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 be-

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tween 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.

Keywords: Phase-field modeling, Droplet formation, Microfluidic flow focusing, Droplet breakup

1. Introduction

Emulsions (or micro-droplets) have a wide range of applications in food industry [1], cosmetics [2], drug delivery [3], and chemical synthesis [4]. Traditional methods of emulsion production, e.g., direct agitation of immiscible
5 fluids, often produces a broad size distributions. Droplet microfluidic technology has shown great potential for production of highly mono-dispersed and micron-sized emulsions [5, 6]. Experimental study of small vesicles generation at a low Reynolds number in micro-devices was performed firstly [7, 8]. Systematic experimental investigation clarifies the mechanism of droplet breakup and formation process in three regimes (squeezing, dripping and jetting) in T-junction
10 geometries [9]. By experimental observations the capillary number, flow rate ratio and geometry are concluded as the major factors from squeezing to dripping regimes [10, 11]. Besides the T-shape design, the cross-junction, co-flowing and flow focusing devices are also common configurations for producing uniform
15 emulsions [12]. The scaling and mechanism of emulsification in cross-junction configuration are discussed to control the monodisperse emulsification process [13]. A flow focusing design with unbounded downstream geometry is reported to investigate the droplet size in W/O emulsions by varying flow rates [14]. The role of geometry and fluid properties in planar flow focusing devices is studied
20 systematically using scaling methods to optimize the control of emulsification process [15]. The emulsions in a confined flow focusing devices are reported and investigated for identifying the mechanism of breakup process [16] and the effects of wetting energy on boundaries [17]. Adding surfactant in fluids alters the surface tension and wetting conditions, and produces both monodisperse

25 O/W (oil in water) or W/O (water in oil) emulsions [18, 19].

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 experiments [20]. Numerical studies of multiphase flows in microfluidic devices are introduced both in discrete and continuous approaches [20]. 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 [21]. The LB benchmarks using the free energy theory and wetting boundary conditions were implemented to study droplet formation in different configurations [22, 23, 24, 25], to describe the flow regimes and the transitions in cross-junction structure [26] and to investigate the dynamic characteristics of water droplet on a hydrophobic gas diffusion layer surface [27]. 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 [28]. The effects of uniform magnetic field on emulsification process in a flow focusing micro-channel are studied numerically using a refined L-S method [29].

The idea of P-F approach can be dated back to the ancient studies a century ago [30, 31]. 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 [32, 33, 34] 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 [35, 36]. 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. [35, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64]).

When the fluid-fluid interface touches a solid wall, it leads to the well known mathematical singularity if no-slip condition is used [65]. 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 [66]. Hydrodynamic slip boundary condition is discussed and several theories are outlined in [67]. The moving contact line problem is studied using phase field equations by considering sharp interface limit [68] and energy relaxation between fluid and solid wall surface [69]. The generalized Navier boundary condition (GNBC) for dynamic moving contact line is introduced mathematically and is verified by comparing with molecular dynamics (MD) simulations [70, 71, 72, 73]. It is also applied to study the moving contact line on heterogeneous surfaces [74, 75]. A similar general form is utilized to study dynamic moving wetting line [76]. 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 [77, 78, 79]. Numerical schemes are updated to improve the stability of moving contact line models [80, 81, 82]. The phase-field method has been employed to study var-

ious multiphase problems, including droplet impact on homogeneous surfaces [83, 84], droplet spreading on partially wetting substrate [85], impingement and spreading process of a micro-droplet [86], electrohydrodynamic multiphase flow [87, 88], as well as droplet formation process in a T-junction configuration [89, 90].

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 micro-channel system is defined as L which is also considered as the characteristic length of this system. There are one

Figure 1: 3D schematic of the flow focusing microfluidic channel. The characteristic length is used as L to show the geometric size of the model. There are one main inlet for the disperse phase (Q_d) and two side inlets for the continuous phase (Q_c).

