Single-phase fluid flow distribution and heat transfer in microstructured reactors


Published in:
Chemical Engineering Science
Research

Single-phase fluid flow distribution and heat transfer in microstructured reactors

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A R T I C L E   I N F O

Article history:
Received 17 December 2009
Received in revised form
20 May 2010
Accepted 26 May 2010
Available online 9 June 2010

Keywords:
Microreactor
Flow distribution
Numbering-up
CFD
Heat transfer
Thermal conductivity

A B S T R A C T

Single-phase microreactors and micro-heat-exchangers have been widely used in industrial and scientific applications over the last decade. In several cases, operation of microreactors has shown that their expected efficiency cannot be reached either due to non-uniform distribution of reagents between different channels or due to flow maldistribution between individual microreactors working in parallel. The latter problem can result in substantial temperature deviations between different microreactors resulting in thermal runaway which could arise from an exothermic reaction. Thus advances in the understanding of heat transfer and fluid flow distribution continue to be crucial in achieving improved performance, efficiency and safety in microstructured reactors used for different applications. This paper presents a review of the experimental and numerical results on fluid flow distribution, heat transfer and combination thereof, available in the open literature. Heat transfer in microchannels can be suitably described by standard theory and correlations, but scaling effects (entrance effects, conjugate heat transfer, viscous heating, and temperature-dependent properties) have often to be accounted for in microsystems. Experiments with single channels are in good agreement with predictions from the published correlations. The accuracy of multichannel experiments is lower due to flow maldistribution. Special attention is devoted to theoretical and experimental studies on the effect of a flow maldistribution on the thermal and conversion response of catalytic microreactors. The review concludes with a set of design recommendations aimed at improving the reactor performance.

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doi:10.1016/j.ces.2010.05.044
1. Introduction

Despite the growing number of realized applications of microchannels in science and engineering, there is still a low level of understanding of the relevant fluid dynamics and heat transfer processes. Among the research that has been carried out within the academic and industrial communities are investigations of the validity of the macro-scale equations of the friction factor and the Nusselt number on the microscale. Over the course of the past two decades, many conflicting publications with the results on the validity of classical macro-scale equations for microchannel fluid flow and heat transfer have been given. Among this research are the investigations of the validity of the macro-scale equations for the friction factor, the transition Reynolds number, and the Nusselt number on the microscale. Some authors claim that new phenomena occur in microchannels, while many others report that there exist several effects, usually neglected in deriving correlations, which lead to larger discrepancies with observed experimental results in microchannels. Usually several assumptions were made in the literature studies when flow and heat transfer were modeled: (i) steady-state fully developed flow; (ii) the thermophysical properties of the fluid do not vary with the temperature; (iii) simplified boundary conditions (constant wall temperature or constant heat flux); and (iv) the fluid heating due to viscous dissipation can be neglected. The influence of those factors, also referred to as “scaling effects” (Herwig and Hausner, 2003), were considered in a recent review paper (Rosa et al., 2009). This overview on the history of the discrepancies between microchannel friction factor measurements in experiments and macro-scale laminar equations shows that it is primarily the early studies which present these contradictions. More recent studies have shown good agreement with laminar theory. Steinke and Kandlikar (2006) reviewed 150 papers specifically addressing “scaling effects” in microchannels. They concluded that the papers that do not account for the singular pressure losses or the developing flow in the microchannel reported significant deviation from theory.

As most microreactors operate at elevated temperatures, it is often the case that temperature differences in microchannels can create a non-uniform flow distribution due to differences in the pressure drop, which is temperature dependent (Chaudhuri et al., 2007). This situation can be observed even when a proper flow distributor is applied. Therefore the problem of flow distribution is closely related to the temperature distribution in a downstream microreactor or micro-heat-exchanger. On the other hand, in most heat transfer and pressure drop calculations of microstructured reactors, it is presumed that the inlet flow and temperature distributions across the reactor cross-section are uniform. These assumptions are generally not realistic under actual operating conditions due to various reasons (Ranganayakulu et al., 1997; Ranganayakulu and Seetharamu, 1999; Rao et al., 2002). These facts are well recognized in the literature and their effects were intensively studied over the last decade. However, the studies considering combined effects of wall heat conductions and inlet flow distribution on the temperature distribution are very limited.

This paper presents a review of the experimental and numerical results on fluid flow distribution, heat transfer and combination thereof available in the open literature. At the moment, there is a general agreement that both fluid flow and heat transfer in microchannels can be accurately described by standard theory and correlations, but scaling effects (entrance effects, conjugate heat transfer, temperature-dependent properties, non-uniform flow distribution, and singular pressure losses) have often to be accounted for in the design of individual parts of microsystems. In the first part of the paper, the main methods to control flow distribution are reviewed. Several different designs of inlet/outlet chambers are presented together with corresponding models used for optimization of flow distribution. The models accounted for the effects of non-uniform fluid distribution and additional pressure losses due to sudden expansion/contraction of the fluid flow in microsystems are discussed. In the second part, recent achievements in understanding of heat transfer in microchannels are presented. Finally, several examples of microreactor performance optimization by creating specific designs accounting for optimal flow and temperature distributions are presented.

2. Flow equalization devices to create flow equipartition

The use of microchannel devices for industrial production requires the arrangement of a large number of channels in a parallel network to enable operation with macro-scale flow rates. Therefore, the numbering-up is generally the main operation concept used in this regard. Usually, this operation is seen as a simple multiplication of an elementary system. However, investigations of a set of parallel channels demonstrated that performance of microstructured devices decrease when flow maldistribution is exhibited. The performance of a single microreactor cannot be repeated in an assembly operating at the industrial scale, without considering the effect of flow maldistribution on process performance. The flow non-uniformity in a microstructured reactor can be divided in gross and channel-to-channel non-uniformity (Mueller and Chiou, 1988). The latter is caused by manufacturing tolerances, the differences in the thickness of catalytic layers, and the local temperature differences in the microchannels, which change the physical properties of the fluid. However, usually such differences do not exceed 5%. On the other hand, the gross flow non-uniformity is mainly associated with poor design of the header configuration, or with improper choice of the drag coefficient of the flow distributor. Especially at low Re numbers (below 10), which are often observed in microreactors, the ratio of the maximum flow velocity to the minimum flow velocity in different channels can be as high as 2–4.

2.1. The importance of scaling effects in microchannels

Despite the existing discrepancies between published results, the most recent results showed that the flow behavior in microchannels is similar to that for larger size channels and standard theories and correlations are suitable. The transition from laminar to turbulent flow occurs, depending on the geometry of cross-section, at a Re number in the range 2100–2300. In several studies the effect of surface roughness on the friction factor in laminar flow was considered as a possible factor for deviations from classical theory. Judy et al. (2002) performed measurements of frictional pressure losses over a range of flow rates corresponding to laminar flow (Re number: 8–2300) in tubes with a diameter between 15 and 150 μm for three different fluids with different dipole moment (water, methanol, and isopropanol) with two tube materials (stainless steel and fused silica) in round and square cross-sectional geometries. Guo and Li (2003) studied flow behavior in capillaries with diameters ranging from 80 to 166 μm. The authors
concluded that in the range of Re numbers below 300 viscous heating of the fluid as a possible source of energy dissipation in small tubes can be neglected. The product of the Darcy friction factor and the Reynolds number, $f$ and Re, was equal to 64. Kleinstreuer and Koo (2004) compared numerical model predictions with several experimental data sets where the surface roughness was significant as compared to pipe diameter. The authors concluded that surface roughness might affect the friction factor and hence the flow parameters in rough channels made of aluminum or stainless steel. However in a more recent study, Ko et al. (2008) investigated the variation of the pressure difference between the inlet and the outlet as a function of fluid flow rate and compared it with the classical theory for channel heights of 23, 39, 43, and 55 μm. A good agreement was observed in the whole range of hydraulic diameters. Their result indicated that there was no reduction or enhancement in the friction on the wall, which might be due to the reduction in viscosity or the surface roughness effect. Furthermore, an earlier onset of transition from laminar to turbulent flow as mentioned by others (Mala and Li, 1999) was not observed in more recent studies.

