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Flow Uniformity in Deep Microchannel Reactor under High Throughput Conditions

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Abstract:

Microreactors have attracted the attention of many engineers in chemical industry, since they can be used to improve yields of various chemical reactions. To use microreactors in an industrial process, it is necessary to increase the throughput. As a strategy to increase the productivity of microreactors, we are studying deep microchannel reactors, which are simple in structure, and thus can be introduced at a lower capital cost. One of the concerns in the application of deep microchannel reactors is the flow maldistribution, which reduces the contact area between two fluids and thus deteriorates the performance as a reactor. It is anticipated that the major cause of low maldistribution is the nonideality in the flow conditions, which can be magnified in a microchannel with a high aspect ratio. In the present study, the impact of nonideality in the channel geometry on the reaction interface was studied for a microchannel with a depth of 400 mm, which is expected to be the typical dimension for industrial-scale productions. A series of computational fluid dynamics simulation was carried out for various flow rates and fluid properties. The results showed that, in a deep microchannel reactor, the effect of nonideality in the channel geometry on the reaction interface is rather small. This result suggests that a uniform contacting of two reactant feeds can be realizable even in deep microchannel reactors.

Introduction

Mixing and heat transfer characteristics are very important parameters of chemical reactors since they strongly influence the yield or selectivity of the product. For a conventional large reaction unit, these characteristics can be controlled to acceptable conditions, only when the mixing and cooling strategies are carefully designed.

Recently, flow reactors with characteristic dimensions of less than 1 mm are attracting the attention of researchers in both academia and industry. Such minute flow channels are called microchannels, and a flow reactor having microchannels is called a microreactor. Flow in microchannels is laminar, and thus computational fluid dynamics simulation can be used to study the mixing and heat transfer behaviors. In microreactors, changes in fluid temperature can be made very fast, and fast mixing can be achievable if the microchannels are properly designed. These features of microreactors lead to improved selectivity or yield of chemical reactions, as compared with conventional macro-sized chemical reactors. In microreactors, for example, the hot-spot temperature can be easily suppressed, and thus formation of undesired side products arising from variation in reaction temperature can be minimized. In fact, there are many reactions that are reported to show improved selectivity or yield in microreactors.[1,2] Moreover, if a batch process is converted to a microreaction process, both plant operation and control of product quality can be easier. Accordingly, microreaction technology is attractive for chemical industries, since it can be used to develop a chemical process with very high efficiency.

It should be noted that because microreactors consists of reaction channels which are smaller than 1 mm in characteristic dimension, the throughput of a single microreactor is typically a few tons per year at maximum. Thus, an industrial process cannot be constructed by simply using one ordinary microreactor. For producing 1000 tons of fine chemical per year, for example, it is necessary to increase the throughput by a factor of 1000 or more. An option for increasing the throughput of a microreactor is to raise the pumping pressure. However, it is not realistic to operate a single reactor by just increasing the pressure by a factor of 1000. It is often stated that parallelization is most suited for constructing a chemical process with a high processing rate. Such an approach is often referred to as "numbering-up" and is considered to be an advantage of microreactors. This is because by parallelizing microreactors, an industrial-scale production can be achieved without scale-up processes, in which a great deal of human resources and time is spent for optimizing the reactor design.

However, the numbering-up approach requires a technique to equally divide a feed stream into more than hundreds of small streams with equal flow rates. It is not realistic to use a control valve for each branched stream. Several researchers have proposed strategies to design flow distributers or manifolds,[3,4] but the typical number of reaction channels they are handling

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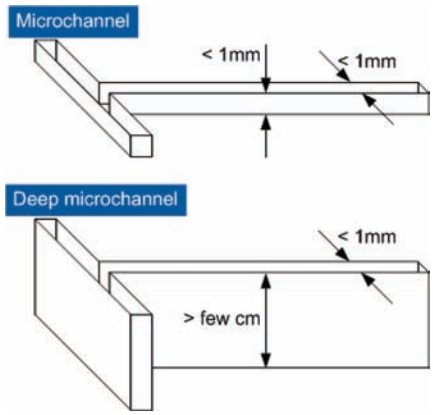

Figure 1. **Deep microchannel reactor (FMR).**

was only in the order of 10. Thus, parallelization of more than 100 microreactors is considered not to be practical.

