Design of Feed Segments in Microreactors: Influences of Arrangements and Shapes of Segments on Product Composition for Multiple Reactions

Nobuaki Aoki, Shinji Hasebe, and Kazuhiro Mae

Department of Chemical Engineering, Graduate School of Engineering, Kyoto University
Nishikyo-ku, Kyoto, Japan, 615-8510
aoki@cheme.kyoto-u.ac.jp

ABSTRACT

In many microreactors, reactant flows are split into many lamination segments and then fed into the reactor to enhance mixing efficiency. This feeding method is applied since mixing in microreactors is mainly driven by diffusion because of low Reynolds and thus the mixing time is proportional to the square of diffusion length. The mixing efficiency of reactants greatly influences the product composition for multiple reactions. In most multiple reaction systems, fast mixing is effective to improve yield and selectivity of desired products. Thus, relations among design factors for the feed segments, mixing efficiency, and product composition of multiple reactions are important to establish a design method of microreactors using this feeding method to produce desired products at high yield and selectivity. We studied influences of the design factors for the segments on mixing efficiency and product composition for a multiple reaction system by using CFD simulations. The design factors studied were arrangements, aspect ratio of the rectangular cross section, and cross-sectional shapes of feed segments at the reactor inlet. The results showed that the cross-sectional arrangements and shapes that enable reactants to diffuse to every direction are effective to fasten mixing and improve yield of the desired product. The mixing efficiency is also improved with increase in the aspect ratio when the cross-sectional area of the segment is fixed. This result indicated that the direction that has shorter diffusion length affects greatly the average mixing efficiency on the plane perpendicular to the axial direction. Consequently, the influence of the diffusion direction and that length to each direction are important when we consider the effect of design factors on the yield and selectivity of desired product of multiple reactions.

KEYWORDS: microreactor; arrangements and shapes of feed segments for feed; mixing by diffusion; yield and selectivity of multiple reactions

INTRODUCTION

The miniaturization of reactors leads to low Reynolds number at each reactor channel and laminar flow. Thus, mixing in microreactors is mainly driven by molecular diffusion. The mixing efficiency of reactants greatly influences the product composition for multiple reactions. In most multiple reactions, fast mixing is effective to improve yield and selectivity of desired products. Reduction of diffusion length is essential for fast mixing in microreactors because mixing time is proportional to the square of diffusion length.

In many microreactors, a micromixer is embedded to shorten the mixing time in the reactor. The reactant fluids are split into many lamination segments at the mixer section, and then both species mutually diffuse and react in the reactor. Micromixers such as the interdigital micromixer (Ehrfeld et al., 1999), static V-micro-jetmixer (Ehlers et al., 2000), and SuperFocus mixer (Hessel et al., 2003; Hardt et al., 2003) are used for this purpose. Ehrfeld et al. demonstrated that the reduction of the lamination width by reducing the micro channel width shortens mixing time. Ehlers et al. evaluated the mixing efficiency of the mixer from the selectivity of the secondary product in the parallel competitive reactions. They also studied the progress of the mixing and the parallel competitive reactions by using CFD simulations. Hessel et al. studied effects of mixer chamber geometries on mixing efficiency by experiments and CFD simulations.

However, few studies have quantitatively addressed relations among design factors of feed segments in microreactors and product composition for multiple reactions. These relations are very important to establish a design method of microreactors to produce desired products at high yield and selectivity. The objective of our study is to clarify the relation and then establish a design method of feed segments.

In the previous study (Aoki et al., 2004), only the mixing at laminated segments has been discussed. When the shape of the inlet channels can be changed arbitrarily, the arrangement and the shape of feed segments can be regarded as the design factors of the microreactors. Extending the previous study, influences of design factors such as arrangements and shapes of feed segments on relations between the yield of the desired product and the conversion of the reactant for a multiple reaction system are studied by using CFD simulations.

