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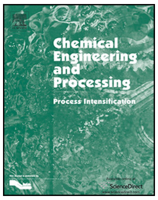

Investigation of a packed bed in a mini channel with a low channel-to-particle diameter ratio: Flow regimes and mass transfer in gas–liquid operation

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ABSTRACT

This work analyzes flow regimes and mass transfer in a miniaturized packed bed reactor under reacting conditions. The reactor consisted of a cylindrical channel with an inner diameter of 2 mm which was filled with spherical catalyst particles having an outer diameter of 0.8 mm. The liquid phase hydrogenation of α-methylstyrene over a $Pd/\gamma\text{-}Al_2O_3$ catalyst was employed as reaction system. Based on experimental reaction rates, overall (gas–liquid–solid) volumetric mass transfer coefficients of hydrogen, which were between $0.5\,s^{-1}$ and $6.0\,s^{-1}$, were computed. Detailed discussions focus on the impact of the flow regime, the gas and liquid superficial velocities and, consequently, their ratio, as well as system pressure on overall mass transfer rates.

1. Introduction

The synthesis of fine chemicals is usually performed with suspended catalysts in batch stirred tank reactors. In these processes, productivity and energy efficiency are limited by set-up times and energy-intensive heating and cooling periods between the batches. Further disadvantages are an elaborate catalyst separation and a varying product quality between the individual batches [1–7]. The transformation of such discontinuous processes into continuous ones, performed in reactors with fixed catalyst packings, could lead to a more efficient and cheaper production, but requires miniaturized reactor systems, so called packed bed mini reactors.

Packed bed mini reactors consist of at least one flow channel with an inner diameter of a few millimeters which is filled with catalysts particles. The combination of relatively large particles and mini channels will provoke high mass transfer rates between the fluid phases and the solid catalyst at a moderate pressure drop. Additionally, catalyst handling and exchange is rather simple. In the past, miniaturized packed bed reactors have been used to examine the performance of conventional catalyst particles, used in industrial scale, in lab scale reactors in order to save operation costs, reduce setup complexity, and experimental time [8–11]. On the other hand, these reactors are considered as efficient chemical production units [12–14]. For both applications, a deep knowledge about the interaction of gas, liquid, and solid phases is substantial, because the flow regime will dictate the catalyst wetting efficiency, phase velocities, interfacial areas, and the residence time distribution, which affect mass and heat transfer in the apparatus and, consequently, conversion and selectivity of the process.

Table 1 summarizes important studies investigating the performance of packed bed mini reactors for heterogeneously catalyzed reactions with gaseous and liquid educts. The list illustrates that such reactors were often used to hydrogenate different liquid educts. Except in [12,15,16], the investigated range of superficial gas and liquid velocities is narrow and orientates toward low fluid velocities in order to meet the space velocities of an industrial trickle-bed reactor. At low liquid throughputs, high residence times and conversions are achieved. However, the catalyst particles may not be completely wetted and external mass transfer may be reduced. In order to create a highly efficient chemical reactor, large superficial velocities should be preferred to achieve high catalyst utilization and production rates.

Abbreviations: AMS, α-methylstyrene; GL, gas–liquid; LS, liquid–solid; GS, gas–solid; HDS, hydrodesulphurization; LS, liquid–solid; PBMR, packed bed mini reactor; PBμR, packed bed micro reactor; PSR, pellet-string reactor.

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Nomenclature

Latin symbols

a_{GL}	specific gas–liquid surface area [$m^2\ m_R^{-3}$]
a_{GS}	specific gas–solid surface area [$m^2\ m_R^{-3}$]
a_{LS}	specific liquid–solid surface area [$m^2\ m_R^{-3}$]
A_{OV}	absolute outer surface area of the catalyst particles [$m^2\ m_R^{-3}$]
a_{OV}	specific outer surface area of the catalyst particles [$m^2\ m_R^{-3}$]
$c_{AMS,0}$	concentration of AMS at the reactor inlet [$mol\ m_L^{-3}$]
$c^*_{L,H2}$	saturation concentration of hydrogen in the liquid phase [$mol\ m_L^{-3}$]
$c_{S,H2,0}$	concentration of hydrogen in the liquid phase located at the catalyst surface [$mol\ m_{CAT,V}^{-3}$]
$c_{S,H2,M}$	Average concentration of hydrogen over the active layer [$mol\ m_{CAT,V}^{-3}$]
d_{CH}	channel inner diameter [m]
d_P	diameter of the spherical catalyst particle [m]
$D_{L,H2}$	diffusion coefficient of hydrogen in the liquid phase [$m^2\ s^{-1}$]
$E_{A,APP}$	apparent activation energy of the reaction [$J\ mol^{-1}$]
$E_{A,DIFF}$	activation energy of the diffusion [$J\ mol^{-1}$]
$E_{A,INT}$	intrinsic activation energy of the reaction [$J\ mol^{-1}$]
k_{APP}	apparent reaction rate constant [s^{-1}]
k_{GL}	gas–liquid mass transfer coefficient [$m\ s^{-1}$]
k_{GS}	gas–solid mass transfer coefficient [$m\ s^{-1}$]
k_{INT}	intrinsic reaction rate constant [s^{-1}]
k_{LS}	liquid–solid mass transfer coefficient [$m\ s^{-1}$]
k_{OV}	overall mass transfer coefficient [$m\ s^{-1}$]
L_B	bed length [m]
m_{CAT}	weight of one catalyst sphere [kg]
N	number of catalyst particles in the reactor [–]
p	absolute pressure [MPa]
$\dot{n}_{H2,OV}$	mean molar flow rate of hydrogen from the gas phase to the catalyst [$mol\ s^{-1}$]
$\dot{n}_{AMS,OV}$	mean molar flow rate of AMS from the liquid phase to the catalyst [$mol\ s^{-1}$]
R	gas constant [$J\ mol^{-1}\ K^{-1}$]
r_{AMS}	reaction rate with respect to AMS [$mol\ s^{-1}$]
$r_{CAT,V}$	volumetric reaction rate per void volume of the catalyst [$mol\ s^{-1}\ m_{CAT,V}^{-3}$]
r_{H2}	reaction rate with respect to hydrogen [$mol\ s^{-1}$]
T	temperature [K]
$u_{G,S}$	superficial gas velocity [$m\ s^{-1}$]
$u_{L,S}$	superficial liquid velocity [$m\ s^{-1}$]
$u_{TP,S}$	superficial two-phase velocity [$m\ s^{-1}$]
$\dot{V}_G$	volumetric flow rate of the gas phase [$m_G^3\ s^{-1}$]
$\dot{V}_L$	volumetric flow rate of the liquid phase [$m_L^3\ s^{-1}$]
V_{CAT}	volume of the active layer of the catalyst in the reactor [m_{CAT}^3]
$V_{CAT,V}$	void volume of the active layer of the catalyst in the reactor [$m_{CAT,V}^3$]
V_P	volume of catalyst particles in the reactor [m^3]
V_R	reactor volume ($V_R = V_P + V_V$) [m_R^3]
V_{SP}	volume of one catalyst particle [m^3]
V_V	void volume of the reactor [m_V^3]
X_{AMS}	conversion of AMS at the reactor outlet [–]

