

Mixing in microreactors: effectiveness of lamination segments as a form of feed on product distribution for multiple reactions

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Abstract

A mixing model in a microreactor was developed by using the principle of lamination segments. For four types of multiple reactions, the effects of lamination width and rate constants on the relation between the conversion of reactant and the yield of desired product were investigated by using CFD simulations. The simulation reveals that the lamination width greatly affects the yield for the desired product. In most cases, when the conversion of the reactant is the same, the yield of the desired product for perfect mixing is higher when reactants are fed with lamination segments. An exception is that the rate constant of the reaction producing the desired product is much smaller than that consuming the desired product, and the order of the former reaction is less than the latter. In this case, the feed with large lamination width shows higher yield of the desired product than the feed with perfectly mixed. In this study, the maximum lamination width which shows almost the same performance as the case of perfect mixing was derived for various multiple reactions and reaction conditions. The results provide effective information for the design of microreactors and of the measurement system of rate constants of multiple reactions. © 2004 Elsevier B.V. All rights reserved.

Keywords: Microreactor; Computational fluid dynamics; Multiple reactions; Conversion; Yield; Selectivity; Diffusion; Multilamination

1. Introduction

Microreactors have the potential to be useful tools for chemical syntheses and kinetics study. The reduction of reactor dimension leads to a large surface to volume ratio of the reaction channel, which increases heat- and mass-transfer efficiencies. This feature allows microreactors to suppress hot spots and to be released from problems of mass-transfer limitation. With these characteristics, microreactors have the possibility of being used for inherently safe production and measurement of intrinsic rate constants. These advantages have been demonstrated for various fast and highly exothermic reactions such as direct fluorination of organic compounds [1], partial oxidation of ammonia [2], ketone reduction using Grignard reagents [3], and hydrocarbon hydrogenation and dehydrogenation [4]. Kinetics parameters of phosgene formation [5] and oxidation of hydrogen [6] were measured by using the feature of the microreactors that the heat- and mass-transfer limitations are removed.

The reduction of reactor dimension also leads to small Reynolds number at each reactor channel and laminar flow.

Thus, mixing in microreactors is mainly driven by molecular diffusion. The degree of mixing of reactants greatly influences the product composition for multiple reactions and the kinetic measurements for a very fast reaction whose reaction time is shorter than the mixing time. The influences of mixing degree on product composition of multiple reactions have been investigated for reactors and mixers of macro- [7–15] and micro-scales [16–18]. Several studies approached this issue by using computational fluid dynamics (CFD) simulations [13–16]. Fluent[®] is an example of such CFD codes and solves the conservation equations for mass, momentum and energy by using the control volume method.

The reduction of the diffusion length is essential for fast mixing in microreactors, since mixing time is proportional to the square of diffusion length. In many micromixers, which are important parts of microreactors, the reactant flow is split into many lamination segments to shorten the mixing time. The interdigital micromixer [17] and the static V-micromixer [18] are examples of mixers that use this mixing principle. It is possible to quickly mix liquids or gases by those mixers. However, few studies quantitatively addressed the relationship between the size of lamination segments in microreactors and the product composition of multiple reactions, and how the size of lamination segments affects the

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preciseness of the measurements of rate constants. Those are very important factors for establishing a design method of microreactors for industrial production.

The objective of the present study is to show the beneficial effects of feeding reactants with a form of lamination segments for multiple reactions, and to show the lamination width in microreactors necessary to exhibit desired performance. For the problem of measurements of rate constants, the proper lamination width needed for precise measurements is also discussed. To this end, a model of lamination segments mixing in a microreactor was developed, and relations among the lamination width, mixing efficiency, rate constants and yields of desired products for multiple reactions were calculated by the use of CFD simulations.

2. Methods for CFD simulations

Fluent[®] is used to calculate the profiles of velocity and concentrations of species in microreactors. The laminar flow and finite-rate model are employed in all of this study. Fig. 1 shows the lamination segments model used to examine the effect of the lamination width on the extent of multiple reactions. Reactants A and B flow into the reactor with the form of lamination and then both species mutually diffuse and react. The width of each lamination flow of reactant A between two parallel plates is hereafter expressed by W , and is the same as that of B for all cases. As the mixing time is proportional to the square of lamination width, the mixing degree can be controlled by changing the lamination width.