### 2.2. Design criteria for application of equalization devices

It is well known that, when the inlet tube of a heat exchanger is small compared to the size of the inlet header, the fluid tends to go preferentially into the microchannel that faces the inlet tube (Rohsenow et al., 1985). The internal (hydraulic) diameter of the inlet flow duct of the microreactor is typically between 1 and 4 mm and the inlet duct is sufficiently long, so that at a certain distance from the inlet region of the duct, a constant parabolic velocity profile develops, with maximum at the axis and with zero at the walls:

$$\frac{w}{w_{\text{max}}} = 1 - y^2$$  \hspace{1cm} (1)

It is easy to evaluate the Coriolis’s coefficient ($N_0$) for this case. Assigning the normalized deviation of velocities from the average value as $\Delta \bar{w} = w/\bar{w}$, it can be derived that

$$N_0 = \frac{1}{A_0} \int_{A_0} (1 + \Delta \bar{w})^2 \, dA = 1 + \frac{3}{A_0} \int_{A_0} \Delta \bar{w} \, dA + \frac{3}{A_0} \int_{A_0} \Delta \bar{w}^2 \, dA$$

$$+ \frac{1}{A_0} \int_{A_0} \Delta \bar{w}^3 \, dA$$  \hspace{1cm} (2)

The second term of this sum equals zero by the definition of the average velocity, while the fourth term is alternating and small, so it can be neglected. Therefore, for a parabolic velocity profile:

$$N_0 \approx 1 + \frac{3}{A_0} \int_{A_0} \Delta \bar{w}^2 \, dA = 1.57$$  \hspace{1cm} (3)

A sheet screen (grid) is the most effective and simple way to distribute fluid flow equally through a whole cross-section of the reactor. Such a grid creates additional fluid flow resistances, which are equally distributed across the whole cross-section of the reactor. The screen-leveling property depends on its geometrical parameters such as the effective (open) cross-section and the thickness of the resistance layer (resulting from screens). The resistance of the grid increases with decreasing effective cross-section or with increasing thickness of the resistance layer. These parameters define the drag coefficient of the screen, expressed as

$$\zeta_{sc} = \frac{2 \Delta \rho}{\rho \overline{w}_{sc}^2}$$  \hspace{1cm} (4)

where $\overline{w}_{sc}$ is the average velocity, $\rho$ the fluid density, and $\Delta \rho$ pressure drop along the screen. A similar expression can be applied to estimate the resistance of the downstream equipment (e.g. microreactor). For example, the critical value of the screen drag coefficient estimated by Eq. (4) is equal to 30 for the ratio of cross-section of the microreactor to the inlet tube of 6. It has to be mentioned that there is a clear difference between planar (or thin-walled) and three-dimensional (thick-walled) screens. The former does not have guiding walls while the latter consists of a number of thick bars, whose thickness in the direction of the fluid flow equals or exceeds the spacing between them.

### 2.3. Distributing headers

In general, the greater the diffuser (header) expansion angle $\alpha$, the steeper the velocity drop at the walls of the diffuser and the more elongated is the velocity profile, i.e. the higher is the ratio of the maximum velocity to the mean velocity, at the entrance to the reactor channels. At constant inlet conditions and constant relative length of the diffuser, there exist four principal flow modes depending on the angle $\alpha$ of the flow diffuser (Table 1).

In microreactors it is desirable to avoid large dead volumes; therefore diffusers of type IV are usually applied. This means that an elongated velocity profile is developed with a ratio of the maximum velocity to the mean velocity larger than 2.5 at the entrance to the upstream channels (Idelchik and Ginzburg, 1974). This observation has been confirmed in a number of studies performed with microfluidic devices. With a conical header, the flow velocity was above average in 1/3 of the central passages (Jiao et al., 2003a). The ratio of the maximum to minimum velocity increased from 2.08 to 2.81 with increasing Re number. Experimental investigation on flow maldistribution in plate heat exchangers has been studied by several authors. Jiao et al. (2003a) studied flow maldistribution in five flow distributors with inlet angles of 65°, 49°, 45°, 41°, 34° (Fig. 1a and b) and compared with the base case ($\alpha = 90°$) under similar inlet flow conditions.

The cross-section of the downstream heat exchanger (not shown) was $140 \times 100 \text{mm}^2$. Due to the small dimensions of the individual flow passages, a more coarse passage-to-passage scheme was introduced, where the flow was measured at the cross-sections of 16 representative zones with a cross-section of $35 \times 25 \text{mm}^2$, assuming that the flow was uniform in each of those zones. The minimum flow non-uniformity was obtained at an inlet angle of 45°, however the difference between the minimum and maximum flow rates was 6%, even in the best

### Table 1

<table>
<thead>
<tr>
<th>Mode</th>
<th>Diffuser angle (deg.)</th>
<th>Flow description</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>Separation free diffusers</td>
<td>$\alpha \leq 4$</td>
</tr>
<tr>
<td>II</td>
<td>Diffusers with local flow separation</td>
<td>$6 &lt; \alpha \leq 14$</td>
</tr>
<tr>
<td>III</td>
<td>Diffusers with significant flow separation</td>
<td>$14 &lt; \alpha \leq 40$</td>
</tr>
<tr>
<td>IV</td>
<td>Diffusers with total flow detachment</td>
<td>$\alpha &gt; 40$</td>
</tr>
</tbody>
</table>
case. The relative flow non-uniformity increased as the Re number increased from 500 to 2000. An empirical correlation between the flow maldistribution parameter as a function of the Re number was proposed. The effects of the inlet pipe diameter, the diameters of the first and second headers and the drag coefficient of the header on the flow distribution in a micro-heat-exchanger have been studied (Jiao et al., 2003b). The geometry was composed of two headers with configurations B and C, Fig. 1c) or without (configuration A) a thin-walled screen between them. The screen had either five openings (D=25; 20; 15; 20; 25 mm, configuration B) or seven openings (D=25; 20; 15; 20; 25; 25 mm, configuration C). The authors calculated the drag coefficient of both screens. The ratio of maximum to minimum velocity decreased from 2.57 to 2.08 when configuration A was replaced with C at Re=1100. The authors introduced the equivalent inlet and outlet diameter for each header (\(D_{in}\) and \(D_{out}\)) in a way that the area of the circle with the diameter \(D\) is equivalent to the sum of all corresponding cross-sectional areas of flow passages. According to this definition, \(D_{in}\) equals the internal diameter of the inlet pipe, \(D_{out} = \frac{A_{in}}{\pi}\), and \(D_{out} = \frac{A_{out}}{\pi}\). In a subsequent study, Jiao and Baek (2005) investigated the effect of the configuration parameter \(h/H\) on flow maldistribution (Fig. 1b). The ratio of maximum to minimum velocity decreased from 2.57 to 2.08 when the \(h/H\) ratio was varied from 0 to 0.2 at Re=1100. Further increase of the configuration parameter to 0.3 resulted in an increase of \(u_{max}/u_{min}\) to 2.21. At the same time, the relative flow non-uniformity also decreased from 0.012 to 0.009 and then increased again to 0.012.

The issue of exit-port shape, although clearly of practical relevance, seems not to have been investigated in the open literature. There is only one study where an attempt was made to compare the performance of a flow distribution manifold for fully developed turbulent flow at three different exit-port geometries: an array of discrete slots, an array of discrete circular apertures, and a single continuous longitudinal slot with the same open flow area (Chen and Sparrow, 2009). The best performance was achieved by the continuous slot outlet. For that case, the overall deviations from the ideal case were within ±5%. The least uniform outflow from the manifold occurred for discrete circular exit ports with deviations from the mean value of ±15%. The performance of the three exit-port geometries was rationalized by their different resistances to fluid flow. The authors concluded that the discrete ports offer a higher resistance to the outflowing fluid as compared to that of the continuous slot.