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 non-wetting condition on the channel wall. These boundary conditions match the wetting properties used in our experiments and will be discussed in §2.3. The relevant physical properties of the two immiscible fluids are: viscosities μ_c and μ_d , densities ρ_c and ρ_d and surface tension σ . The droplet is formed by the squeezing force on the disperse phase from the continuous phase. The normalized droplet volume $V^* = V_d/L^3$ is a physical dimensionless parameter and depends on the following dimensionless numbers: the Reynolds number $Re = \rho_c Q_c / L \mu_c$, and the Capillary number $Ca = \mu_c Q_c / L^2 \sigma$. The flow rate ratio between the two binary fluids $Q = Q_d / Q_c$ is also an important factor in this droplet formation regime.

In our study, we use two specific immiscible fluids with given physical properties to numerically investigate the emulsification process. These two immiscible fluids will also be used in our experiments. Our goal is not to discuss the flow

transition from shear stress dominated regime to surface tension dominated regime, but to investigate the mechanism of droplet formation in the squeezing regime by using phase field equations. In our model, the value of Capillary number is small enough ($Ca \approx 0.016$) so that the flow regime can be confirmed
140 as surface tension dominated. The Capillary number can be considered as a constant since the flow rate of continuous phase is fixed. Only the flow rate of disperse phase is varied in order to control the size of droplet. Thus the effect of capillary number variation on the droplet size can be neglected.

2.2. Mathematical formulation of phase field equations

145 In the phase field models [54], the physical sharp limit interface is replaced by a diffuse-interface which has a non-trivial thickness in which the physical properties and the interfacial tension force are smoothly and continuously distributed. The governing equation we choose for the phase variable is known as the Cahn-Hilliard equation which can be seen as a kind of advection-convection
150 equation [38]:

$$\begin{aligned} \frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi &= M \nabla^2 G, \\ G &= -K \nabla^2 \phi + f'(\phi), \end{aligned} \tag{1}$$

where ϕ is the phase variable and its value could be varying on $[-1, 1]$. The phase variable means the pure continuous phase when $\phi = 1$ and the pure disperse phase when $\phi = -1$. The nonlinear term in the chemical potential is $f'(\phi) = u\phi^3 - r\phi$. M is called the phenomenological mobility parameter. r, u are the computational parameters obtained from MD simulations and have a relationship of $r = u = \frac{K}{\varepsilon^2}$, where K, ε are the mixing energy density and interfacial thickness, respectively [70]. G is the chemical potential representing the change rate of free energy at the interface and it is calculated as functional derivative of the free energy $G = \delta F / \delta \phi$, where $F(\phi)$ is the free energy. The first term in G represents the interfacial energy, and the second term represents

the bulk free energy. Then the free energy can be expressed as:

$$F(\phi) = \int_{\Omega} \left[\frac{1}{2} K (\nabla \phi)^2 + f(\phi) \right] d\mathbf{r}. \quad (2)$$

The mixing energy density K is related to the surface tension coefficient σ and the interfacial thickness ε as follows,

$$\sigma = \frac{2\sqrt{2}}{3} \frac{K}{\varepsilon}. \quad (3)$$

This formula clearly shows that the surface tension force is proportional to the energy density at the interface between two immiscible fluids. It also indicates the connection between the energy density and the sharp interface limit. As the interfacial thickness ε shrinks to zero, the energy density K should decrease to
155 zero in order to keep the surface tension as a constant.

Then the Cahn-Hilliard equation transforms into:

$$\begin{aligned} \frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi &= \frac{MK}{\varepsilon^2} \nabla^2 \Psi, \\ \Psi &= -\varepsilon^2 \nabla^2 \phi + \phi (\phi^2 - 1), \end{aligned} \quad (4)$$

where Ψ is an auxiliary variable to separate the fourth order equation into two second order equations. The mobility parameter M determines the relaxation time of interface and the timescale of diffusion in Cahn-Hilliard equation [40].
160 This parameter is proportional to square of the interfacial thickness $M = \chi \varepsilon^2$, where χ is the mobility tuning parameter [86].