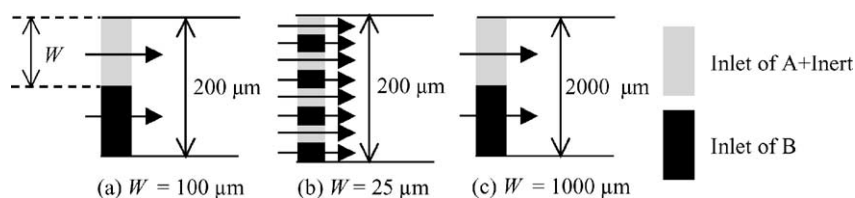
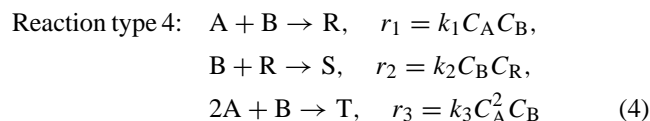
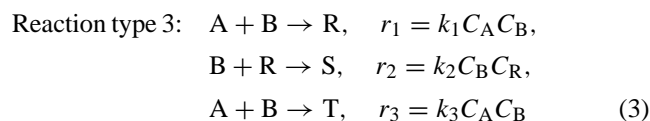
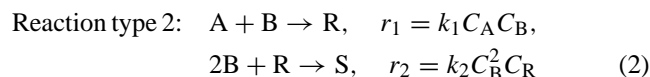
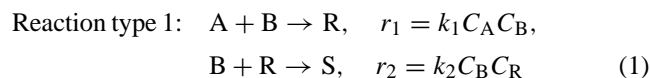


Fig. 1. Schematic representation of reactor inlet.

When the interval of two parallel plates is 200 μm , the width of each lamination flow, W , is 25 or 100 μm . When the interval is 2000 μm , W is 1000 μm . The case where the reactants in the feed are perfectly mixed is also adopted as a case of an ideal mixing. This case is hereafter called “mixed feed”. For all cases, the reactor length, L , is 1 cm.

To discuss the effects of reaction types, four different types of reaction systems are adopted in this research. The reaction formulas and the rate equations of multiple reactions are as follows:



where R is the desired product; S and T are the by-products; r_i and k_i the reaction rate and the rate constant of i th step, respectively; and C_j the molar concentration of species j .

Table 1
Simulation settings

Reaction types	The ways reactants are fed	Rate constants ^{a,b}	C_{A0} (kmol/m ³)	C_{A0}/C_{B0}^c
1	$W = 100$ or $25 \mu\text{m}$ or mixed	$k_1 = k_2 = 0.01$ or 1 or 100	13.9	0.50
	$W = 100$ or $25 \mu\text{m}$ or mixed	$k_1 = 0.01$ and $k_2 = 1$	13.9	0.50
2	$W = 100$ or $25 \mu\text{m}$ or mixed	$k_1 = k_2 C_{B0} = 0.01$ or 1	9.23	0.33
	$W = 100$ or 25 or $1000 \mu\text{m}$ or mixed	$k_1 = 0.01$ and $k_2 C_{B0} = 1$	9.23	0.33
3	$W = 100$ or $25 \mu\text{m}$ or mixed	$k_1 = k_2 = k_3 = 0.01$ or 1	18.5	0.67
	$W = 100$ or $25 \mu\text{m}$ or mixed	$k_1 = k_2 = 0.01$ and $k_3 = 1$	18.5	0.67
4	$W = 100$ or $25 \mu\text{m}$ or mixed	$k_1 = k_2 = k_3 C_{B0} = 0.01$ and 1	27.7	1.0
	$W = 100$ or $25 \mu\text{m}$ or mixed	$k_1 = k_2 = 0.01$ and $k_3 C_{B0} = 1$	27.7	1.0

^a The unit of rate constant is $\text{m}^3 \text{kmol}^{-1} \text{s}^{-1}$ except for the following two cases: that is, the unit of k_2 in Reaction type 2 and that of k_3 in Reaction type 4 are $\text{m}^6 \text{kmol}^{-2} \text{s}^{-1}$.

^b C_{j0} is the concentration of species j at the inlet of the reactor for the whole feed.

^c $C_{B0} = 27.7 \text{ kmol/m}^3$ for all cases.

Dominant assumptions used in CFD simulations are as follows: (1) Physical property of every reactant fluid is the same, that is, the density is $9.982 \times 10^2 \text{ kg/m}^3$, molecular weight is $1.802 \times 10^{-2} \text{ kg/mol}$, molecular diffusion coefficient, D , is $10^{-9} \text{ m}^2/\text{s}$, and viscosity is 0.001 Pa s ; (2) the reactants react isothermally; and (3) the inlet velocity of each reactant fluid, u , is the same.