SIMULATION METOHDS

First, the settings for all CFD simulations of this study are explained. The microreactors applied in the CFD simulations are that reactant fluids split into multiple segments are fed into the reactor and then reactants mutually mix by diffusion and react. We simulated the reaction process in the reactor for various arrangements and shapes of the feed segments that will be mentioned in the subsequent sections. The reaction formulas and the rate equations of the multiple reactions proceeding in the microreactors were as follows:

$$\left.\begin{array}{ll}\mathrm{A}+\mathrm{B}\rightarrow\mathrm{R}, & r_1=k_1C_\mathrm{A}C_\mathrm{B}\\ \mathrm{B}+\mathrm{R}\rightarrow\mathrm{S}, & r_2=k_2C_\mathrm{B}C_\mathrm{R}\end{array}\right\}, \tag{1}$$

where A and B are the reactants; R is the desired product; S is the by-product; r_i and k_i are the reaction rate [$\mathrm{kmol\cdot m^{-3}\cdot s^{-1}}$] and the rate constant of the ith step [$\mathrm{m^3\cdot kmol^{-1}\cdot s^{-1}}$], respectively, and C_j is the molar concentration of component j. The reactants react isothermally, and $k_1 = k_2 = 1\ \mathrm{m^3\cdot kmol^{-1}\cdot s^{-1}}$. The initial concentrations of the reactants are as follows: $C_{\mathrm{A}0} = 27.7\ \mathrm{kmol^{-1}\cdot m^3}$; $C_{\mathrm{B}0} = 55.4\ \mathrm{kmol^{-1}\cdot m^3}$. Dominant assumptions used in the CFD simulations were as follows:

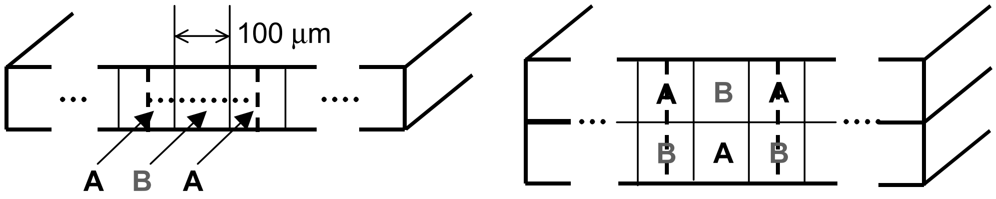
- The diffusion coefficient of every component, D, is $10^{-9}\ \mathrm{m^2\cdot s^{-1}}$.
- The reactor length, L, is 1 cm; the inlet velocity of reactant fluid, u, is $0.005\ \mathrm{m\cdot s^{-1}}$. That is, the average residence time of the reactants in the microreactors is 20 s.
- The vessel dispersion number, D/uL. is 2×10^{-4}, and the influence of axial dispersion is negligible (Levenspiel, 1998).

A commercial CFD code Fluent 6.1® was used for the CFD simulations. This code solves mass, momentum, and energy conservation equations by the control volume method. The laminar flow and finite-rate model were employed. The SIMPLE algorithm was used to solve pressure-velocity coupling equation; the second order upwind scheme was used to solve mass and momentum conservation equations.

Using the settings mentioned above and the design factors explained in the subsequent sections, we calculated relations between the yield of the desired product R, Y_R and the conversion of the reactant A, x_A in the microreactors. Y_R and x_A were calculated from the mass-weighted average concentration of each component on the plane perpendicular to the axial direction.

