquid feeding. In fact, there is very limited work on the spatial and temporal evolution of induced pulses (e.g. Boelhouwer et al., 2002) as well as on the effect of cyclic feeding frequency on basic fluid-mechanical parameters of TBR operation. The need for such work is pointed out in a recent review on the subject (Nigam and Larachi, 2005). In a recent paper, Giakoumakis et al. (2005) present a systematic study of induced pulsing flow parameters (for ON–OFF feeding), including holdup variation and mean pressure drop as well as pulse celerity and intensity data, the latter employed to characterize in a quantitative manner the evolution of pulses as they move from bed-top to bottom. In that study, to facilitate data interpretation and to set the stage for subsequent work, *the existence in the packed bed, at any time, of at least one pulse* is proposed as a criterion to differentiate *fast* from slow mode of induced pulsing. According to this criterion, as discussed in more detail by Giakoumakis et al. (2005), in the fast mode the characteristic time period T of a pulse should be smaller than the ratio of bed height z over the pulse celerity C; i.e. $T \leqslant (z/C)$. In essence this condition poses a lower limit on the frequency of induced pulses for fast-mode pulsing operation. It is pointed out that at considerably smaller frequencies than the "limiting" one, the bed would operate at two different and alternating *transient states*, i.e. at a liquid-rich and gas-rich state; this situation is considered different than the fast mode of pulsing.

The study of Giakoumakis et al. (2005) was carried out using a bed packed with uniform 6 mm glass spheres. The present paper is a continuation of that work, using two types of porous alumina particles, i.e. nearly uniform 3 mm spheres and 1.5 mm dia. cylindrical extrudates. These are two of the most common catalyst-support particles in TBR applications. The comparable (equivalent) size of these particles allows an evaluation of *particle-shape* effects on fluid-mechanical characteristics; furthermore, by comparing the data from 6 and 3 mm spherical packings, the effect of *particle size* on induced pulsing operation can be assessed.

It is noted that the effect of particle shape has been very inadequately treated in the literature so far, even for the simpler case of steady trickling flow. A parallel study on the latter by Trivizadakis et al. (2006), using the aforementioned particle types, shows that particle-shape effects exert a very significant influence on the key fluid-mechanical parameters of trickle beds. It is, therefore, of importance to study forced periodic liquid flow through beds packed with these two kinds of particles, given also their practical significance. It will be added that most of this work was carried out in the context of a collaborative R&D project (CYCLOP, 1999–2003) supported by the European Commission, with the participation of several well-known industrial and research organizations, aimed at exploring potential industrial applications of induced pulsing.

In this presentation, experimental data are reported first on the effects of cyclic feeding frequency, and of particle shape and size, on common hydrodynamic parameters (holdup and pressure drop), and then on the typical characteristics of induced pulses (evolution, celerity, intensity). Pulse attenuation properties are discussed next. Based on these results, tentative recommendations are made regarding preferred conditions for possible applications to industrial TBR.

2. Experimental conditions

Tests are carried out in a cylindrical column of 0.14 m i.d. and 1.24 m height. The experimental equipment is described by Giakoumakis et al. (2005). Air and water are employed in the tests, covering the range of flow rates 2.4–6.13 $\mathrm{kg/m^2\,s}$ for liquid and 0–0.37 $\mathrm{kg/m^2\,s}$ for gas. Pressure drop measurements are made using three pressure transducers along the bed. Instantaneous holdup measurements along the column are obtained, using a conductance technique (Tsochatzidis et al.,