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New method for simultaneous measurement of hydrodynamics and reaction rates in a mini-channel with Taylor flow

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

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ABSTRACT

Current knowledge on the design and performance of three-phase monolithic reactors and mini-structured reactors is limited by a lack of knowledge on hydrodynamics and mass transfer while a chemical reaction takes place. Therefore, a new method for the simultaneous measurement of hydrodynamics and reaction rates in a mini-channel with catalytically active walls at pressures up to 5 MPa is presented. A newly developed setup has led to an investigation of the influence of the operating conditions and of the gas–liquid feeding on the characteristics of Taylor flow and on chemical conversion. The detected parameters include gas bubble length, liquid slug length, bubble flattening in the cross-sectional area, change in the gas bubble velocity due to conversion and overall macrokinetics. The hydrogenation of α-methylstyrene was used as a test reaction system at a pressure of 1 MPa and a temperature of 343 K.

1. Introduction

Research from the last decades indicates that micro- and mini-structured reactors will play an important role in the future to enhance multiphase reaction processes, which are present in 95% of all chemical processes [1]. Micro-reactors and monolithic reactors, both containing straight flow channels with hydraulic channel diameters in the range between 50 and 1000 μm, are promising tools for process intensification. The flow of gas–liquid mixtures at superficial velocities below 1.0 m s^{-1} leads to so-called Taylor flow, which is characterized by a sequence of elongated gas bubbles separated by liquid slugs. Key features of these novel reactor types operated in Taylor flow include the reduction of mass transfer resistances due to miniaturization and the feasibility of scale-up by numbering up. Additional benefits over conventional reactor technologies include the high surface-to-volume ratio, high volumetric productivity, sharp residence time distribution and low pressure drop [1–7].

The route to design and operate micro- or monolithic reactors with highest outcome includes the following steps: (a) the derivation of correlations for mass transfer and heat transfer that are directly related to hydrodynamic properties such as two-phase velocity, gas bubble and liquid slug length as well as material properties; (b) the identification of optimal hydrodynamic parameters for the planned chemical reaction network by numerical simulation and (c) the development of design rules for a feeding system to create favorable hydrodynamics in a multichannel array including considerations to quantify the variation between the individual channels of the block.

The studies of Behl and Roy [8], Roy and Al-Dahhan [9], and Bauer et al. [10], among others, reveal that there is a relatively narrow operating window of the gas–liquid velocities for which a reasonable distribution with conventional technology can be obtained for a multichannel system with channel diameters on the millimeter scale. Consequently, experiments using multichannel arrays result in a gas–liquid maldistribution and therefore in incorrect performance indicators. Furthermore, it is complicated to separate the effects of the feeding system and the effects of the gas–liquid flow characteristics based on the observed mass transfer rates because a broad range of flow velocities is usually covered. Therefore, experiments in a single channel are important to further enhance the fundamental understanding of hydrodynamics, heat and mass transfer and overall reaction kinetics.

For studies in single channels, special attention needs to be given to the gas–liquid feeding system, which has a strong impact on the characteristics of Taylor flow such as the gas bubble length, the

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Nomenclature

A_{Bub}	bubble cross sectional area [mm^2]
A_C	channel cross sectional area [mm^2]
Ca	Capillary number ($Ca = \mu_L u_{TP} \sigma^{-1}$)
$c_{AMS,in}$	concentration of α-methylstyrene at the channel inlet [mol l^{-1}]
$c_{AMS,out}$	concentration of α-methylstyrene at the channel outlet [mol l^{-1}]
c_{Sat}	saturation concentration of hydrogen in α-methylstyrene [mol l^{-1}]
d_h	hydraulic channel diameter [mm]
d_{II}	inner diameter of the injector [mm]
d_{IO}	outer diameter of the injector [mm]
$\Delta n_{Pass,R}$	change in amount of substance per pass due to chemical reactions [mol]
$\Delta n_{Pass,S}$	change in amount of substance per pass due to physical absorption of hydrogen [mol]
Δu_B	change in bubble velocity due to hydrogen consumption [cm s^{-1}]
$\varepsilon_{G,in}$	gas holdup at the channel inlet [$cm_G^3\ s^{-1}(cm_G^3\ s^{-1} + cm_L^3\ s^{-1})^{-1}$]
G	gas
L	liquid
L_B	gas bubble length [mm]
L_{BF}	length of bubble flattening ($L_{BF} = [1 - \tan(\phi)]0.95R_C$) [μm]
L_{CF}	length of constant film thickness ($L_{CF} = \tan(\phi)0.95R_C$) [μm]
$L_{UC,in}$	unit cell length at the channel inlet [mm]
Mo	bare monolith
$n_{B,in}$	amount of substance in one gas bubble [mol]
p	pressure [MPa]
R_C	hydraulic channel radius [mm]
R_{Diag}	diagonal bubble radius [mm]
R_{Side}	orthogonal bubble radius [mm]
Re_{GC}	channel gas Reynolds number ($Re_{GC} = \rho_G d_h u_{GS} \mu_G^{-1}$)
Re_{LC}	channel liquid Reynolds number ($Re_{GC} = \rho_L d_h u_{LS} \mu_L^{-1}$)
T	temperature [K]
$u_{B,in}$	bubble velocity at the channel inlet [cm s^{-1}]
$u_{G,S}$	gas superficial velocity [cm s^{-1}]
$u_{L,S}$	liquid superficial velocity [cm s^{-1}]
u_{TP}	two-phase velocity [cm s^{-1}]
V_G	gas volumetric flowrate [ml/min]
V_L	liquid volumetric flowrate [ml/min]
WC	washcoat

