Chemical Engineering Science 64 (2009) 3329 – 3343

Contents lists available at ScienceDirect

Chemical Engineering Science

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

New insights into transient behaviors of local liquid-holdup in periodically operated trickle-bed reactors using electrical capacitance tomography (ECT)

Guozhu Liu[a,*], Jiang-an Lan[b], Yanbin Cao[a], Zibin Huang[b], Zhenmin Cheng[b,*], Zhentao Mi[a]

[a]Key Laboratory of Green Chemical Technology of Ministry of Education (MOE), School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, China
[b]State Key Laboratory of Chemical Engineering, East China University of Science and Technology, Shanghai 200237, China

A R T I C L E   I N F O

Article history:
Received 17 December 2008
Received in revised form 17 March 2009
Accepted 3 April 2009
Available online 14 April 2009

Keywords:
Trickle-bed reactor
Electrical capacitance tomography
Periodic operation
Liquid holdup
Liquid distribution
Maldistribution
Transient response
Imaging

A B S T R A C T

Transient behavior of local liquid-holdup of air–kerosene fluids in periodically operated trickle-bed reactor (TBR) is investigated in an acrylic column (140 mm ID and a height of 980 mm) packed with 3.6 mm glass spheres using a nonintrusive technique of electrical capacitance tomography (ECT). Local liquid-holdups determined from ECT images of normalized permittivity are experimentally calibrated under the steady-state operation with that from the classic drainage method. The instantaneous ECT images are captured at several axial positions along the column for the periodically operated TBR with slow-mode. The effect of periodic operation parameters (split, period, and time-average flow rate) on the instantaneous profiles of local liquid-holdup is firstly examined compared with the previous results. Transient variations of radial distributions of liquid and their maldistribution factor are calculated and further analyzed to provide more liquid distribution information under periodic operations.

1. Introduction

Trickle-bed reactor (TBR) is broadly used in the petrochemical and petroleum industries (the processes of hydrodesulphurization, hydrocracking, catalytic dewaxing of gas oil and lube oils, etc.), as well as biochemical processes. Due to their large production volumes involved, there is a considerable economic incentive to improve and intensify TBR performances (Dudukovic et al., 1999, 2002; Nigam and Larachi, 2005; Dudukovic, 2007). A novel process intensification technology on the TBR performance, periodic (or unsteady-state) operation, has received a special attention over the last decade in view of its remarkable improvement on the reaction rate and operational life (Khadilkar et al., 1999; Silveston and Hanika, 2002; Lange et al., 2004; Gancarczyk and Bartelmus, 2007; Hamidipour et al., 2007; Liu et al., 2008).

The most important feature of the periodic operation is to operate TBR in a transient mode by periodically modulating the gas or liquid phase supply, which leads to non-steady state fluid hydrodynamics, chemical reaction and heat effects associated in a three phase reactor (Khadilkar et al., 1999). The potential for performance enhancement arises from periodically renewal of wetted catalyst surface, more intensive interaction between gas and liquid phases, the competition between the gas and liquid phases in supplying reactants to the catalyst, as well as the interaction between fluid dynamics, catalyst wetting, and reaction. So far, this performance enhancement has been observed and verified for many reactions by many groups with several modulation strategies, as shown in several valuable surveys of the periodic operation literatures (Silveston and Hanika, 2002; Schubert et al., 2006; Liu et al., 2008). Recently, Tukac et al. (2007) reported an increase of pilot reactor productivity of styrene hydrogenation by 30% for periodic operation in comparison with steady-state one, which is an encouraging achievements for the promising industrial applications of periodic operation. However, complex interactions between hydrodynamics, mass transfer and reaction phenomena bring an almost insurmountable obstacle in the design and industrial scale-up of periodically operated TBR.

