Systematic Approach for Thermal-Fluid Design of Microreactors

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ABSTRACT
Recent advances in computational fluid dynamics (CFD) enable us to know flow and temperature distribution in a microreactor precisely without conducting any experiments. However, it is not a practical way to apply CFD simulations directly to the optimal design problem of a microreactor, since it requires too much time and efforts. In this research, the systematic approach to the thermal optimal design problem of a microreactor is proposed. In the proposed approach, a heat transfer model in a microreactor is expressed by combining a large number of small lamped parameter systems called thermal compartments. The proposed approach is applied to a simple design problem and results show that a microchannel with varying width is very effective to realize the uniform temperature distribution. The new design approach is also proposed to derive the robust design of a microreactor under the model parameter uncertainties. The optimal design obtained by the proposed approach can be used as the initial design of a microreactor for further shape optimization based on CFD simulations.

KEYWORDS
microreactor, optimal design, thermal compartment model, model uncertainty

INTRODUCTION
The industrial scale production using a microreactor requires a large number of parallel microchannels, since each microchannel provides only a small amount of products. However, the inadequate scale-up of microreactors causes poor uniformity in the residence time and temperature distribution among microchannels, which may make product quality worse.

At present, most microdevices are designed and fabricated by trial and error. Therefore, it takes much time and effort to reach the optimal design. To achieve further breakthrough in micro chemical plants’ technologies, a systematic design approach for microreactors needs to be established.

Recent advances in CFD-based design approach enable us precise estimation of flow and temperature patterns in a microreactor without conducting experiments. Tonomura et al. (2003) proposed the CFD-based optimization method for design of plate-fin type microdevices. However, it seems to be an impractical way to apply directory the CFD-based design approach to the optimal design of a microreactor, since it requires high skill in mesh generation and long computational time.

Commenge et al. (2002) examined the specific features of fluid flow through multi-plate microchannel reactors by an approximate pressure drop model. The approximate pressure drop model, which is based on pressure drop calculations through a resistive network of ducts, provides rapid calculation of the fluid distribution. However, temperature distribution in the microreactor, which may affect product quality drastically, is not evaluated in the model.

In this study, a new thermal compartment model is developed to provide rapid calculation of the
temperature distribution in a microreactor. Then, the systematic approach for thermal-fluid design of microreactors based on the pressure drop and thermal compartment models is proposed. The proposed approach is applied to the optimal design problem of a microreactor with uniform temperature distribution. Then, the effect of the model uncertainty on the microreactor design is discussed. Finally, the modified design approach is proposed to realize the robust design under the model uncertainties.

OPTIMAL DESIGN PROBLEM OF MICROREACTORS

In this paper, a microreactor is divided into three parts, namely, inlet manifold for flow distribution, parallelized microchannel part for reaction and outlet manifold for mixing, as shown in Fig. 1. If reactions do not occur in the inlet and outlet manifolds, fluid design and thermal design of microreactors can be separated into independent optimization problems. Table 1 summarizes the optimal fluid and thermal designs of microreactors. In the fluid design, manifold shape and number of microchannels are determined to minimize the residence time of a microreactor under constraints related to the maximum pressure drop and residence time distribution. In the thermal design, the shape of microchannel and coolant temperature is determined to minimize the temperature distribution in the microreactor under the constraints related to yield or selectivity.

In this paper, we focus only on the thermal design problem of microreactors. The perfect flow distribution among microchannels is assumed in the thermal design problem. Therefore, cyclic boundary condition can be applied to each microchannel. In the next section, thermal compartment model is proposed to provide rapid calculation of the thermal distribution in a microreactor.

Table 1 Optimal design problem of microreactors

<table>
<thead>
<tr>
<th></th>
<th>Fluid design</th>
<th>Thermal design</th>
</tr>
</thead>
<tbody>
<tr>
<td>Objective function</td>
<td>Minimum residence time</td>
<td>Minimum temperature distribution</td>
</tr>
</tbody>
</table>
| Optimization variables | Manifold shape  
Number of microchannels | Microchannel shape  
Flow rate  
Coolant temperature |
| Constraints          | Maximum pressure drop        | Maximum pressure drop  
Yield  
Selectivity |

THERMAL COMPARTMENT MODEL

The rigorous fluid dynamics of a microreactor is expressed by Navier-Stokes equations. However, it is difficult to solve Navier-Stokes equations under the complex boundary conditions. In order to approximate the pressure distribution in the microreactor, the pressure drop compartment is introduced by Commenge et al. (2002). The pressure drop compartment has three dimensions (Length, Depth, and Width) and the pressure drop in the compartment is calculated by solving an equation for laminar flow through a rectangular channel. Then the total pressure balance of microreactor is described by combining pressure compartments and solved under the given boundary conditions.