liquid slug length and their uniformity [11,12]. A second important fact is the measurements at elevated temperature and pressure and for industrially relevant chemicals such as aromatic hydrocarbons. Studies focused on the hydrodynamics and gas–liquid mass transfer are usually conducted under mild conditions and are limited to superficial gas and liquid velocities above 5 cm s^{-1} [11,12]. However, process simulations applying the mass transfer model of Kreutzer et al. [13] reveal that especially lower velocities lead to high mass transfer rates because of the reduced thickness of the liquid film between the gas bubble and the channel wall. Additionally, the residence time increases with the reduction of the flow velocity and will result in a higher conversion per pass, contributing to the utilization of the reactor in continuous operation processes. There is a serious lack of knowledge on whether stable Taylor flow velocities lower than those previously determined can be achieved.

There are various publications on multiphase reaction studies in single channels [14–23]. Table 1 summarizes the details about the studies concerning single channels on the millimeter scale. The studies can be divided into those without visual observation [14–16] and those with visual observation of the two-phase flow [17–23]. The latter is highly favorable because it allows the detection of any flow irregularities such as pulsations affected by the pressure fluctuations of leaving gas bubbles and liquid slugs at the channel outlet or by clearance volumes between channel connections [24]. Additionally, these setups are able to study the effect of Taylor flow parameters such as liquid slug length and gas bubble length on reaction channel performance, but they are limited to virtually atmospheric pressure, except in the studies by Liu et al. [17], who studied the hydrogenation of 2-ethylanthraquinone at a pressure of 0.4 MPa. Low system pressure will cause low conversion per pass, and using the recycle mode is compulsory to measure reaction rates with low experimental error. Therefore, the inlet concentration is a function of time, and gauged reaction rates can differ from the results of experiments with a constant inlet concentration (steady-state mode), especially if the reaction rate is strongly coupled to the reactant concentration in the liquid phase.

For a deeper understanding of the behavior of mini-reactors with straight flow channels in the steady state, a new experimental setup was built to simultaneously study the hydrodynamics and overall macrokinetics in a single monolithic channel at elevated pressure and temperature. The main part of the setup is a new reactor having transparent observation chambers at the inlet and at the outlet in addition to a variable gas–liquid feeding system. The section between the transparent zones was made of a monolithic catalyst forming a straight, single channel. For the first time, such a setup can evaluate the hydrodynamics under pressures higher than 0.4 MPa and can examine specific phenomena, such as the gas bubble size decrease due to hydrogen consumption by the chemical reaction and bubble breakup or coalescence in the opaque monolithic catalyst section. The characteristics of Taylor flow and the specific mass transfer areas can be modified using different needle injectors to feed the gas in the axial direction into the mini-channel. Using this setup, new experimental data on the characteristics of Taylor flow and overall macrokinetics are presented for the hydrogenation of α-methylstyrene.

2. Materials and methods

The challenge was to develop an experimental setup that couples hydrodynamic studies and chemical reaction studies at elevated pressure and temperature. A flexible gas–liquid feeding system that could manipulate the specific mass transfer areas was designed to investigate the influence of the Taylor flow characteristics on conversion, focusing on low gas and liquid superficial velocities.

2.1. Experimental setup

The main component of the experimental setup is the newly developed semitransparent mini-channel reactor, which consists of a single-channel observation chamber at the inlet, a single-channel reaction chamber and a single-channel observation chamber at the outlet. The observation chambers consist of a glass capillary (Hilgenberg GmbH, square cross section, d_h = 1.0 mm), which was embedded in a resin to enhance the mechanical strength. Tests revealed that the developed system can handle system pressures of at least up to 5 MPa; and temperatures up to 363 K. The reaction chamber contains the catalyst. Further details will be given in Section 2.4.