

# Design of Two-phase Slug Flow Microreactors Using Reduced Order Flow Model

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Two-phase slug flow microreactors (SFMRs) are known to be capable of achieving more efficient mass and heat transfer than conventional reactors. However, no method for designing SFMRs has been established so far. The goal of this study is therefore to develop a model-based methodology for deriving optimal design and operating variables for SFMRs under constraints on production. Although computational fluid dynamics (CFD) modelling and simulation is a powerful tool for analyzing the characteristics of multiphase flow, it is not realistic to directly apply CFD to optimal design problems of SFMRs, because CFD requires huge amount of computational time. In this study, a reduced order flow model based on the idea of time averaging of void fraction in two-phase flow and mass transfer rate between two phases was developed to efficiently simulate the heat and mass transfer processes involving chemical reactions in SFMRs. And then, a two-stage design method was proposed for design of SFMRs, in which a single channel is designed and then a multichannel device is designed. Finally, the usefulness of the proposed design method was assessed through a numerical case study.

## 1. Introduction

Microreactors gained interests not only from academic investigations but also from chemical and pharmaceutical industry. Advancements made in the R&D of microreactors over the last two decade has been reported in review articles (e.g., Yao et al., 2015, Verdnik et al., 2022). When two mutually immiscible fluids, such as gas and liquid, are simultaneously fed into a microchannel, various flow patterns such as annular, dispersed and slug flows are generated as to the design and operating conditions (Shao et al., 2009). Among these, slug flow, also called Taylor flow or segmented flow, is especially attractive in a broad range of applications (Suryawanshi et al., 2018), due to its small residence time distribution and large interface area per fluid volume (Kreutzer et al., 2008). In addition, rapid mass transfer between two phases is achieved because the liquid interface is constantly renewed by the circulation flow inside the liquid slug (Kashid et al., 2005). These characteristics of two-phase slug flow can bring benefits to reaction system design and operation. For example, the space-time yields of the oxidation of ethyl lactate for producing ethyl pyruvate in two-phase slug flow microreactors (SFMRs) were ten times larger than those in conventional reactors (Yasukawa et al., 2011). The formation of by-products in the direct fluorination of toluene was reduced by using SFMRs (Jahnisch et al., 2000). To increase the production scale of SFMRs, a numbering-up method for setting up multiple channels in parallel was proposed and its fluid distribution performance was evaluated (Rawashdeh et al., 2012). As shown in previous studies, the slug length is recognized as one of the important process parameters. Therefore, a slug length monitoring method has been reported (Miyabayashi et al., 2015). In addition to the above previous studies, research into a design method for an SFMR that achieves the desired slug length and reaction field, especially a systematic model-based design method, is also essential for the practical application of SFMRs. However, there is, to our knowledge, no established the SFMR design method.

In recent years, with the advancement of computer and numerical analysis technology, it is becoming possible to perform simulations using detailed models of multiphase flows, including gas-liquid two-phase flows. A representative example of this is computational fluid dynamics (CFD), which is used to analyze gas-liquid two-phase slug flows in microchannels (e.g., Santos et al., 2010). CFD can provide a more detailed description of the internal flow patterns within the liquid slugs and the transfer of the dissolved chemical species within and

across the slugs. However, to solve the complete slug flow system for mass and heat transfer with chemical reaction using CFD requires extensive computational resources. Therefore, it is currently not realistic to directly use CFD in design by combining it with an optimization method, because repeated CFD involves a large computation (Khatouri et al., 2022). In addition, the use of a two fluid model can be considered as a simpler model than CFD for describing two-phase flow in capillaries (Cao and Luo, 2012). This model considers gas and liquid as two continuous media, and the movement of gas and liquid phases can be described simultaneously by two different control equations. To the best of our knowledge, the two fluid model has only been used for hydraulic studies and has not been applied to the study of two-phase flow systems involving interphase mass transfer, reaction, and heat transfer. In this study, a reduced order flow model is constructed to efficiently simulate the heat and mass transfer processes involving chemical reactions in SFMRs. Then, a developed model-based methodology is proposed to derive the optimal design and operating variables of SFMRs under constraints on production. It consists of two stages: first, the design of a single channel, followed by the design of a multichannel device. Finally, the usefulness of the proposed method is assessed through a numerical case study.