For various combinations of k_i , W , and the reaction type shown in Eqs. (1)–(4), CFD simulations are conducted to calculate the relation between the mass average conversion of reactant A at the reactor exit, x_A , and the yield of the desired product R, Y_R . Table 1 summarizes the cases simulated in this study. To derive the results of different conversions of reactant A, simulations with different residence times are also executed by changing the inlet velocity. The longer the residence time, the larger is the vessel dispersion number, D/uL . In other words, the longer the residence time is, the larger is the influence of axial dispersion. In this study, to ensure that the axial dispersion is negligible, this dimensionless number is kept not larger than 0.01 for all cases [19]. Except for some cases in Reaction type 1, the number of mesh elements for the simulation domain is 3000 for the cases of mixed feed or $W = 100 \mu\text{m}$, is 6600 for $W = 25 \mu\text{m}$, and is 12,000 for $W = 1000 \mu\text{m}$. For the cases of Reaction type 1 with $k_1 = k_2 = 100 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$, the number of mesh elements is 12,000 for $W = 100 \mu\text{m}$, and is 105,600 for $W = 25 \mu\text{m}$.

3. Results and discussions

3.1. Effects of lamination width and rate constants

First, how the relation between Y_R and x_A is affected by the rate constants and the lamination width is discussed for the cases of Reaction type 1. The rate constants of both steps are assumed to be the same, that is, $k_1 = k_2 = k$. Fig. 2 shows the relation between Y_R and x_A for different combinations of rate constant and lamination width. Reactants A and B are fed stoichiometrically for the overall reaction,

$A + 2B \rightarrow S$. Thus, the reactants are completely converted to the end product S when $x_A = 1.0$. For perfectly mixed feed, the relation between Y_R and x_A depends only on the ratio of k_2 to k_1 . Thus, the profile does not depend on the value of k as shown in Fig. 2.

The dimensionless number for n th order reactions, $\phi_i = k_i C_{B0}^{n-1} W^2 / D$, represents the ratio between the reaction rate and the diffusion rate. It is said that this number gives a criterion to determine the controlling step of reactions: the reaction is the controlling step when $\phi_i < 1$, and the diffusion is the controlling step when $\phi_i > 10^4$ [20,21]. In a low reaction rate such that $k = 0.01 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$, ϕ_i is 2.8 when $W = 100 \mu\text{m}$, and is 0.17 when $W = 25 \mu\text{m}$. Thus, the intrinsic reaction is the rate controlling step when W is $25 \mu\text{m}$. The lamination width of $100 \mu\text{m}$ is narrow enough to achieve almost comparable relation with that obtained from the case of mixed feed. For greater rate constants, the diffusion of reactants affects the reactions, that is, ϕ_i becomes larger. The yields at the same conversion of A decrease with the increase of the lamination width and/or the rate constants. When $k = 100 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$ and $W = 100 \mu\text{m}$, ϕ_i takes a large value of 2.7×10^4 . In this case, the yield of desired product R is much lower than those of other cases. This is because the primary reaction is strongly affected by the mixing as the diffusion length between A and B is longer than that between B and R.

The results shown in Fig. 2 suggest that the rate constants calculated by assuming perfect mixing are strongly affected by W and k . In other words, for faster reactions, a narrower lamination width is needed to measure rate constants precisely from a perfect mixing model. It is said that the relation between Y_R and x_A is comparable to that for the case of mixed feed when $\phi_i < 1$ [20,21]. The results in Fig. 2 verify their proposition. When $\phi_i > 10$, the results in Fig. 2 differ largely from that for the case of mixed feed. Thus, a precise model including CFD should be used to calculate rate constants if $\phi_i > 10$.

Fig. 3 shows the lamination width that satisfies $\phi_i = 1$ for second order reactions and $D = 10^{-9} \text{ m}^2/\text{s}$, which is the

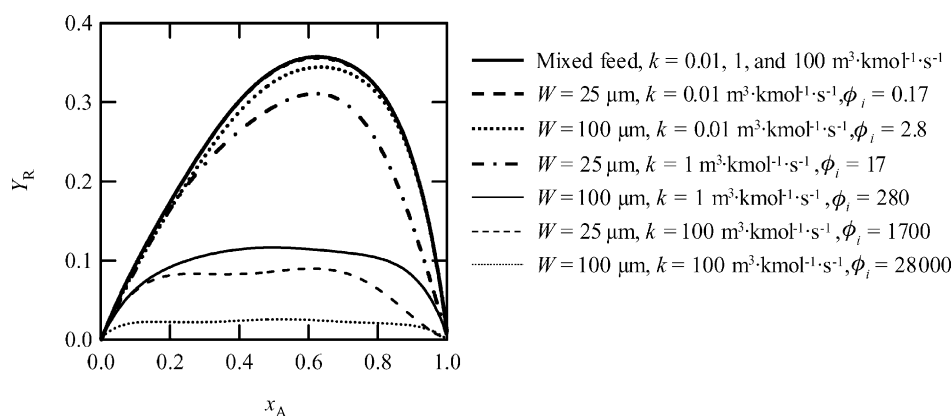


Fig. 2. Relation between yield of R and conversion of A with different rate constants and lamination widths (Reaction type 1).