Recently, several different types of high-throughput microreactors have been developed, and some of them are commercially available. Werner and co-workers have developed a StarLam series of microreactors,[5] while another high-throughput type microreactor called LH-1000 has been developed by Ehrfeld Mikrotechnik BTS GmbH.[6]

We are currently conducting a study to use deep microchannel reactors for realizing high-throughput micro chemical plants.[7] In what follows, deep microchannel reactors are referred to as FMR, which stands for Fukamizo (deep channel in Japanese) Micro Reactor. FMR is very simple in concept, as shown in Figure 1. The microchannels used in ordinary microreactors are less than 1 mm in both depth and width. After being brought into contact, the reactants flow side by side through the main channel, and mix by molecular diffusion. The mass transfer takes place only in the transverse direction. When the *Z*-dimension, or depth, of such a microchannel is extended, the mass flux remains unchanged and the throughput increases. Since FMR is simple in structure, it can be fabricated easily. The capital cost of a micro chemical plant with FMR would be smaller than that with conventional microreactors. Also the pressure drop can be changed by extending the depth. The pressure drop tends to be high when the flow rate through the microreactor is large. Extension of depth results in lower linear velocity and thus a smaller pressure drop. Thus, FMR is considered to be a promising technique for constructing a micro chemical plant.

One of the technical issues of FMR is flow maldistribution in the deep microchannel. Since the aspect ratio of deep microchannel is very large, the reactant distribution in the deep microchannel may appear to be nonuniform. The nonuniformity in the distribution gives rise to a variation in mixing ratio, which decreases the average reaction yield. The maldistribution is caused by nonideality in channel geometry and flow conditions. In our previous study, a preliminary study was conducted to examine the effect of channel geometry, or fabrication error, on the flow distribution.[7] An FMR with a channel depth of 2 mm was taken as an example in the simulation study, and a

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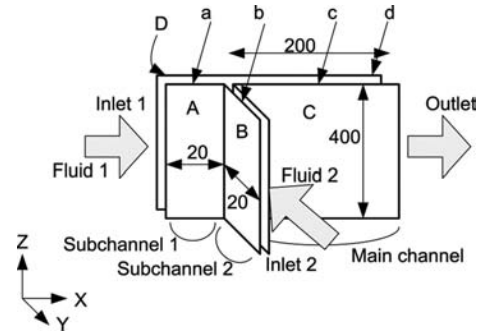

Figure 2. **Geometry of the deep microchannel for simulation: dimensions in mm. Nominal width was 0.1 mm.**

channel geometry in which fabrication error gives the least effect on the flow distribution was studied. Although the experimental study was conducted using a microreactor with a depth of 26 mm, the numerical investigation to explore the effect of various fabrication errors on the flow was carried out only for FMR with a depth of 2 mm. Obviously, this is far too small for use in industrial production. When the target throughput of a micro chemical plant is 1000 times greater than a microreactor with a depth of 100 μm, the depth of the deep micro channel should be greater than 100 mm.

At present, a research project is being carried out to examine the validity of FMR in an industrial-scale production. A micro chemical plant was developed in this study. A pump with capacity of 60 L/h and three with 48 L/h are installed so that four different types of fluid can be handled. The total pumping capacity amounts to over 1000 t/y. The plant was designed so that FMR can be replaced and performance of various FMR can be examined. The largest FMR fabricated in this study was 0.1 mm in width and 400 mm in depth, and the aspect ratio was 4000.