Liquid holdup and the liquid distribution over the particles play a crucial role in determining many key design parameters, such as catalyst wetting efficiency, radial heat evacuation, gas–liquid mass transfer of gaseous reactant, solvent evaporation, etc. (Brkljac et al., 2007; Cheng et al., 2001; Cheng et al., 2007b; Zhou et al., 2004; Zhou et al., 2005). In this view, the impact of liquid modulation needs to be addressed in detail. In general, two modes of unsteady-state operation can be distinguished in view of the cycle period: fast- and slow-modes. So far much work has been done addressing on the instantaneous variation of local liquid-holdup under the periodic operation with the two modes on over past decade. Xiao et al. (2001)

* Corresponding authors. Tel.: +86 22 2789 2357; fax: +86 22 2740 2604 (G. Liu), Tel.: +86 21 6425 3529; fax: +86 21 6425 3528 (Z. Cheng).
E-mail addresses: gliu@tju.edu.cn (G. Liu), zmcheng@ecust.edu.cn (Z. Cheng).

0009-2509/$ - see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2009.04.008

studied the effect of gas-induced pulsing flow on the hydrodynamics with conductive techniques, and observed more uniform radial and axial liquid distributions but a significant decrease in the liquid holdup during gas-induced pulsing flow. Boelhouwer et al. (1999, 2001, 2002) investigated liquid holdup and the pulse properties during the fast- and slow-mode cyclic operation flow compared with the spontaneous pulsing flow regime. They observed that the pulse duration in spontaneous pulsing flow is longer than in induced pulsing flow, and that the shock waves decay while moving down the bed. Borremans et al. (2004, 2007) observed only marginal effects of periodic operation on bed cross-wise liquid flow distribution with a collector, and that the cyclic variation of the liquid mean residence time is a function of split ratio, cycle period, and bed height in liquid cyclic operation. Giakoumakis et al. (2005) studied the effect of cyclic frequency (i.e., reciprocal of cycle period), and gas and liquid flow rates on the dynamic liquid hold-up under the fast-mode operation with several conductimetric probes at various locations along the packed bed, and obtained quantitative information is on the axial propagation and attenuation of induced pulses. They also proposed a characteristic attenuation factor as a phenomenological treatment of the pulse decay process and an expression to correlate pulse celerity data. Trivizadakis et al. (2006) and Trivizadakis and Karabelas (2006) provided some useful detailed information on the axial propagation and attenuation of pulses under the fast-mode cyclic operation, from instantaneous, cross-sectional averaged holdup measurements, and observed lower liquid pulse attenuation rate and spread of local mass transfer coefficients with spherical particles as compared to cylindrical extrudates. Bartelmus et al. (2006), Gancarczyk and Bartelmus (2007) and Gancarczyk et al. (2007) observed the shift of trickle to—induced pulsing transition boundary to lower superficial liquid velocities with increasing liquid viscosities by the fast- and slow-mode of base-impulse periodic liquid feeding, and developed a dimensionless equation to correlate dynamic liquid hold-up for the liquids differing in physicochemical properties. Aydin et al. (2006, 2007a,b, 2008) and Aydin and Larachi (2008) studied temporal variations of the liquid holdup in a mini-pilot scale trickle-bed reactor cold-mockup for both Newtonian and non-Newtonian liquids using a conductimetric technique with probes that mimic the packing. They found the aggravation in the collapse of the liquid holdup pulses with increasingly temperatures and pressures. Brkljac et al. (2007) developed a model based on the relative permeability concept to predict the two-phase pressure drop and dynamic liquid hold-up during max/min and on/off periodical operation. They successfully predicted unsteady-state hydrodynamics by selecting suitable permeability parameters. Ayude et al. (2007) studied temporal variations of the liquid holdup in a mini-pilot scale trickle-bed reactor under ON-OFF liquid flow modulation strategy with a conductimetric technique, using probes that mimic the packing. The effects of the bed depth and the cycling parameters on the liquid holdup modulation were examined for a wide range of conditions. They also developed an exponential function for slow and intermediate cycle periods to reconstruct of the time dependence decay of local liquid-holdup along the column. Indeed, all of those works make great contributions in understanding the transient behaviors of liquid holdup under the periodic operations. Unfortunately, few of them address the instantaneous liquid distribution under the cycling operations due to the limitation of the conductimetric and collector techniques for the determination of local liquid-holdup and its distribution. To further understand the complex hydrodynamic phenomenon of TBR under liquid flow modulation, further researches are still required with a novel measurement technique to obtain instantaneous liquid holdup and its distributions.