Greek symbols

Φ	Thiele modulus [–]
ε_B	external porosity of the packed bed [$m_V^3\ m_R^{-3}$]
ε_{CAT}	inner porosity of the active layer [$m_{CAT,V}^3\ m_{CAT}^{-3}$]
$\varepsilon_{G,S}$	gas feed ratio [$\dot{V}_G(\dot{V}_G + \dot{V}_L)^{-1}$]

With regard to mass transfer in these systems, only little information is published. Based on the data of Satterfield et al. [8], Kallinikos and Papayannakos [10], and Metaxas and Papayannakos [17], mass transfer is independent of liquid throughput. In contrast, Bauer and Haase [12], Langsch et al. [15], and Haase et al. [16] found that mass transfer increases with raised liquid throughput. All studies agree that a raised superficial gas velocity enhances mass transfer [8,10,12,15–17]. The varying trends might trace back to the different hydrodynamic conditions inside the reactor. As most of the research was performed in opaque channels, the phenomena could not be related to hydrodynamic conditions, e.g. the flow regimes. At identical conditions, Kallinikos and Papayannakos [11] achieved a stratified flow of the gas and liquid in horizontal flow direction, whereas Bauer et al. [18] observed an axially segmented two-phase flow in vertical downflow, which has a similar characteristic as slug flow in empty channels. It is clear that the differing flow regimes affect drastically deviating interfacial areas and phase velocities which will influence achievable mass transfer and reaction rates. This example shows that a constructive comparison of experimental data is complicated if the hydrodynamics in the system are unknown.

Flow regimes in packed bed mini reactors with a vertical flow direction were reported by Langsch et al. [15], Bauer et al. [18], and Vonortas et al. [19]. Vonortas et al. [19] analyzed three different reactor dimensions with an identical channel-to-particle ratio of 1.4 in upflow operation mode with superficial gas and liquid velocities below 0.08 m s^{-1} and 0.05 m s^{-1}, respectively. The channel diameters were 2.5 mm and 4.0 mm and channels with circular as well as square cross sections were employed. They classified the nitrogen–water flow according to the continuous phase into two main flow regimes: (a) film flow and (b) bubble flow. Film flow is characterized by a continuous gas phase and thin liquid films on the particles as well as on the channel wall. This regime dominated at high superficial gas velocities. At lower superficial gas velocities, the liquid formed the continuous phase and gas flows in form of small, dispersed bubbles inside the liquid around the particles. The authors divided this regime into isolated and continuous bubble flow. In isolated bubble flow, a cluster of dispersed bubbles is separated from the next cluster by a pure liquid slug. In continuous bubble flow, these pure liquid slugs are absent and small gas bubbles are observed in the whole reactor.

These classifications are in good agreement with the observations of Bauer et al. [18] who investigated a two-phase downflow in a square channel (d_{CH} = 1.0 mm) packed with catalyst spheres (d_P = 0.8 mm). The experimental plan of the latter researchers also covered higher superficial gas ($0.04 < u_{G,S} < 50$ m s^{-1}) and liquid velocities ($0.02 < u_{L,S} < 1.00$ m s^{-1}). Despite bubble and film flow regimes, as observed also by Vonortas et al. [19], they introduced slug flow, churn flow, as well as several sub- and transition regimes to classify the observed flow conditions. Slug flow was characterized by gas-free liquid slugs separated by large gas bubbles covering almost the whole cross section of the channel [18]. This regime was not detected by Vonortas et al. [19] which could be caused by the large channel diameters. From hydrodynamic studies in empty flow channels, it is well known that slug flow develops only in channels with diameters smaller than a critical diameter which was specified by Chen et al. [20] in the range of 2.0 mm. Langsch et al. [15] studied flow regimes in a round channel (d_{CH} = 1.5 mm) and observed