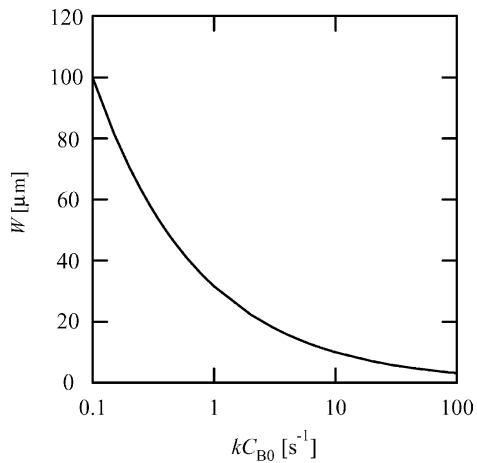


Fig. 3. Lamination width which satisfies $\phi_i = 1$.

order of molecular diffusion coefficient of liquids phase. By drawing such a figure, it is possible to know the lamination width at which the flow into the reactor can be treated as perfectly mixed flow.

3.2. Influences of reaction order of each reactant

In this section, the influences of the reaction order of C_B are discussed from the simulation results on Reactions types 1 and 2. First, the rate constants are changed under the condition that $k_1 = k_2 C_{B0} = k$ for Reaction type 2. Then, the ratio of the rate constants is changed. The same investigation is also conducted for exploring the effects of reaction order of C_A for Reaction types 3 and 4.

3.2.1. Effects of reaction order of C_B

Under the condition that $k_1 = k_2 C_{B0} = k$, the relation between Y_R and x_A is derived for Reaction type 2 (the results are summarized in Fig. 4). As long as the rate constant satisfies the condition that $k_1 = k_2 C_{B0} = k$, the dependence of the lamination width on the relation between Y_R and x_A and that of rate constants on the relation between Y_R and x_A

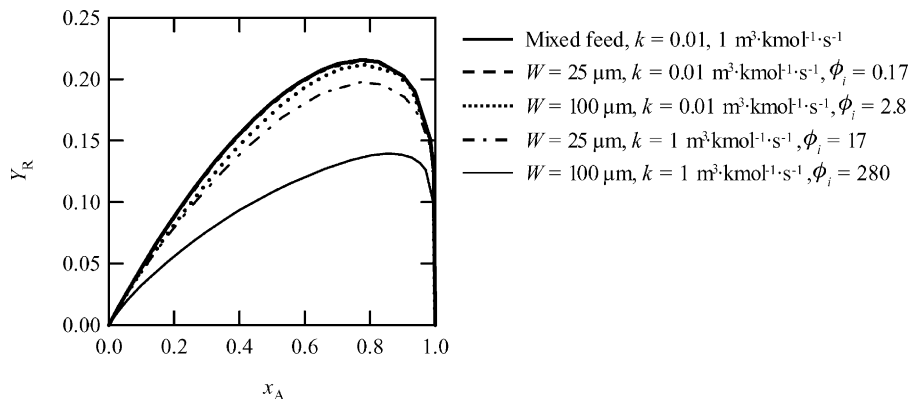


Fig. 4. Influence of the order of C_B on the relation between Y_R and x_A for Reaction type 2.

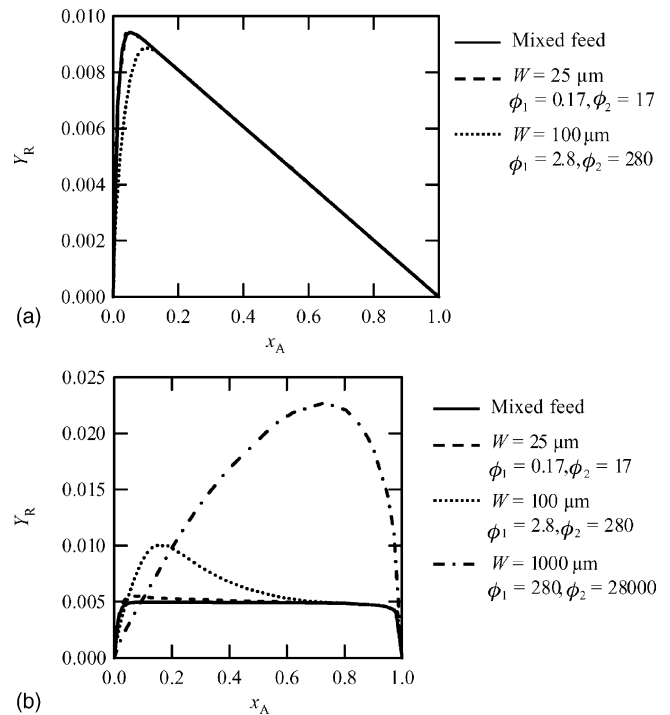


Fig. 5. Influence of the order of C_B on the relation between Y_R and x_A : (a) $k_2/k_1 = 100$ for Reaction type 1; (b) $k_2 C_{B0}/k_1 = 100$ for Reaction type 2.

are similar to those for Reaction type 1 which are shown in Fig. 2.