2.4. Thin wall screens (meshes, gauzes)

A thin, uniformly perforated grid is often introduced in the inlet header to improve flow distribution (Lalot et al., 1999; Norton et al., 2006). Lalot et al. (1999) experimentally investigated the effect of flow non-uniformity on the performance of plate heat exchangers. It was possible, depending on the drag coefficient of the screen, to modify the ratio of maximum to minimum velocity from 4 to 1.5–2. Further improvement was not possible due to a reverse flow in the section downstream from the screen. This result was in a good agreement with earlier studies performed by Idelchik (1991, 1993) who demonstrated that fluid flow equalization cannot be achieved beyond a screen critical value (\(\gamma_{cr}\)):

\[
\gamma_{cr} = 3.5 \left( \frac{N_1A_{sc}}{A_0} - 1 \right)
\]
In this expression, also known as Idelchik’s correlation, $N_0$ is the Coriolis' coefficient, $A_{sc}$ and $A_0$ are the open cross-sections of the screen and the inlet flow duct, respectively. On the other hand, there is an optimum value for the drag coefficient of the flat screen

$$z_{opt}^{sc} = N_0 \left( \frac{A_{sc}}{A_0} \right)^2 - 1$$

(7)

It is easy to see that for a planar screen the design criterion $z_{opt}^{sc} < z_{cr}^{sc}$ is satisfied only in a narrow range at $A_{sc}/A_0 < 2.9$. Otherwise a planar screen can even amplify the flow non-uniformity downstream of it.

2.5. Thick wall screens

The problem of a critical value for the drag coefficient does not exist in the case of thick wall screens, as the degree of velocity equalization is virtually the same at all cross-sections downstream from them. In this case, the nature of the distribution of the inlet flow under a given set of flow conditions is a function only of the shapes and geometric parameters of the downstream reactor and the inlet flow duct of the screen. Several examples of applications of thick-walled screens for flow equalization were reported in the literature. Schrenk et al. (1992) used a flow distributor which consists of three functionally distinct sub-elements. The first sub-element divides a fluid stream into branch streams. Then, the second sub-element expands the branch streams. Finally, the third sub-element performs the function of contracting the branch streams, independently adjusting their flow rates and recombining them into an integrated whole. A feature of common significance is the symmetrical expansion and contraction of the divided and stacked branch streams. Two studies are related to improvement of the performance of plate-fin heat-exchangers operating in the turbulent mode (Li and Zhang, 2003; Wen and Li, 2004). The authors studied the fluid flow maldistribution using computational fluid dynamics (CFD) simulations. The effects of the configuration of header and distributor on the flow distribution have been investigated. Numerical investigation showed that application of a two stage distribution structure (header+screen, see Fig. 2) improved the flow distribution (Wen and Li, 2004).

Comparing to the applications mentioned above we presented a configuration of the header intended for operation in laminar mode with low $Re$ numbers ($Re < 10$) resulting in a negligible pressure drop across the reactor. The header consists of a fluid flow diffuser and a thick-walled screen with a thickness of the parallel bars in the micrometer range (Rebrov et al., 2007a). A schematic view of the header is shown in Fig. 3.

The header consists of a cone diffuser and a thick-walled screen positioned in front of the microreactor. The thick-walled screen consists of two sections positioned with a 90° turn relative to each other. The first section ($U$) comprises a set of $m$ elongated parallel upstream channels, and the second section ($D$) comprises a set of $n$ elongated parallel downstream channels positioned at an angle of 90° with respect to the upstream channels. A schematic view of the header is shown in Fig. 3.

Fig. 3. The header consists of a cone diffuser and a thick-walled screen positioned in front of the microreactor. The thick-walled screen consists of two sections positioned with a 90° turn relative each other. The upstream section ($U$) comprises a set of $m$ elongated parallel upstream channels, and the downstream section ($D$) comprises a set of $n$ elongated parallel downstream channels positioned at an angle of 90° with respect to the upstream channels. Adapted from Rebrov et al. (2007a).
other. Both the upstream and downstream sections comprise a set of elongated parallel channels. The geometry is defined by several parameters, among them parameter $a$, the minimum length between two neighboring downstream channels, and parameter $b$, the distance in cross-sectional view between a top wall of the first downstream channel and a side wall of the upstream channels. These parameters were found to have the highest impact on the flow equalization. A correlation is proposed for the design of the geometry of the screen (Rebrov et al., 2007b):

$$b = 0.5a + 0.1678c + P3(c, d)$$

where $c$ is the width of the upstream channels and $d$ the height of the downstream channels.

An additional distance ($b > a/2, P3 > 0$) was required to equalize a difference in the hydraulic conductance for the fluid flow between the side and the middle portions of the upstream channels. In this case, if the flow is equally distributed between the first and second downstream channels, flow separation between all subsequent downstream channels would always happen in corresponding equidistant planes. The design of the thick-wall screen was experimentally validated using laser Doppler anemometry, which is a direct method for the determination of the fluid velocity in the separate microchannels (Mies et al., 2006, 2007a). In these experiments the gas was replaced by a liquid as flowing medium to maintain the same Reynolds number. The flow non-uniformities in the reactor channels remained below 2% in the complete range of Reynolds numbers of 6–113.

2.6. Reaction plates with quadrant shape (constant distance between inlet and outlet ports)

The use of both macroscopic momentum balances and detailed CFD models to design flow distribution systems has been presented by various groups. Application of CFD simulations to optimize design of a microreactor requires too much computational time. Several groups used a pressure drop compartment model in the fluid design stage to estimate the pressure distribution over a microreactor including inlet and outlet manifolds. In the pressure drop compartment model, the relation between the inlet and outlet pressures is described as a function of the design parameters and flow velocity. Through the fluid design, the shapes of the inlet and outlet manifolds and the number of parallel microchannels are determined to optimize a given performance index such as the minimization of the total residence time of the microreactor.

Amador et al. (2004), Tonomura et al. (2004), and Delsman et al. (2004a) studied manifold structures with quadrant shape used for even flow distribution in plate-fin microreactors (Fig. 4a).

Fig. 4. (a) Schematic view of a reaction plates with quadrant shape. Adapted from Delsman et al. (2004a), (b) CFD simulation of a geometry with one inlet and two outlets. Adapted from Balaji and Lakshminarayanan (2006), and (c) equivalent electrical resistance network model. $R$ is the flow resistance, and $Q$ is the fluid flow rate. Adapted from Pan et al. (2008).
All authors concluded that the flow uniformity in the microchannel is dependent on the manifold shape (the section of the plate between the inlet and the microchannels), length and location of fins and inlet flow rate. In several geometries, the highest pressure drop was localized to cause uneven flows along the microchannels. Delsman et al. (2004a) and Griffini and Gavriilidis (2007) used 3D CFD models to study the flow distribution over plate-fin microreactor plates. The CFD calculations showed that 2D simulations can be misleading in assessing flow misdistribution. The latter study revealed the presence of a critical value of the Reynolds number corresponding to a transition from a flow regime fully determined by viscous forces, where fluid distribution is independent of flow rate, to a regime affected by inertial effects. Two designs employing parallel fins with a length of 135 and 45 mm were studied to improve the flow distribution as compared to the plate without fins. The depth and width were kept constant at 0.41 and 7.1 mm, respectively. Increasing the length of the fins and/or decreasing the plate width considerably improved the flow distribution.

A 2D CFD model has been used by Balaji and Lakshminarayan (2006) to improve the design by addition of multiple inlet (or outlet) ports. From the results obtained, the authors concluded that the plate geometry with one inlet and two outlet ports provided the most uniform flow distribution under a wide range of flow conditions (Fig. 4b). However, no 3D simulation results were presented confirming this. Tomonura et al. (2004) used a 3D CFD model and applied an algorithm for optimization of the manifold shape. After a CFD simulation, the performance index was calculated. While the result is not optimal, the shape was updated and the mesh was automatically determined by a mesh generator. However, this approach still required substantial computational time. Huang et al. (2008) found that a new design using small guide vanes equally spaced around the feed header of the double-inlet/single-outlet module produced the highest degree of flow uniformity.

In an attempt to reduce computational time, several authors used approximate models based on a simplified description of the reactor as a network of equivalent rectangular ducts. Commenge et al. (2002) developed an analytical model for microreactor plates based on wall friction in the distribution chambers and the channels when the inlet was perpendicular to the microchannels. Since they neglect the influence of inertia forces, their model was only valid at low Re numbers. Amador et al. (2004) proposed an analytical model which is analogous to the electrical resistance networks. This model can be used at low Reynolds numbers to study the effect of manufacturing tolerances and channel blockages on the flow distribution in the downstream microchannels.

