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A novel inclined rotating tubular fixed bed reactor concept for enhancement of reaction rates and adjustment of flow regimes

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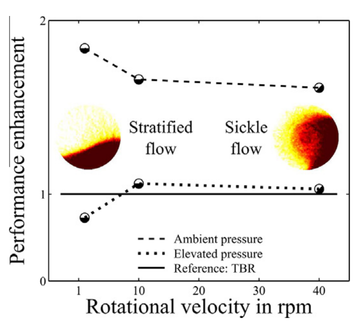
HIGHLIGHTS

- A tubular reactor concept with an inclined and rotating fixed bed is presented.
- Performance enhancement is achieved at severe limitations of the gas mass transfer.
- Reactor inclination and rotation allow for a flexible adjustment of flow regimes.
- The stratified flow regime enhances catalyst utilization and accessibility.

GRAPHICAL ABSTRACT



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ABSTRACT

The inclined rotating tubular fixed bed reactor has been introduced recently as a novel reactor concept for multiphase processes, especially for heterogeneously catalyzed gas–liquid reactions with a mass transfer limitation of the gas phase (Härting et al., 2015; DOI: http://dx.doi.org/10.1016/j.ces.2015.02.008). It is based on the adjustment of a beneficial gas–liquid distribution in the cross section of the fixed bed that allows for complete utilization of the fixed bed accompanied by periodic wetting and draining of the catalyst.

The hydrodynamics in gas–liquid co-current downflow are studied by applying a compact gamma-ray computed tomography system for different organic liquids and gas phase properties as well as various fixed bed packing materials. Four different flow regimes with stratified, sickle, annular and dispersed flow patterns are identified. Pressure drop and liquid saturation are presented as a function of reactor inclination and rotation.

Inclination of the reactor is applied to force phase separation and the superimposed rotation of the clamped fixed bed results in a favorable wetting intermittency via periodic catalyst immersion. A significant rate enhancement of the hydrogenation of alpha-methylstyrene to cumene under severe limitations of the mass transfer of the gas phase is observed in the novel reactor concept at separated flow conditions. In addition, the potential for an efficient quenching of hotspots for exothermic reactions is demonstrated.

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Nomenclature

A	area (m^2)
a	Redlich–Kister-parameter (–)
c	molar concentration (mol L^{-1})
D	reactor diameter (m)
E	enhancement factor (–)
h	filling level of circular segment (m)
L	reactor length (m)
n	reactor rotational velocity (rpm)
N	amount of substance (mole)
p	pressure (MPa)
Q	volumetric flow rate (L s^{-1})
R	reaction rate (mol s^{-1})
r	radius of reactor cross section (m)
S	split (–)
t	time (s)
T	absolute temperature (K)
u	fluid superficial velocity (m s^{-1})
v	sonic velocity (m s^{-1})
X	yield (–)
x	molar fraction (mole $mole^{-1}$)

Greek symbols

α	reactor inclination angle against the vertical (°)
β	liquid saturation ($m_L^3 m_V^{-3}$)
γ	reactant limitation criterion (–)
Δp	pressure drop (kPa)
η	ratio between liquid-solid and gas-solid mass transfer coefficient (–)
θ	center angle (°)
ϑ	temperature (°C)

Subscripts

0	inlet
AMS	alpha-methylstyrene
C	cumene
CS	circular segment
drain	drained
dyn	dynamic
G	gas
H_2	hydrogen
HD	hydrodynamics
imm	immersed
L	liquid
LS	liquid-solid
on	liquid on duration
Mix	binary mixture
off	liquid off duration
p	period
R	reaction
sat	saturation
TBR	trickle-bed reactor
tot	total
v	void

1. Introduction

The trickle bed reactor (TBR) is a common multiphase reactor type in the chemical, biochemical and petrochemical industry. Typical processes are the wet oxidation of alcohols, hydrogenations and refining of petroleum such as hydrodesulphurization and hydrodemetallization of crude oil [1,2]. Gas and liquid flow co-currently downwards through a random fixed bed of catalyst particles at moderate superficial velocity of the liquid, hence the eponymous trickling flow arises. The resulting high liquid residence time counterbalances the slow intraparticle diffusion rates. The low gas–liquid–solid interactions result in moderate pressure drops but also in rather low heat and mass transfer rates [3]. In particular, liquid films at the surface of the catalyst act as a barrier for the mass transfer of the gas phase to the active sites of the catalyst [4]. Thus, TBR are often operated at high temperature and elevated pressure to increase the reaction rates, to ensure sufficient solubility of gas in the liquid phase and to mitigate catalyst deactivation caused by hydrogen depletion [5]. Furthermore, selection and positioning of the liquid distributor and re-distributors is crucial in TBR to avoid severe loss of operation capacity that is caused by liquid maldistribution [6]. In order to increase mass and heat transfer rates and consequently conversion, TBR can be operated in the natural pulse flow regime [7,8]. This can be achieved at high gas and liquid superficial velocities given a confined diameter of the reactor, i.e. sufficient pressure drop [9]. Due to the higher fluid superficial velocities, the pressure drop in the pulse flow regime increases, whereas the effect of shortened residence time is to be evaluated according to the kinetics of the reaction system.

Alternatively, flow rate modulation has been proposed to improve the mass transfer of the gas phase and to subsequently enhance the overall reaction rate. Here, the liquid feed at the inlet of the reactor is periodically cycled between two flow rates. This binary modulation can be implemented by cyclic interruptions of the liquid flow rate in an *on*/*off*-mode or by cycling between a low and a high liquid flow rate, also referred to as *base*/*peak*-mode. The *split* (S), see Eq. (1), defines the ratio between the duration of the *on* or *peak* time, respectively, and the period length, see Eq. (2). Periodic operation was studied in lab-scale reactors for several processes with period times ranging from 3 s (*fast mode*) to a few minutes (*slow mode*):

$$S = \frac{t_{on}}{t_{on} + t_{off}} \tag{1}$$

$$t_p = t_{on} + t_{off} \tag{2}$$

The reaction system used in many of these studies and also in this study is the Pd-catalyzed hydrogenation of alpha-methylstyrene (AMS) to cumene. This system is representative for exothermic noble metal catalyzed hydrogenations involving low-volatility liquids. It exhibits severe limitations of the mass transfer of the gaseous reactant due to the low solubility of hydrogen at low pressures [10] and the typically applied excess of AMS. The reactions system is widely used as a benchmark system to study the TBR performance enhancement at periodic operation conditions as summarized in Table 1.

Castellari and Haure [11] and Gabarain et al. [12] reported a performance enhancement, i.e. $E = X_{unsteady}/X_{steady}$, of 4.5 as a result of *on*/*off* liquid cycling in a *slow mode* operation at ambient pressure. They observed temperature oscillations with a rise of up to 40 K in a 1.5 cm thin catalyst layer. Lange et al. [13,14] studied the *on*/*off* as well as the *peak*/*base* cycling mode in a similar temporal manner at ambient pressure and observed an enhancement of 1.4 independent from the cycling mode applied. Urseanu et al. [15] reported a performance enhancement of the same magnitude at elevated pressure in the *peak*/*base* cycling mode with a *peak*