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Application of hot-wire anemometry for experimental investigation of flow distribution in micro-packed bed reactors for synthesis gas conversion

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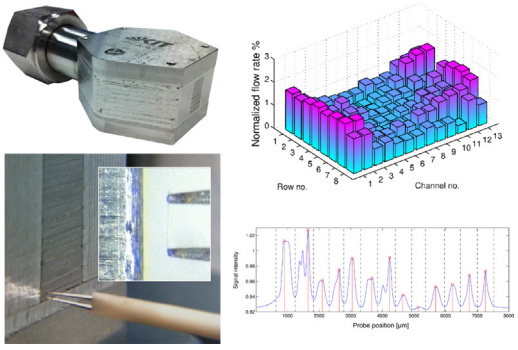
HIGHLIGHTS

- Hot-wire anemometry (HWA) flow distribution measurement applied to micro-packed bed reactors.
- Measurements of flow distribution as a function of particle size distribution and particle size enabled.
- Potential bottlenecks of catalyst particles packing in microreactor identified.

GRAPHICAL ABSTRACT



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ABSTRACT

The knowledge of the flow distribution inside microstructured reactors is valuable e.g. for improving the reactor design, developing flow distributors, and optimizing the catalyst loading method. The applicability of the hot-wire anemometry (HWA) technique for experimental determination of the flow distribution inside a multi-stack micro-packed bed reactor is demonstrated for the first time. The anemometry data is then evaluated in relation to the reactor performance for methanol synthesis under relevant industrial operating conditions. A 400 μm long hot-wire connected to a constant temperature anemometer was applied for scanning the flow out of a specially designed reactor model, clamped on a motorized table equipped with a precise positioning system. The anemometry measurements revealed a nonuniformity in the catalyst packing, partially resulting from the reactor design. The flow distribution is poorer for smaller particles and for wider particle size distributions, and worsens as the superficial flow velocity (and the pressure drop) decreases. The effects of the packing nonuniformity on the reactor performance appear minor under methanol synthesis conditions. They could, however, turn out significant upon pushing the overall conversion in the whole reactor towards equilibrium as the synthesis reaction is exothermic, and temperature increase does alleviate the problem. The reactor design should be modified instead.

1. Introduction

Micro process technology has opened up new opportunities for process intensification through equipment size reduction, process simplification and process integration. Miniaturized flow

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dimensions and high surface-to-volume ratio in micro-units are the origin of promising characteristics such as enhanced heat and mass transfer and improved safety (Pohar and Plazl, 2009; Mills et al., 2007; Borovinskaya and Reshetilovskii, 2008; Matlosz and Commenge, 2002). Portability, modularity, flexibility and high performance of microstructured reactors may facilitate the development of compact natural gas-, coal-, and biomass-to-liquid (XTL) technologies which are safe and economic in small scales and suitable for remote and offshore applications. This effort has been pioneered by e.g. Karlsruhe Institute for Micro Process Engineering (IMVT), Mainz Institute of Microtechnology (IMM), Pacific Northwest National Laboratory (PNNL), etc. and taken further to commercialization by companies like CompactGTL and Velocys. While there is a trend towards building larger GTL plants to benefit from economies of scale, such technologies may be the breakthrough required for competitive production of the easily-transportable fuels (e.g. methanol, dimethyl ether or Fischer-Tropsch liquids) from stranded/associated natural gas or gas produced from highly distributed bio-based feedstock that is not otherwise economically feasible to exploit.

Microstructured reactors can normally be considered as parallel arrays of identical channels. In general, uniform distribution of the flow among the microchannels is highly desirable in order to maximize the overall performance of the reactor (Commenge et al., 2002). Similarity of the size and geometry of channels (which directly affect the pressure drop) and proper design of the reactor inlet are the most important factors for obtaining a uniform flow distribution. To emphasize the significance of identical channel size to the flow distribution between different channels and reactor performance, Pfeifer and Schubert (2007) made a simple comparison between two parallel rectangular channels with 80 and 90 μm width, and their estimations showed 27% higher flow rate, 11% lower heat transfer area and 25% lower overall heat flux for the wider channel, both channels being connected to the same manifold. Such variations in channel size may be an outcome of fabrication tolerances or e.g. uneven catalyst coating. In the latter case, lower catalyst mass (thinner coating) in the channel with higher flow rate can lead to a higher load on the catalyst (2.5 times higher flow rate per catalyst mass for the previous example, assuming 10 and 5 μm m thick coatings) (Pfeifer and Schubert, 2007). In the case of microstructured reactors packed with catalyst particles, a similar approach can be used to evaluate the significance of non-uniform catalyst packing for flow distribution among channels connected to the same manifold. Considering two identical parallel microchannels or also microsized slits with different bed porosities of 0.5 and 0.4, the flow rate through the channel with the higher void fraction is approximately 2.8 times higher than the one with the lower porosity, as estimated from the Ergun equation (Eq. (1)) (Ergun, 1952) under laminar flow regime and conditions of identical pressure drop, where the second term is supposed to be negligible. Identical pressure drop for both channels is valid, as both are connected to a common inlet and outlet.