The fluid flow is modeled using the Navier-Stokes equation and the equation of continuity:

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right] = -\nabla p + \nabla \cdot \mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T) + G \nabla \phi + \mathbf{f}_{ext}, \quad (5)$$

$$\nabla \cdot \mathbf{u} = 0, \quad (6)$$

where $\mathbf{u}$ is the velocity vector, p is the pressure and $\mathbf{f}_{ext}$ is the external body force exerting on the fluids such as gravity or buoyancy. In our study the

two-phase flows are incompressible in the microfluidic channel and there is no other external force field applied in this system, thus the external body force can be neglected safely. The fluid properties of the two phases are different and vary with the phase variable ϕ especially at the sharp limit interface. The density in disperse phase is ρ_d corresponding to $\phi = -1$ and the density in continuous phase is ρ_c corresponding to $\phi = 1$. The viscosity in disperse phase is μ_d corresponding to $\phi = -1$ and the viscosity in continuous phase is μ_c corresponding to $\phi = 1$. Therefore the density we used in our computation can be expressed as $\rho = \frac{1}{2}[(1 - \phi) \cdot \rho_d + (1 + \phi) \cdot \rho_c]$ and the viscosity in our computation is expressed as $\mu = \frac{1}{2}[(1 - \phi) \cdot \mu_d + (1 + \phi) \cdot \mu_c]$. The term $\nabla \cdot \mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T)$ represents the effect of viscosity; and $G \nabla \phi$ is a body force transformed by using the divergence theorem to represent the surface tension force. Other forms of representations have been used to represent the surface tension force using the free energy theory [54]. However, in our study the most commonly used expression $G \nabla \phi$ is selected to calculate the interfacial tension force. The chemical potential G is expressed by the surface tension coefficient σ and the interfacial thickness ε :

$$G = \frac{3\varepsilon\sigma}{2\sqrt{2}} \left(-\nabla^2 \phi + \frac{\phi(\phi^2 - 1)}{\varepsilon^2} \right). \quad (7)$$

It can be seen that the chemical potential of the fluids varies directly with the interfacial thickness. The interfacial thickness is usually determined to satisfy the sharp interface limit and energy equilibrium [38, 66]. Therefore the surface
165 tension body force does not have a specific constant value and this variation should be balanced by the phenomenological mobility in the Cahn-Hilliard equation as a timescale relaxation parameter to control the interface movement according to the transport of binary fluids and the interfacial diffusion. In order to fully understand the theory, these governing equations are non-dimensionalized
170 to identify four dimensionless numbers. The width of micro-channel system L is chosen as the characteristic length. The other non-dimensional variables are $\mathbf{u}^* = \mathbf{u}L^2/Q_c$, $t^* = tQ_c/L^3$, $p^* = pL^4/\rho_c Q_c^2$, $\rho^* = \rho/\rho_c$, $\mu^* = \mu/\mu_c$ and $\nabla^* = L\nabla$. Then the non-dimensional equations are

$$-\frac{\partial \phi}{\partial t^*} + \mathbf{u}^* \cdot \nabla^* \phi = \frac{3}{2\sqrt{2}} \frac{1}{Pe} \nabla^{*2} [-Cn^2 \nabla^{*2} \phi + \phi(\phi^2 - 1)], \quad (8)$$

$$\nabla^* \cdot \mathbf{u}^* = 0, \quad (9)$$

$$\begin{aligned} \rho^* \left[\frac{\partial \mathbf{u}^*}{\partial t^*} + \mathbf{u}^* \cdot \nabla^* \mathbf{u}^* \right] = & -\nabla^* p^* + \frac{1}{Re} \nabla^* \cdot \mu^* (\nabla^* \mathbf{u}^* + \nabla^* \mathbf{u}^{*T}) + \\ & \frac{3}{2\sqrt{2}} \frac{1}{CnCaRe} [-Cn^2 \nabla^{*2} \phi + \phi(\phi^2 - 1)] \nabla^* \phi. \end{aligned} \quad (10)$$

