Simulation of Heat Transfer Enhancement Effects in Microreactors

S. Hardt*, W. Ehrfeld*, V. Hessel* and K. M. Vanden Bussche**

*Institut für Mikrotechnik Mainz GmbH, Carl-Zeiss-Str. 18-20,
D-55129 Mainz, Germany, hardt@imm-mainz.de
**UOP, 25 East Algonquin Road, Des Plaines, IL, USA, kvandenb@uop.com

ABSTRACT

Sate-of-the-art microstructuring techniques allow to model the interface for heat and mass transfer in miniaturized reactions systems on a micrometer scale. In this context, two different approaches for heat transfer enhancement are presented. The ideas put forward rely on entrance flow effects and inertial flows in meandering channels, respectively. It is found that a heat transfer enhancement of at least one order of magnitude can be achieved compared to unstructured channels. On this basis, a miniaturized heat-exchanger reaction system is investigated, where a kinetic model of an endothermic, heterogeneously catalyzed gas-phase reaction is used. The miniaturized heat-exchanger reactor, both with and without heat transfer enhancement, is subsequently benchmarked against conventional fixed bed technology. It is shown that, for the reaction system under study, a substantial reduction of the required amount of catalyst can be achieved in microsystems.

Keywords: Microreactors, Microstructuring Techniques, Heat Transfer Enhancement, Computational Fluid Dynamics.

1 INTRODUCTION

A limiting factor for many reactions employed in the chemical process industry is heat and mass transfer to the regions where the chemical conversion occurs. An approach to overcome these limitations is based on microreactors, where high temperature and concentration gradients build up in small-scale channels [1]. In addition to the benefits due to miniaturization, a further enhancement of heat and mass transfer is obtained by applying microstructuring techniques. Structuring the interface for heat and mass exchange results in an increase of the specific surface area. Furthermore, special flow patterns are created which promote a fast exchange of heat and matter.

For many chemical processes heterogeneously catalyzed gas phase reactions are of major importance. In such reactions mass transfer is often inhibited by internal transport processes within the porous catalyst medium, whereas mass transfer within the gas phase is comparatively fast. The opposite is found for the heat transfer problem, where the transport resistance is mainly due to the gas phase. Correspondingly, this work is focused on heat transfer enhancement by microstructuring techniques. However, due to the fact that heat and mass transfer are governed by the same dynamics described by an advection-diffusion equation, the results are easily to be translated to a corresponding mass transfer problem.

2 DESIGN CONCEPTS FOR MICROSTRUCTURED HEAT EXCHANGERS

In the following two different approaches for heat transfer enhancement by microstructuring techniques are presented. In both cases the typical dimensions of the microchannels were chosen to be compatible with the process application discussed in the second part of this work and with constraints set by microstructuring techniques for small series fabrication. Note, however, that dimensionless numbers were used to characterize heat transfer, allowing the results to be translated to a different length scale.

The simplest channel geometry is an arrangement of infinite parallel plates with fluid flow in between. The velocity and temperature distributions for the corresponding flow pattern, the so-called Poisson flow, are well known and allow to compute the heat transfer to the channel walls [2].

The first approach to be considered as a modification of the parallel-plates arrangement is a folding of the channel walls. On the one hand folding increases the specific surface area of the heat exchanger. On the other hand the fluid is forced to follow a curved path, as opposed to the purely longitudinal, fully developed Poisson flow.

For the computation of temperature and velocity fields the commercial CFD code CFX4 of AEA Technology was used. CFX4 is based on a finite volume method for structured, body-fitted grids and was applied in all the CFD calculations to be reported in this work.

As a specific realization of folded channel walls a sine-wave shape was considered. For simplicity isothermal walls were assumed. The wavelength of the sine-wave and the offset between the channel walls were chosen 1 mm, as indicated in Fig. 1. A 2D computational model with air as the fluid was used. In order to characterize a heat exchanger with such meandering channels, local Nusselt (Nu) numbers, which in general depend on the Reynolds *(Re)* number, were computed.

It is found that, for the range of Reynolds numbers considered, the Nusselt numbers only mildly depend on the position in the channel, with the exception of the entrance region.