The basic concept of thermal compartments for energy balance is the same as that of the pressure drop compartments. The thermal compartment has three dimensions (Length, Depth, and Width) and heat transfer coefficients between connected thermal compartments. The mathematical model of the thermal compartment is defined by Eq.(1)

\[
C_p(F_i^{in}T_i^{in} - F_i^{out}T_i^{out}) - \sum Q_{ij} + \Delta Hr_j = 0
\]
where $F_i$, $T_i$, $C_p$ and $\Delta H_{ri}$ are flow rate, temperature, specific heat and reaction heat generated in the compartment $i$. $Q_{ij}$ is heat transfer rate from the thermal compartment $i$ to the surrounding thermal compartment $j$. The total heat balance of the microreactor is solved under given boundary conditions.

The biggest difference between the pressure drop compartment and the thermal compartment is the heat transfer through the wall needs to be considered in the thermal compartment model. Figure 2 shows the thermal compartment model for one microchannel with two side walls. Cyclic boundary condition between connected microchannels is assumed under the perfect uniform flow distribution condition.

![Thermal compartment model](image)

Fig. 2 Thermal compartment (left) and thermal compartment model (right)

Figure 3 shows the flowchart of the optimal design approach of a microreactor. The flowchart mainly consists of two parts, i.e. the thermal design (Left) and fluid design (Right). In the thermal design part, thermal compartment model is generated according to the design specification. The derived optimal design using the thermal compartment model is evaluated through CFD simulation. If the derived design does not satisfy the given design specification, model parameters in the thermal compartment model are updated and thermal compartment model is regenerated. In the next section, the proposed optimal design approach is applied to the design problem of a microreactor with uniform temperature distribution.

![Flowchart](image)

Fig. 3 Flowchart of optimal design approach

**MICROREACTOR DESIGN WITH UNIFORM TEMPERATURE DISTRIBUTION**

Keeping temperature in a microreactor uniform at specified reaction temperature is one of the most important design specifications, since the temperature distribution in a microreactor may affect the product quality drastically. Therefore, in this section, the proposed optimal design approach is applied to a microreactor design problem with uniform temperature distribution.
Case Study

A very simple exothermic liquid phase reaction \( A \rightarrow B \) shown by Eq. (2) is taken up in this case study. As this reaction has very huge reaction heat, keeping temperature in a microreactor uniform is the most important design issue.

\[
A \rightarrow B + 10^6 \text{J/mol} \quad (2)
\]

Figure 4 illustrates a schematic diagram of a channel part taken up in the case study. In this case study, it is assumed that the perfect flow distribution among microchannels can be achieved as a result of the optimal fluid design, which means that all microchannels have same temperature profiles axially. Therefore, cyclic boundary conditions among microchannels can be introduced into the optimal thermal design problem of the microreactor. These conditions can reduce the design region from the whole channel part to the one microchannel with a surrounded wall which is emphasized with a solid line in Fig. 4. In the case study, the basic design region is defined as a rectangular glass whose dimension is 1000 µm in thickness and 1000 µm in width. In order to express the temperature distribution, the basic design region is divided into the fifty inner thermal compartments representing flow in the microchannel and fifty outer thermal compartments representing the wall.

The optimal design problem is defined as follows:

**Objective function:** Minimizing the integrated temperature error along a microchannel

**Optimization variables:** Length, depth of a microchannel as constant values
Width of a microchannel along a microchannel
Feed flow rate

**Constraints:** Minimum yield

In the optimal design problem, the width of the microchannel is assumed to be a piecewise linear continuous function which has eleven sections. The integrated temperature error \( I.E. \) is calculated by Eq. (3) where \( x \) means the axial position in a microchannel and \( T_{set} \) is a specified reaction temperature. In case of a conventional macro reactor, it is not common to optimize tube diameter as a function of the tube position. However, microchannel with varying width is not so difficult to fabricate.

\[
I.E. = \int (T(x) - T_{set}) \, dx
\]  

The other simulation conditions are summarized in Table 2.
Table 2 Simulation conditions for case study

<table>
<thead>
<tr>
<th>Feed composition of A</th>
<th>100 %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feed temperature</td>
<td>600 K</td>
</tr>
<tr>
<td>Reaction temperature</td>
<td>700 K</td>
</tr>
<tr>
<td>Coolant temperature</td>
<td>600 K</td>
</tr>
<tr>
<td>Heat transfer coefficient</td>
<td>10000 J/(m²K)</td>
</tr>
<tr>
<td>Physical properties of A&amp;B</td>
<td>Water</td>
</tr>
<tr>
<td>Reaction rate of A</td>
<td>- 0.01Cₐ mol/(m³s)</td>
</tr>
<tr>
<td>Minimum yield</td>
<td>70 %</td>
</tr>
</tbody>
</table>