The four dimensionless numbers in the governing equations are:

$$Re = \frac{\rho_c Q_c}{L \mu_c}, Cn = \frac{\varepsilon}{L}, Ca = \frac{\mu_c Q_c}{L^2 \sigma}, Pe = \frac{\varepsilon Q_c}{ML\sigma}, \quad (11)$$

where Q_c and μ_c are the flow rate and viscosity of the continuous phase. The
175 Reynolds number Re describes the ratio of inertial force to viscous force. The
Cahn number Cn is defined as the ratio of interfacial thickness between the
binary fluids to the characteristic length L , thus it is also known as the di-
mensionless interfacial thickness. The Capillary number Ca shows the ratio of
viscous force to surface tension force. The Péclet number Pe represents the
180 ratio of advection to diffusion in the two-phase flows. And the Péclet number
 Pe in our model differs from the usual Peclet number for mass transfer since
it is specifically for the phase field model. Because the physical properties and
geometric parameters are chosen to be constant, Ca and Re in our model have
specific values $Re \approx 0.01$, $Ca \approx 0.016$. The Péclet number and Cahn num-
185 ber are both functions of interfacial thickness ε which is used to approximate
the physical sharp limit interface. The phenomenological mobility M in Pe
is an artificial parameter introduced into the Cahn-Hilliard equation by using
interfacial energy theory [35].

2.3. Discussion of Boundary conditions

To save the computational time, we only simulate one quarter of the whole
domain since both of the micro-channel geometry and the flow pattern are sym-

metric. Therefore the symmetric boundary conditions are used for all of the symmetric surfaces in the system:

$$\mathbf{u} \cdot \mathbf{n} = 0, \quad \boldsymbol{\delta} \mathbf{n} - (\boldsymbol{\delta} \mathbf{n} \cdot \mathbf{n}) \mathbf{n} = 0, \quad (12)$$

190 where $\boldsymbol{\delta}$ is the stress tensor matrix and $\mathbf{n}$ is the unit normal vector.

There are three inlets in the micro-channel: the main inlet is for the disperse phase and the two side inlets are for the continuous phase. Fluid flow in the model belongs to laminar flow ($Re < 1$), thus the boundary conditions for all the three inlets are defined as the fully developed inlet flow rate. The mean flow rates are Q_d for the disperse phase and $0.5Q_c$ for the continuous phase. All the inlet velocity distributions of the fully developed laminar flow satisfy the parabolic profile and the maximum velocities in each inlet are located on the center point of the 2D inlet cross section domains:

$$\nabla_t \cdot \mathbf{u} = 0, \quad (13)$$

$$L_{ent} \nabla_t \cdot [-p \cdot \mathbf{I} + \boldsymbol{\delta}_t] = -p_{ent} \mathbf{n}, \quad (14)$$

where L_{ent} is the length of entrance before the inlet and $\nabla_t, \boldsymbol{\delta}_t$ represent the tangential operators. Non-viscous, pressure constraint outflow is applied as the boundary condition of the outlet, i.e., the pressure on the outlet is fixed as zero

$$\boldsymbol{\delta} \mathbf{n} = 0, \quad p = 0. \quad (15)$$

All of the above boundary conditions can be found and performed in the phase-field module using Comsol-Multiphysics.

However, the boundary conditions related to the moving contact line or fluid-wall interactions, have to be implemented based on theory of free energy [73]. The total free energy of the fluid-fluid-wall triple contact system can be described as

$$\Upsilon = \int_{\Omega} f_m(\phi, \nabla \phi) \, d\Omega + \int_{\partial\Omega} f_w(\phi) \, dS, \quad (16)$$

where Ω is the domain of two-phase fluids and $\partial\Omega$ is the area of solid wall surface. f_m is the fluid-fluid mixing energy and this expression can be written as

$$f_m(\phi, \nabla\phi) = \frac{K}{2} |\nabla\phi|^2 + \frac{K}{4\epsilon^2} (\phi^2 - 1)^2. \quad (17)$$