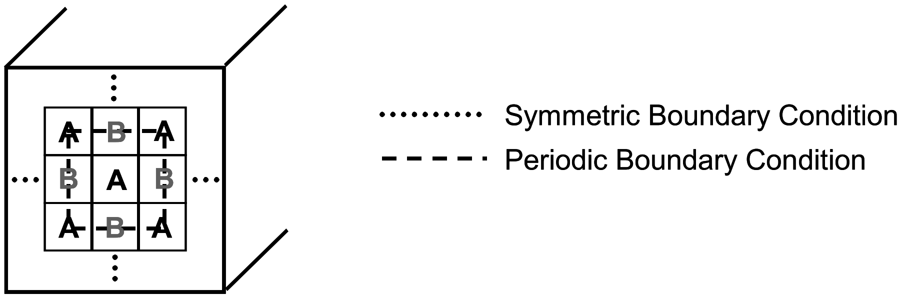
Segment arrangements

The three types of reactors that have different segment arrangements were simulated. The three segment arrangements are shown in Figure 1. In Arrangement 1, the segments that have square cross section at the reactor inlet are aligned periodically in a line; in Arrangement 2, the segments are aligned periodically in two lines; and in Arrangement 3, the segments are arranged regularly in two-dimensional directions. The side length of the square cross section of each segment for all Arrangements is 100 μm. Dashed lines in the figure represent the periodic boundary conditions and dotted line represents the symmetric boundary conditions. The simulation domain for Arrangement 1 was the volume surrounded by the periodic boundaries, the symmetric boundary, and the wall of the reactor. The domain for Arrangement 2 was the volume surrounded by the periodic boundaries and the walls of the reactor. Effects of the reactor walls of the vertical sides were neglected in the simulations for Arrangements 1 and 2. The domain for Arrangement 3 was the volume surrounded by the periodic boundaries, and effects of the reactor walls of the vertical and horizontal sides were neglected. The numbers of mesh elements in simulation domains were 40,000 for Arrangement 1 and 80,000 for Arrangement 2 and 3. The shape of the mesh elements was cuboid.



(a) Arrangement 1: segments are aligned periodically in a line.

(b) Arrangement 2: segments are aligned periodically in two lines.

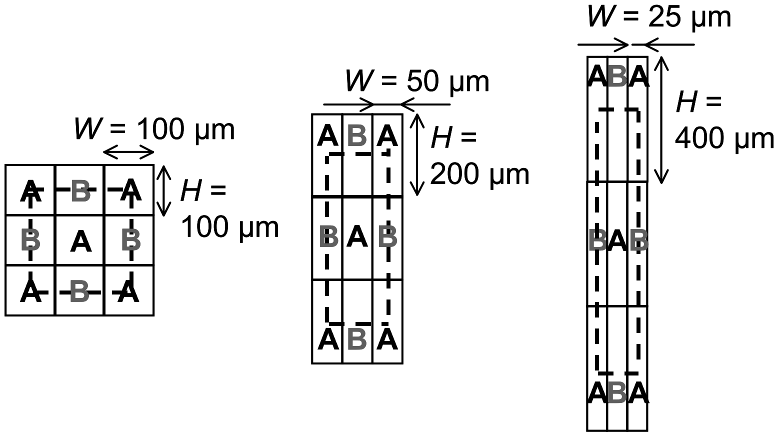


(c) Arrangement 3:
Segments are regularly arranged in two dimensional direction.

Figure 1. Arrangements of feed segments that have the square cross section.

Aspect ratio of the rectangular cross section of the segments

For Arrangement 3 in Figure 1, the influence of aspect ratio of the rectangular cross section on the relation between Y_R and x_A was examined. Figure 2 shows the segment sizes. The aspect ratio of the rectangular cross section was 1 (width, W = 100 μm, height, H = 100 μm), 4 (W = 50 μm, H = 200 μm), or 16 (W = 25 μm, H = 400 μm). Each rectangular cross section of these three aspect ratios takes the same cross-sectional area, 10^{-8} m^2. The simulation domain for each aspect ratio was the volume surrounded by the periodic boundaries represented by dashed lines in Figure 2. The number of mesh elements was 80,000 for all cases. The shape of the mesh elements was cuboid.