When the ratio of rate constants, $k_2 C_{B0}/k_1$, is changed to 100, ϕ_2 , which represents ϕ of the second step, becomes larger, and the second step becomes closer to diffusion control condition. Fig. 5 shows the relation between Y_R and x_A for the case where the rate constant of the first step is different from that of the second step. The results of Reaction types 1 and 2 are shown in Fig. 5(a) and (b), respectively. In the simulation, $k_2/k_1 = 100$ for Reaction type 1 and $k_2 C_{B0}/k_1 = 100$ for Reaction type 2. k_1 is $0.01 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$ for all cases. For Reaction type 2, a wider lamination width shows higher yield of the desired

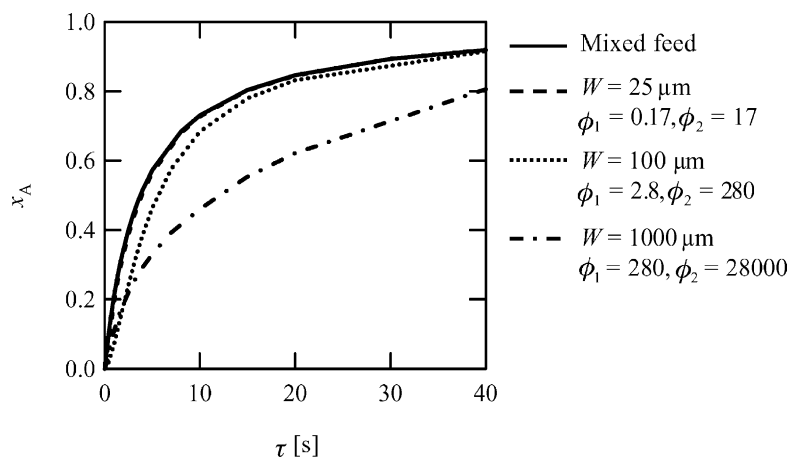


Fig. 6. Relation between x_A and the average residence time τ for Reaction type 2; $k_2C_{B0}/k_1 = 100$.

product. This is because the reaction rate of the higher order reaction is more strongly affected by the concentrations than that of the lower order reaction, and the higher order reaction proceeds only in the region where the reactants are well mixed. The ratio of this region to the whole reactor volume decreases as the increase of lamination width. Therefore, the wider the lamination width, the more preferable the production of R. The result shows that the feeding of reactants at wide lamination width is effective for obtaining the desired

products in some multiple reactions. However, wider lamination width reduces the mixing rate by diffusion and the average reaction rate. The relations between the conversion of reactant A, x_A , and the average residence time, τ , are shown in Fig. 6.

3.2.2. Effects of reaction order of C_A

Fig. 7 shows the relation between Y_R and x_A for Reaction types 3 and 4. In this figure, the lamination width is changed

