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Use of 'smart interfaces' to improve the liquid-sided mass transport in a falling film microreactor

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ABSTRACT

It has been shown in the past, that the use of a falling film microreactor is advantageous for operation conditions, during which conventional processing equipment reaches its limits. The reactor design facilitates the development of well controlled, stable menisci. The very large specific gas/liquid interface (up to 20 000 m^2/m^3) provides excellent mass transfer capabilities between the phases. Nevertheless, despite the excellent gas/liquid mass transfer that occurs the chemical reactions are limited by the mass transfer within the phases. Commonly, the rate limiting step is the diffusive mass transport within the liquid side.

This study investigates the potential of falling film microreactors equipped with structured channels to enhance the mass transfer within the liquid phase. To do this, four different reaction plates have been fabricated and are experimentally examined. Besides two reaction plates with straight, unstructured channels (channel width: 600 or 1200 μm), one plate with fins and one plate with additional grooves in straight 1200 μm wide channels forming a so-called staggered herringbone mixer are used.

Taking carbon dioxide absorption as benchmark reaction it is shown that structured channel walls can significantly enhance the mass transfer within the liquid phase. This leads to an increase of the overall performance of the benchmark reaction. Properly chosen channel geometry can increase the conversion by up to 42%. Hence, by using an optimal reaction plate it is possible to more than double the flow rate, without any loss in conversion.

1. Introduction

1.1. Gas–liquid contacting

Contacting gas and liquid phases is an important issue in chemical process engineering. Usually, the crucial point of such gas–liquid contacting is to ensure a good mass transfer between the two phases. For this operation, the use of microreactors is advantageous compared to conventional reactors. Mass and heat transfer can be especially enhanced due to the very large specific surfaces and interfaces offered, allowing for reactions which cannot be realized within conventional reactors. For example, highly exothermic direct fluorinations can be safely operated in microreactors (Jähnisch et al., 2000) whereas controlled direct fluorinations of aromatic substrates in conventional macroscopic reactors could only be achieved at extremely low temperatures and concentrations (−70 °C; 0.01 mol/L) (Grakauskas, 1970; Cacace and Wolf, 1978; Conte et al., 1995). It is possible to realize these reactions in a microreactor with 50 vol% of fluorine at temperatures up to −15 °C. Moreover, the space–time yield can be increased by more than one order of magnitude (Löb et al., 2004). In addition to the heat and mass transfer capabilities, improved safety is another advantage of microreactors. This stems from the very small reactant hold-up and the shift of explosion limits due to the small channel dimensions (Kestenbaum et al., 2002; Veser, 2001).

In general, two principles for gas–liquid contacting within microreactors exist. The first is to separate both streams from each other and using the microreactor to create the interface between them. In this way two continuous phases are generated. This concept is realized, e.g. within falling film microreactors, microreactors with overlapping channels or mesh microreactors. The advantage of this contacting principle is that phase separation

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is facilitated and a controlled, well-defined known interface is assured. The higher technical requirements to prevent phase intermixing is the disadvantage of this type of contacting.

The second principle is to disperse one phase within the other, leading to distinct microfluidic flow patterns such as bubbly, Taylor, churn, and annular flow, as known from gas/liquid flows in e.g. annular ducts. A frequently used flow operation mode with a highly regular distributed interface and highly agitated convective mixing in the phases is segmented (Taylor) flow within micro-bubble columns or foam microreactors. The advantage of this method lies in the relatively low technical expenditure for fabricating and operating these reactors. On the other hand subsequent separation of the phases is needed and a scale out is difficult. Further, this contacting principle shows a size distribution of the relevant flow properties such as the interfaces, diffusion distances within one phase, and the possible presence of mixed flow patterns when numbering-up.

Both microbubble columns and the falling film microreactor (FFMR) can be operated in such a manner that very high specific surfaces can be obtained (up to 20 000 m^2/m^3). The advantage of the FFMR is that over a wide range of liquid and gas flow rates the flow patterns remain the same compared to the microbubble column, which shows a variety of flow patterns (Hessel et al., 2000). This limits optimal operation of this device to specific flow conditions. Compared to a conventional packed column, both microdevices exceed the performance of the conventional device by orders of magnitude.