Pan et al. (2008, 2009) studied the influence of triangle manifold geometries on the flow distribution between microchannels by an analytical model and an equivalent electrical resistance network model (Fig. 4c). The authors developed an optimization procedure to calculate the manifold geometries and dimensional variations to obtain flow equipartition in microchannels. The inlet and outlet manifolds were divided in several rectangular channels based on the analysis of pressure distributions in the manifolds performed by numerical simulation. Then an equivalent resistance network model was applied to establish the relationships between the velocities and pressure drops in microchannels and rectangular channels. Similar to Ohm’s law, the relationship of pressure drop ($\Delta p$), flow rate ($Q$) and flow resistance ($R$) was defined as follows:

$$\Delta p = RQ$$

(9)

The flow resistance was defined as

$$R = \frac{32\mu L_{NC}}{D^2A}$$

(10)

where $\mu$ is the fluid viscosity, $L$ the channel length, $\lambda_{NC}$ the non-circular coefficient (Commenge et al., 2002), $A$ the cross-sectional area of rectangular channel, and $D$ the channel diameter. The rectangular channels of inlet distributing manifold and outlet collecting manifold were numbered up from 1st to Nth, respectively. The pressure drops via different flow channels should be equal (Amador et al., 2004), therefore $N$–1 equations were expressed as follows:

$$R_{in}(j)Q_{in}(j) + R_{c}(j)Q_{c}(j) = R_{out}(j-1)Q_{out}(j-1) + R_{i}(j-1)Q_{i}(j-1)$$

(11)

for $j=2, 3, \ldots, N$. The results demonstrated that the division of the manifold into a number of rectangular channels is a valid approach at low $Re$ numbers between 0.6 and 6. Additional corrections for singular losses should be introduced at $Re$ numbers in the range from 60 to 300 (Pan et al., 2009). This correction factor accounts for additional losses due to flow turning in the manifold and sudden expansion/contraction in the inlet/outlet sections. In this case velocity distributions were asymmetrical, with higher values of velocities in the microchannels further away from the inlet port. With further increasing Reynolds number towards 600, the resistance network model fails to predict the results of numerical simulations. The microchannel width was found the most important parameter that influences the optimization procedure.

Bogojevic et al. (2009) studied the influence of channel blockage on flow distribution. The authors performed CFD simulations in order to systematically assess the influence of channel blockage in different locations of the microchannel array consisting of 20 channels. Blockages in the middle channels had the smallest influence on flow distribution, while side channel blockages made flow distribution more uneven.

2.7. Bifurcation approach

Amador et al. (2004) studied flow distribution in a bifurcation manifold (Fig. 5). The authors concluded that the bifurcation structure produced flow equipartition when all channels at the same level have exactly the same geometrical characteristics, the length of the straight channel after each bend is sufficient to produce a fully developed velocity profile and there is no variation in the channel diameters due to manufacturing tolerances.

2.8. Multiscale networks

O-Charoen et al. (2007) studied the effects of microreactor depth, microchannel depth and width, and microreactor inlet width and length on the flow uniformity and pressure drop in microfluidic microarray devices. Due to limited computational resources, the geometry was scaled down from thousands of microreactors in the microarray device to 105 square-shaped...
microreactors. These microreactors were divided into 7 columns as shown in Fig. 6.

A parametric study was performed on the designed geometric variables to study their effects on the flow and pressure profiles. The microreactor depth and microchannel width were found to have the most significant effects on the flow pattern inside the microfluidic platform. Consequently, the flow uniformity was significantly improved by altering these parameters. As a result of the CFD study, an improved geometry of the microfluidic assembly was proposed and fabricated. The numerical results were verified by flow visualization with a fluorescent tracer. The authors observed a good agreement between the experimental results and the simulations.

Saber et al. (2009, 2010) proposed a model for rapid estimation of flow distribution in a complex network comprising several different scales. They found that a two-scale configuration of the flow distributor yielded the most uniform flow distribution and the smallest total pressure drop (Saber et al., 2010). Tong et al. (2009) performed a systematic study applying several individual strategies, which were considered to be promising for flow equalization in manifolds. Those strategies included: (i) enlargement of the cross-sectional area of the distribution manifold, (ii) linear or non-linear tapering of the cross-sectional area of the distribution manifold or both the distribution and collection manifolds, (iii) variation of the cross-sectional areas of the microchannels, (iv) tailoring of the inlet duct, and (v) contouring of the shapes of the inlets of the microchannels. At this stage, the authors did not present the design rules, however they concluded that the first approach was the most promising one for further investigation.

2.9. Constructal distributor

Bejan and Errera (1997) found that a fractal tree-like network structure (observed widely in natural structures such as cracks in a dry ground, lungs, arteries or veins) not only gave flow uniformity but also minimized flow resistance of a volume-to-point path. Bejan (2000a, b) and Chen and Cheng (2002) demonstrated that the optimal path to distribute a fluid between a volume (several sets of microchannels) and a point (inlet tube) providing minimal global resistance is a tree-like network. The channel network has the

Fig. 6. (a) Cross-sectional view of the full model and (b) one unit of the microreactor model: (top) 3D geometry of a microreactor unit; (bottom left) top view (x-y plane); (bottom right) front view (x-z plane). The parameters are (a) length of microreactor inlet/outlet, (b) width of microreactor inlet/outlet, (c) microchannel width, (d) microreactor depth, and (e) microchannel depth.
Adapted from O-Charoen et al. (2007).

Fig. 7. Constructal distributor with 4 layers, 8 scales and 256 outlets: (a) 3D view and (b) schematic view of a cross-section perpendicular to the flow direction.
Adapted from Tondeur and Luo (2004).
structure of a sequence of several generations of T-bifurcations (Fig. 7).

The inlet channel is split perpendicularly into two opposing channels (generation 1), and each of these is again split into two channels (generation 2), such that channels of generations 1 and 2 are coplanar. These three successive generations of channels form the basic pattern, the elementary cell, which is reproduced at smaller scales. The same network was obtained by minimizing the time of discharge in the flow from a volume to one point. Ondez et al. (2003) extended the constructal approach from simple to more complex structures. A solution was proposed for optimal flow paths for flow distribution between one point and 3, 8, and 24 points positioned equidistantly in a square pattern in a plane.

Tondeur and Luo (2004) established the optimal scaling relation between the channel radii of successive generations of $2^{1/3} \approx 1.26$. They noticed that the pressure drop in a constructal distributor is not governed by Poiseuille's law, because of direction changes and flow splitting. The authors proposed a new approach in which they accounted for pressure losses ($\Delta p_k$) using a semi empirical power law function of the channel radius ($r_k$) (Luo and Tondeur, 2005):

$$ f_k = \frac{f_0}{2^k} $$ (12)

$$ \Delta p_k = \frac{a \mu f_0^2 l_k}{\pi} \left( \frac{r_k}{r_1} \right)^{m} $$ (13)

where $l_k$ is the channel length in generation $k$, $f_k$ the volumetric flow rate, and $a$, $p$, and $q$ the empirical constants. This form of pressure drop expression is analogous to Poiseuille's law at $k=1$, $p=4$, $a=8$. Another important observation from this study was that small manufacturing imperfections may generate rather large differences in flow distribution over a large surface area. This showed that the "optimal" geometric design is not sufficient to ensure flow equipartition as this type of design is not robust with respect to local geometry imperfections that are often a case during microreactor fabrication. They concluded that constructal distributors can be adapted to square cross-sectional areas. A straightforward extension to rectangular surfaces is possible, when the length is an integer multiple of width. For surfaces of different shapes, e.g. circular, a different topology must be introduced.

Luo et al. (2008) examined experimentally the effects of constructal distributors and collectors, built on a binary pattern of pores, on flow distribution in a micro-heat-exchanger. Both the thermal performance and pressure drop were determined with different assembly configurations of constructal and pyramid distributors and the inlet and outlet of the micro-heat-exchanger. The introduction of constructal distributors at the inlet and outlet sections increased the overall heat transfer coefficient by 30% due to better flow distribution while the pressure drop was increased by 1.7 times. The application of a constructal distributor at the inlet and a conventional pyramid distributor at the outlet increased the overall heat transfer coefficient by 15%. However, the application of a constructal distributor at the outlet and a conventional pyramid distributor at the inlet increased the overall heat transfer coefficient by 28%, while the pressure drop was increased by 1.3 times in both cases at $Re$ number in the range 500–700. The authors suggested that further multi-objective optimization is necessary to reach a compromise between flow equidistribution, void volumes, dispersion and pressure drop.

Senn and Poulikakos (2004) applied constructal distributors for flow distribution in fuel cells with the goal to optimize the geometry with respect to maximum electric power output at a minimum flow resistance. The authors concluded that the tree network system provided reduced the pressure drop at the same net power density.