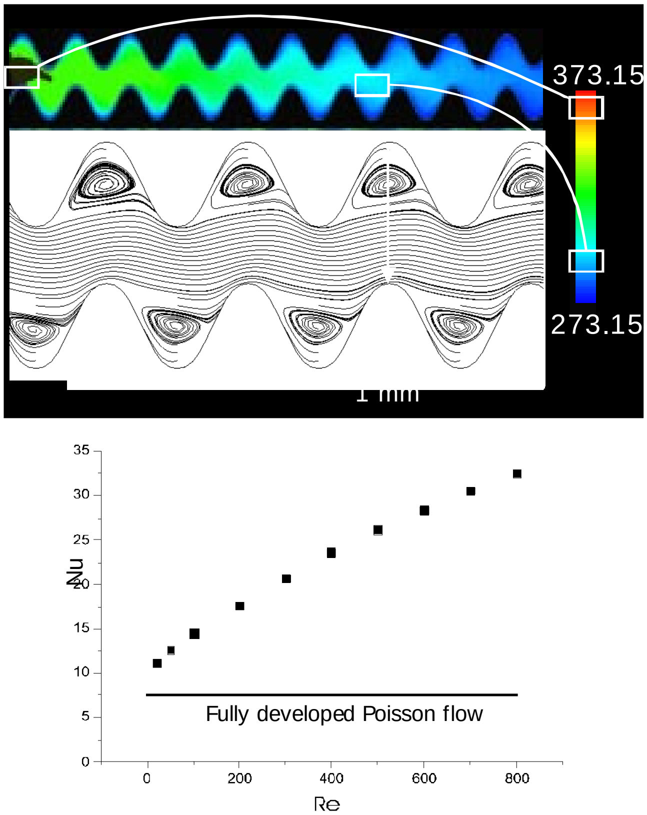

Figure 1: Temperature Distribution, streamlines and Nusselt numbers in the sine-wave heat exchanger.

In Fig. 1 Nu is plotted against Re, where in case of doubt (i. e. a weak dependence of Nu on position) the Nusselt number was evaluated close to the channel outlet. For comparison to a parallel plates heat exchanger, the horizontal line indicates the Nusselt number for a fully developed Poisson flow. Note that due to the finite length of the computational model and the corresponding entrance flow effects, the actual Nusselt number for a parallel-plates arrangement will be slightly higher than that given by the Poisson flow, especially for large Reynolds numbers. However, this effect is not very pronounced, giving $Nu \approx 10$ close to the outlet for $Re = 800$. The sine-shaped walls affect the flow pattern in such a way that a substantial heat transfer enhancement is achieved, reflected in Nusselt numbers of up to 32.

In the upper part of Fig. 1 the temperature distribution and the streamlines characteristic for comparatively high Reynolds numbers (≥ 500) are displayed. The hot gas is entering the channel from the left. It is apparent that the fluid is following the meanders only to some extend, where large vortices are formed close to the convex sections of the channel walls. Even for smaller offsets between the two boundary walls the fluid is driven to a path with smaller curvature than the meanders. This effect is due to the convective term in the Navier-Stokes equation, which accounts for inertial forces on a moving fluid element and becomes important for high Reynolds numbers.

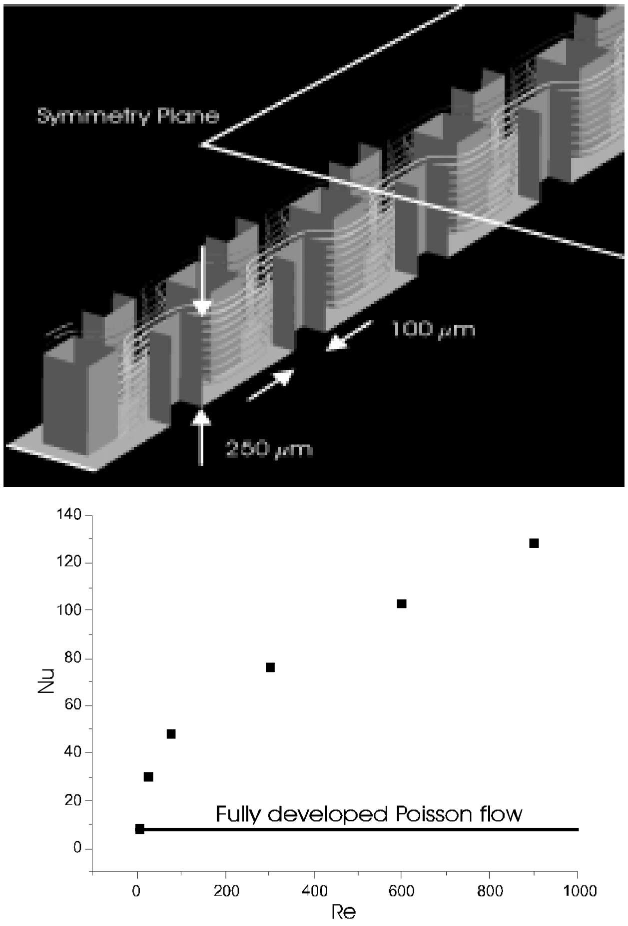

Figure 2: Streamlines and Nusselt numbers in a heat exchanger with microfins.

The second design considered for heat transfer enhancement is based on an entrance flow effect. It is well known that for a hydrodynamically and thermally developing flow the Nusselt number can take very high values [2]. By continuously splitting and recombining the fluid flow a permanent entrance flow effect can be generated. A corresponding geometry is shown in Fig. 2.