1.2. Falling film microreactor

The falling film microreactor is a well studied and purchasable microreactor for gas/liquid contacting developed by the Institut für Mikrotechnik Mainz GmbH (IMM). With this reactor, liquid films of a few tens of micrometers thickness and interfacial areas up to 20 000 m^2/m^3 can be obtained. Beside this, the advantage of the microreactor design compared to conventional falling film reactors is that the microchannels stabilize the liquid film and prevent film break-up. The FFMR has been widely applied for different chemical reactions for example for photochemical chlorination (Ehrich et al., 2002), ozonization of olefines (Steinfeldt et al., 2007), sulfonation (Müller et al., 2004) and catalyzed hydrogenation (Yeong et al., 2003). Additionally, a variety of detailed studies concerning the reactor itself have been previously published (Wille, 2000). Yeong et al. (2006) for example provide a deeper understanding of the liquid film development in the microchannels to estimate the influence of the channel width on the mass transfer. Additional knowledge on the gas phase residence time grants even more detailed interpretations (Commenge et al., 2006).

Zanfir et al. (2005) investigated the absorption of carbon dioxide in a falling film microreactor through the use of computational fluid dynamics. The model developed neglected the liquid meniscus shape. Despite this, it was in good agreement with experimental results for NaOH concentration of 0.1 and 1 mol/L. Additionally, it was found that the major limiting step is on the liquid side. The authors demonstrated that the carbon dioxide is consumed within a short distance from the phase border.

Al-Rawashdeh et al. (2008) extended this model by a realistic description of the meniscus shape. Investigations on the influence of flow distribution on conversion showed that an uneven flow distribution lowers the conversion compared to ideal equal distribution only by 2%. Additionally the influence of fabrication imprecisions on the overall performance were investigated. Again the impact was found to be very small (about 2% deviation between maximum and minimum conversion). Investigations on the influence of the wettability of the reaction plate on the conversion depicted that a hydrophobic surface decrease the conversion by almost 20% in comparison to a hydrophilic surface due to a reduction of the relevant interfacial area.

Despite the fact that microreactors offer a variety of (important) advantages (Roberge et al., 2005) the main challenges lie in enhancing the throughput in order to enable production scale processing. Therefore, different concepts were developed including ‘true’ (internal) numbering-up (equaling-up) or scale-out by smart dimensional increases, while ‘numbering-up’ relevant flow features such as the channels. For gas–liquid contactors this was recently demonstrated. By increasing the number and the length of microchannels in a falling film microreactor, the absolute surface of the interface was increased by an order of magnitude. This was realized with the help of two different design approaches: a cylindrical and a flat one (Vankayala et al., 2007). Another major challenge is to maximize process intensification by finding suitable novel process windows. Overcoming limiting steps of the reaction rate allows for an increased conversion and to operate with higher flow rates resulting in a higher throughput. This can, for example, be realized by purposefully induced fluid–structure interactions. According to the Center of Smart Interfaces (2008) such interactions can be called ‘Smart Interfaces’, which are defined as follows:

“[…] ‘Smart Interfaces’ refers to fluid–solid boundary interfaces that have been designed or built for achieving a specific purpose, such as enhancement or controllability of mass, momentum or heat transfer. […]”

With this a reasonable approach to overcome limiting steps is to develop process equipment with the intention to influence the fluids.

This paper intends to demonstrate the potentials for process intensification, which arise from the use of structured reaction plates in a falling film microreactor. Therefore four different reaction plates are examined by the use of a benchmark reaction, i.e. the absorption of carbon dioxide in a basic solution.

2. Investigated process system

2.1. Reactor

Fig. 1 shows the essential parts of a (disassembled) FFMR. In order to adapt the reactor to different operation requirements, it is possible to change the reaction plate, specifically the number and dimensions of the microchannels. The exact design of the microstructured plates impacts mass transfer by changing the

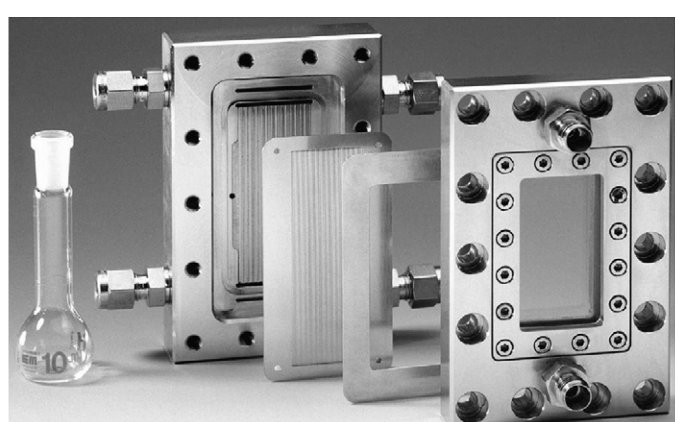

Fig. 1. Falling film microreactor.