$$\frac{\Delta P}{L} = 150\frac{\mu u_s(1-\varepsilon)^2}{d_p^2\varepsilon^3} + 1.75\frac{\rho u_s^2(1-\varepsilon)}{d_p\varepsilon^3} \tag{1}$$

where ΔP is the pressure drop, L is the length of the channels, μ and ρ are respectively the viscosity and density of the fluid, ε is the void fraction (porosity) of the catalyst bed, u_s is the superficial velocity, and d_p is the average diameter of the catalyst particles. At the same time, according to Eq. (2), there is ~16% less catalyst mass in the channel with the higher void fraction and higher flow rate:

$$\varepsilon = 1 - \frac{\rho_b}{\rho_p} \tag{2}$$

where ρ_b and ρ_p are respectively bed (bulk) density and catalyst particle density. Overall, these simple calculations suggest that an increase in the bed porosity from 0.4 to 0.5, can lead to almost 240% increase in the flow rate per catalyst mass between the two channels with common manifold. The total performance loss depends also on other factors such as reaction kinetics, endo/exothermicity, and thermodynamic limitations, and may be significant.

Knowledge about the flow distribution in microstructured reactor is useful for prediction of the reactor behavior and optimization of reactor design, inlet geometry and catalyst loading method. Laser Doppler anemometry (LDA) is an experimental technique that has been applied for determination of flow distribution in microstructured reactors, supplementary to CFD simulations (Rebrov et al., 2007; Mies et al., 2007). However, for the small measurement volumes and the low velocities corresponding to micro-units, application of LDA to gas streams is complicated and not very accurate. Therefore, to apply the method for studying the flow in micro-units, the gas stream may be replaced by liquid flow with identical Reynolds numbers, as suggested by Schouten et al. (Rebrov et al., 2007; Mies et al., 2007). Apart from the complications associated with the use of liquid and possible errors that may be introduced by fluid substitution, such approaches may not be appropriate for micro-packed bed reactors where liquid flow may affect the catalyst packing or even deagglomerate the catalyst particles and wash them out of the reactor.

Pfeifer et al. (2004) developed an experimental technique based on hot-wire anemometry that is accurate for investigation of gas flow distribution in microstructured units. The technique makes use of a precise positioning system and a small hot-wire (probe) connected to a constant-temperature anemometer. The probe is being moved in front of and parallel to the reactor outlet to scan the flow passing through the individual channels. The applicability of the method has been demonstrated for quantitative analysis of the flow distribution in one- and two-dimensional arrays of wall-coated microchannels (Pfeifer et al., 2004; Pfeifer and Schubert, 2007). In addition, the method can be used for non-destructive evaluation of the catalyst coating uniformity.

Hot wire (thermal) anemometry is a well established experimental technique (see e.g. Bruun, 1995) where a thin wire, heated by electric current, is exposed to a stream of fluid (with constant temperature and composition) and the power needed for heating the wire correlates with the heat loss from the wire, which in turn is a function of the flow velocity. The principle governing equation can be written as follows:

$$P = Q_{cv} + Q_{co} + Q_r \tag{3}$$

where P is the power for Joule heating and is equal to V^2/R (with V being the voltage drop across the wire and R the wire electrical resistance), and Q_{cv}, Q_{co} and Q_r are convective, conductive and radiative heat losses from the hot-wire, respectively. King (1914) suggested a simple relation between the flow velocity and the conductive heat transfer coefficient (h) from an infinite cylindrical body in a flow at low Reynolds number, known as King's law (Eq. (4)),

$$Q_{cv} \propto h \propto a + bu^{0.5} \tag{4}$$

where a and b are constants and u is the flow velocity. In constant-temperature anemometers, the hot-wire is connected to a Wheatstone bridge, and a servo amplifier keeps the bridge in balance by controlling the current to the probe (keeping the electrical resistance (R) of the hot-wire and hence its temperature constant). At constant fluid temperature, and when heat loss by radiation and conduction from two ends of the wire are either negligible or independent of the flow velocity, Eq. (3) can be rewritten as;