In recent years, applications of process tomography as a robust non-invasive tool for direct analysis of the characteristics of multiphase flows have increased in number. Some examples of techniques allowing cross-sectional imagining of the liquid flow in trickle beds are the tomographic measurements by photon attenuation (X- and γ-ray tomography) and magnetic resonance imagining (Basavaraj and Gupta, 2004; Basavaraj et al., 2005; Gladden, 2003, 2006, 2007; Koptyug et al., 2004; Mantle and Sederman, 2003; Toye et al., 2005; van der Merwe et al., 2007; Wu et al., 2007). These techniques give access to the phase distribution in a section of the reactor with higher spatial resolution. However, the major disadvantage of photon attenuation measurements and magnetic resonance imagining are their high cost and safety considerations. In addition, the speed of the technique to capture real-time data of highly fluctuating flow systems may be one of the most important concerns for the purpose of imagining TBR under periodic operation. For example, X-ray computer tomography (X-ray CT) only could capture up to several image frames per second. In this regard, electrical capacitance tomography (ECT) is considered to be the most powerful tool among other available tomography techniques because of its high-speed capability (100 frames per second), low construction cost, high safety and suitability for large vessels (Bolton et al., 1999; Makkawi and Ocone, 2007; Ostrowski et al., 2000; Tibirna et al., 2006; Wang et al., 2008; Warsito and Fan, 2003). Regardless of the successful application of ECT in the three-phase fluidization bed reactor, there are only several primary applications in studying the hydrodynamics of TBR due to the high difficulty in the ECT image reconstructions (Cheng et al., 2007a; Reinecke and Mewes, 1996, 1997; Tibirna et al., 2006).

The objective of this work is to study the transient behaviors of the liquid holdup of air–kerosene system in a periodically operated TBR packed with glass spherical particles with ECT. Steady-state liquid holdups are measured by the classic drainage methods to calibrate the ECT response. The instantaneous ECT images at several axial positions are captured for the TBR modulated with the slow-mode, and the influences of bed height and cycling parameters on the transient behaviors of liquid holdup are addressed. Furthermore, the transient radial liquid distributions and maldistribution factor are further calculated form the ECT images to show the liquid distribution under periodic modulation.

2. Experimental

2.1. Trickle-bed reactor setup

A schematic diagram of the experimental apparatus is shown in Fig. 1. The setup consists of an acrylic column of 140 mm ID and a height of 980 mm packed with the glass spherical pellets (d_p = 3.5 mm). The pellets are supported by an acrylic sieve-tray at the bottom of the column. Beneath the sieve-tray, there is a gas–liquid separator that vents the gas and returns the liquid to a reservoir. A coil heat exchanger is placed in the reservoir to ensure the constant temperature of liquid. The liquid is fed from a reservoir with a peristaltic pump controlled by a rotor velocity regulator, which is commanded by a programmable logic controller. Gas flow rate is measured by a rotameter and regulated by means of a needle valve. Liquid and gas enters the column from the top through a perforated plate distributor. Kerosene is selected as the liquid phase fluids due to its similarity to the hydrocarbon streams subjected to hydrotreating, as well as its chemical stability and its low vapor pressure. The details of the reactor and the packing properties are given in Table 1.

Before starting the experiments, the column is flooded with liquid to ensure complete wetting of the particles and reproducible observations. Then, adjusts the liquid flow rate to the one corresponding to the wet period considering the desired mean liquid velocity, u_L, and set the cycling parameters and gas flow rate. Afterwards, the system is operated at the desired cycling strategy for five cycle periods to reach an invariant cycling state before capturing ECT images. All the experiments are performed with an