## 2. Modeling and design method for SFMR

The constructed design model and the design procedure are explained.

### 2.1 SFMR to be designed

Figure 1 shows an example of SFMR with numbering-up structure, which means that the channels are parallelized. Gas and liquid feed streams are evenly distributed among the parallelized channels that are located on reaction plates. Each channel consists of a gas-liquid junction followed by a reaction section. The typical channel shape at the gas-liquid junction is T-shape, and the mixing process there determines the slug length. The generated slugs flow through the reaction section, and the slug lengths change due to reaction and mass transfer between the two phases. The streams leaving the channel outlets join together to form the product stream. Cooling plates are arranged on both sides of each of reaction plates to control the reaction temperature. The number of channels is determined by dividing the given total production volume by the production per channel. The number of stacked reaction plates is determined by considering the size constraints of one plate given by the designer and the required channel length. In other words, when the channel length exceeds the plate length constraint, the channel is folded as shown in Figure 1, and the reaction plates are stacked to achieve the required channel length. Engineers need to determine the best design and operation variables such as channel sizes, coolant temperature and inlet flow rates, to realize desirable reaction conditions.

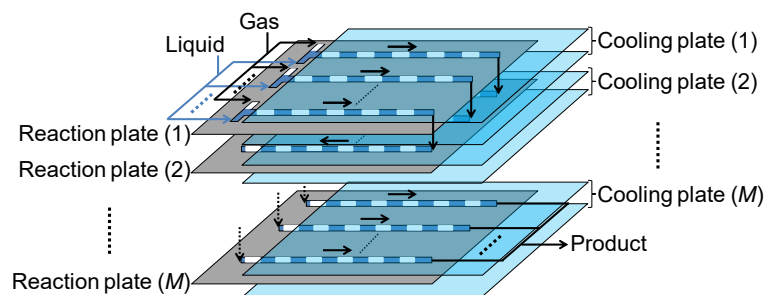


Figure 1: A schematic diagram of SFMR

### 2.2 Reduced order flow model

Models are important for the rational design of chemical production devices. For SFMRs, the following two models are required: one model (model A) needs to explain the relationship between the design and operating conditions of the junction and the length of the slugs that are formed, and another model (model B) needs to predict state variables such as the slug length and the component concentration in the slug that change over the reaction section. Since many correlation equations developed in previous studies, mainly based on experiments, have been reported, they can be used as model A instead of CFD. On the other hand, for model B, a reduced order flow model with lower dimensions than CFD was developed in this study. To predict the change in the slug length and the component concentration caused by reactions and mass transfer between the two phases, in this study, the gas-liquid slug flow shown in Figure 2a was represented by a conceptual diagram shown in Figure 2b.

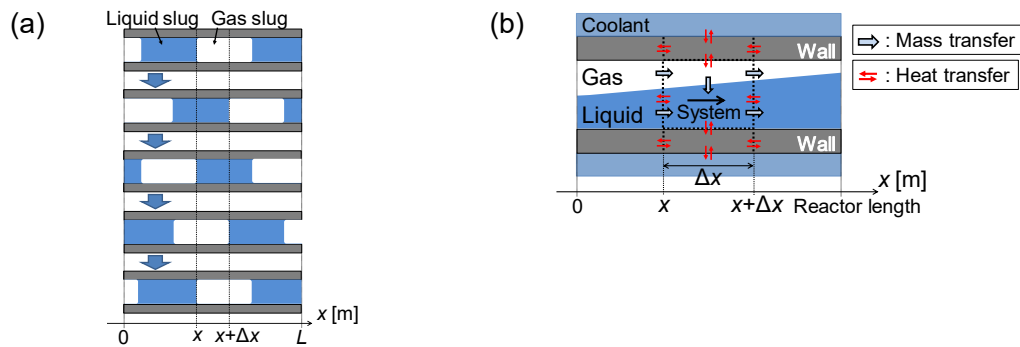


Figure 2: Reduced order flow modelling. (a) Gas-liquid slug flow and (b) conceptual diagram of the model developed in this study

