Chemical Engineering Science 128 (2015) 147–158

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Hydrodynamics of co-current two-phase flow in an inclined rotating tubular fixed bed reactor – Wetting intermittency via periodic catalyst immersion

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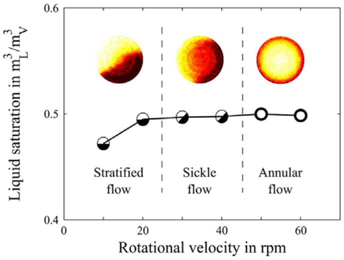
HIGHLIGHTS

- A new tubular reactor concept for process intensification is introduced.
- Reactor inclination and rotation allow for wetting intermittency via periodic catalyst immersion.
- Flow regimes can be adjusted by reactor inclination and rotation independent of liquid saturation.
- Lower pressure drops can be achieved in comparison to trickle bed reactors.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 29 September 2014
Received in revised form
2 February 2015
Accepted 8 February 2015
Available online 12 February 2015

Keywords:
Fixed bed hydrodynamics
Process intensification
Flow regimes
Multiphase flow
Gamma-ray computed tomography

ABSTRACT

The hydrodynamics of an inclined rotating tubular fixed bed reactor operated with gas–liquid co-current downflow are studied. Reactor inclination is applied to force phase segregation, while the superimposed rotation of the reactor results in a wetting intermittency via periodic catalyst immersion. The fixed bed is clamped to avoid abrasion of the catalyst. The inclined rotating reactor is presented as a new reactor concept for process intensification of heterogeneous catalytic reactions requiring enhanced mass transfer of the gaseous phase and partial catalyst wetting. Four different flow regimes with stratified, sickle, annular and dispersed flow patterns are determined experimentally by applying a compact gamma-ray computed tomography system. The effects of (i) gas and liquid superficial velocities, (ii) inclination angle and rotational velocity of the reactor and (iii) physico-chemical properties of the liquid phase on the occurrence of the flow regimes are investigated. The results of these investigations are illustrated with flow maps. In addition, pressure drop and liquid saturation depending on the operating conditions are shown.

1. Introduction

Trickle bed reactors (TBR) are widely applied in the chemical industry for heterogeneous catalytic reactions, e.g. for selective hydrogenation of olefins, oxidation of glucose, hydrodesulphurization of crude oil and the oxidation of phenol (Dudukovic et al., 2002). They are operated in co-current gas–liquid downflow mode with the liquid phase trickling slowly through the fixed bed

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http://dx.doi.org/10.1016/j.ces.2015.02.008

formed of randomly filled catalyst particles. The trickle flow regime is preferred to provide high residence times to compensate for slow intra-particle diffusion rates (Ranade et al., 2011), to reduce phase interactions and thus, to lower pressure drop. However, these reactors exhibit shortcomings such as liquid maldistribution with only partial utilization of the catalyst fixed bed. The mass transfer of the gas phase to the active sites of the catalyst is low due to stagnant liquid films at the surface of the catalyst that act as a barrier (Khadilkar et al., 1996). Furthermore, maldistribution and partial wetting in the trickle regime lower significantly the heat transfer with the risk of hotspot formation (Beaudry et al., 1987). Operating the reactor in the pulse flow regime can homogenize the liquid distribution and enhances the heat removal to silence hotspots; however, this is accompanied by the expense of higher pressure drops and shortened residence times.

Liquid flow modulation was proposed as an unsteady-state operation concept to enhance mass transfer, gas–liquid distribution and removal of reaction heat, which allows at the same time control of the mean residence time by tuning the liquid pulses with respect to frequency, amplitude and shape (Silveston, 1987). Mostly, liquid flow modulation with a constant gas flow and a binarized (*on*/*off*) liquid flow was applied to enhance the reaction rates of gas–limited reactions. Basically, the packing drains during the *off* period, the liquid films at the catalyst surface are thinned and the ratio of the wetted surface decreases, which in turn enhances the accessibility of the catalyst surface to the gas phase and boosts the reaction. In the subsequent *on* phase, the liquid phase removes the reaction heat and the reaction products are quickly swept away from the vicinity of catalyst sites (Castellari and Haure, 1995).

A recent review by Atta et al. (2014) summarizes the systematic studies on flow rate modulation searching for optimal operating conditions. This strategy has been proven to enhance the performance of different processes compared to steady-state operation. For example, Lange et al. (1994) observed an increase in the time-average conversion up to 15% for the hydrogenation of alpha-methylstyrene (AMS) to cumene with a Pd/alumina catalyst, while Borren et al. (2010) found a relative increase in the gas–liquid mass transfer up to 38%.

