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Liquid backmixing in an inclined rotating tubular fixed bed reactor – Augmenting liquid residence time via flow regime adjustment

Hans-Ulrich Härting [a], Ronny Berger [a], Rüdiger Lange [b], Faïçal Larachi [c], Markus Schubert [a,*]

[a] Helmholtz-Zentrum Dresden-Rossendorf, Institute of Fluid Dynamics, Bautzner Landstraße 400, 01328 Dresden, Germany
[b] Technische Universität Dresden, Institute of Process Engineering and Environmental Technology, 01062 Dresden, Germany
[c] Department of Chemical Engineering, Laval University, Québec, G1 V 0A6, Canada

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ABSTRACT

The liquid residence time and the backmixing in an inclined rotating tubular fixed bed reactor operated with gas–liquid co-current downflow are studied experimentally. This novel reactor concept is introduced to extent the process intensification strategies of chemical multiphase reactors. The intermittent catalyst immersion due to rotation induces a continuous refreshment of the liquid at the catalyst surface and enhances the access of the gas phase to the catalyst in the drained section of the fixed bed. Depending on inclination angle and rotational velocity, different flow regimes are observed. In particular, the flow regimes with stratified gas–liquid flow can be utilized to enhance the performance of the reactor for heterogeneous catalytic reactions.

The backmixing study is based on the method of the imperfect tracer pulse and the propagation of the tracer is measured by low-intrusive wire-mesh sensors. Compared to conventional trickle bed reactors, liquid residence time and axial dispersion are increased by the inclination and rotation. The effects of reactor inclination angle and rotational velocity as well as of particle size and liquid superficial velocity on the liquid backmixing in the inclined rotating tubular fixed bed reactor are shown in detail.

1. Introduction

Trickle bed reactors (TBR) are widely applied for heterogeneous catalytic multiphase reactions, such as the desulphurization of heavy crude oil or the hydrogenation of olefins [1]. Liquid and gas phase are fed at the top of the fixed bed, which is formed by randomly packed catalyst particles. TBR are usually operated at low liquid flow rates resulting in rivulet and film flow at the particle surface to ensure sufficient residence times of the reactants and at elevated pressure to compensate for low diffusivity and solubility of the gas phase [2]. However, the performance of TBR suffers from partial utilization of the fixed bed due to poor liquid irrigation and from stagnant liquid films at the catalyst surface that act as a barrier for the mass transfer of the gaseous reactant to the active sites of the catalyst [3].

For process intensification, the forced periodic variation of the liquid inlet flow was proposed to enhance the mass transfer, to homogenize the cross-sectional liquid distribution and to control hotspots [4]. Several studies revealed the benefits of periodic flow rate modulation like an increase in the time-averaged reaction rates [5], which is mainly attributed to higher mass transfer rates [6] and the potential to decelerate catalyst deactivation by reducing its overheating [7]. A drawback of this operation strategy is the intensity decay of the liquid pulses downstream the reactor [8,9], especially at elevated temperature and pressure to which most industrial applications are subjected [10]. Thus, the beneficial impact of this approach in large scale reactors might be reduced.

The inclined rotating tubular fixed bed reactor is an alternative and novel reactor concept for process intensification of preferably exothermic gas-limited heterogeneous catalytic reactions. It is based on the adjustment of a favorable gas–liquid distribution and constitutes an advanced approach of flow rate modulation. The principle of the new reactor concept is shown in Fig. 1a. The reactor

* Corresponding author. Tel.: +49 (0) 351 260 2627.
E-mail addresses: h.haerting@hzdr.de (H.-U. Härting), r.berger@hzdr.de (R. Berger), ruediger.lange@tu-dresden.de (R. Lange), faical.larachi@gch.ulaval.ca (F. Larachi), m.schubert@hzdr.de (M. Schubert).

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Nomenclature

a Constant
c Concentration (mol/L)
d Diameter (m)
D Dispersion coefficient (m^2/s)
g Transfer function (1/s)
i,j Pixel index
L Axial distance between the wire-mesh sensors (m)
n Rotational velocity (rpm)
p Pressure (bar)
s Complex variable
t Time (s)
u Fluid superficial velocity (m/s)
U Voltage (V)
z Axial coordinate (m)

Greek symbols
α Inclination angle (°)
ε Liquid holdup (m_L^3/m_V^3)
σ Conductivity (S/m)
τ Residence time (s)
ϑ Temperature (°C)