In this study, therefore, the effect of nonideality in the flow condition on the distribution of the reaction interface was investigated for FMR with an aspect ratio of 4000 by using computational fluid dynamics (CFD) simulation. Such a simulation was not possible with ordinary personal computers since the numerical grid is very large. Thus, a high-performance computer (Intel Xeon 3 GHz, 32 GB) was introduced into this study to facilitate the computation. In the previous study, the effect of a tilted wall on the flow distribution was investigated. In reality, however, there are other possible causes that affect the flow distribution such as flow rate and fluid properties. The influences of these factors were also studied using CFD simulation.

Simulation Conditions. Figure 2 shows the geometry of the deep microchannel assumed in the present study. Unless stated otherwise, the depth and the width were 400 mm and 100 μm, respectively. The length of the subchannel connecting the each reactant inlet and the intersection was 20 mm, while the main channel was 200 mm in length. The mesh size at the intersection was 0.1 of the channel width, and typically 10 μm. To reduce the total mesh size, coarser mesh was used near the inlets and outlet, and the mesh size in the depth direction was 1 mm. This is considered reasonable because the velocity and concentration gradient in the depth direction is very small. The number of nodes of a typical numerical grid was 1.4 million.

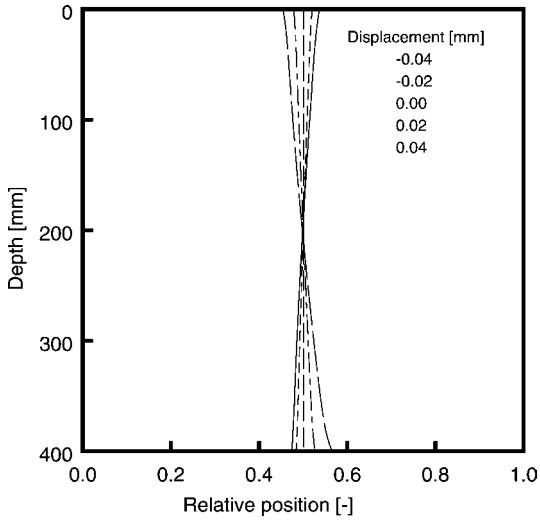

Figure 3. **Effect of displacement in edge a in the *Y* direction on the relative position of the reaction interface.**

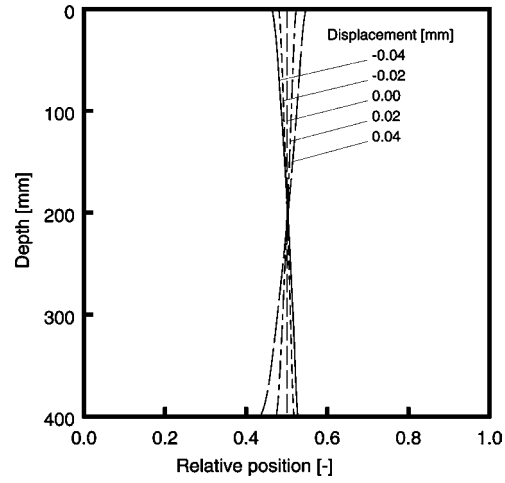

Figure 4. **Effect of displacement in edge b in the *X* direction on the relative position of the reaction interface.**

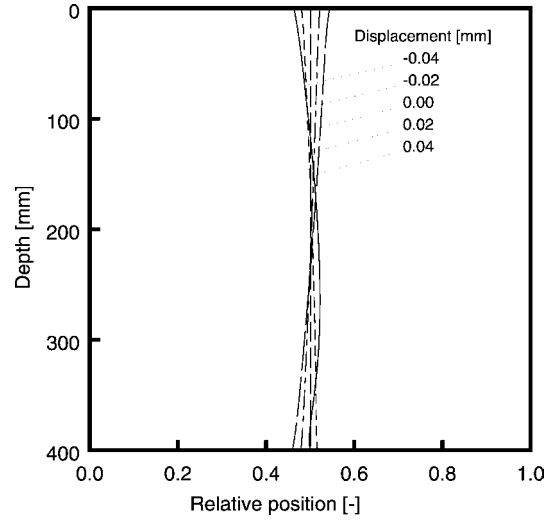

Figure 5. **Effect of displacement in edge d in the *Y* direction on the relative position of the reaction interface.**

The simulation was carried out using commercial CFD software, FLUENT, version 6.3.

