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Three dimensional phase-field investigation of droplet formation in microfluidic flow focusing devices with experimental validation

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ABSTRACT

In this paper, the droplet formation process at a low capillary number in a flow focusing micro-channel is studied by performing a three-dimensional phase field benchmark based on the Cahn–Hilliard Navier–Stokes equations and the finite element method. Dynamic moving contact line and wetting condition are considered, and generalized Navier boundary condition (GNBC) is utilized to demonstrate the dynamic motion of the interface on wall surface. It is found that the mobility parameter plays a very critical role in the squeezing and breakup process to control the shape and size of droplets. We define the characteristic mobility M_c to represent the correct relaxation time of the interface. We also demonstrate that the characteristic mobility is associated with the physical process and should be kept as a constant as the product of the mobility tuning parameter χ and the square of interfacial thickness ε^2. This criterion is applied for different interfacial thicknesses to correctly capture the physical process of droplet formation. Moreover, the size of the droplet, the velocity of the droplet along the downstream, and the period of droplet formation are compared between the numerical and experimental results which agree with each other both qualitatively and quantitatively. The presented model and criterion can be used to predict the dynamic behavior and movement of multiphase flows.

1. Introduction

Emulsions (or micro-droplets) have a wide range of applications in food industry (Muschiolik, 2007), cosmetics (Gallarate et al., 1999), drug delivery (Yamaguchi et al., 2002), and chemical synthesis (Odian, 2004). Traditional methods of emulsion production, e.g., direct agitation of immiscible fluids, often produces a broad size distributions. Droplet microfluidic technology has shown great potential for production of highly mono-dispersed and micron-sized emulsions (Shah et al., 2008; Teh et al., 2008). Experimental study of small vesicles generation at a low Reynolds number in micro-devices was performed firstly (Nisisako et al., 2002; Thorsen et al., 2001). Systematic experimental investigation clarifies the mechanism of droplet breakup and formation process in three regimes (squeezing, dripping and jetting) in T-junction geometries (Garstecki et al., 2006). By experimental observations the capillary number, flow rate ratio and geometry are concluded as the major factors from squeezing to dripping regimes (Christopher et al., 2008; Xu et al., 2008). Besides the T-shape design, the cross-junction, co-flowing and flow focusing devices are also common configurations for producing uniform emulsions (Christopher and Anna, 2007). The scaling and mechanism of emulsification in cross-junction configuration are discussed to control the monodisperse emulsification process (Tan et al., 2008). A flow focusing design with unbounded downstream geometry is reported to investigate the droplet size in W/O emulsions by varying flow rates (Anna et al., 2003). The role of geometry and fluid properties in planar flow focusing devices is studied systematically using scaling methods to optimize the control of emulsification process (Lee et al., 2009). The emulsions in a confined flow focusing devices are reported and investigated for identifying the mechanism of breakup process (Garstecki et al., 2005) and the effects of wetting energy on boundaries (Li et al., 2007). Adding surfactant in fluids alters the surface tension and wetting conditions, and produces both monodisperse O/W (oil in water) or W/O (water in oil) emulsions (Xu et al., 2006a; 2006b).

On the other hand, numerical simulation is a powerful means not only for understanding the complex physical processes, but also for predicting and guiding the practical designs of experi-

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ments (Wörner, 2012). Numerical studies of multiphase flows in microfluidic devices are introduced both in discrete and continuous approaches (Wörner, 2012). As a typical discrete model, the Lattice Boltzmann (LB) method is suitable and efficient for modeling micro-scale binary viscous fluids by simulating the interactions among a number of lattice particles. The first three dimensional LB model of multiphase flow was formulated twenty years ago (Martys and Chen, 1996). The LB benchmarks using the free energy theory and wetting boundary conditions were implemented to study droplet formation in different configurations (Gong et al., 2010; Graaf et al., 2006; Gupta and Kumar, 2010; Liu and Zhang, 2009), to describe the flow regimes and the transitions in cross-junction structure (Liu and Zhang, 2011) and to investigate the dynamic characteristics of water droplet on a hydrophobic gas diffusion layer surface (Hao and Cheng, 2009). In contrast to discrete methods, continuous methods are usually utilized to capture the dynamic behaviors of the interface between multiphase fluids, including the volume of fluid method (VOF), the level set method (L-S) and the phase field method (P-F). Numerical simulation of droplet formation in a T-junction configuration using the VOF method is validated by comparing the experimental visual results (Sivasamy et al., 2011). The effects of uniform magnetic field on emulsification process in a flow focusing micro-channel are studied numerically using a refined L-S method (Liu et al., 2011).

The idea of P-F approach can be dated back to the ancient studies a century ago (Rayleigh, 1892; van der Waals, 1893). The free moving interface between multiple material components is considered as a continuous, but steep change of some physical material properties, for instances, density or viscosity, thus a continuous phase field variable is introduced and the interface is represented by a thin but smooth transition layer. One major advantage of this method is that the free interface can be automatically tracked without imposing any mathematical conditions (e.g. Young–Laplace junction condition (Edwards et al., 1991; Krotov and Rusanov, 1999; Probstein, 1994) in other sharp interface model) on the moving interface, thus it provides an easy treatment of topological variations at the interface.