In the gas-liquid slug flow, the gas and liquid slugs pass alternately through a small section  $\Delta x$  of as shown in Figure 2a. Therefore, the conventional plug flow reactor (PFR) model, which is intended for a homogeneous system, cannot be directly applied to the gas-liquid slug flow. The conceptual diagram in Figure 2b shows the time-averaged void fraction ( $\alpha$ ) of gas-liquid slug flow in the  $x$  direction, which corresponding the reactor length. Based on this conceptual diagram, PFR model was applied to the gas flow and liquid flow, respectively, and further, the incorporation of a gas-liquid mass transfer term into the PFR models was considered. The mass transfer rate between the gas and liquid phases was time-averaged at a certain position. The time  $t_1$  required for one gas-liquid slug system to pass through the section  $\Delta x$  is calculated by Eq.(1), and the time  $t_2$  that the gas-liquid interface exists in the section  $\Delta x$  is given by Eq.(2). When calculating  $t_1$  and  $t_2$ , it was assumed that the influence of the wall film and the meniscus on them was negligible.

$$t_1 = \frac{L_{\text{gas}} + L_{\text{liq}}}{u} \quad (1)$$

$$t_2 = \frac{2\Delta x}{u} \quad (2)$$

Using Eqs.(1) and (2), the time-averaged mass transfer rate between the gas and liquid phases at a certain position can be expressed as Eq.(3).

$$A_{\text{gas-liq}} N_A \frac{t_2}{t_1} = A_{\text{gas-liq}} N_A \frac{2\Delta x}{(L_{\text{gas}} + L_{\text{liq}})} \quad (3)$$

When the cross-sectional area ( $A$ ) of the channel is constant, the void fraction and the volume of a liquid slug can be expressed by Eqs.(4) and (5).

$$\alpha = \frac{L_{\text{gas}}}{L_{\text{gas}} + L_{\text{liq}}} \quad (4)$$

$$V_{\text{liq}} = AL_{\text{liq}} \quad (5)$$

Using Eqs.(4) and (5), Eq.(3) can be transformed into Eq.(6).

$$A_{\text{gas-liq}} N_A \frac{t_2}{t_1} = 2A_{\text{gas-liq}} N_A \frac{A\Delta x(1-\alpha)}{V_{\text{liq}}} \quad (6)$$

The reason for this transformation is that in two-phase flow, void fraction is the main parameter affecting mass and heat transfer. Assuming that reactions in the gas phase can be ignored, the final models constructed based on the above ideas are shown below:

$$\frac{dF_{i,\text{gas}}(x)}{dx} = -2A_{\text{gas-liq}} N_A(x) \frac{A(1-\alpha(x))}{V_{\text{liq}}} \quad (7)$$

$$\frac{dF_{i,\text{liq}}(x)}{dx} = 2A_{\text{gas-liq}} N_A(x) \frac{A(1-\alpha(x))}{V_{\text{liq}}} + A \sum v_{i,k} r_k(x)(1-\alpha(x)) \quad (8)$$

$$\frac{dF_j(x)}{dx} = A \sum v_{j,k} r_k(x)(1-\alpha(x)) \quad (9)$$

Eq.(7) corresponds to the gas-phase PFR model for component  $i$ , Eq.(8) corresponds to the liquid-phase PFR model for component  $i$ , and Eq.(9) corresponds to the liquid-phase PFR model for component  $j$ . The constructed model represents the gas-liquid slug flow accompanied by interphase mass transfer and reactions and enables a reduction in the computational load compared to CFD.

### 2.3 Two-stage design

A two-stage method is proposed to design an SMFR for given production conditions. The production conditions can be divided into two categories: those related to the equipment and those related to the reaction system. The former include the material of the equipment, the channel structure at the gas-liquid junction, and the cross-sectional shape of the channel. The latter include product specifications, reaction equations, reaction rate equations, and fluid properties. In the proposed two-stage design method, in the first stage, a design problem is formulated to determine the design and operating conditions of a single channel to satisfy the production conditions, and in the second stage, the number of channels is calculated from the results obtained at the previous stage and the total production volume, and the SFMR is designed taking into account constraints such as the size of one plate, the channel length, and the width between the channels. In the first stage, the objective functions are set, e.g., yield, selectivity, and production rate, and the design and operational variables to be optimized include channel diameter and length, coolant temperature, and gas and liquid feed flow rates. The reduced order flow model shown in the previous section is used to derive the solution. If temperature control is required, it is necessary to formulate the heat balance equation for the slug flow and the wall in addition to the reduced order flow model.