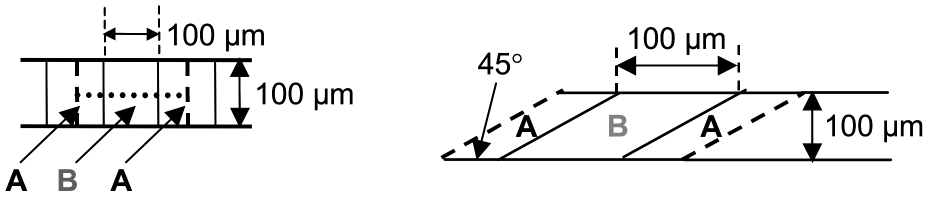


(a) Aspect ratio 1 (b) Aspect ratio 4 (c) Aspect ratio 16

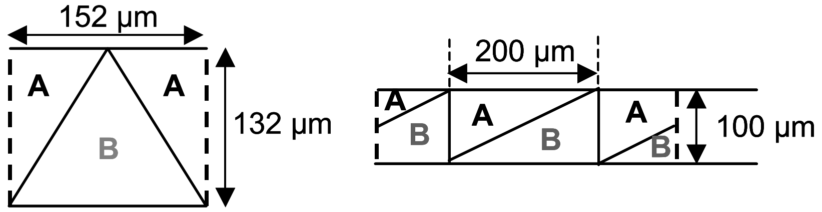
Figure 2. Shapes of fluid segments at the reactor inlet having different aspect ratios.

Cross sectional shapes of the segments

The influences of the cross-sectional shapes of feed segments at the reactor inlet on the progress of the multiple reactions were then investigated. Rectangular, parallelogram, regular triangle, and right-angled triangle were employed as the cross-sectional shape as shown in Figure 3. The cross-sectional areas of the segments for all shapes take the same value, 10^{-8} m^2. The segments having each cross-sectional shape are periodically aligned in a line. In Figure 3, dashed lines represent the periodic boundary conditions, and dotted line represents the symmetric boundary conditions. Table 1 summarizes the cross-sectional shapes and the numbers of mesh elements for the simulation domains.



(a) Rectangle (b) Parallelogram



(c) Regular Triangle (d) Right-angled Triangle

Figure 3. Cross sectional shapes of feed segments at the reactor inlet.

Table 1. Number and shape of mesh elements for each simulation domain.

Cross-sectional shapes of feed segments	Shape of mesh elements	Number of mesh elements
Rectangle	Cuboid	40,000
parallelogram	Hexahedron	40,000
regular triangle	Cuboid and Hexahedron	59,200
Right-angled triangle	Hexahedron	67,600

RESULTS AND DISCUSSION

Segment arrangements

Figures 4 shows the relations between Y_R and x_A for each reactor having different arrangement of the segments. The yield of R improves with increasing the number of the feed segments to the vertical direction. This tendency can be explained by considering the direction of diffusion for each arrangement. For Arrangement 1, each reactant mainly diffuses to the horizontal direction. For Arrangement 2 and 3, each reactant diffuses to the vertical direction as well as the horizontal direction. Thus, the reactants in Arrangement 2 and 3 diffuse faster than in Arrangement 1. Moreover, the reactants in Arrangement 3 diffuse both upward and downward direction, while the reactants in Arrangement 2 diffuse to upward or downward direction. Therefore, Arrangement 3 is the most effective for fast mixing. Since faster mixing improves the yield of R in this multiple reaction system as shown in our previous study (Aoki et al, 2004), Arrangement 3 gives the highest yield of R.

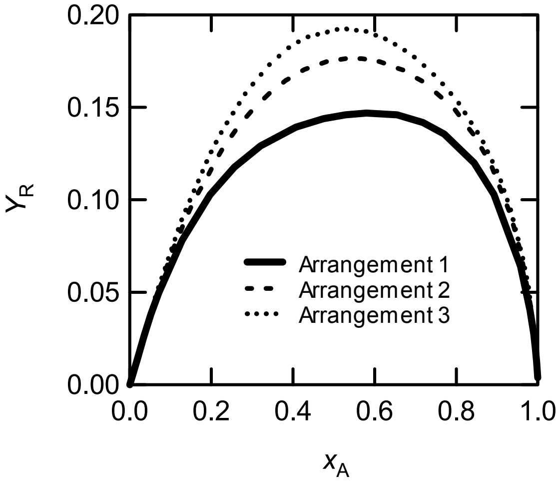


Figure 4. Comparison of relations between Y_R and x_A for the three segment arrangements.

Aspect ratio of the rectangular cross section of the segments

Figure 5 shows the relation between Y_R and x_A for each aspect ratio of the rectangular cross section. The yield of R at the same conversion of A improves with increasing the aspect ratio. This result also means that increasing the aspect ratio enhances the average mixing efficiency on the plane perpendicular to the axial direction. When the aspect ratio is increased, the diffusion length to the horizontal direction decreases while the diffusion length to the vertical direction increases. In other words, increasing aspect ratio enhances the efficiency of mixing by diffusion to the horizontal direction and lowers it to the vertical direction. This indicates that the direction that has shorter diffusion length greatly affects the average mixing efficiency.