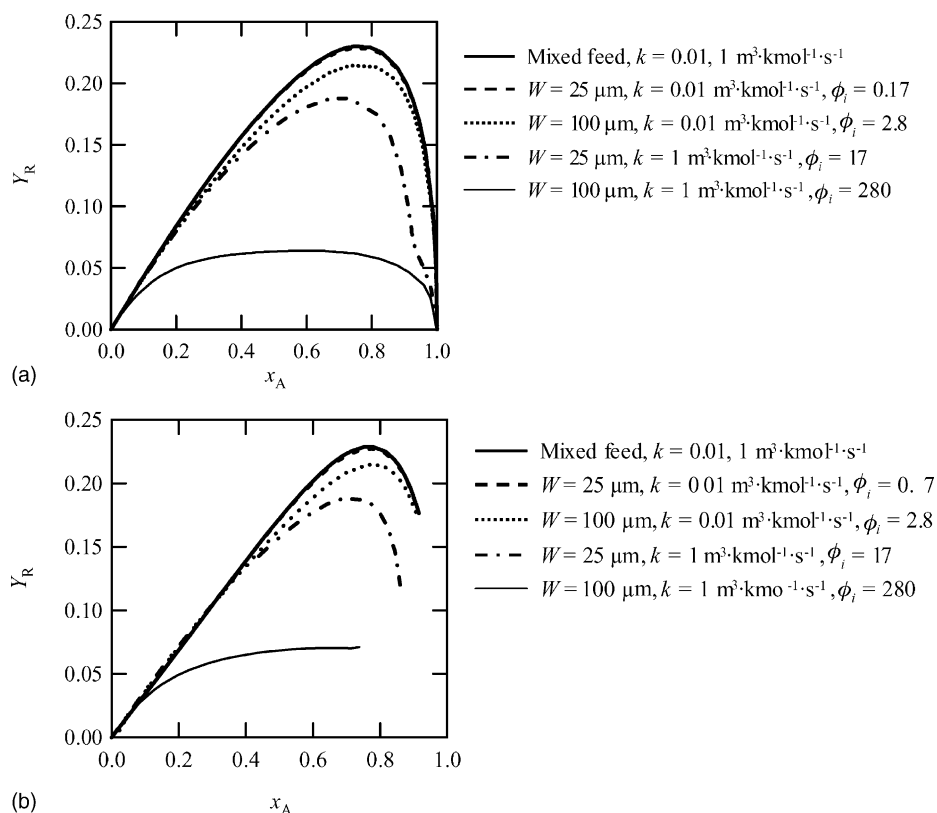


Fig. 7. Influence of the order of C_A on the relation between Y_R and x_A : (a) $k_1 = k_2 = k_3 = k$ for Reaction type 3; (b) $k_1 = k_2 = k_3 C_{B0} = k$ for Reaction type 4.

under the condition that $k_1 = k_2 = k_3 = k$ for Reaction type 3 and that $k_1 = k_2 = k_3 C_{B0} = k$ for Reaction type 4. For all cases, k_1 is $0.01 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$. Similar tendencies are obtained from the simulations of the above conditions as those shown in the previous subsection discussing the influence of the order of C_B . For Reaction type 4, the conversion of reactant A does not reach 1.0. The reason is as follows. In this case, the initial rates of the first and third steps are equivalent as $k_1 = k_3 C_{B0} = k_3 C_{A0}$. The rate of the third step decreases faster than that of the first step because the order of the third step is higher than that of the first step, and thus the third step is slower than the first step. Therefore, the first step proceeds favorably, which makes the second step proceed favorably. Since the moles of both reactants fed into the reactor are the same, and reactant B is favorably consumed by the overall reaction of the first and second steps, $A + 2B \rightarrow S$, reactant B is consumed completely while reactant A still remains. The ultimate conversion of reactant A at the end of the line in Fig. 7(b) becomes smaller as ϕ_i increases. This result shows that the relative consumption of reactant B to reactant A becomes larger as ϕ_i increases, and that the first step of Reaction type 4 proceeds more favorably.

Fig. 8 shows the relation between Y_R and x_A for the case where the third step of Reaction types 3 and 4 is much faster

than the others. The results in Fig. 8 are qualitatively similar to those in Fig. 5. That is, the wider lamination width shows lower yield of desired product for Reaction type 3 and higher yield for Reaction type 4. From the discussion so far, we can conclude that feeding reactants with wider lamination width provides higher yield and selectivity of desired product at the same conversion when both the reaction order and the rate constant for the reaction producing by-products are higher than those of the reaction producing the desired product. However, the reactions proceed slower compared to the case where reactants in the feed are perfectly mixed.

The results of this section show the following advantages of the mixing of reactants at regular lamination segments: one is that the rapid diffusion via lamination segments of reactant fluids increases production rate for fast reactions, and the other is that the mixing of reactants at proper lamination width increases the selectivity of desired product. The second advantage is typical for microreactors in which reactants are fed in the form of lamination segments. Precise control of mixing is difficult in a conventional stirred tank reactor. On the other hand, in the microreactor the size of lamination segments can be selected arbitrarily by properly designing the mixing device. In other words, the selection of proper lamination width is essential for the microreactor to increase the efficiency.