When generating a numerical grid, one of the edges a−d was assumed to be displaced as a result of fabrication errors. The variation in the channel dimension results in the tilted wall. The tilt of wall A in Figure 2, for example, arises due to a displacement of edge a towards the *Y* direction. The maximum magnitude of displacement was assumed to be 40 μm. Since the nominal channel width was 100 μm, the real displacement is expected to be smaller than this.

The density and viscosity of each fluid were assumed to be respectively 1000 kg/m^3 and 0.001 Pa·s. The mean velocity of the inlet was 0.050 m/s. In the present case, the mean velocity of 0.050 m is equivalent to 250 mL/min, which is rather small for industrial application. The reason for choosing small values for the mean velocity was that the effect of the tilt was greater at a lower flow rate, as explained later. In the simulation using FLUENT, each inlet was treated as a "mass-flow-inlet" type, and the mass flow rate equivalent to the mean velocity of 0.050 m/s was specified.

Results and Discussion

Figure 3 shows the effect of tilts of wall A on the reaction interface at the outlet. The vertical axis represents the depth from the top surface. The horizontal axis shows the relative position of the reaction interface, which was calculated by dividing the distance between the interface and the channel wall by the channel width. Since the property and flow rate of the fluids are the same, the interface should appear halfway between the channel walls, and the relative position should be 0.5 throughout the depth. That is, when the fluids contact ideally, the interface should appear as a vertical line. The figure shows that the reaction interface was distorted due to the effect of tilted walls. The maximum variation of the position from 0.5 in Figure 3 was 0.06 in relative position, which appeared at the bottom when the displacement was 40 μm. When fabricating deep microchannel reactors with a width of 100 μm, fabrication errors in edge position are expected to be smaller than 40 μm. This means that under realistic fabrication errors, the change in relative position of a reaction interface is expected to be less than 10%.

The effect of other channel walls on the reaction interface was studied by comparing Figures 3, 4, and 5. The change in each interface position was comparable to each of the others. It is remarkable that the tilt of wall C does not affect the interface position at all. Although wall D has the largest surface area among all walls, the change in interface position is similar to the others.

The tilt of walls gives rise to a change in local flow resistance, which leads to variation in the position of reaction interface. It is difficult to develop a first-principles model describing the relationship between the change in the flow resistance and the distribution of the reaction interface. In the present study, therefore, a simple model was developed to discuss the qualitative behavior of the interface. The channel geometry assumed in building the model is shown in Figure 6. The channels consisted of parallel walls. The following equation describes the relationship between the pressure drop, ΔP, and the flow rate per unit height, q, of a flow between parallel walls.

$$\Delta P = RqR = \frac{12\mu L}{W^3} \quad (1)$$

μ [Pa·s], L [m] and W [m] are the viscosity of fluid, the length, and the width of the channel, respectively. R [kg/m·s] can be interpreted as a flow resistance of the channel. This equation is applicable to a flow with a parabolic velocity distribution. In the present study, fluids with different properties exist in the main channel, and thus the velocity distribution is not always parabolic. However, there are many cases where eq 1 can be applied. For example, when each reactant is diluted in water, the fluid properties are nearly equal to each other, and the velocity distribution is almost parabolic. In this study, eq 1 was assumed to be valid and applied to each branch of the channel shown in Figure 6.

$$\Delta P_1 = R_1 q_1 \quad (2)$$

$$\Delta P_2 = R_2 q_2 \quad (3)$$

$$\Delta P_3 = R_3 q_3 \quad (4)$$

The subscripts 1−3 denote the subchannel for fluids 1 and 2, and the main channel, respectively. The following equation shows that the flow rate in the main channel is the sum of flow rates of subchannels 1 and 2.