### 3. Case study: problem setting, results and discussion

The usefulness of the developed model and design method is verified through a case study. The case study focused on a parallel gas-liquid reaction in which the main and side reactions are given by Eqs.(10) and (11), respectively.



Here, A and B are the raw materials, R is the target product, and S is a by-product. The reaction rates  $r_1$  and  $r_2$  of Eqs.(10) and (11) are given by Eqs.(12) and (13), respectively.

$$r_1 = 5.0 \times 10^{13} \exp\left(-\frac{19700}{RT}\right) C_{A,\text{liq}} C_B \quad (12)$$

$$r_2 = 1.6 \times 10^{18} \exp\left(-\frac{45000}{RT}\right) C_{A,\text{liq}} C_B \quad (13)$$

The rate equation for the main reaction,  $r_1$ , was determined with reference to the liquid-phase oxidation of benzaldehyde at room temperature. The rate equation for the side reaction,  $r_2$ , was determined so that the side reaction would proceed more dominantly than the main reaction when the reaction temperature reached 293 K or higher. In addition, the following assumptions were made: the material of the reactor is stainless steel, the channel structure at the gas-liquid junction is T-shaped, and the channel cross section is square. Eqs.(14) and (15) (Qian and Lawal, 2006) were used in this study as model A. In addition, the mass transfer coefficient  $k_{\text{liq}}$  between the gas and liquid slugs and the heat transfer coefficient  $h_{\text{wall-liq}}$  between the wall and the slug flow were given by Eqs.(16) (Yue et al., 2009) and (17) (He et al., 2010), respectively.

$$L_{\text{liq},0} / d = 1.637 \alpha(0)^{-0.893} (1 - \alpha(0))^{-0.05} Re(0)^{-0.075} Ca(0)^{-0.0687} \quad (14)$$

$$L_{\text{gas},0} / d = 1.637 \alpha(0)^{-0.107} (1 - \alpha(0))^{-1.05} Re(0)^{-0.075} Ca(0)^{-0.0687} \quad (15)$$

$$k_{\text{liq}}(x) \frac{A_{\text{gas-liq}}(x)}{V_{\text{liq}}(x)} = \frac{2}{d} \left( \frac{D_A u(x)}{L_{\text{gas}}(x) + L_{\text{liq}}(x)} \right)^{0.5} \left( \frac{L_{\text{gas}}(x)}{L_{\text{gas}}(x) + L_{\text{liq}}(x)} \right)^{0.3} \quad (16)$$

$$\frac{h_{\text{wall-liq}}(x)d}{\kappa_{\text{liq}}} = 24.2 + 0.54 \left( \frac{\rho_{\text{liq}}(x)u(x)dc_{p,\text{liq}}}{\kappa_{\text{liq}}} \right)^{0.45} \left( \frac{L_{\text{liq}}(x)}{d} \right)^{-1.34} \quad (17)$$

In the case study, the objective function was set to be the production rate per single channel volume. The main design and operation conditions are as shown in Table 1, and the selectivity of the target product was set to be 0.55 or more as a design specification. Using MATLAB, the design and operation variables that maximize the objective function were derived on the basis of the model-based design method described in Section 2. The results are shown in Table 1. Figure 3 shows the temperature profile under the optimum design conditions. Table 1 shows that the derived  $W$ ,  $T_c$ , and  $H_w$  are the lower limit values, which indicates that the progress of side reaction is suppressed by improving the heat removal performance of the channel in which the reaction proceeds. In this design, it is believed that  $Q_{\text{gas}}$  and  $Q_{\text{liq}}$  were derived to increase the production rate per single channel volume, which is the objective function, while taking into account the removal of reaction heat that affects product selectivity. This is the result of the first-stage design. In the second stage, the SFMR was