The wall relaxation energy f_w is a function of phase field variable. The energy equilibrium of static contact angle can be described by no-slip condition at the wall and the corresponding boundary conditions to demonstrate the stationary energy balance at solid wall are called the natural boundary conditions [40]. However, the dynamic behavior of the moving contact line at solid wall should be stated by the minimum energy dissipation principle. To demonstrate the energy dissipation of moving contact line on the solid surface, the generalized Navier boundary condition (GNBC) is introduced [70]. The GNBC is formulated by stating the Navier slip which is proportional to the total tangential stress on the solid surface. The total tangential stress consists of two parts: the viscous surface stress and the uncompensated Young stress. The wall relaxation energy f_w is formulated as [66]

$$f_w(\phi) = -\frac{1}{2}\sigma \cos\theta_s \sin\left(\frac{\pi}{2}\phi\right). \quad (18)$$

Since no slip boundary condition is not adequate to describe the three phase contact line at the wall surface [71, 74], the partial slip [91] at wall surface in tangential direction is used in GNBC as the Navier slip and the equilibrium contact angle θ_s of the binary fluids can be determined by Young-Dupré's equation which demonstrates the balance of surface energy near the triple phase contact line:

$$\cos(\theta_s) = \frac{\sigma_{da} \cos(\theta_{ds}) - \sigma_{ca} \cos(\theta_{cs})}{\sigma_{dc}}, \quad (19)$$

where σ_{da} , σ_{ca} and σ_{dc} are the disperse phase-air, continuous phase-air and disperse phase-continuous phase surface tension respectively. And θ_{ds} and θ_{cs} are the static contact angle between disperse phase and air and the angle between continuous phase and air on solid surface, respectively. Then the GNBC can be written as a combination of the viscous component and the uncompensated

stress component. Since there is no mass flux across wall boundary according to the Eq.(12) and then the diffusion across wall is also equal to zero. Therefore the expression of GNBC with the impermeability conditions $\mathbf{u} \cdot \mathbf{n} = 0$ and $\partial G / \partial \mathbf{n} = 0$ is

$$\beta(\phi) \mathbf{u}_\tau = -\delta \cdot \mathbf{n} + L_w(\phi) \nabla \phi, \quad (20)$$

where $\beta(\phi)$ is the slip parameter and this value depends on the contact characteristics of wall surface. $\mathbf{u}_\tau$ is the small slip velocity on the surface in tangential direction. δ is the viscous stress tensor. The first term in the right side of this equation is the viscous part and the second term is the uncompensated Young stress with $L_w(\phi) = K \partial_n \phi + \partial f_w(\phi) / \partial \phi$ as the surface chemical potential. Here the parameter K is the mixing energy density which is the same as K used in the free energy. Then we can obtain the transport boundary condition with the wall relaxation dynamics of ϕ

$$\frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi = -\Gamma L_w(\phi), \quad (21)$$

where Γ is a positive phenomenological parameter which represents the wall relaxation in the transport process [69]. Then the wall boundary conditions are coupled with the GNBC and the wall transport equation using Navier slip and wall relaxation energy.

3. Experimental setup and method

The schematic of the microfluidic channel is depicted in Fig. 1. The geometry used in experiments is the same as that used in the computational work. The characteristic length of this geometry is L , which is designed as $70\mu\text{m}$. All of the main inlet and the two side inlets have the cross-square section with the characteristic length L and the height of outlet is about $1.6 L$. The width of this micro-system is the characteristic length L in the z direction. Due to the limitations of the fabrication technique [92], there are some fillets in the corners of the micro-device. The radius of fillets at the cross intersection of inlets

is measured to be $R = 17.5\mu\text{m}$, and the radius of fillets at the expansion of downstream is measured to be $r = 10.5\mu\text{m}$, as shown in Fig. 1.