$$q_3 = q_1 + q_2 \quad (5)$$

From eqs 2, 3, 4, and 5, the fraction of fluid 1 in the main channel can be written as follows.

$$\frac{q_1}{q_3} = \frac{r_2 - p + 1}{r_1 p + r_2} \quad (6)$$

where r_1, r_2, p are defined as follows.

$$r_1 = \frac{R_1}{R_3}, \quad r_2 = \frac{R_2}{R_3}, \quad p = \frac{\Delta P_2 + \Delta P_3}{\Delta P_1 + \Delta P_3} \quad (7)$$

The parameter p represents the ratio of the pressure difference between inlet 2 and the outlet, to that between inlet 1 and the outlet. In cases where the driving pressures are equal to each other, the fraction of fluid 1 in the main channel q_1/q_3 can be reduced to the following equation.

$$\frac{q_1}{q_3} = \frac{r_2}{r_1 + r_2} = \frac{R_2}{R_1 + R_2} \quad (8)$$

These equations cannot describe the detailed flow behavior in a deep microchannel, but it helps us understand the qualitative effect of tilts on the flow. The tilt of wall A gives rise to a change in flow resistance R_1, and the fraction of fluid 1, q_1/q_3, changes. Similar change happens when wall B is tilted, as shown in Figure 4. Our numerical results also showed that the tilt of wall C does not affect the distribution of the reaction interface. This behavior is described by eq 8, which indicates that the fraction of fluid 1 q_1/q_3 does not depend on the resistance in the main channel R_3.

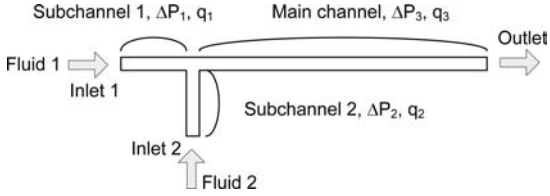

Figure 6. **Channel geometry assumed in the model development.**

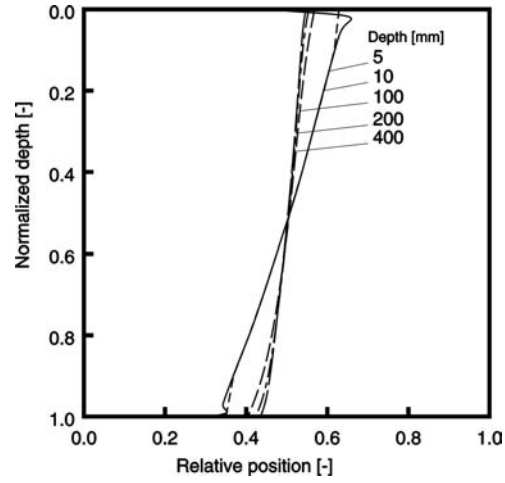

Figure 7. **Dependence of the relative position of the interface on the channel depth.**

Wall D has the largest surface area than the other walls, and thus its tilt is expected to give a large change in flow distribution. The tilt gives rise to a change in both R_1 and R_3, but only R_1 affects the flow distribution. This accounts for the fact that the variations in flow distribution resulting from tilts of wall A, B and D are similar to each other.

In the following, it was assumed that edge b was shifted by 40 μm in the X direction, and the influences of channel geometry, flow conditions, and fluid properties on the distribution of the reaction interface were studied. The effect of channel depth on the interface is depicted in Figure 7. The vertical axis represents the distance from the top surface normalized with the channel depth. The variation in the interface appears to be smaller in a deep channel. The effect of channel width is summarized in Figure 8. Displacement of 40 μm in edge b gives a smaller effect on the flow resistance with extending the width, since the relative dimension error in the width decreases.

The effect of the main channel length on the reaction interface is illustrated in Figure 9. The position of the reaction interface is not affected by the length. This is because the resistance in the main channel R_3 does not influence the flow distribution, as indicated by eq 8.