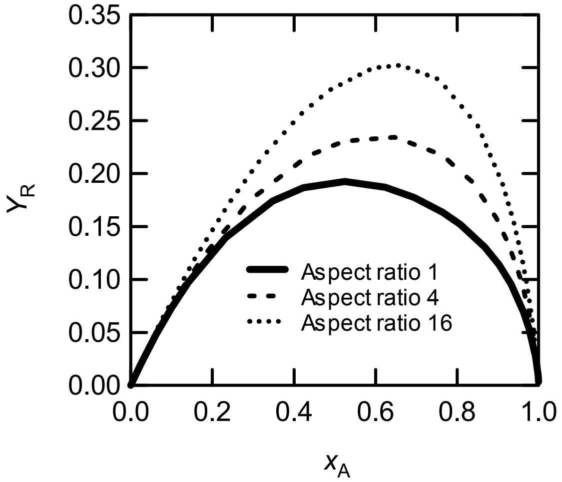


Figure 5. Influence of the aspect ratio of the rectangular cross section on the relation between Y_R and x_A.

Cross-sectional shapes of the segments

Figure 6 shows the relation between Y_R and x_A for each reactor having different cross-sectional shape of the segments at the reactor inlet. The yield of R for the reactor having the feed segments of the square cross section is lower than that of the other shapes of the cross section. This tendency can also be explained by considering the direction of diffusion for each cross-sectional shape. The reactants diffuse to only the vertical direction when the cross-sectional shape is the square, while the reactants diffuse to the horizontal and vertical direction when the cross section is the other shape. Consequently, the cross-sectional shapes other than the square cross section enable reactants to diffuse faster than the square cross section and thus improve the yield of R.

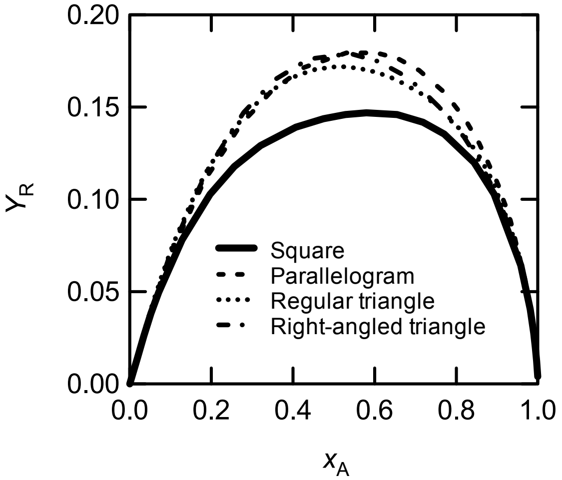


Figure 6. Influence of cross-sectional shapes on the progress of the multiple reactions.

From the results in this section, the influence of the diffusion direction and that length to each direction are important when we consider the effect of design factors on the yield and selectivity of desired product of multiple reactions. Quantitative study on effects of arrangements and cross-sectional shapes of the feed segments on diffusion length to each direction and progress of multiple reactions is needed to establish a method for designing microreactors where reactants are split into segments and then fed.

CONCLUSION

We studied the influences of the design factors for the feed segments on the mixing efficiency and the progress of the multiple reactions by using CFD simulations. The design factors discussed in this study were arrangements, aspect ratio of the rectangular cross section, and cross-sectional shape of the segments at the reactor inlet. The arrangements and cross-sectional shapes of segments at the reactor inlet that enable reactants to diffuse to every direction are effective to produce the desired product for the multiple reactions if the cross-sectional area of the feed segments is fixed. The mixing efficiency is also improved with increase in the aspect ratio when the cross-sectional area of the segment is fixed. This result indicated that the direction that has shorter diffusion length greatly affects the average mixing efficiency on the plane perpendicular to the axial direction. The influence of the diffusion direction and that length to each direction are important when we consider the effect of the design factors on the yield and selectivity of desired product of multiple reactions.

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