Chemical Engineering Science 146 (2016) 126–134

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Numerical study of double emulsion formation in microchannels by a ternary Lattice Boltzmann method

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HIGHLIGHTS

- Established a ternary Lattice Boltzmann method based on color-gradient model for multiphase flow simulations.
- Simulated contacting conditions of two immiscible droplets in another fluid by ternary color-gradient model.
- Predicted the flow regimes for double emulsion formation in microchannels.
- Presented the effects of viscosities of middle and inner fluids on the double emulsion formation.

ARTICLE INFO

Article history:
Received 5 July 2015
Received in revised form
13 January 2016
Accepted 24 February 2016
Available online 2 March 2016

Keywords:
Microfluidics
Droplet
Double emulsion
Ternary Lattice Boltzmann method
Color gradient method

ABSTRACT

A ternary LBM model was successfully established to numerically investigate the double emulsion formation process in a flow-focusing microfluidic device. The model was validated by comparing the simulated interfacial phenomena in three-phase systems with the theoretical solutions, where physical properties of each fluid can be independently defined in the newly established LB model. Meanwhile, the numerical simulations showed good agreement of the double emulsion formation in different flow regimes with the experimental data in the literature. Especially for the three representative flow regimes, i.e., dripping, jetting and middle jet containing monodispersed inner drop, the model predictions show that the emulsion size follows scaling law in dripping regime. Effect of the viscosities of middle and inner fluids on the occurrence of flow regimes was further discussed, indicating that smaller viscosity of either middle or inner fluid should be beneficial to the double emulsion production.

1. Introduction

Emulsion is a system in which two or more immiscible fluids disperse in another continuous liquid phase (Pal, 2011). Specifically, double emulsion, usually called “droplet in droplet” (Nisisako et al., 2005), is known as a small droplet completely engulfed by another emulsion. Due to its unique core–shell structure, double emulsion has great potential in the applications related with drug delivery (Zhao, 2013), functional material (Chen et al., 2009; Fernández-Nieves et al., 2007; Kim and Kim, 2014; Lorenceau et al., 2005; Peng et al., 2008; Zhang et al., 2013) and food sciences (Douaire and Norton, 2013), or is taken as a micro-reactor to implement mixing or reaction processes precisely (Duncanson et al., 2012). Thus it is important to develop appropriate micro-devices to fabricate highly monodispersed and uniform double emulsions. Since last century, microfluidic technology has been fast advanced to enable the controlled formation of nano- or micro-particles (Lone and Cheong, 2014), multi-cored capsules (Chu et al., 2007) or vesicles (Huebner et al., 2008) in tiny volume liquid. Particularly, in a variety of micro-systems such as T-junctions (Okushima et al., 2004), co-flowing (Cramer et al., 2004) and flow-focusing (Anna et al., 2003) devices to disperse liquids and form single emulsions uniform and monodispersed double emulsions, especially those containing high viscosity or non-Newtonian fluid, can be continuously produced (Abate et al., 2011). The means can be categorized into two-step method (Chang et al., 2009; Chu et al., 2007; Okushima et al., 2004) and one-step method (Chen et al., 2012; Kim et al., 2013; Shao et al., 2013; Utada et al., 2005). The former one is: forming the inner droplets first, and then encapsulating the inner droplets in a second emulsification step; the latter one is: using a single step to fabricate double emulsions or even multiple emulsions. It should be noted that by controlling the inner droplet formation position through slightly changing the device geometry, the same channel is able to implement both the two-step method and the one-step method (Chang et al., 2009; Shao et al., 2013).

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http://dx.doi.org/10.1016/j.ces.2016.02.036
0009-2509/

Many researchers have provided valuable empirical knowledge of the double emulsion formation in microchannels in their extensive studies (Zhao, 2013). For example, two distinctive flow mechanisms of double emulsion formation are illustrated as dripping and jetting modes (Utada et al., 2005), and the corresponding flow behaviors are accordant with the flow conditions (Kim et al., 2013). Several empirical equations summarized from experiments (Chang et al., 2009; Shao et al., 2013; Utada et al., 2005) have been put forward to predict the size of double emulsions, where Capillary number (Ca=viscosity force/interface tension) and Weber number (We=inertia force/interface tension) are two of the most important dimensionless numbers to characterize the system for emulsion productions. Double emulsions are formed by shear-rupturing mechanism at appropriate Capillary number of outer flow. Shearing force is brought up by a competition between viscous force and interface tension at the local flow field (Nisisako et al., 2005; Utada et al., 2007).