The flow rate of each fluid is an important operation variable that determines the fluid behavior in a microchannel. The ratio of the flow rates varies the location of the reaction interface. As a first step to study the effect of flow rate variation on the interface, the flow rates of fluids were assumed to be equal to each other. A series of simulations was carried out by varying the mean velocity, and the results are shown in Figure 10. It was observed that with decreasing the flow rate, the position of the reaction interface showed a greater variation, and thus the mixing ratio of two fluids was less uniform. Thus, operation with a higher flow rate is preferred for obtaining uniform flow distribution.

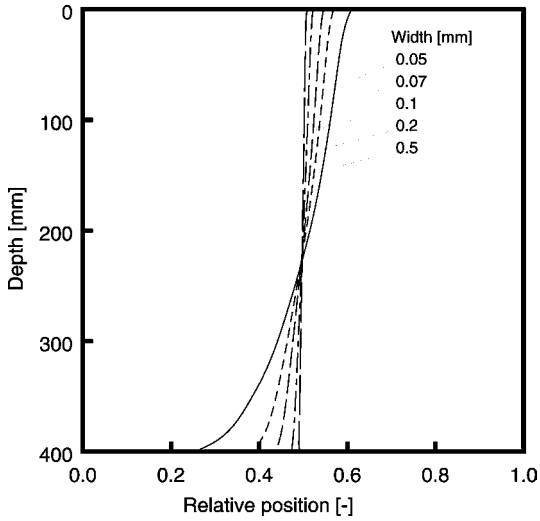

Figure 8. **Effect of channel width on the reaction interface: channel depth = 400 mm.**

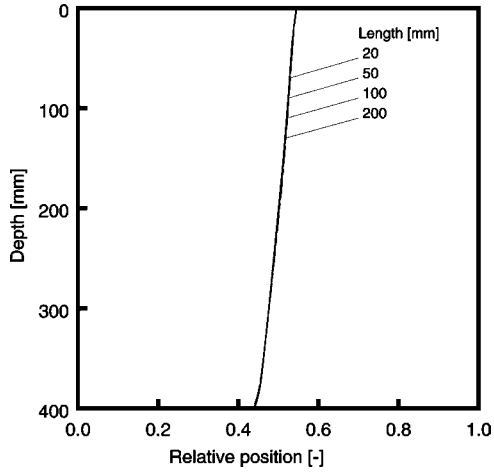

Figure 9. **Effect of main channel length on the reaction interface.**

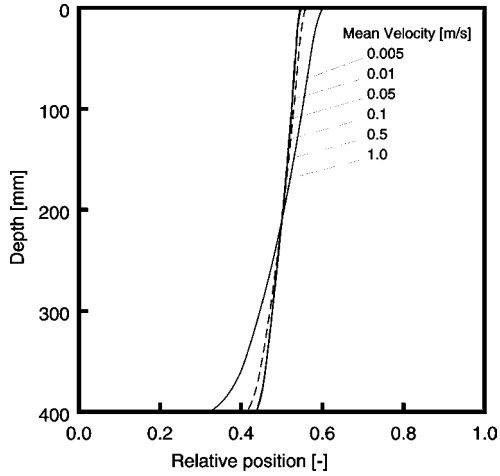

Figure 10. **Effect of mean velocity on the reaction interface.**

The mean velocity of each fluid was changed in the range of 0.01−0.5 m/s, and the effect of flow ratio on the distribution of the reaction interface was studied. As shown in Figure 11, the position of the reaction interface moved, depending on the ratio of flow rates. However, the gradient of the reaction