The commonly used phase field model for two phase fluids system couples an advection-diffusion equation, which represents the evolution of the phase variable, with the Navier–Stokes equations by introducing an extra interfacial stress term induced from the chemical potential. The evolution equation for the phase variable is derived from the energetic variational of the action functional of the Landau-Ginzburg free energy which uses the flux of chemical potential to demonstrate the non-uniform phase system of binary fluids (Cahn, 1959; Cahn and Hilliard, 1958). One great advantage of the phase-field model is that it leads to well-posed nonlinearly coupled systems that satisfies thermodynamically consistent energy dissipation laws due to the energy-based variational formalism. Recently, it has been successfully employed in many fields of science and engineering and become one of the major modeling and computational tools for the study of microfluidic interfacial phenomena (cf. Boyer et al., 2010; Cahn and Hilliard, 1958; Chen and Wang, 1996; Chen et al., 2015; Du et al., 2004; Feng, 2006; Feng et al., 2007; 2016; Feng and Wise, 2012; Han and Wang, 2016; Jacqmin, 1999; Kim, 2012; Kim and Lowengrub, 2005; Liu and Shen, 2003; Lowengrub et al., 2009; Miehe et al., 2010; Nochetto et al., 2014; Shen and Yang, 2009; 2010; 2014; 2015; Spatschek et al., 2010; Wang and Wise, 2011; Wise, 2010; Yang et al., 2013b; Yue et al., 2004; Zhao et al., 2016a; 2016b; 2016c).

When the fluid-fluid interface touches a solid wall, it leads to the well known mathematical singularity if no-slip condition is used (Cox, 1986). The P-F model can overcome this singularity by introducing a diffuse-interface and demonstrate the fluid-wall interactions by using chemical energy diffusion instead of shear stress (Jacqmin, 2000). Hydrodynamic slip boundary condition is discussed and several theories are outlined in Blake (2006). The moving contact line problem is studied using phase field equations by considering sharp interface limit (Yue et al., 2010) and energy relaxation between fluid and solid wall surface (Yue and Feng, 2011). The generalized Navier boundary condition (GNBC) for dynamic moving contact line is introduced mathematically and is verified by comparing with molecular dynamics (MD) simulations (Qian, 2006; Qian et al., 2003; 2004; 2006). It is also applied to study the moving contact line on heterogeneous surfaces (Qian et al., 2005; Wang et al., 2008). A similar general form is utilized to study dynamic moving wetting line (Carlson et al., 2009). Another type of slip boundary condition for moving contact line is formulated by analyzing the balance of physical friction forces on triple-phase contact line and is also validated by MD simulations (Ren and E., 2007; 2011; Ren et al., 2010). Numerical schemes are updated to improve the stability of moving contact line models (Gao and Wang, 2012; 2014; Shen et al., 2015). The phase-field method has been employed to study various multiphase problems, including droplet impact on homogeneous surfaces (Khatavkar et al., 2007a; 2007b), droplet spreading on partially wetting substrate (Gao and Feng, 2011), impingement and spreading process of a micro-droplet (Lim and Lam, 2014), electrohydrodynamic multiphase flow (Lin et al., 2012; Yang et al., 2013a), as well as droplet formation process in a T-junction configuration (De Menech, 2006; De Menech et al., 2008).

Most of the existing works on phase field models are based on theoretical and mathematical study, which is important and can be further validated by experimental results. In this paper, we use the P-F model with the finite element method to study the droplet formation process in a 3D microfluidic flow focusing device and compare the results with the corresponding lab experiment results. Our aim is to clarify the physical process of droplet formation, and identify the role of energy diffusion and the relationship between time relaxation parameter and diffuse interface based on physical and mathematical scaling. Through systematic numerical studies, we investigate the effect and physical meaning of several characteristic model terms and parameters on the droplet and formation process, and derive a criterion for the parameter selection in order to consistently and correctly capture the physical phenomenon.

The rest of the paper is organized as follows. In §**2**, we describe the model system and illustrate the phase field equations coupled with the generalized Navier boundary condition (GNBC). Then we describe the experimental details, including the configuration of microdevice, and fluids, in §**3**. The implementation and verification of the P-F model will be presented, and important findings based on the systematic numerical investigation in comparison with the experimental data will be emphasized in §**4**.

2. Mathematical formulation and description of the model system

2.1. Description of the model system

The three dimensional geometry of a flow focusing configuration in micro-device is shown in Fig. 1. The width of this microchannel system is defined as L which is also considered as the characteristic length of this system. There are one main inlet and two side inlets. Each of inlets has a square cross-section with the width L. In the downstream there is only one outlet and its height is $1.6L$. Two immiscible fluids are injected into the microfluidic system. The disperse phase is injected into the channel through the main inlet, and the continuous phase is injected through the two side inlets. The flows at the inlets are assumed as fully developed laminar flows with the flow rates of disperse phase Q_d and continuous phase Q_c. We assume the continuous phase has a perfect wetting condition on the channel wall and the disperse phase has