Nonetheless, the benefits of periodic operation seem not yet sufficiently convincing to force its industrial implementation, which is probably due to additional demands on the control infrastructure in continuously operated production environments with upstream and downstream processes. Although flow modulation was discussed to enhance liquid maldistribution and catalyst wetting (Khadilkar et al., 1999), a recent study with local heat-probes and collector techniques revealed contradicting results (Borremans et al., 2004). Furthermore, it was experimentally observed that the liquid pulse intensity of a binarized liquid inlet flow decays rapidly downstream the reactor (Boelhouwer et al., 2002; Schubert et al., 2010b). Dietrich et al. (2005) investigated the hydrogenation of AMS by a particle-scale model with dynamic wetting of the particle surface. For the attenuation of the pulses along the reactor, they found a conversion enhancement of only 17% compared to 29% if assuming ideal binarized pulses. Thus, it appears that the beneficial impact of periodic operating in large scale reactors vanishes after some entrance length. An additional simulation study by Dietrich et al. (2012) on the AMS hydrogenation at particle-scale considered local liquid holdups and revealed that a pronounced performance enhancement can only be achieved for an elevated amount of dynamically changing liquid holdup. Accordingly, the authors suggested to develop reactor concepts for a forced manipulation of the local liquid distribution as an alternative to feed modulation.

A new process intensification strategy, which does not require inlet flow rate modulation, was proposed by Schubert et al. (2010a). It was shown that tilting the reactor induces partial phase segregation due to gravitational forces, thus minimizing the liquid-rich area and pressure drop. Imposing pulse flow caused occasional passages of liquid-rich pulses over the entire bed cross-section that renewed the liquid fraction retained in the gas–rich area of the packing and removal of reaction heat and products. This bed obliquity was unveiled as a new artifice to pulse flow modulation with possible prospects for catalytic reactions requiring antagonistically high-interaction regime mass transfer and partial catalyst wetting, which would be beneficial for gas–limited catalytic reactions. The frequency of pulse passage can be specified via flow rate assignments and bed inclination adjustment. However, the desired pulse inception in the inclined reactor configuration requires higher flow rates compared to TBRs, thus lowering the reactant mean residence time in the reactor and limiting the applicability of this concept.

An alternative reactor configuration for process intensification, which potentially brings advantages over the existing configurations, is the inclined rotating tubular fixed bed reactor shown schematically in Fig. 1. The superimposed rotation of the previously introduced inclined reactor results in a catalyst wetting intermittency via periodic immersion of the catalyst packing into the liquid phase, which is accumulated nearby the reactor bottommost wall area. The catalyst is fixated in the tubular reactor by means of mesh screens to avoid attrition of the particles. The periodic immersion refreshes the liquid at the catalyst surface and silences efficiently hotspots, while the drained packing section provides excellent access to the gas phase, thus favoring this concept for process intensification of exothermic gas–limited heterogeneous catalytic reactions. Further possible advantages of this new reactor concept are (i) an extended catalyst lifetime due to the even utilization of the fixed bed, (ii) a stable wetting intermittency along the whole reactor length at moderate flow rates without the need for inlet flow modulation and (iii) a higher flexibility for liquid residence time adjustment via control of inclination angle and rotational velocity. The careful adjustment of the flow regime is the key to success of the claimed benefits from a reaction perspective. However, currently detailed knowledge on the hydrodynamics for gas–liquid flows in such an inclined rotating tubular fixed bed reactor is not available.

Rotating tubular fixed beds are so far only known from horizontally rotating biological contactors, where axially stacked circular disks are partially immersed in a liquid reservoir (Patwardhan, 2003). Further biological contactor configurations consist of horizontally rotating cages, which are completely or partially filled with random packings (Mathure and Patwardhan, 2005; Patwardhan, 2003). In addition, there exists a vertically rotating TBR concept for biogas purification (Gai et al., 2001).

A basic flow pattern study in a rotating horizontal packed bed was performed by Karapantsios et al. (1993) with a ring conductance probe. The authors observed an increase of the liquid spreading in the cross-section of the fixed bed with increasing rotational velocity, starting with a stratified liquid distribution for low rotational velocities, turning over to an intermediate sickle-shaped liquid distribution that eventually transformed into an annular liquid distribution for the highest rotational velocity. However, the effect of a superimposed reactor inclination is not yet addressed.

It is worth mentioning that other rotating multiphase reactor concepts exist such as the spinning disc reactor (Oxley et al., 2000; Visscher et al., 2012) or the rotating packed bed reactor (Burns et al., 2000; Ramshaw, 1993; Llerena-Chavez and Larachi, 2009) also known as high-gravity (HiGEE) reactor. Here, elevated shear forces are generated via high centrifugal fields in order to increase the specific mass transfer area, to improve mixing, to overcome flooding of the packed bed and to avoid countercurrent flow limitations. Compared to the HiGEE reactors, where mean accelerations of up to 500*g* can be achieved (Burns et al., 2000), the inclined rotating tubular fixed bed reactor introduced here rotates at moderate rotational velocities up to 60 rpm for a distinctive catalyst wetting intermittency.

In this work, the hydrodynamics of an inclined rotating tubular fixed bed reactor operated with gas–liquid co-current downflow are