This microfluidic device was fabricated in PDMS using a soft lithography technique, and the Master molds were manufactured in a dry film photoresist (MM540, $35\mu\text{m}$ thick, DuPont) by lithographic patterning. The details of
210 fabrication process can be found in earlier experimental studies [92, 93]. To study the droplet formation process, the two fluids, mineral oil (Sigma M5904, Sigma-Aldrich) with 2 % surfactant Span 80 (Sigma-Aldrich) and distilled water, were delivered into the inlets separately using two precision syringe pumps (NE-
215 300, New Era and KDS 200, KDS Scientific). Since the material of PDMS is hydrophobic, mineral oil was chosen as the continuous phase and distilled water was the disperse phase. The physical properties of the two immiscible fluids were measured experimentally. The mineral oil has a density of $\rho_c = 840\text{ kg/m}^3$ and a viscosity of $\mu_c = 23.8\text{ mPa}\cdot\text{s}$. The distilled water has a density of $\rho_d = 1000$
220 kg/m^3 and a viscosity of $\mu_d = 1\text{ mPa}\cdot\text{s}$. The surface tension coefficient between the binary fluids is about $\sigma = 5\text{ mN/m}$.

The equilibrium contact angle θ_s of the two immiscible fluids we used in our experiments can be calculated using Young-Dupré’s equation [17]. The difference of the surface energy between the two immiscible fluids on PDMS
225 surface is $\sigma \cdot \cos(\theta_s)$. Based on the calculation, the value of $\cos(\theta_s)$ is -6.2 and it is less than -1, which means that the distilled water is the de-wetting phase and does not touch the PDMS wall surface, and the oil mineral is a completely wetting phase. This energy difference at the PDMS surface could be expressed by the generalized Navier boundary condition (GNBC) in numerical simulations.

In the droplet formation experiments, the flow rate of continuous phase (mineral oil) was fixed at $Q_c = 1\mu\text{L/min}$ while the flow rate of disperse phase (distilled water) was varied from $Q_d = 0.1\mu\text{L/min}$ to $1\mu\text{L/min}$. To maintain
230 stability of the two phase flow, gas tight glass syringes were used to reduce the effect of motor’s step motion. Droplet formation process in different flow ratio was monitored with an inverted microscope (IX73, Olympus) and a high-speed
235 camera (Phantom Miro M310, Vision Research). We used ImageJ to analyze

the experimental results, and extracted the images to compare them with the numerical results.

In the experiment, the droplet velocity was analyzed and determined by using software ImageJ and MATLAB. First, the area center position (x_c, y_c) of a droplet at different image frames (i.e., time) were extracted by using ImageJ. The time-position data were then linearly fitted with MATLAB built-in curve-fitting function. The slope of the best fitted curve gave the corresponding droplet velocity. The experiment images were taken after five minutes when the flow rates were changed, so that the flow conditions were stable. And for each experiment condition, a total of 70 to 80 droplets were analyzed to determine the average droplet velocity.

4. Results and discussions

4.1. Numerical implementation of the phase field model

Ideally as the thickness of the diffuse interface (ε) decreases, less energy dissipation will occur, and better approximation will be achieved in phase field models. However, the proper selection of thickness of diffuse interface needs to take computational cost and capability into consideration to achieve accurate solution of the phase variable ϕ . The principal criterion is that numerical results of ϕ should be smooth enough to express the diffuse interface and the simulations can lead to numerical convergence both in the bulk of fluids and on the wall surface. An important criterion is proposed for the sharp interface limit when the diffuse-interface contacts a wall surface [68]

$$Cn_c = 4S, \quad S = \frac{\sqrt{M\mu_e}}{L}, \quad \mu_e = \sqrt{\mu_c\mu_d}, \quad (22)$$

where μ_e is the equivalent viscosity for the binary fluids. Cn_c is the critical Cahn number for the convergence of sharp interface limit, S represents the bulk diffusion of the two-phase fluids and $\sqrt{M\mu_e}$ reflects the diffusion length at the contact line. This criterion is proposed according to the stability of numerical convergence using the Galerkin finite element method. Thus Cn varies with the

255 bulk diffusion for a sharp-interface limit and this sharp-interface limit can be approached by reducing Cn while keeping S constant.

