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Comparison of structured trickle-bed and monolithic reactors in Pd-catalyzed hydrogenation of alpha-methylstyrene

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This paper is dedicated to Professor Dr.-Ing. habil. Ruediger Lange on the occasion of his 60th birthday, March 14th 2011.

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ABSTRACT

The so-called structured trickle-bed reactors (STBRs) represented by the combination of a tubular reactor and a structure with straight flow channels packed with catalyst particles have been tested for carrying out multiphase reactions.

The highly efficient reactor performance of these reactors is observed in hydrogenation of alpha-methylstyrene by virtue of intensive gas–liquid–solid mass transfer even at moderate gas and liquid velocities.

Different variants of STBR have been proven and compared with each other as well as with the conventional reactors. The comparison shows that STBR, especially monoliths packed with particles, demonstrate the highest space-time yield, but at the expense of an elevated pressure drop.

1. Introduction

Recent research has shown that monolithic reactors with a gas–liquid flow in small regular channels with an active component deposited on the walls can lead to performance enhancement in comparison with such conventional multiphase reactors as trickle-bed [1–3] and slurry reactors [4,5]. The performance enhancement is mainly attributed to the more intensive contact between all phases and better mass transfer inherent in the slug flow, which is characterized by the passage of elongated gas bubbles being separated by liquid slugs [6].

As a rule, research on monolithic reactors is focused on two different options with regard to practical realization. The first one is the application of monolithic systems as alternative to batch reactors, where a fixed catalyst (instead of a suspended catalyst) is used at superficial velocities needed for maximum conversion [7]. The second one is the utilization of monolithic catalysts in the column type reactors, which usually employ randomly packed catalyst particles [8]. The latter approach has so far shown no clear benefit with regard to the reaction rate and selectivity.

The parallel flow channels of a monolith packed with catalyst particles represent a novel and promising approach [9]. For this new reactor type, Kapteijn and co-workers [10] introduced the term structured trickle-bed reactor (STBR). Dautzenberg and Mukherjee [11] used the more general term composite structured packing (CSP).

For a single-phase gas flow in such a composite structure, the pressure drop, the flow profile [12] and the particle-to-fluid heat and mass transfer [13] were studied numerically and experimentally. However, there are no data devoted to the characteristics of the concurrent gas–liquid flow in particle-packed channels in the available literature.

As well known, one of the gas–liquid patterns realized in small channels can be slug flow. Operating such a mini-structured fixed-bed reactor under gas–liquid slug flow leads to a periodic variation of the external wetting of the particles. When an elongated gas bubble goes through the packed channel, the particles are not wetted for a moment. The following liquid slug covers the particles and removes reaction products as well as heat. In the flow direction, the open frontal area changes periodically due to the curvature of the catalyst spheres and causes periodic fluctuations in velocity. In this way, turbulences are created inside the gas bubbles and liquid slugs, resulting in the enhancement of mass transfer to both the monolith walls and the particles.

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Nomenclature

a_C	intrinsic catalyst activity (mol/(s g_{Pd}))
a_{exp}	catalyst activity (experimental conditions) (mol/(s g_{Pd}))
a_{st}	catalyst activity (standard conditions) (mol/(s g_{Pd}))
c_H	concentration of hydrogen in the bulk liquid (mol/m^3)
c_H^*	saturation concentration of hydrogen (mol/m^3)
cpsc	channels per square centimeter
d_h	hydraulic diameter (mm)
d_{sphere}	particle diameter (mm)
ΔP	total pressure drop gradient (bar/m)
E_A	activation energy (J/(mol K))
GSA	geometric surface area (m^2/m^3)
k_0	Arrhenius preexponential factor (mol/(s g_{Pd}))
K_H	adsorption equilibrium constant of hydrogen (m^3/mol)
m_{Pd}	mass palladium (mg)
OFA	open frontal area
p	pressure (bar)
R	universal gas constant (8.3145 J/(mol K))
T	temperature (K)
t_{wall}	wall thickness (mm)
t_{WC}	Washcoat thickness (μm)
$u_{G,S}$	superficial gas velocity (m/s)
$u_{L,S}$	superficial liquid velocity (m/s)
V_{Bed}	bed volume (m^3)
x_{Pd}	palladium loading of catalyst (wt%)
X_{Pd}	palladium loading of fixed bed (kg/m^3)
STY	space-time-yield (mol/(s m^3))
STBR	structured trickle-bed reactor

It has been already demonstrated that the periodic variation of the hydrodynamic conditions on the catalyst surface can lead to higher gas mass transfer rates and, hence, better performance of trickle-bed reactors [14,15].