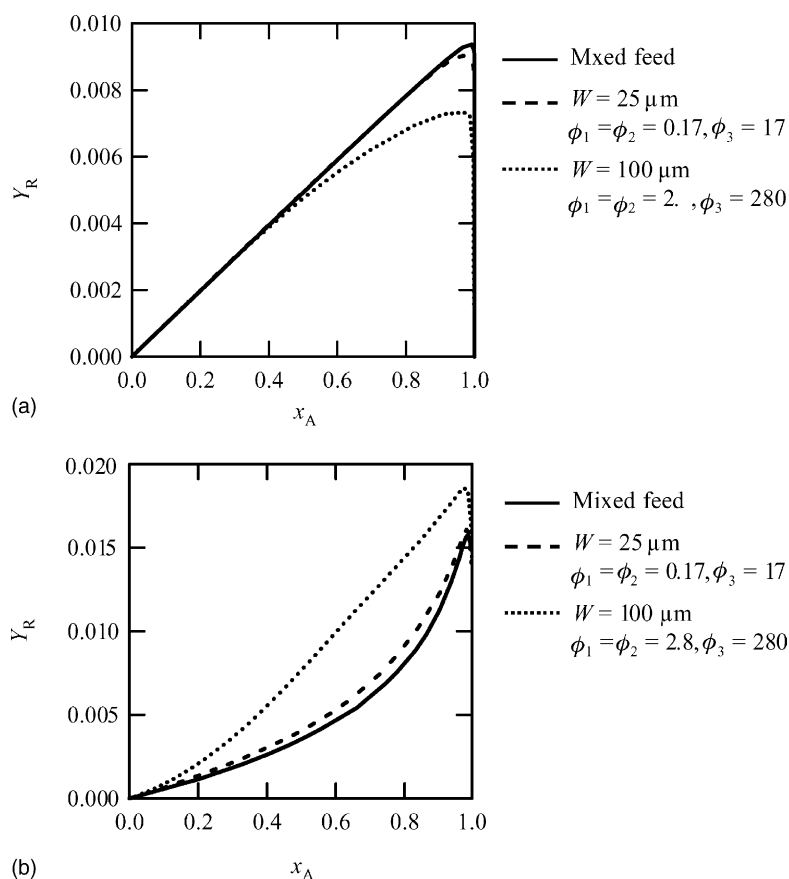


Fig. 8. Influence of the order of C_A on the relation between Y_R and x_A : (a) $k_3/k_1 = 100$ for Reaction 3; (b) $k_3 C_{B0}/k_1 = 100$ for Reaction 4.

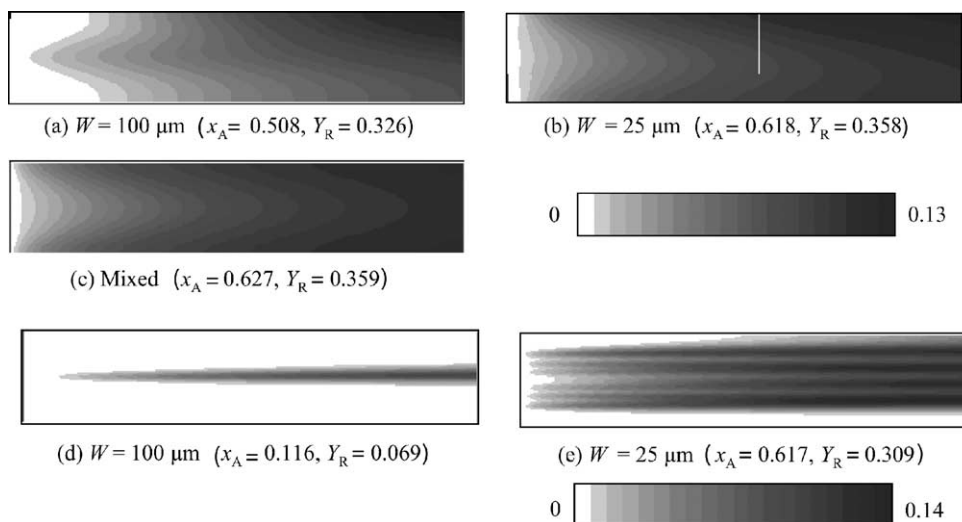


Fig. 9. Mole fraction of R in the reactor for Reaction type 1. For (a)–(c), $k_1 = k_2 = 0.01 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$; the inlet velocity is 0.002 m s^{-1} ; and the average residence time is 5 s. For (d) and (e), $k_1 = k_2 = 1 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$; the inlet velocity is 0.1 m s^{-1} ; and the average residence time is 0.1 s.

3.3. Design of microreactors for efficient production and separation of desired product

In a microreactor, reactants flow regularly. Using this feature, structures of reactors having separation function are discussed in this section. When microreactors are designed with a separation function, the distribution of the product mole fraction in the reactor provides us with useful information. Fig. 9 shows contour plots of the mole fraction of the desired product for Reaction type 1. For all cases, the inlet velocity is 0.1 m s^{-1} , and the average residence time is 0.1 s. For cases (a)–(c), $k_1 = k_2 =$

$0.01 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$, and for cases (d) and (e), $k_1 = k_2 = 1 \text{ m}^3 \text{ kmol}^{-1} \text{ s}^{-1}$.

When rate constants are small, reactions proceed after reactants are well mixed in the whole area of the reactor as shown in Fig. 9(a)–(c). When rate constants are large and the lamination is wide, reactions proceed before reactants are well mixed. In this case, the first reaction step occurred only near the contact surface of reactants A and B as shown in Fig. 9(d). In this case, the mixing efficiency is quite low. When the lamination width is small, even if rate constants are large, R is produced in the whole area of the microreactor as shown in Fig. 9(e).