The fluid is entering from the left into a checkerboard array of microfins with a square cross section of 100 × 100 μm^2. The upper and lower boundary of the channel are parallel plates 500 µm apart. The reflection symmetry and periodicity of the structure in transverse direction was used to reduce the extent of the computational model. The fabrication of such geometries requires 3D microstructuring techniques. Again, a flow of air was considered in combination with isothermal wall boundary conditions.

Fig. 2 shows that the streamlines form a very regular pattern in the channel. Very high Nusselt numbers are achieved with the permanent entrance flow effect, as apparent by comparison to the corresponding value for a fully developed Poisson flow. In the *Nu-Re* relation the Nusselt number is defined via the surface area including the fins and the Reynolds number by the hydraulic diameter of the parallel plates arrangement without fins. Similar to the meandering channel the Nusselt numbers, which display only a weak position dependence, were evaluated close to the outlet. Again, due to the finite extension of the computational model, there is some heat transfer enhancement at high Reynolds numbers even in a parallel plates arrangement without fins, which is negligible for practical purposes. While the Nusselt number measures heat transfer per unit area, an additional enhancement factor is due to the increased surface area of the microfin arrangement. For the geometry shown the surface area is by a factor of 13/4 larger than that of a parallel-plates heat exchanger.

3 HEAT EXCHANGER REACTION SYSTEMS

In the following the effects of heat transfer enhancement for heterogeneously catalyzed gas phase reactions are investigated. The idea is to study a continuous flow heat-exchanger reaction system with alternating layers of heating and reaction gas. The two gases are fed into the reactor in counter-current mode, where the heating gas provides energy for the endothermic reaction. The channel walls of the reaction gas channel are coated with a nanoporous catalyst, in which the conversion of the educt gas to the product occurs.

Figure 3: Concentration map of product gas P1, in arbitrary units, over the channel geometry.

The computer simulations were carried out for an example endothermic reaction. In the course of this reaction two feed components, E1 and E2, are converted into a product P1 and two by-products, P2 and P3. The reaction kinetics are of the Langmuir-Hinshelwood type with a rapidly increasing reaction rate at high temperatures, i. e. featuring a high activation energy.

Although the simulations of the heat-exchanger reactor were performed for a specific reaction, many other processes will exhibit a similar behavior when a heating gas channel is thermally coupled to the reaction gas feed. The characteristic prerequisites for improvements due to heat transfer enhancement are a fast, heat-transfer limited reaction and/or a reaction rate which is rapidly increasing with temperature.

For the CFD simulations of the heat-exchanger reactor a 2D model as displayed in Fig. 3 was considered. A heating gas channel and a reaction gas channel (500 µm channel height) are arranged as alternating layers, separated by thermally conductive walls with a thickness of 200 µm. On the reaction gas side the walls are coated with a 100 µm catalyst layer. The thickness of the catalyst layer is determined by fabrication constraints as well as limitations set by the mass transfer efficiency within the porous medium. The chosen value for the layer thickness is compatible with both constraints. Note that only the lower/upper half of the upper/lower channel was modeled, since the flow is symmetric with respect to a symmetry plane at half the channel height. The velocity, pressure, temperature and concentration fields are simultaneously solved for. The CFD method employed accounts for compressibility effects, temperature dependent material properties as well as heat conduction in the wall material. As a simplifying assumption, the multicomponent diffusion scenario of the system with five different chemical species was approximated by Fick diffusion.

In contrast to most other approaches, the transport equations for matter, momentum and heat are also solved within the porous catalyst layer. Effective transport equations for porous media are obtained when the advection-diffusion equation is averaged over the pore structure of the medium [3]. For momentum transport, a generalized version of Darcy's law was considered. In the transport equation for heat the effective thermal diffusivity of the weighted average model of Hadley [4] was used. Mass transfer was modeled according to the effective mass diffusivity approach of Neale and Nader [5]. The pore size of the catalyst is such that diffusive mass transfer occurs in the transition region between Fick and Knudsen diffusion, which was accounted for by the Dusty Gas Model [6].

As a result of the simulations, concentration fields for the different chemical species are obtained. A typical spatial distribution of product P1 is displayed in Fig. 3. In the enlargement of the reaction gas outlet high product concentrations are visible inside the catalyst layer. Due to the temperature dependence of the reaction rate, most of the conversion occurs in this high temperature region close to the outlet.

