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# Three-dimensional simulation of mixing performance inside droplets in micro-channels by Lattice Boltzmann method

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## HIGHLIGHTS

- Developed 3D LBM models for mixing process in droplets with experimental validation.
- Verified influence of micro-channel geometry on droplet generation (size and shape).
- Demonstrated strong dependence of mixing in droplets on initial generation process.
- Revealed sensitivity of recirculation effect to the mixing inside a micro-droplet.
- Proposed potential solutions to precisely control mixing process in micro-droplets.

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## ABSTRACT

Three-dimensional multiphase/multicomponent Lattice Boltzmann model (LBM) was developed to simulate the complex hydrodynamics inside micro-droplets generated in micro-channels. Special attention was paid to the ways to generate droplets, which determined the initial liquid concentration and velocity distribution inside the droplets and accordingly made a remarkable contribution to the following mixing performance along the flow development in the micro-channels. Flow characteristics and mixing performance under different geometries and how the differences in configuration influenced the initial status of droplets were illustrated by LBM simulations. The results showed that the effective way to intensify the mixing process was to break up the bilateral symmetric fluid distribution as well as to take full advantage of the internal recirculation inside the droplet during its generation process. It is expected that general guidelines to manipulate the droplet formation and its inside mixing can be brought up through this work. Furthermore, flexible solutions can be simply provided to control the mixing process more precisely by just changing the key geometry of the microchannel.

## 1. Introduction

Droplet-based microfluidics has drawn more and more attention in both academics and industry. The small channel dimension results in a high surface to volume ratio, and micro droplets in such channels are able to provide a more confined space, which can greatly intensify the external and internal transport phenomena [1]. In particular, the unique characteristics of micro-droplets enable the well-controlled reactions by compartmentalizing reactions inside the droplets with about nano-liter to micro-liter volumes to provide rapid mixing of the reagents [2]. At present, droplet-based microfluidics has been successfully applied to a wide variety of areas, such as enzymatic kinetics [3,4], protein crystallization [5,6] and chemical or biological synthesis [7,8].

Zhao et al. [9] and Dessimoza et al. [10] performed detailed researches on the liquid–liquid two-phase flow patterns in micro-channels and classified them into mono-dispersed droplet flow, droplet population flow, slug flow, parallel flow, and annular flow according to the process conditions such as wetting properties, flow velocities, fluid viscosities and geometrical features. Droplet formation mechanism of liquid–liquid systems has also been extensively studied in recent years [11,12]. In this kind of system, droplets or slugs are generated by the combination of two immiscible phases, where the spontaneous generation process is a result of the competition of the viscosity force and the interfacial tension at the liquid–liquid interface or Laplace additional pressure caused by the interfacial tension. The continuous phase has better wettability to the channel and spreads out well over the channel wall. Jovan et al. [12] investigated the hydrodynamics and the pressure drop of liquid–liquid slug flow in round micro-capillaries, and developed the stagnant and moving film models. Typically, there are three types of micro-channel flow geometries to generate droplets, namely cross-flowing streams, flow-focusing streams and co-flowing streams [13], which would be discussed in this work.

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**Nomenclature**

| | |
|---|---|
| $C$ | normalized concentration of the species |
| $Ca$ | Capillary number |
| $f$ | probability density functions of fluid $\sigma$ |
| $F$ | interaction force, lattice units |
| $G$ | interaction strength, lattice units |
| $IOS$ | intensity of segregation |
| $Q$ | volumetric flow rate, $m^3/s$ (in experiment) and lattice units (in simulation) |
| $p$ | pressure, Pa (in experiment) and lattice units (in simulation) |
| $R$ | the radius of the droplet |
| $u$ | velocity, m/s (in experiment) and lattice units (in simulation) |
| $w$ | channel width, m (in experiment) and lattice units (in simulation) |
| $h$ | channel height, μm (in experiment) and lattice units (in simulation) |
| $l$ | flow distance along the flow direction, μm (in experiment) and lattice units (in simulation) |
| $e$ | the direction vector in the simulation |
| *Greek letters* | |
| $\gamma$ | interfacial tension, N/m (in experiment) and lattice units (in simulation) |
| $\mu$ | dynamic viscosity, Pa s (in experiment) and lattice units (in simulation) |
| $v$ | kinematics viscosity, $m^2/s$ (in experiment) and lattice units (in simulation) |
| $\rho$ | density, $kg/m^3$ (in experiment) and lattice units (in simulation) |
| $\sigma$ | index for fluid components |
| $\tau$ | relaxation time |
| $\omega$ | weighting coefficient |
| $\psi$ | interaction potential |
| $\Omega$ | collision term |