3. Heat transfer in microchannels

Heat transfer in large-sized channels has been extensively studied during the last fifty years and there exist well-established engineering correlations for calculation of heat-transfer coefficients. For example, for fully developed laminar flow, the Nusselt number ($Nu$) is a constant whose value depends only on cross-sectional geometry and boundary conditions. For circular tubes, $Nu=3.66$ and 4.36 for isothermal wall and constant heat flux boundary conditions, respectively (Shah and London, 1978). More recently, many correlations have been proposed in the literature for heat transfer, based on experimental investigations on liquid and gas flow in mini- and micro-channels. Sobhan and Garimella (2001) presented a comprehensive review of these investigations carried out between 1990 and 2000. Many experimental studies have shown considerable deviations from conventional theory for heat transfer (e.g. Weisberg et al., 1992; Peng and Wang, 1993).

Other authors found that their experimental results were adequately predicted by conventional correlations (Webb and Zhang, 1998; Qu and Mudawar, 2002). With the availability of enhanced computational capabilities, more accurate simulations of fluid flow and heat transfer in microchannels of different dimensions and aspect ratios have been reported over the last decade. It should also be noted that in those experimental investigations in microtubes the axial heat flux was not measured. The axial heat conduction in the channel wall in large pipes can indeed be neglected because the wall thickness is usually very small compared to the channel diameter. However, heat transfer in a microchannel is a combination of axial heat conduction in the solid and convection to the cooling fluid.

3.1. Application of 1D heat transfer correlations

Harms et al. (1999) studied heat transfer of water in rectangular microchannels of 251 $\mu$m width and 1000 $\mu$m depth. In the laminar regime, the measured local $Nu$ numbers agreed well with classical developing flow theory. Guo and Li (2003) concluded that experimental results can only be described with $Nu$ number below 3 when a 1D heat transfer model did not take into account heat conduction in the reactor wall. Tiselj et al. (2004) studied the effect of axial heat flux on heat transfer in a microchannel at $Re$ numbers in the range 3–160. The authors experimentally determined the axial heat flux values to verify their numerical calculations performed with a model which included a silicon chip with 17 parallel triangular microchannels with a length of 15 mm and with steel inlet and outlet collectors. The cross-section of each channel was an isosceles triangle with a length of 15 mm and with steel inlet and outlet collectors. The cross-section of each channel was an isosceles triangle with a base of 310 $\mu$m and the angles at the base of 55°. The hydraulic diameter was 160 $\mu$m. They showed that dependence of the $Nu$ number as a function of axial channel position, $z$, has a singular point at 0.012 $m$ (Fig. 8).

At this point, the difference between the wall temperature and the fluid temperature becomes negative and the heat flux is directed from the fluid to the wall. The singular point shifted towards the channel outlet as the $Re$ number increased. Taking this into account, the heat flux was described by conventional heat transfer correlations. The experimental results on temperature distribution were in good agreement with the numerical calculations when isothermal boundary conditions were applied. A similar observation was reported by van Male et al. (2004) who studied the heat transfer in a square microchannel that is heated from one side. The microchannel was contained in a silicon wafer
and was covered by a thin silicon sheet. At the top side of this sheet, heating elements were positioned which emulated the heat that is produced as a result of an exothermic chemical reaction. A correlation for Nusselt number was derived as a function of the Graetz number both for laminar and plug-flow conditions. The authors reported that the Nusselt number has a singular point, in which the heat flux through the side and bottom walls changed direction when the gas inside the channel was heated above the temperature of these walls.

Lee et al. (2005) performed an experimental study of heat transfer in rectangular microchannels with a channel width from 194 to 534 μm and with the depth of five times the width in each case using both a full 3D conjugate approach and a simplified thin wall heat transfer model. The results obtained with the thin wall model were in good agreement with the experimental data when boundary conditions comprising uniform temperature and axially uniform heat flux with circumferential temperature uniformity (so-called thin wall boundary conditions) were applied. In case of either constant temperature or constant heat flux boundary conditions, the deviations from the 3D full conjugate analysis were 12.4% and 7.1%, respectively, demonstrating the importance of setting accurate boundary conditions in the simulations. For this reason, the authors recommended the use of numerical simulations with thin-wall boundary conditions, instead of 1D correlation, to predict the performance of microchannel heat exchangers.

3.2. CFD modeling of axial heat conduction

Recent modeling studies of micro- and minichannel networks have argued the importance of solid-phase axial heat conduction. Fedorov and Viskanta (2000) investigated heat transfer in a heat sink with rectangular microchannels with a 3D numerical model for fluid flow and the heat conduction in the silicon substrate. The authors obtained a good agreement with experimental results and found rather complex heat flux patterns due to a strong coupling between convection in the fluid and conduction in the silicon substrate that can only be resolved by a detailed 3D simulation. Extremely large transverse and longitudinal temperature gradients have been predicted within the solid wall in the immediate vicinity of the channel inlet. Deshmuck and Vlachos (2005a, b) emphasized the significance of reactor framework material in thermally integrated combustor–reformer microreactors. They studied coupled catalytic ammonia decomposition and homogeneous propane combustion using a 2D CFD model with an objective to recover power from the propane/air combustion to produce the maximum flow rate of hydrogen via ammonia decomposition. The decomposition reaction was modeled using a microkinetic model over a Ru catalyst while the combustion reaction is modeled as an irreversible one-step reaction using a power law kinetic model. The authors found that operation of coupled microdevices is feasible but is limited to high conductivity materials that could decrease temperature gradients in the combustion section. However, the choice of the best operational mode (co-current or counter-current) at the microscale depends on the ratio between the convective and conductive heat fluxes and may differ from that of large-scale devices. To clarify whether or not it is justified to omit the axial heat conduction term in the fluid, Hardt and Baier (2007) performed an analysis of corresponding terms in the heat-transfer equations using a ratio of channel diameter to channel length of 0.01, $Re=10$, $Pr=1$. They obtained a ratio of the conductive to the convective terms of $10^{-3}$ in microchannels of 100 μm hydraulic diameter and showed that axial heat conduction in the fluid phase may usually be safely neglected, compared with convection. There are a few exceptional cases where this ratio can be higher, for example in reactors into which a non-preheated fluid is fed to the hot entrance region of the reactor and being heated up rapidly while undergoing an exothermic reaction (Stutz and Poulikakos, 2005). In the latter study, the effect of axial heat conduction in an autothermal methane microreformer was studied numerically using insulated wall and conductive wall boundary conditions. The former model neglected wall conductivity. The authors showed with increasing the wall thermal conductivity, that the maximum wall surface temperature is decreased. The axial variation of the wall temperature was not monotonic. The maximum methane conversion is obtained if the channel wall is not conductive. A conductive channel wall yielded methane conversion up to 16% lower.

Norton et al. (2006) proposed a tunable microreactor in which effective thermal resistance of the channel wall can be changed. In this design metallic plates of different thermal conductivities and thicknesses could be placed between the reactor walls and an outer alumina insulating jacket (Fig. 9).

This design allowed for a systematic investigation of the effect of wall thermal conductivity on temperature uniformity during the highly exothermic combustion of hydrogen or propane in air. A copper plate was used to obtain a very low thermal resistance, while no thermal plate, i.e., an air gap between the reactor wall

![Fig. 8. Average water and silicon wall temperature distribution in the microreactor. Adapted from Tiselj et al. (2004).](image)

![Fig. 9. Schematic view of the experimental set-up for study of the effect of wall thermal conductivity. Adapted from Norton et al. (2006).](image)
and the insulating jacket, provided a very high thermal resistance. The inlet and outlet sections were also considered in the modeling. As expected the highest temperature gradient of more than 300 K was observed without using additional metal plates. When a copper plate was used, the thermal zone in the reactor was almost isothermal. The flow rate was a less significant factor in determining thermal uniformity. The authors pointed out that the thermal conductivity of the reactor material should be tailored depending on particular applications. The thickness of the layer between the channels with exothermic reaction and the heat sink channels should also be controlled. Depending on the flow rate and composition, between 60% and 80% of the heat generated by the exothermic reaction was lost to the surroundings even although the system was well-insulated. The authors assigned these large heat losses to inherently large surface area-to-volume ratios in microreactors and concluded that these losses can only be avoided by application of special insulation techniques, such as vacuum isolation. Therefore, single-channel prototypes, even with insulating packaging, cannot resemble the thermal performance of a larger microreactor.