**Result of Optimal Design**

Figure 5 illustrates the result of the case study explained in the previous section. The solid line in the upper graph shows the optimal channel width along the microchannel and dotted line shows the channel width which is optimized as constant value. The optimal microchannel has expanding channel width from the inlet to the outlet of the microchannel. The expanding channel width is very effective to keep temperature distribution uniform along the microchannel since narrow channel near inlet shortens residence time for faster reaction rate zone and wide channel near outlet makes residence time longer for slower reaction rate zone. The lower graph of Fig. 5 illustrates the temperature profile of the microchannel. As shown in this graph, a hot spot occurs near the inlet of the microreactor when the straight channel is used even if it is optimized. On the other hand, the temperature is almost kept constant along the microchannel when the proposed design approach is applied. In case of the optimal design, the maximum temperature error, which is defined as temperature difference between the highest temperature in the microchannel and the reaction temperature, is about 7.6 K.

![Optimal Channel Width and Temperature Profiles](image)

In order to evaluate the effectiveness of the proposed design approach, the conventional design approach is also applied to the case study. In this paper, the conventional design approach of a microreactor is defined as follows: The feed and coolant temperatures are kept at 700 K which is equal to 550 K.
the reaction temperature and the square cross section of a microchannel is assumed. Figure 6 shows the maximum temperature error for various channel widths. As shown in Fig. 6, the width of the microchannel must be less than 50 µm to make temperature error lower than that of the optimal design. In this case, the throughput of each microchannel is only 2% of the optimal design. It can be concluded that the conventional design approach may cause unnecessary miniaturization.

![Fig. 6 Maximum temperature difference of conventional design](image)

**OPTIMLA DESIGN UNDER MODEL PARAMETER UNCERTAINTY**

The biggest problem of the proposed design approach is model parameter uncertainties. For example, if the heat transfer coefficient between fluid and wall is uncertain, the CFD simulations needs to be repeated in order to estimate the heat transfer coefficient. These simulations require long computational time. Therefore, in this section, the optimal design approach proposed in the previous section is modified in order to design the robust microreactor under the model parameter uncertainties.

The modified design approach is as follows: First, the reaction heat generated per unit heat transfer area of each compartment is equalized by changing channel width. In this case, heat transfer coefficient is not necessary for design. Second, the coolant temperature is optimized as manipulated variable to minimize temperature error using CFD simulation. The inlet temperature of the fluid is set to the coolant temperature. This approach does not require mesh regeneration in CFD simulation, since coolant temperature is a boundary condition. The modified design approach requires less information and efforts.

In order to evaluate the effectiveness of the modified design approach, it is applied to the same case study. Figure 7 illustrates temperature contours for both of the conventional and optimal designs. As shown in the conventional design, there is a hot spot near inlet of the microchannel. On the other hand, in the optimal design, temperature distribution is more uniform than that of the conventional design. The lower graph of Fig. 7 shows the result of the CFD simulation for both designs. As seen in this graph, the cross sectional average temperature is kept almost constant by optimizing the coolant temperature, which is 660 K in this case. It can be concluded that reaction temperature is controlled by changing coolant temperature without disturbing uniformity of the temperature distribution when the modified design approach is applied to the optimal thermal design problem of a microreactor.
In the remaining part of this section, the effectiveness of the proposed design approach to the uncertainty in the reaction rate constant is discussed. At first, the microreactor are designed using conventional and proposed approaches for rate constant $k = 0.1$. Then, the temperature distribution of the proposed microreactors is simulated for various rate constant $k$. In Fig. 8, the standard deviation of the temperature error is plotted for various $k$. As seen in the graph, the microreactor designed by proposed approach is more robust than the conventional design, when uncertainty exists in the reaction rate constant. This is because, generated heat distribution along the channel is still almost uniform even if rate constant $k$ changes. Therefore, temperature distribution along the channel can be controlled only by changing coolant temperature and feed flow rate. It can be concluded that proposed approach is also effective for keeping temperature distribution uniform under reaction rate model uncertainty.
CONCLUSION
The optimal design approach based on thermal compartment model is developed. The thermal compartment model based on heat transfer between lumped parameter systems provides rapid calculation of the temperature distribution of a microreactor. The proposed model is applied to the optimal design problem of a microreactor with uniform temperature distribution. The obtained results show that a microchannel with varying width is very effective to keep the temperature distribution in the microreactor uniform when highly exothermic reactions occur in microchannels. The CFD simulations confirm the validity of the optimal design.

The thermal compartment model also offers a qualitative understanding of the influence of uncertainties of the model parameters such as heat transfer coefficient and reaction model parameters on the temperature distribution in the microreactor. The proposed design approach is also very effective to the optimal design under the model parameter uncertainties.

The thermal compartment model is very simple but powerful tool to shorten the computational time of microreactor design. It can be concluded that the proposed design approach has potential for being widely applied to the optimal design problem of microreactors with various constraints.

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REFERENCES