However, many factors, such as material and geometry of the micro-channel, flow conditions and physical properties of fluids, could influence the emulsion formation process. Parametric studies of such processes play a key role in distinguishing the flow condition and adjusting the double emulsion system. The theoretical description of double emulsion formation systems, to our best knowledge, is still far from being well established. Zhou et al. (2006) have shown the double emulsion formation in a flow-focusing device and summarized the effect of viscous and Capillary number in the jetting regime by expanding finite element framework. Park and Anderson (2012) applied a ternary Navier–Stokes/Cahn–Hilliard model with consideration of thermodynamics to predict multiple drops situation. They indicated that the free energy parameters should be properly set up, and the model was limited to low viscosity ratio. Chen et al. (2015) successfully used the ternary VOF method to simulate double emulsion formation in both dripping and jetting regimes, where numerical studies have been devoted to carefully examine the effects of viscosity ratio, interface tension and flow conditions. To sum-up, some efforts have been made to understand the fundamental fluid dynamics in flow-focusing devices captured by Utada et al. (2005). However, the numerical method in the literature could not reproduce important flow regimes such as the middle jet containing monodispersed inner drops in the same 2D devices (Kim et al., 2013). These motivate us to develop a theoretical method to describe the dynamics of double emulsion formation in a general ternary system.

The present work aims to establish a ternary Lattice Boltzmann method (LBM) for microfluidics simulation with double emulsion formation. LBM, a promising method based on Boltzmann equation and kinetic theory, has inherent advantages for tracking the deformable interfaces by incorporation of microscopic interactions. Several model frameworks (Wörner, 2012) have been established for immiscible multiphase flows, which can be classified as color-gradient method (Grunau et al., 1993; Latva-Kokko and Rothman, 2005), pseudo-potential method (Shan and Chen, 1993) and free energy model (Briant and Yeomans, 2004; Swift et al., 1995). These methods have been well applied to simulate multiphase flow behaviors in micro-channels (Wang et al., 2011). Geometry effect of droplet generation was studied by using Shan-Chen model (Wang et al., 2011; Zhao et al., 2012). Liu and Zhang (2009) and Van der Graaf et al. (2006) showed that droplet detachment behavior in T-junctions is promoted with the increase of Ca of outer fluid and the regime transition depends on the critical Ca number by the free-energy model. As for the color-gradient method, great effort has been made to model two-phase flows by the pioneers (Grunau et al., 1993; Latva-Kokko and Rothman, 2005; Leclaire et al., 2013a, 2013b; Lishchuk et al., 2003; Reis and Phillips, 2007), where the computational load is greatly reduced and negative probability distribution problem has been solved. For example, by using the color-gradient model, Liu and Zhang (2011) demonstrated that transition of three flow regimes (i.e. dripping, jetting and parallel flows) is dependent on the Capillary number and flow rates of two immiscible fluids at a cross-junction device. It should be mentioned that the model can be adopted for describing multiphase systems with large density and/or viscosity ratio, low interface tension, etc. However, little work on a three-phase or multi-phase model for microfluidic application was reported so far. Dupin et al. (2006) re-defined the fluid phase color gradient for each interface with improved capability for N-phase simulation. Detailed analysis was presented by Spencer et al. (2010), in which Young–Laplace Law was recovered and multiphase droplet contact behavior was further studied. Based on their work, Leclaire et al. (2013a, 2013b) developed an enhanced N-phase LB model for high density ratios, e.g., O (1000), and viscosity ratios, e.g., O (100), which were testified by using a ternary phase Poiseuille flow.

In this work, the improved ternary LB model (Leclaire et al., 2013a, 2013b) is further advanced to describe the double emulsion behaviors in micro-channels. This model couples the thermodynamics and hydrodynamic variables of each phase, in which the interface tension and viscosity of each phase in micro-channels can be defined with clear physical meaning. Firstly, we investigate the contact of droplets with the same volume under the condition of various interface tensions in order to underline the importance of interface property. Next, a way to set wetting property is to be illustrated, which corresponds to the means in experiments to control hydrophilicity or hydrophobicity property of micro-channels. After that, the one-step flow-focusing geometry, which is similar to the experiments of Utada et al. (2005), further developed by Kim et al. (2013), will be modeled to testify the predictive capability on the detailed hydrodynamics under several different flow regimes, such as jetting, dripping and middle jet containing monodispersed inner drop(s). At last, the effect of the viscosity ratio on the double emulsion formation will be further discussed.

2. Mathematical model and algorithm

The current lattice Boltzmann method for simulating multiphase flows can be shown as a simple algorithm, derived from the Boltzmann equation. Here we used the model framework established by Leclaire et al. (2013a, 2013b).

As for the multiphase color-gradient method, the discretized particle distribution function, noted as N_j^k, is adopted for describing the fluid dynamics, where j represents the discretized direction and k is for different fluids, that is, $k=o,m,i$ for three immiscible fluids. The uniform Lattice Boltzmann governing equation is:

$$N_j^k(\boldsymbol{x}+\boldsymbol{e}_j\delta t, t+\delta t) = N_j^k(\boldsymbol{x}, t)+\Omega_j^k+G_j^k \tag{1}$$

where Ω_j^k is the collision operator, combined with three sub operators (see Eq. (6)), G_j^k represents the external force, such as gravity, electric and/or magnetic force, though external force can be ignored in pressure-driven flows in micro-channels. In this work, the simulations are 2-dimensional and follow D2Q9 scheme (i.e., 2 dimension and 9 velocities), where $\boldsymbol{e}_j$ describes the lattice velocity, defined in Eq. (2), δx and δt are the lattice unit and time step, usually, with $\delta x=\delta t=1$, and the lattice sound speed c_s will be