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Design of microfluidic slug mixing based on the correlation between a dimensionless mixing rate and a modified Peclet number

Wiroon Tanthapanichakoon, Kazuo Matsuyama, Nobuaki Aoki, Kazuhiro Mae*

Department of Chemical Engineering, Graduate School of Engineering, Kyoto University, Kyoto-daigaku Katsura, Nishikyo-ku, Kyoto 615-8510, Japan

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Abstract

A method to design mixing in microfluidic slugs using a modified Peclet number, $Pe^* = U_s d_s^2/lD$, has been reported by the authors, but it was limited to mixing at constant diffusivity D. This paper reports an improved method to quantitatively determine the effect of D on a relation between Pe^* and mixing rates. Computational fluid dynamics (CFD) simulations were used for the investigation. We introduce D into the mixing rate term in the relation between Pe^* and mixing rates, and found that (mixing rate $\times d_s^2/D$) becomes a function of only Pe^*. Thus, slug mixing can be designed using the new dimensionless number, (mixing rate $\times d_s^2/D$), and Pe^*. This allows us to use mixing rate data at any value of D to estimate mixing rates at another value of D. Though Pe^* includes effects of D, l, d_s, and U_s, effects of initial arrangements of reactants inside a slug and slug cross-sectional shapes are not considered. Thus, the relations between (mixing rate $\times d_s^2/D$) and Pe^* (referred as Pe^* correlation) are quantitatively determined to cover the effects of these parameters. Furthermore, we used the Pe^* correlation to show theoretically that channel contraction is an effective microfluidic operation to enhance mixing in slugs.

Keywords: Multiphase flow; Laminar flow; CFD simulation; Mixing; Modified Peclet number; Mixing enhancement by channel contraction

1. Introduction

Use of liquid slugs in microfluidic systems simultaneously allows rapid mixing and narrow residence time distribution under the laminar flow regime (Song and Ismagilov, 2003; Zheng et al., 2003). Slug mixing is generally developed for immiscible liquid–liquid systems. Main characteristics of the microfluidic liquid slug are that the slug internal fluid circulates very effectively owing to high shearing interactions with the channel wall. The ordered laminar flow in microchannels also allows the precise control of liquid slug transport. Nanoparticle synthesis (Shestopalov et al., 2004) and reaction kinetic measurement (Song and Ismagilov, 2003) are some of the applications of microfluidic liquid slugs. Mixing performance is an important factor determining product yields and selectivities (Ehlers et al., 2000; Aoki et al., 2004; Yoshida et al., 2005). Thus, methods to determine the slug length, diameter and velocity to achieve desired mixing rates are crucial. This is because mixing rates need to be adjusted according to the diffusivity of solution and reaction kinetics in order to achieve efficient reaction operations and accurate reaction rate measurements.

In our previous paper (Tanthapanichakoon et al., 2006), Pe^* was proposed for designing mixing in slugs at a constant diffusivity D. However, when D changes, the relation at the old value of D between (mixing rate $\times d_s^2$) and Pe^* becomes invalid, and we need to re-establish the relation between mixing rates and Pe^*. D in slug operations depends on many factors such as reaction phases, reactants, and solvents, etc. In this paper, the method is improved to introduce D into the mixing rate term in the relation between mixing rate and Pe^*. This leads to a new dimensionless number, (mixing rate $\times d_s^2/D$). The new dimensionless number and Pe^* allow us to use mixing rate data at any value of D to estimate mixing rates at another value of D. Though Pe^* includes effects of slug length, diameter, velocity, and diffusivity on mixing rates, effects of initial arrangement of reactants inside a slug and slug cross-sectional shape are not considered. Thus, the relation between mixing rates and Pe^* needs to be determined for each reactant arrangement and each slug cross-sectional shape to cover the influences of these

* Corresponding author. Tel.: +81 75 383 2668; fax: +81 75 383 2658.
E-mail address: kaz@cheme.kyoto-u.ac.jp (K. Mae).

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parameters. Furthermore, the relation between Pe^* and mixing rates was used to design a new microfluidic operation for mixing enhancement in slugs by channel contraction.

2. Simulation methods

As shown in Fig. 1, each slug was modeled as either two-dimensional (2D) or three-dimensional (3D) single-phase flow domain. Slug A is simulated using 2D simulations, which corresponds to a slug in a slit channel. Slug B is simulated using 3D simulations, which correspond to slugs in a rectangular channel with a square cross section. Straight slug edges were used to eliminate the influence of slug edge curvatures on mixing rates when varying slug lengths and diameters. In actual slug flow, the slug edge shapes are determined mainly by the properties of two liquids inside and outside the slugs. The internal circulating flow is generated by moving the slug sidewalls in the negative x-direction. The slug traveling at a velocity U_s in the positive x-direction can be simulated by moving the sidewalls in the negative direction with velocity $U_{\text{wall}} = -U_s$. In slug B, all four slug sidewalls (upper, lower, left, and right) were moved in the negative x-direction to generate an internal circulating flow. The front and back edges of the slugs were set as the non-moving walls. The simulations correspond to the mixing inside a slug flowing through a straight channel. We assumed that a slug fully occupies the channel cross section and its cross-sectional shape is determined by the channel cross-sectional shape.