Rapid mixing is of great significance for most reactions. However, the fluid in micro-channel is always kept laminar flow under low *Re* numbers and hardly changed into turbulence so that the mixing is dominated by molecular diffusion. A variety of methods have been developed to intensify the mixing in the microfludics. Novel channel geometry design is one category of them. Stroock et al. [14] used bas-relief structures on the bottom of the channel to introduce velocity fluctuations in order to better homogenize fluids. This idea of chaotic advection was extensively developed by Song et al. [15]. They changed the straight main channel into a winding shape. Flowing along the channel, the interface between the two halves of the plug inside the droplet is reoriented by the winding channel and is stretched and folded by internal recirculation, which largely accelerates the molecule diffusion.

Besides changing the structure of the main channel to intensify the mixing process, Tice et al. [16] pointed out that in straight microchannels the initial distribution of unmixed fluids within the plug has a great influence on the mixing behavior, and this initial distribution is governed by the generation process of the plug. Their experiments indicated that when the two fluids to be mixed are initially located separately in the front and back halves of the plug, the recirculation results in a much more efficient mixing along the channel than in the case that two fluids are symmetrically distributed in the left and right halves. Therefore, understanding the flow phenomenon inside a droplet during its generation process could help to find effective methods to intensify the mixing.

In our previous work, three-dimensional simulation based on a multiphase, multi-components LBM approach has been established to investigate the basic principles such as flow patterns, droplet sizes and generation frequencies in the liquid–liquid two-phase flow in microchannels [17]. This work aims to study the influence of different droplet generating processes on the mixing behaviors inside the droplets by LBM simulation. Special attention is paid to reveal the mixing intensification during the initial generation of droplets under various micro-device designs.

## 2. Mathematical model and experimental setup

### 2.1. Introduction to LBM

LBM is developed from the discretized fluid model Lattice Gas Automata (LGA) [18,19]. The computation model is based on the Lattice Boltzmann equation, which is a promising new method for fluid simulations owing to its excellent numerical stability and constitutive versatility. The basic governing equations could be directly derived from the continuous Boltzmann equation discretized in both time and space dimensions [20]. Several LBE models in both two- and three-dimensional space have been developed with the single relaxation time BGK approximation to study hydrodynamics of fluids. Compared with the conventional Navier–Stokes equations, LBM considers fluids as a collection of pseudo-particles with certain possible spatial positions, which could be described by a velocity distribution function. And the description of microscopic momentum is simplified from continuous to discretized. The position of these particles is confined to the nodes of the lattices where the interaction between particles like collision and streaming takes place [21]. In contrast to the conventional CFD method, LBM is positioned between the continuum level (described by Navier–Stokes equations) and the microscopic (molecular) level, which is an ideal approach for meso-scale and scale-bridging simulations such as the fluid flow in micro-channels, especially successful in fluid flow applications involving interfacial dynamics and complex boundaries. The commonly used mesh types of Lattice Boltzmann method are D2Q9 (i.e., 2-dimension 9-direction model) and D3Q19 (i.e., 3-dimension 19-direction model). Most of the simulation results of this work are based on the D3Q19 model.

At present, LBM has been expanded to describe complex reaction-free multi-phase flows with the corresponding approaches as the chromodynamic (color) [22,23], the pseudo-potential [24,25] and the free-energy [26,27] models. Among all the approaches, the pseudo-potential model is further investigated in microfluidic devices. This model introduces a non-local interaction force between different kind of particles to describe the non-ideal fluidics developed by Shan and Chen [24] and Shan and Doolen [28,29].

### 2.2. Multi-component Lattice Boltzmann equations

In this work, the multi-component Lattice Boltzmann model proposed by Shan and Chen [24] was adopted to simulate the droplet generation process in multiple phase flows inside micro-channels. The main equations of the multiphase LBM model are listed below:

$$f_i^\sigma(x + e_i\delta t, t + \delta t) = f_i^\sigma(x, t) + \Omega_i^\sigma(x, t) \tag{1}$$