On the other hand, the value of interfacial thickness must be selected based on the mesh size. For 3D cubic mesh, the interfacial thickness should be equal to or larger than half of the mesh size so that the phase function ϕ is described smoothly. In our model, the cubic mesh is utilized and the mesh size is 0.05 L . Therefore the thickness ε should be equal or greater than $0.025L$. In our model, four different thicknesses $0.025L$, $0.03L$, $0.035L$, $0.04L$ are selected for numerical analysis and the corresponding values are $Cn = 0.025$, $Cn = 0.03$, $Cn = 0.035$, $Cn = 0.04$. According to the criterion of numerical convergence
260 for the sharp-interface limit, this relationship (Equation 22) is determined by two parameters: the interfacial thickness (ε) and tuning mobility (χ). Thus for $\varepsilon = 0.025L$ the value of minimum mobility is determined by the corresponding tuning parameter $\chi = 12.8m \cdot s/kg$. It should be noted that this criterion only determines the lower limit of mobility. The reasonable value of χ in numerical
265 simulations depends on real physical systems, and can be found by comparison with experimental results. Once determined, the same value will be valid for other flow conditions provided that the same fluids are used in channels of the same surface properties.

The generalized Navier boundary condition (GNBC) is used to demonstrate
275 the dynamic interaction between wall surface and fluids. In GNBC the Navier slip term is equal to the sum of viscous stress term and the uncompensated Young stress. The Navier slip term $\beta(\phi) \mathbf{u}_\tau$ is the product of the Navier slip velocity $\mathbf{u}_\tau$ and the boundary friction coefficient $\beta(\phi)$. This friction coefficient depends on the fluid-wall interaction at the contact line and has the same unit
280 as viscosity [78]. The value of this coefficient varies from 0.05 to 4 for various strength of fluid-wall interaction. In our simulation we estimate the value of friction coefficient as $\beta = 2$ in the LJ unit(Lennard-Jones potential unit) according to the expression of contact line velocity acquired by MD simulation [67, 71, 77]. The phenomenological parameter Γ in the boundary transport equation of phase
285 variable ϕ indicates the energy dissipation and represents the corresponding re-

Figure 2: The 2D cross section schematic of droplets formation for numerical results in four different values of mobility ($M = \chi\varepsilon^2$). The 2D cross section is in the middle of the channel, which will be clarified in Fig. 3. The thickness of interface is fixed: $\varepsilon = 0.025L$. The tuning mobility parameter are $\chi = 12.8(a), 204.8(b), 820(c), 1280(d)$ (unit: $m \cdot s / kg$) respectively. The flow rate of the disperse phase (blue) and the continuous phase (red) are $0.5\mu L/min$ and $1\mu L/min$ respectively. The length of arrows indicates the velocity of fluids.

laxation time on wall surface. This wall relaxation parameter can be determined by dimensional analysis using the relation of $[M] = [\Gamma][Length]^3$ [73], where the $[Length]$ represents the scaled length in the phase field system and links the two parameters M and Γ . The order of this length is proportional to the interfacial thickness. In our computation, this length is chosen to be the same as the interfacial thickness ε since the variation of ϕ is located in the diffuse interface.

To obtain accurate numerical results, the mesh size should be small enough and the interfacial thickness is selected to satisfy the sharp-interface limit criterion. Considering the computational usage in the 3D phase field model the mesh size is chosen as $0.05L$. Since we use the symmetry of micro-channel to compute one quarter of the geometry, there are 10×10 cubic mesh cells in the one quarter square section of the main inlet and 20×10 cubic mesh cells in one half section of each side inlet. The total number of cubic mesh elements in one quarter geometry is over 16000 and all the fillets are mapped by non-cubic mesh. In this study we use the quadratic finite elements for velocity field and linear finite elements for pressure. The time step size is 0.0001 second and the

simulation stops after 5000 steps.