A commercial CFD code, Fluent 6.2.16, was used in the simulations. The laminar flow model was used. This code solves

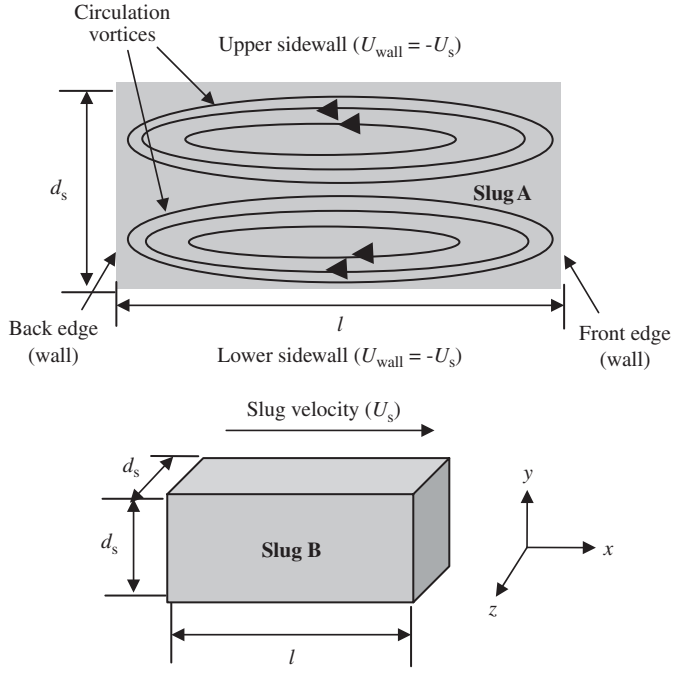

Fig. 1. Slugs as single-phase flow domains where internal circulating flow was generated by moving the slug sidewalls in the negative x-direction: two types of slugs simulated in the simulations.

Table 1
The number of meshes and the time step size

Slug type	The number of meshes
A	15,000
B	54,000 ($d_s \leqslant 250\,\mu$m, $U_s \leqslant 100$ mm/s)
	96,000 ($d_s > 250\,\mu$m, $U_s > 100$ mm/s)
Reactant arrangement	Time step size (s)
Axial	5×10^{-5}
Radial	1×10^{-4}

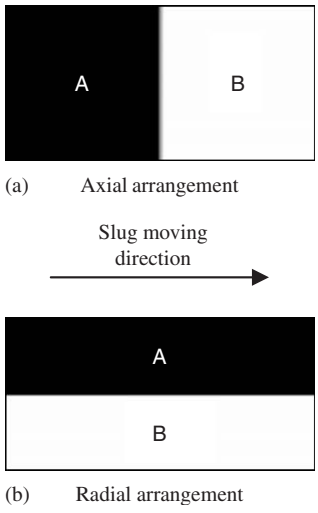

Fig. 2. Two initial arrangements of reactants investigated in the simulations: (a) axial arrangement, (b) radial arrangement.

mass, momentum, and energy conservation equations by the control volume method. The third-order MUSCL scheme was used to solve mass and momentum conservation equations because this scheme is superior to the other schemes available in Fluent for the smallest numerical diffusion. Mixing of reactants in the slug over flow time was calculated using unsteady simulations. Table 1 summarizes the number of meshes and the time step sizes used in the simulations. Details of the simulation method were described in our previous paper (Tanthapanichakoon et al., 2006).

We considered the mixing of two reactants (A and B) where each reactant has an equal density and volume in the slug. The density and viscosity were set at 998.2 kg/m^3 and 0.001 Pa s, respectively. Viscosity of fluids affects mixing performance. Since the shear force to circulate fluid in the slug increases with viscosity, mixing for high viscosity reactants is supposed to be slow especially until the internal circulation flow becomes steady. We investigated the two initial arrangements of reactants, axial arrangement (Fig. 2a) and radial arrangement (Fig. 2b). Table 2 summarizes all the simulation conditions. The simulation conditions were selected to cover Pe^* range of 125–25,000. The diffusion coefficient D was varied to 10^{-8} and 10^{-9} m^2/s to confirm effects of D on the relation between Pe^* and mixing rates. The diffusion coefficient of 10^{-8} m^2/s was used in the other simulations to establish