The importance of the axial heat flux in a wall-coated methanol steam reformer microreactor with thick porous catalytic coatings has been studied by Chein et al. (2009). The governing equations for fluid flow, heat transfer and mass transfer with chemical reaction were simultaneously solved using the CFD COMSOL software. The authors noticed that a non-linear temperature profile along the reformer length was established. Along the reformer length, the heat transfer coefficient dropped to a lower value in the regions close to the inlet and outlet of the reformer. The authors observed that the heat flux and temperature distribution were affected by the thickness of the catalyst layer. Based on the numerical results, the minimum value of the heat transfer coefficient was deduced.

### 3.3. Effect of wall thermal conductivity on microreactor performance

In a multichannel reactor assembly, which is not designed as a heat-exchanger reactor, a large portion of the heat created or consumed in the reaction is transported by conduction through the wall material. Thus, the reactor should be designed to ensure fast enough heat conduction to guarantee reaction conditions that are as uniform as possible over the multichannel domain. The importance of axial conduction in the solid wall in a counter-current flow microreactor/heat-exchanger has been reported by Peterson (1999) and Stief et al. (1999). They demonstrated that low thermal conductivity materials (e.g. glass) are necessary to achieve high thermal efficiencies by reducing axial conduction losses (Fig. 10).

Substrate thermal conductivity may be selected either to create a large thermal gradient or a localized hot-spot or to achieve isothermal operation by controlling the rate of axial heat conduction (Fig. 11).

Packaging materials likewise may be selected to either facilitate or hinder heat removal to further manipulate system thermal efficiency (Moreno et al., 2008). Microreactors have been fabricated from a variety of substrates ranging from high thermal conductivity materials such as silicon (Srinivas et al., 2004; Tiggelaar et al., 2005; Quiram et al., 2007), aluminum (Rebrov et al., 2001, 2003b; Ismagilov et al., 2008), molybdenum (Mies et al., 2004; 2007b) to low thermal conductivity ceramics (Alm et al., 2005, 2007; Mitchell and Kenis, 2006; Schmitt et al., 2005) and glass (Dietrich et al., 2005). Microreactors with non-insulating packaging and high thermal conductivity materials were applied for isothermal and safe operation of highly exothermic reactions: a singlet oxygen generator (Hill et al., 2007), direct hydrogen peroxide synthesis (Inoue et al., 2007) and isothermal kinetic studies: CO oxidation (Ajmera et al., 2002, 2003; Srinivas et al., 2004), ammonia oxidation (Rebrov et al. 2001, 2002, 2003b), water gas shift reaction (Rebrov et al., 2007c), complete oxidation of n-butane, ethanol, isopropanol, and 1,1-dimethylhydrazine (Ismagilov et al., 2008), complete oxidation of propane (Men et al., 2009) and preferential CO oxidation (F. Chen et al., 2009; Nikolaidis et al., 2009; Vahabi and Akbari, 2009). On the other hand, microreactors with insulating packaging have been reported for efficient methane steam reforming coupled with methane complete oxidation (Kolios et al., 2002); methanol steam...
reforming coupled with hydrogen oxidation (Delsman et al., 2004b, c; Terazaki et al., 2005; Shah et al., 2005; Shah and Besser, 2007); water gas shift reaction (Mukherjee et al., 2007; Dubrovskiy et al., 2009). Terazaki et al. (2005) used a vacuum glass package for thermal insulation of microreactors. In some cases, thermal conductivity of catalytic coatings should also be considered in the design of catalytic microstructured devices (Chein et al., 2009; Dubrovskiy et al., 2009). For example, in case of Mo2C catalytic coatings with a thickness of 50 μm deposited on a Mo substrate with a thickness of 100 μm (Dubrovskiy et al., 2008), the overall axial heat transfer rate will be substantially higher as compared to that over a Mo plate without coating.

Moreno et al. (2008) investigated the effect of thermal conductivity of reactor material and insulation, on the performance of an integrated microreactor/heat-exchanger both in co-current and counter-current flow modes. A power law kinetic model was combined with a plug-flow reactor model assuming the absence of external and internal mass transfer limitations. The authors compared two ideal cases of (i) isothermal and (ii) adiabatic packaging of the microreactor. Isothermal boundary conditions allow free heat exchange between the microreactor material and the packaging layers. Under these conditions, the wall temperature was equal to the corresponding inlet fluid temperature at each boundary for counter-current systems and it was equal to the weighted-average temperature of the two fluids at each boundary for co-current systems: $T_w = \text{fixed at } z=0, L$. These boundary conditions have also been used by Peterson (1999) and Stief et al. (1999) to study heat transfer between non-reacting fluids in microchannel systems. In adiabatic boundary conditions, heat conduction in and out of the system was not allowed: $dT_w/\text{dz} = 0$ at $z=0, L$. The reaction kinetics was described by the Damköhler number ($Da$). Analysis of heat transfer in the absence of chemical reaction was also performed by these authors to demonstrate the importance of axial conduction on thermal efficiency in heat transfer microsystems. Following previous analysis of non-reacting fluids by Peterson (1999) and Stief et al. (1999), fluid–solid heat transfer was described by the number of transfer units (NTU):

$$NTU = \frac{ha}{\rho C_p}$$

where $h$ is the heat transfer coefficient, $a$ the surface area-to-volume ratio, $\tau$ the residence time, $\rho$ the density, and $C_p$ the fluid specific heat capacity. The solid-phase axial conduction was described by the conduction parameter (CP):

$$CP = \frac{\lambda}{\rho \alpha \nu \rho C_p}$$

where $\lambda$ is the thermal conductivity in the solid phase, $L$ the reactor length, $u$ the superficial fluid velocity. In their analysis, the authors used symmetric conditions ($NTU_1 = NTU_2$, $CP_1 = CP_2$, $T_1 = T_2$). In this case, the heat exchanger effectiveness ($\varepsilon$) was calculated from the resulting temperature profile as follows:

$$\varepsilon = \frac{T_{out,2} - T_{in,2}}{T_{1,in} - T_{2,in}}$$

Heat exchanger efficiencies are shown as a function of NTU for the cases of low, intermediate, and high solid-phase thermal conductivities in isothermal (Fig. 12a) and adiabatic cases (Fig. 12b). The effectiveness increases with an increase of NTU and it does not depend on the conduction parameter for the case of isothermal packaging conditions (Fig. 12a). However, as CP increases, substantial increases in conduction heat losses occur.
Following similar analysis, Stief et al. (1999) identified an optimal range of CP values between 0.1 and 0.5 which correspond to thermally insulating materials with λ of 0.4–2 W m⁻¹ K⁻¹. At adiabatic conditions, the effectiveness deceased at high CP, despite the removal of conductive heat losses (Fig. 12b). At high CP, the system approached the limit of thermal equilibration between the two fluids and the wall, equivalent to co-current operation with a maximum efficiency of 50%. Low thermal conductivity materials allowed creating a substantial temperature gradient along the heat-exchanger length.

The above analysis was also performed for the case of one exothermic reacting fluid exchanging heat with a second non-reacting fluid via the solid wall. The Damköhler number was chosen to ensure complete conversion. Both fluids were fed at the same temperature in order to make the exothermic reaction as the sole heat source in the system. In this case, the system thermal efficiency was defined as

\[
\varepsilon = \frac{T_{2,\text{out}} - T_{2,\text{in}}}{\Delta T_{\text{ad}}} \quad (17)
\]

Increasing NTU over the range 1–5 results in increasing effectiveness regardless of CP, while a further increase in NTU reduced the effectiveness (Fig. 12c). The maximum effectiveness decreased from 0.24 to 0.003 as the conduction parameter increased from 0.1 to 10. The latter case represents an “isothermal slab” limit corresponding to negligible heat transferred between fluids, while the majority of reaction heat is removed to the environment. In the absence of conduction losses via packaging (Fig. 12d), the effectiveness is a weak function of the conduction parameter in the range of NTU from 1 to 5. At NTU above 5, the effectiveness decreased from 0.64 to 0.5 as CP increased from 0.1 to 10. In both cases, low solid-phase thermal conduction creates a localized hot-spot in the solid-phase along the reactor length. Under adiabatic packaging condition, this results in thermal efficiencies up to 65%. The authors demonstrated the importance of both materials and packaging selection for achieving microreactor design goals. A combination of isothermal packaging and high thermal conductivity materials should be applied in applications where hotspot formation is undesirable and heat utilization is not a critical issue. On the contrary, adiabatic packaging and/or low thermal conductivity substrates should be used in microreactor applications where heat utilization is of primary importance.