designed, i.e., how many channels should be parallelized and how many reaction plates should be stacked were derived. Under the constraints of a total production volume of 100 g/day or more, a reaction plate aspect ratio  $K = 3$  to 4, and a design parameter for channel spacing  $W_w = 5.0$  mm (see Figure 4), an SFMR with five parallel channels and six stacked reaction plates was derived that could achieve 117 g/day, where the width of one reaction plate  $W_p = 5.3$  cm, its length in the flow direction  $KW_p = 16.2$  cm, and its aspect ratio  $K = 3.1$ . The results of this case study, as shown in Table 2, suggested that the developed model can be used sufficiently as an alternative to CFD for SFMR design.

Table 1: Upper and lower limits and optimal values for design and operating conditions

Design and operating conditions	Lower limits	Upper limits	Optimal values
Channel width $W$ [mm]	0.50	1.0	0.50
Channel length $L$ [m]	0.10	4.0	0.81
Coolant temperature $T_c$ [K]	278	303	278
Channel distance $H_w$ [mm]	3.0	5.0	3.0
Gas feed flowrate $Q_{gas}$ [m <sup>3</sup> /s]	2.5E-8	2.5E-7	9.2E-8
Liquid feed flowrate $Q_{liq}$ [m <sup>3</sup> /s]	2.5E-8	2.5E-7	1.2E-7
Objective function $Obj$ [mol/m <sup>3</sup> /s]	-	-	9.72

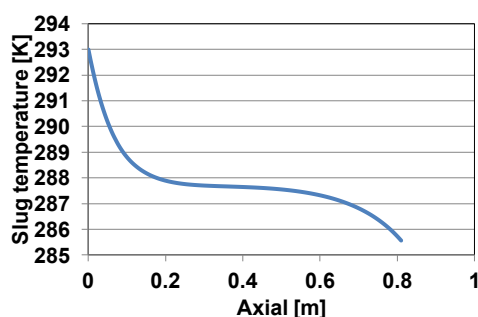


Figure 3: Slug temperature profile

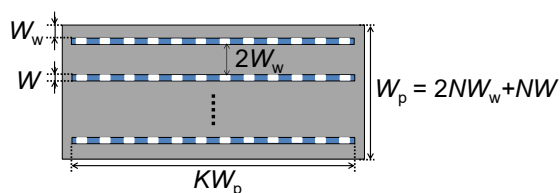


Figure 4: Size constraints on a reaction plate

Table 2: Comparison of the proposed model and the CFD model for SFMR design

Model	Detail	Calculation load	Availability for optimal design
CFD	High	Heavy	Unrealistic
The developed model	High enough	Low	High

#### 4. Conclusions

A simple model capable of expressing gas-liquid slug flow with the heat and mass transfer and chemical reactions in a microchannel was constructed, and a two-stage design method for slug flow microreactors using the constructed model was proposed. The usefulness of them was confirmed through a numerical case study on parallel exothermic reactions. The proposed model and design method can be applied to reaction systems different from the case study. The validity of the design results will be verified by comparison with experimental data in the future.

## Nomenclature

$A$ – cross-sectional area of channel, $m^2$	$r$ – reaction rate, $mol/m^3/s$
$A_{gas-liq}$ – gas-liquid interfacial area, $m^2$	$t_1$ – time, s
$C$ – concentration, $mol/m^3$	$t_2$ – time, s
$Ca$ – Capillary number, -	$u$ – slug velocity, m/s
$c_{p,liq}$ – specific heat capacity, J/kg/K	$V_{liq}$ – liquid slug volume, $m^3$
$d$ – channel diameter, m	$\alpha$ – void fraction, -
$k_{liq}$ – mass transfer coefficient, m/s	$\kappa_{liq}$ – thermal conductivity of liquid, W/m/K
$L_{gas}$ – gas slug length, m	$\nu$ – stoichiometric coefficient, -
$L_{liq}$ – liquid slug length, m	$\rho_{liq}$ – liquid density, $g/m^3$
$F$ – flow rate, mol/s	$\Delta H_i$ – standard reaction enthalpy, kJ/mol
$N_A$ – mass transfer rate, $mol/m^2/s$	
$Re$ – Reynolds number, -	

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