Since there are no available publications devoted to the use of monolithic reactors packed with catalyst particles for multiphase reactions, this paper is focused on the experimental evaluation and comparison of the fixed-bed reactors employing four different types of the catalyst systems: (i) Bed A—monolithic inert substrate with catalyst particles packed inside channels, (ii) Bed B—monolithic catalyst with active sites fixed on the walls and catalyst particle packed, (iii) Bed C—monolithic catalyst only with active sites fixed on the walls, and (iv) Bed D—typical catalyst particles used in trickle-bed reactors.

The superficial gas and liquid velocities that provide maximum overall reaction rates per bed volume were identified for each fixed-bed reactor. Pressure drop measurements were made to assess the energy loss for the operation. Finally, the studied fixed-bed reactors were compared with each other and other reactor types with respect to space-time yield and pressure drop using the data presented in the literature.

2. Experimental

2.1. Setup

Because of the low conversion per pass in monolithic reactors of small lengths, the experiments were conducted in a semi-batch mode of operation with a circulating liquid flow (Fig. 1).

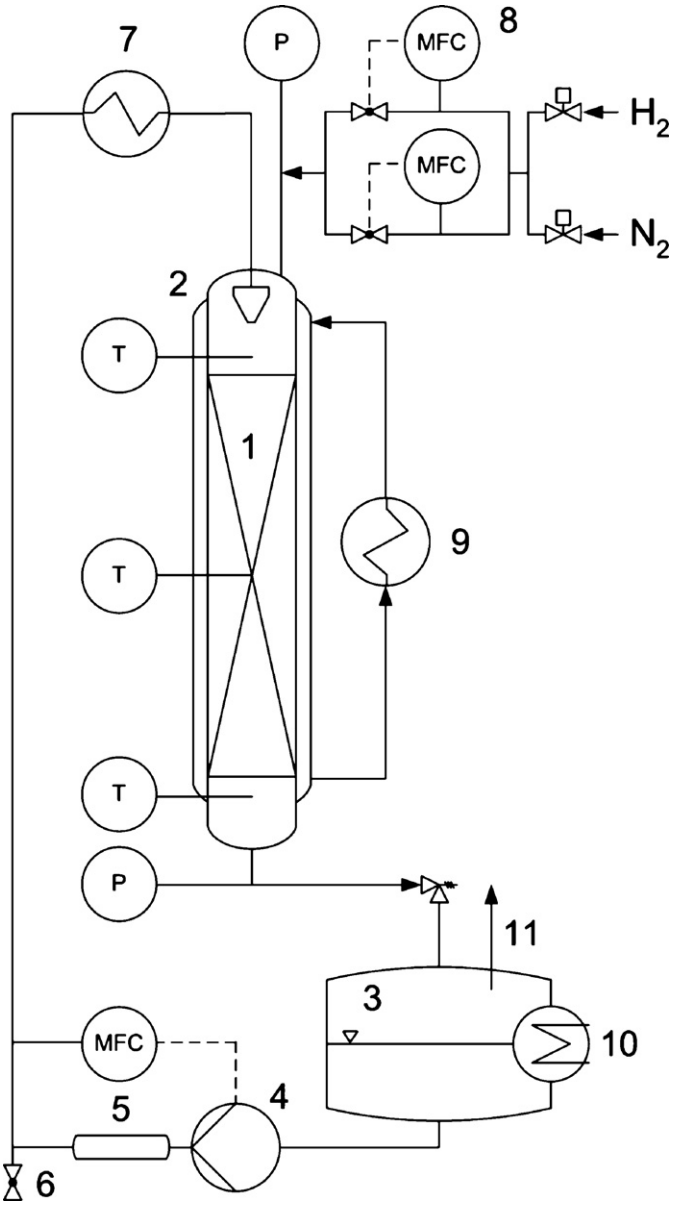

Fig. 1. Experimental setup: (1) fixed bed, (2) double jacket reactor, (3) gas/liquid separator, (4) pump, (5) pulsation damper, (6) sampling valve, (7) electrically heated micro heat exchanger, (8) mass flow controllers, (9) thermostat, (10) cryostat, (11) gas vent.

The liquid feed was pumped from a reservoir vessel into the head of the reactor. A pulsation damper was applied to ensure constant liquid flow rates and to avoid pressure fluctuations. The liquid feed was pre-heated to the reaction temperature by a micro heat exchanger (provided by the Karlsruhe Institute of Technology, Institute of Micro Process Engineering, model: 1440-A-5.2).

A commercial spray nozzle (Duessen-Schlick, model: 553) was used to distribute liquid over the reactor cross-section uniformly. Hydrogen was fed into the head of the reactor by means of mass flow controllers that provided a broad range of throughputs.

The stainless steel, double-jacked reactor had a length of 50.0 cm and an inner diameter of 1.7 cm and could accommodate the different fixed beds (Table 1). The temperature of the reactor was measured at the entrance, in the middle, and at the bottom of the bed by thermocouples.

The pressure above and underneath the catalyst bed was monitored by pressure transducers and adjusted with an overflow valve