The major aim of this work is to investigate the effects of heat transfer enhancement. Performing a simulation similar to the one presented here for a heat-exchanger reactor with 3-D structures as shown in Fig. 2 would be a very time-consuming task due to slow convergence of the

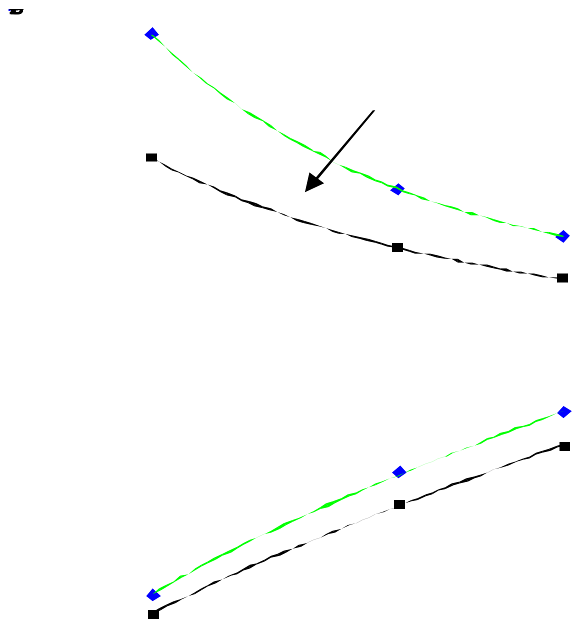

Figure 4: Outlet concentration (above) and specific molar flux (below) of P1, in arbitrary units, as a function of the E2 space velocity.

nonlinear iteration. For this reason heat transfer enhancement effects characteristic for microstructured channels were mimicked with a 2-D geometry by increasing the thermal conductivity of the heating gas. Based on the calculations above a Nusselt enhancement factor of 4 was assumed.

The results of the comparison between structured and unstructured microchannels are displayed in Fig. 4. The goal of the industrial process is to achieve a specific mole fraction of the product P1 at the channel outlet while maximizing the specific molar flux, i. e. the molar flux of P1 per unit reactor volume. The curves indicate that substantially higher product concentrations are achieved with microstructured channels. A fixed design target, i. e. a specific product concentration at the outlet, is reached at higher values of the E2 inlet space velocity when an efficient heat transfer mechanism is available. The corresponding space velocities for unstructured and structured channels, x_1 and x_2, are then used to determine the specific molar flux of the product, as indicated in the lower part of Fig. 4.

By employing microstructuring techniques the specific molar flux can be increased by at least a factor of 2, leading to a decrease of system size and a reduced requirement for catalyst. Instead of reducing the catalyst mass and the overall size of the system the objective could be to increase the conversion to the product. As apparent from Fig. 4, the microstructured reactor could make regions of high product concentration accessible that cannot be reached with unstructured microchannels.

While the previous results indicate a superior performance of microstructured versus unstructured channels, they do not define microreactor performance in absolute terms. In order to assess the benefits of microreaction technology, microreactors have to be benchmarked against conventional approaches. For this purpose, the catalyst mass needed to reach the target conversion was compared to the corresponding value obtained with the conventional fixed bed technology. Even though the volume fraction of catalyst is significantly smaller in microreactors than in conventional equipment, a substantial reduction of vessel size is expected when a reactor composed of layers of microchannels is used. In case of the process under study a reduction of reactor volume by about one order of magnitude is predicted, underlining the increase of specific performance by means of miniaturization.

4 CONCLUSIONS

In the first part of this work two different approaches for heat transfer enhancement were set forth and assessed by CFD methods. It was found that especially channels equipped with microfins allow for a rapid exchange of heat. Such designs exhibit a potential to construct very compact heat exchangers and lend themselves as components of heat-exchanger reaction systems.

In the second part a model of a heat-exchanger reactor was set up. The numerical results clearly showed a superior performance of the reactor containing microstructured heat-exchanger channels compared to unstructured channels. Furthermore, microstructured reactors allow the product concentration to be increased.

Subsequently, the model of the heat-exchanger reactor, both with and without heat transfer enhancement, was benchmarked against a model of the conventional fixed bed technology. The simulations indicate that the catalyst mass can be significantly reduced in microreactors and even further when microstructuring techniques are applied. Correspondingly, when compared to conventional technology, a substantial reduction in system size is predicted.

REFERENCES

[1] W. Ehrfeld, V. Hessel and V. Haverkamp, “Microreactors, Ullmann’s Encyclopedia of Industrial Chemistry”, Wiley-VCH, 1999.

[2] J. P. Hartnett and M. Kostic, Adv. Heat Transfer 19, 247, 1989.

[3] M. Kaviany, “Principles of Heat Transfer in Porous Media”, Springer, 1995.

[4] G. R. Hadley, Int. J. Heat and Mass Transfer 29, 909, 1986.

[5] G. H. Neale and W. K. Nader, AICHE J. 19, 112, 1973.

[6] R. J. Wijngaarden, A. Kronberg and K. R. Westerterp, “Industrial Catalysis”, Wiley-VCH, 1998.