### 3.4. Steady-state multiplicity in thermally integrated microstructured reactors with insulating packaging

The effect of thermal conductivity (CP) on the system performance and the presence of steady-state multiplicity was studied by Moreno et al. (2008) (Fig. 13). Performance of thermally integrated microreactors constructed with adiabatic packaging (Arana et al., 2003; Terazaki et al., 2005) and/or insulating materials (Frauhammer et al., 1999) have been reported only for limited operating conditions with little information provided regarding startup/shutdown or transient behavior. A high value of the thermal conductivity, the system was stabilized at the expense of substantial heat losses for the case of isothermal packaging (Fig. 13a). For the case of adiabatic packaging, the range of Da corresponding to multiple steady states is unaffected by CP (Fig. 13b).

The residence time (Da) necessary to achieve complete conversion under isothermal packaging was consistently higher than that required for adiabatic packaging under identical conditions due to reaction quenching via conductive heat losses. Steady-state multiplicity due to choice of packaging and reactor materials showed the importance of these two parameters in the design of a thermally integrated microsystem. Experimental studies related to the prediction of multiple steady states and ignition/extinction behavior in thermally integrated microreactors have not been reported in the literature. The microsystems
are usually constructed from materials with a high thermal conductivity or to insure substantial heat losses to the environment via fittings. Multiplicity behavior was similar for co-current and counter-current adiabatic cases while counter-current operation removed steady-state multiplicity in the isothermal case.

4. Influence of flow and temperature non-uniformity on microreactor performance

Delsman et al. (2005) proposed a method to predict the influence of flow maldistribution and manufacturing tolerances on the performance of a microreactor with a large number of parallel channels. In this method, a variable reactor parameter (e.g., diameter) is considered as a random parameter with a mean value and standard deviation \( \bar{e}_i \) and the microreactor is considered as a plug-flow reactor. Then, a number of relationships between the variable parameter and efficiency as compared to the ideal case are presented. The pressure drop as a function of the mean channel diameter is given by Eq. (18):

\[
\Delta p = \frac{128\mu Fv}{\pi n \bar{d}^4 (1 + 6\sigma_d^2)}
\]

where \( L \) is the channel length, \( \mu \) the fluid viscosity, \( \bar{d} \) the mean channel diameter, \( F \) the flow velocity, \( n \) the number of channels and \( \sigma_d = \sigma / \bar{d} \) the relative standard deviation of the channel diameter. This equation shows that a variation of the channel diameter results in a decrease in the overall pressure drop over the reactor, when the total flow rate is kept constant. A small deviation in channel diameter does not result in a large difference in the pressure drop: a standard deviation of 10% gives a pressure drop of 6% lower as compared with the ideal case.

The residence time of the fluid in a microchannel varies also between the channels. The residence time in the single channel is a function of both the fluid flow rate and the channel volume. It can be expressed as

\[
\tau = \frac{\tau_0 (1 + 6\sigma_d^2)}{1 + \sigma_d^2}
\]

(19)

Since the residence time varies between the channels, a tracer pulse at the inlet of the microreactor will be broadened as in the case for a tubular reactor with dispersion. In a first approximation, the relative standard deviation in the residence time is twice the relative standard deviation in the channel diameter:

\[
\sigma_r^2 = (2\sigma_d^2)
\]

(20)

The influence of different process and geometry parameters on conversion was estimated in case of an irreversible first-order catalytic reaction. The influence of temperature non-uniformity (when temperature varies between the channels) has the largest impact on conversion in a non-ideal reactor as compared to non-uniform flow distribution and non-equal catalyst amount in the channels.

Obtained correlations were used to estimate the influence of a variable channel diameter on the conversion in a microreactor for a heterogeneous first-order reaction. It was found that the conversion in 95% of the microchannels varies between 59% and 99% at \( \sigma_d = 0.1 \) and Damköhler number of 2. Fig. 14a shows conversion as a function of Damköhler number for an ideal microreactor and a microreactor with variations in the channel diameter \( \sigma_d = 0.1 \). It can be seen that although the conversion in individual channels can vary considerably, the effect on the overall reactor conversion is smaller. The lower conversion in channels with a larger flow rate is partly compensated by a higher conversion in channels with a lower flow rate. Due to the non-linear relation between the channel diameter and the flow rate, the effects do not cancel completely and a decrease in reactor conversion is observed.

The influence of temperature non-uniformity between different channels was also considered.

In this case the channel diameter and the amount of catalyst per channel were considered to be equal in all channels, but the temperature was not the same in different channels. The temperature influences both the molar gas flow rate (and therefore the pressure drop) in a channel and the reaction rate coefficient. It was found that the temperature has a much larger influence on the reactor conversion than the channel diameter. The influence of temperature deviations on the conversion was a function of the activation energy of the reaction \( (E_a) \), which was expressed via the parameter \( \gamma = E_a / RT \) assuming that the reaction rate coefficient is proportional to \( \exp(-\gamma (T/T_i)) \). Fig. 14b shows the influence of the parameter \( \gamma \) on the reactor conversion. The influence of the temperature increases with an increase in the parameter \( \gamma \), which means for reactions with a high activation energy and/or a low reaction temperature.

Fig. 14. (a) Influence of channel diameter variations on the reactor conversion; ideal reactor (thick line), reactor with \( \sigma_d = 0.1 \) (thin line) and (b) influence of the parameter \( \gamma \) on the conversion in microreactor with \( \sigma_d = 0.01 \); \( \gamma = 15, 30, \) and 60.

Adapted from Delsman et al. (2005).
The effect of transient behavior of a multi-pass micro-heat-exchanger with an unequal flow distribution between channels and axial dispersion was studied by Srilhari and Das (2008). The flow distribution was taken from the model of Bassiony and Martin (1984). They reported that the nature of the response in the transient regime was dependent on the flow maldistribution, flow arrangements, number of channels and Peclet number. The time required to reach the steady state larger for multi-pass arrangements when compared to single-pass heat-exchanger. These data are in a good agreement with predictions of the Delsman’s model (Delsman et al., 2005).

5. Improvement of microreactor performance by optimization of fluid flow and heat transfer

Makarshin et al. (2007) studied the effect of reactor design on the effective reaction rate constant of methanol steam reforming in microreactors with different geometry: rectangular, cylindrical and tubular. The highest reaction rate constant was observed in the rectangular microreactor due to the smallest temperature gradient over the reactor length. The authors concluded that limited thermal conductivity of the reactor material reduced the effective reaction rate by 1.3 and 1.65 times, respectively, in the cylindrical microreactor with 10 coated disk plates and in a tubular microreactor with a fixed bed catalyst.

Hardt and Baier (2007) proposed a mean-field model for calculation of temperature distribution in multichannel micro-reactors. Their model incorporated heat exchange between the fluid and the solid phases, which were considered as interpenetrating continua, via a heat-transfer coefficient. The volume fractions of each phase were determined from the geometric parameters of the microreactor. The authors introduced a thermal conductivity tensor, which accounted for geometry of the channel walls in the microreactor. The conductive heat transfer in the fluid phase was neglected, but convective heat transfer was taken into account. The results obtained by the mean-field model were in good agreement with those obtained by 3D numerical simulations. The authors reported that a maximum difference between two models did not exceed 3 K on a scale of 35 K.

In our laboratory (Rebrov et al., 2003a), the effect of distance between cooling and reaction channels in a cross-flow micro-reactor/heat-exchanger (MRHE) on the temperature distribution using isothermal boundary conditions has been studied by numerical simulations. Aluminum was chosen as the reactor material because of its high thermal conductivity and ease of micromachining. The heat transfer characteristics of four different cross-flow designs of a MRHE were studied using ammonia oxidation as a model reaction. The simulation was based on the 3D geometry of a single periodic microreactor/heat-exchanger unit with a distance between two sets of channels of 125, 270, 470, and 670 μm (Fig. 15).