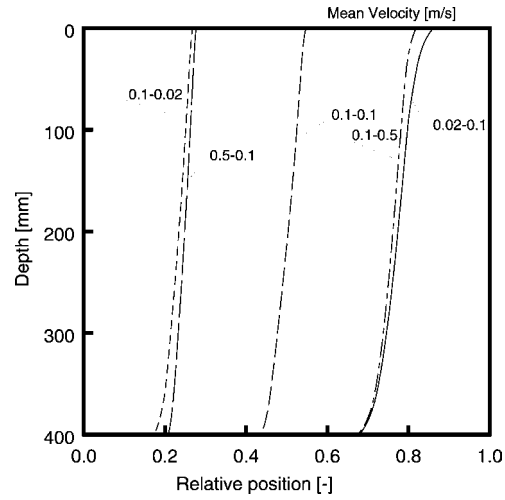

Figure 11. **Ratio of flow rates and the reaction interface: numerals show mean velocities of fluid 1 and fluid 2 in m/s.**

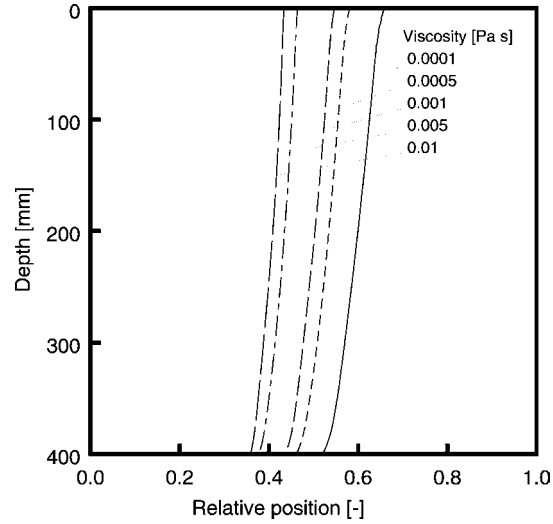

Figure 12. **Viscosity of fluid 2 and the reaction interface.**

interface was almost unchanged. This indicates that the ratio of the flow rates does not affect the position of reaction interface.

Generally, properties of fluids strongly influence the flow field. When the viscosities of fluids are different from each other, the location of the reaction interface varies, and the velocity distribution in the main channel is no longer parabolic. Thus, eq 8 does not hold for such a case. The fluid with higher viscosity tends to expand the thickness of the fluid layer in the main channel. Figure 12 shows the distribution of the interface position for various viscosities of fluid 2. The viscosity of fluid 1 was assumed to be 0.001 Pa·s throughout the simulation. With increasing the viscosity, the influence of the tilt affects the fluid distribution more strongly.

It is difficult to build a simple mathematical model describing the variation in the profile of the reaction interface when the fluid properties are different. It would be necessary to carry out a CFD simulation to gain full understanding of the relationship between fabrication errors and fluid distribution.

Concluding Remarks

Deep microchannel reactors (FMR) can be used to build a plant for industrial-scale production. To increase the throughput

while keeping the property of microchannel reactors, it is necessary to realize an ideal flow distribution. The flow maldistribution is expected to arise from the nonideality in channel geometry and flow conditions. This study was devoted to clarify the effect of fabrication errors on the reaction interface under various flow conditions. When the channel wall is tilted due to fabrication errors, the distribution of the reaction interface varies. The simulation results showed that the change in the location of the reaction interface is less than 10% of the channel width for most of the cases. The fabrication errors assumed in the present study are expected to be larger than that in real FMRs. These results prove that the FMR can be used to construct a high-throughput micro chemical plant having the reactor characteristics equivalent to microreactors.

Equation 8 was developed on the basis of several assumptions, but it helped us understand the fluid distribution in FMRs. In particular, the qualitative account for the effect of channel walls on the flow distribution was obtained. For deriving a design methodology, a model must be modified so that quantitative behavior of the reaction interface can be predicted.

The plant developed in the present research project is equipped with an FMR and is capable of pumping fluids at a rate greater than 1000 t/y. It is difficult to conduct an experimental study on the flow distribution in the FMR. The results of the present numerical study indicate that the fluid distribution in FMR is nearly uniform. Experimental examination of the performance as a chemical reactor is yet to be done.

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