Subscripts
ax Axial
G Gas
L Liquid
P Particle
R Reactor

inclination induces partial phase segregation due to gravity, whereas the superimposed rotation of the inclined reactor results in a wetting intermittency via periodic immersion of the fixed bed into the liquid phase, which is accumulated at the reactor bottommost wall area, see Fig. 1b. The periodic immersion refreshes the liquid at the catalyst surface, which could dampen hotspots efficiently, while the opposite drained packing section provides enhanced access of the gas phase to the active sites of the catalyst. Further possible advantages are an extended catalyst lifetime due to complete and even utilization of the fixed bed and a stable wetting intermittency along the whole reactor length at moderate flow rates without the need for inlet flow modulation. A comparison between the previous periodic operation via flow rate modulation and the novel reactor concept suggests itself. The former is a temporal modulation strategy, whereas the latter aims at the modulation in the spatial domain. It is expected that the combined control of the inclination angle and rotational velocity allows for a more flexible adjustment of liquid residence time and backmixing at fixed flow rates to improve the conversion and selectivity of chemical reactions.

The experimental estimation of the residence time distribution (RTD) is based on stimulus-response techniques and the evaluation of differential macro-mixing models, like the axial dispersion plug flow model (ADM) presented in early works of Danckwerts [11], van der Laan [12] and Aris [13]. Based on the assumption of a Fickian type of dispersion, the ADM provides a theoretical framework for the axial dispersion coefficient and lumps all deviations from the ideal plug flow in this parameter. Using time-domain curve-fitting techniques, the mean residence time and the axial dispersion coefficient are obtained. The analysis of the RTD allows for diagnosis of reactor ills like dead spaces, short-circuiting or internal recirculation. Given knowledge of the RTD and a chemical reaction with first-order kinetics, the conversion of any reactant can be obtained analytically [14]. This enables the direct evaluation of a chemical reactor regarding the increase in reactor volume due to the decreasing conversion caused by axial dispersion. Furthermore, the liquid holdup as another crucial hydrodynamic parameter of multiphase reactors can also be estimated from RTD data as demonstrated by Larachi et al. [15].

Detailed knowledge on the residence time distribution and the axial dispersion in the inclined rotating tubular fixed bed reactor is currently not available. Therefore, this study is proposed as a contribution to highlight the potential opportunities of this novel reactor concept and to widen the arsenal of process intensification strategies of chemical multiphase reactors via periodic flow rate modulation.

2. Experimental setup and data processing

2.1. The inclined rotating tubular fixed bed reactor

The experimental setup is shown in Fig. 2a together with the wire-mesh sensor (WMS) for the detection of the tracer pulses in Fig. 2b and the multipoint gas–liquid distributor for the TBR configuration in Fig. 2c. The segmented reactor has an inner diameter of 100 mm and a total length of 1140 mm. Two wire-mesh sensors are flange-mounted between the reactor segments at 310 mm and 930 mm, respectively, downstream of the reactor top. Further details on the measurement principle of the WMS and the stimulus-response technique are given in the next section. The fixed bed consists of spherical glass particles of 2.5 mm and 4.0 mm diameter, respectively, which are clamped between two mesh

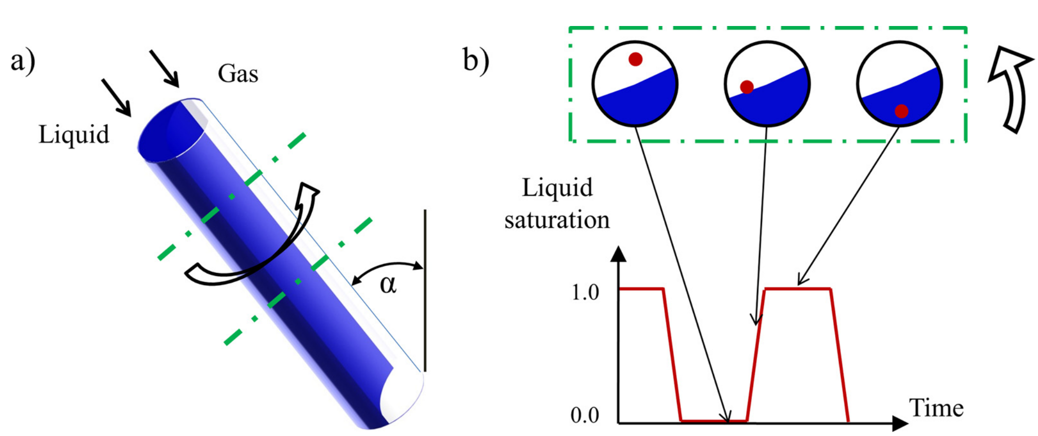

Fig. 1. (a) Principle of the inclined rotating tubular fixed bed reactor, (b) liquid saturation of the particle during rotation as function of time.