Large heat fluxes to the environment in a microreactor mean that a non-uniform coolant distribution should be applied to withdraw more heat in the center of the MRHE and less near the outer surface of it. We showed that the axial temperature gradient in the reaction channels can be controlled by the temperature and the flow distribution of coolant in the cooling channels and the distance between the reaction and cooling channels (Fig. 16).

Optimization of the key parameters affecting the temperature distribution in the MRHE is a complex problem, as these parameters are intricately related and thus cannot be controlled independently. As a first step, we used an optimization procedure to determine the coolant flow distribution that results in the most uniform temperature profile at the position of the catalyst. Then an analysis was carried out to find an optimal geometry of the inlet/outlet reactor chambers to obtain the selected coolant distribution. In this way, the performance of several designs was assessed in terms of the differences in flow velocity between the different channels and compared to the optimal flow distribution. It was shown that a non-uniform coolant distribution improves the ammonia conversion at the optimal aluminum thickness. Our results showed that the catalytic oxidation reaction could be performed at near-isothermal conditions in the optimized reactor assembly. As a result, the selectivity towards the target product was considerably improved. Moreover, experiments were in a good agreement with the CFD simulations.

ProxHeatex is a microstructured reactor/heat-exchanger designed to perform the preferential CO oxidation of carbon monoxide in a hydrogen-rich reforming gas (Delsman et al., 2004b). It consists of two heat exchangers and a cooled reactor in series as shown in Fig. 17a.

Each part of the reactor was made of microstructured plates which were stacked together. Two flanges with eight screws at the top and the bottom of the device compress the gaskets between each plate which ensure the whole tightness. The reaction zone was made of 19 reactor plates and 10 cooling plates whereas each heat exchanger was made of 2 process plates and 4 cooling plates. Insulating plates were inserted between the reaction zone and the heat exchangers to thermally separate these parts. The performance of this reactor was deteriorated by heat transfer via the connecting screws. In an attempt to improve this design, a second generation ProxHeatex device was constructed using welding of separate parts (Delsman et al., 2004c). As material for the second reactor prototype aluminum was considered, which would result in a smaller temperature gradient over the reactor of 20 K instead of 40 K for stainless steel. However, aluminum was discarded as an option, due to practical problems concerning its welding. In the plate design, a 3D CFD model of the gas flow on a single microstructured plate was used to optimize the geometry of the flow distribution chambers to ensure equal flow rates in the microchannels. The complete microdevice was insulated by a 5 cm thick layer of glass wool material (Fig. 17b). This microreactor heat-exchanger was able to reduce the CO concentration of a realistic methanol reformate gas to 10 ppm in combination with a high heat recovery efficiency of 90%.
Ismagilov et al. (2008) studied complete oxidation of unsymmetrical dimethylhydrazine (UDMH) in a microreactor made of a highly conductive AlMgSiCu1 alloy (6082 series, Al51st) (Fig. 18). UDMH is a component of the high-energy propellant for liquid-fueled rockets used in Russia, China, and the U.S. The catalyst section was assembled of 63 microstructured plates with catalytic coating. In each plate of 416 mm thickness, 45 semicylindrical microchannels of 208 mm in radius with a distance in between of 150 mm were electrodischarge machined to provide enough cross-section for heat transfer from the reaction zone to the environment. UDMH vapors were supplied from a saturator with a helium flow and diluted with oxygen in the mixer to form reactant mixture. The reactor demonstrated stable operation in a mixture containing UDMH with concentration in the range 0.16–0.4 mmol/L. The reactor performance was considerably improved by 7% by positioning of a thick-walled screen for uniform reactant distribution.

Ponyavin et al. (2008) developed a three-dimensional conjugate heat transfer and fluid flow numerical model to assess the thermal performance of the high temperature micro-heat exchanger in which a hot helium flow from a nuclear reactor ($T = 975 \, ^\circ C$) was used to heat the reactant stream (H$_2$O, H$_2$SO$_4$, SO$_3$) to create required conditions for the sulfuric acid decomposition reaction ($T > 850 \, ^\circ C$). The authors used adiabatic boundary conditions for the front and back sides of the microreactor. It was found that the effectiveness of the heat exchanger can be increased by up to 7% relative to the baseline design developed by Rebrov et al. (2003a).

**Fig. 16.** Temperature distribution at the catalyst position (in plane A–A). The reagents enters from the bottom, the nitrogen gas enters from the left. Cases A, B, and C represent uniform, stepwise, and coolant non-uniform flow distribution, respectively. The distance between the reaction and cooling channels was 125, 270, 470, and 670 µm in designs A, B, C, and D, respectively. Adapted from Rebrov et al. (2003a).
Ceramatec Inc. (Salt Lake City, USA), by optimization of geometrical parameters of the channels.

Arzamendi et al. (2009) studied the effect of the flow arrangement (co-current, counter-current, cross-flow) in a microstructured reactor/heat-exchanger where methanol steam reforming was combined with methanol combustion. The authors modeled a single periodic unit consisted of a sheet of $20 \times 20 \text{ mm}^2$ with a depth of 1 mm. The geometry of the inlet and outlet manifolds was not included in the simulations. The position of the hot-spot, generated in the metal plate, depended on the mode of operation. The highest hot-spot of 19 K was observed in the counter-current operation near the inlet of the combustion channels. However, the authors did not present experimental results confirming their CFD calculations.

In case of a conventional reactor, it is not common to change the tube diameter along the axial tube position. However microchannels with varying width or diameter can be easily fabricated (Noda et al., 2004). The optimal microchannel has expanding channel width from the inlet to the outlet of the microchannel (Fig. 19).

The expanding channel width is very effective to keep the temperature distribution uniform along the microchannel since a
distribution equipment is still in an early stage of development. In the case of multiphase flow design of a microreactor/heat-exchanger to reduce efficiently non-uniform coolant flow distribution can be created in a cross-temperature profile, which in turn can influence the fluid flow and the heat flux is directed from the fluid to the wall. The choice of flow equalization is usually adequately tackled by using 3D numerical models resembling the geometry of the inlet and outlet diffusers and reaction channels. 2D CFD models could not adequately predict flow distribution in inlet/outlet chambers and flow distribution manifolds. An attempt is made to develop more simple models based on effective hydraulic resistance of the diffusers and microchannels. These models can predict pressure drop and flow distribution at low Reynolds numbers. However, singular pressure losses should be taken into account for accurate modeling at Re number > 300.

There is often a substantial heat flux via fittings in micro-reactors. Its value depends on the thermal conductivity of the reactor material, material cross-sectional area, and boundary conditions at the wall. It should be mentioned that the axial conductive heat flux is often overlooked parameter which results in a lower mean value of the heat transfer coefficient in microchannels as reported by several authors. This is due to the fact that the difference between the wall temperature and the fluid temperature might become negative along the reactor length and the heat flux is directed from the fluid to the wall. The choice of basic reactor material plays a crucial role in managing the temperature profile, which in turn can influence the fluid flow distribution. The thickness of the channel wall should be adjusted to reach the desired value of the thermal resistance. In addition, a non-uniform coolant flow distribution can be created in a cross-flow design of a microreactor/heat-exchanger to reduce efficiently temperature non-uniformity.

At the moment, the problem of (conjugated) fluid flow distribution and heat transfer in a single-phase flow in a microreactor can be solved with a high accuracy by using various commercially available numerical codes. In the case of multiphase flows, such as those involving gas–liquid, the design of flow distribution equipment is still in an early stage of development. Most of the new developments are mainly occurring as a result of experimental studies. Much opportunity exists to demonstrate the scale-out of catalytic microstructured reactors as a robust enabling technology for the development of fine chemicals synthesis and other sectors of chemical industry.

6. Conclusions and outlook

The examples considered in this review demonstrate that there are several approaches used by different research groups to achieve flow equalization in microstructured reactors. The issue of flow equalization is usually adequately tackled by using 3D numerical models resembling the geometry of the inlet and outlet diffusers and reaction channels. 2D CFD models could not adequately predict flow distribution in inlet/outlet chambers and flow distribution manifolds. An attempt is made to develop more simple models based on effective hydraulic resistance of the diffusers and microchannels. These models can predict pressure drop and flow distribution at low Reynolds numbers. However, singular pressure losses should be taken into account for accurate modeling at Re number > 300.

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References