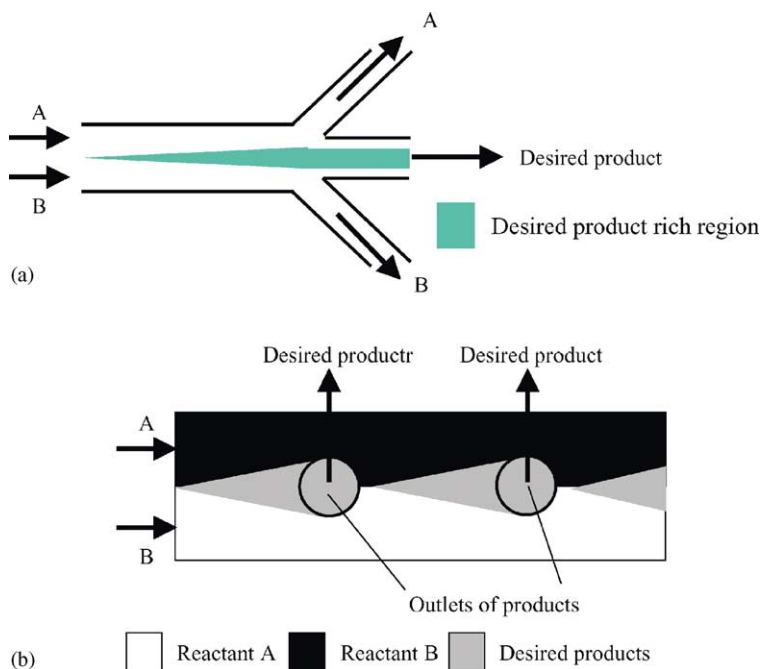


Fig. 10. Reactor with multiple exits: (a) reactor having three-fork-shaped exits; (b) reactor having multiple intermediate exits.

For the case in Fig. 9(d), the desired product is concentrated in the center of the reactor. Thus, the mixing of the flow at the exit of the reactor is not suitable from the viewpoint of the separation of the desired product. A reactor having three-forked-shape outlets shown in Fig. 10(a), for instance, is effective for collecting the desired product from the middle outlet when the product distribution in the reactor is shown in Fig. 9(d). If the residence time is relatively long, products may be effectively separated from the reactants by installing exits on the way of the reactor as shown in Fig. 10(b). This result suggests that new reactor designs can be generated which use feature of the imperfect mixing among reactants and products effectively.

Our next step of the study is to validate the results of this paper by experiments. For this purpose, we are developing a micro-device into which reactants flow with the form of lamination and a method to measure concentration profiles of reactants and products in a reactor. After validating the simulation results, we will develop a measurement system of rate constants of multiple reactions by using the developed device and the measurement method of concentration profiles. We also design reactors having separation function as shown in this section by using CFD. The reactor design will be combined with controlled mixing by the design of lamination width to develop a device that efficiently produces and separates a desired product.

4. Conclusions

One of the dominant characteristics of microreactors is that the mixing condition in the reactor can be controlled by the design. Thus, a proper mixing condition should be decided for each reaction system before a microreactor is designed. In this research, for the case where reactants flow into the reactor with the form of lamination, the relation between the conversion of reactant and the yield of desired product was investigated by using CFD simulations. For four types of reaction systems each of which consists of multiple reactions, the effects of lamination width and rate constants on the yield of the desired product was investigated.

The results of exhaustive simulations show that the lamination width greatly affects the yield of desired product, and that in most cases the perfect mixing of the feed reactants shows better results than do the other cases. Thus, the lamination width which can be regarded as the perfect mixing is useful information for the design of the microreactor. The only exception is the case where the rate constant of the reaction producing the desired product is much smaller than that consuming the desired product and the order of the former reaction is less than the latter. In this case, the feed with large lamination width shows higher yield of the desired product than the feed with perfectly mixed. However, the reactions proceed slowly when reactants are fed with wider lamination segments. The concentration contour plots suggest that imperfect mixing has the possibility of being used

in the device that executes the reaction and the separation simultaneously.

When rate constants are measured under the assumption that the reactants in the feed are perfectly mixed, the degree of mixing affects the precision of the obtained results. The simulation results show that the larger the rate constants are, the smaller the necessary lamination width for precise measurement of rate constants. When the shape of a microreactor is simple, it is possible to estimate the mixing condition in the reactor by using CFD simulation. For such a case, there is the possibility of developing a system which can precisely measure the rate constants without executing the reactions at perfectly mixed condition.

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