4.2. The phenomenological mobility in Cahn-Hilliard equation

Similar phase field models in T-shape micro-channels have been reported using finite difference method [89, 90]. These studies focus on the transition from squeezing to dripping regime by varying the capillary number Ca . A recent study of rising bubbles suggested the importance of the mobility parameter on affecting the bubble rise velocity[94]. However, the role of the diffuse term in Cahn-Hilliard equation remains unclear. Here, we keep the flow rate of continuous phase constant, which means the capillary number is constant ($Ca = 0.016$), and investigate the effect of mobility in the diffusion term, $M\nabla^2 G$. The Péclet number $Pe = \frac{\varepsilon Q_c}{ML\sigma}$ represents the ratio of advection to diffusion. According to the expression of Pe , the mobility and the interfacial thickness are important variables to reflect this ratio. First the effect of mobility will be tested in the phase field model.

Fig. 2 shows the comparison of emulsification with four different values of mobility M ($\varepsilon = 0.025L$), and all of the results satisfy the criterion of numerical convergence ($\chi \geq 12.8m \cdot s/kg$). In this figure we use 10×10 cubic mesh cells in one quarter of square section in the main inlet and 20×10 cubic mesh cells in half of each side inlet as the original mesh case to be mapped for the initial numerical tests. The color legend bars of phase variable ϕ are shown in Fig. 2. The undershoot of ϕ is found in the results, however, the lower bound is relatively small (the lower bound is around $\phi = -1.044$). We can observe that as the value of mobility increases the size of droplet in micro-channel becomes larger. This means that as the relaxation time of interface, the mobility M plays a very critical role in the two-phase flow: a higher value of mobility may cause the increase of relaxation time and the diffusion will be stronger. This value of M also reflects the effect of interfacial energy on the fluid flow. Thus the mobility should be selected such that it is neither too low for satisfying the numerical convergence and nor not too large for ensuring the diffusion not to overly damp the flow [40]. There is no clear criterion for the selection of mobility

Figure 3: The comparison between numerical and experimental results in the characteristic mobility ($M_c = \chi\epsilon^2$, $\chi = 820m \cdot s/kg$). (a)The experimental result, (b)the numerical result in 2D cross section and (c)the 3D numerical result. The 2D cross section is in the middle of the channel, which can be observed in (c). T_0 in (b) represents the experimental time span before the current process in this figure. In this figure we use refined mesh cases with 20×20 cubic mesh cells in square section of the main inlet and 40×20 cubic mesh cells in each side inlet. The thickness of interface is fixed: $\epsilon = 0.025L$. The flow rate of the disperse phase and the continuous phase are $0.5\mu L/min$ and $1\mu L/min$ respectively.

to justify the correctness of simulations. However, we can use the experimental results to determine this value.

Fig. 3 is the comparison between the numerical and experimental results of emulsification with the flow rates $0.5\mu L/min$ for disperse phase and $1\mu L/min$ for continuous phase. In Fig.3 we try to use 20×20 cubic mesh cells in square section of the main inlet and 40×20 in half section of each side inlet. The size of the droplet in experiments matches the numerical results well. The interfacial thickness in this case is $\epsilon = 0.025L$. The radius of droplet is about $R_e = 51.5 \mu m$ in the experiments, and $R_n = 51.2 \mu m$ in numerical simulations. Thus we identify this value of mobility ($M_c = \chi\epsilon^2$, $\chi_c \approx 820m \cdot s/kg$) as the characteristic value that can correctly reflect the real physical emulsification process in our model using distilled water and mineral oil as disperse and continuous phase respectively. When $M < M_c$, the droplet size in numerical simulations will be

Figure 4: The process of droplet formation in the characteristic mobility ($M_c = \chi\varepsilon^2$, $\chi = 820m \cdot s/kg$). (a)The numerical results; (b)the experimental results in 2D cross section. The 2D cross section is in the middle of the channel. The thickness of interface is fixed: $\varepsilon = 0.025L$. T_0 in (a) represents the experimental time span before the current process in this figure. The flow rate of the disperse phase and the continuous phase are $0.5\mu L/min$ and $1\mu L/min$ respectively.

345 smaller than the experimental result; and when $M > M_c$, the droplet size will be larger. It should be noted that this value of mobility is an approximation for the characteristic mobility, and we find this value by matching experimental results. The corresponding Péclet number satisfying the characteristic mobility with the interfacial thickness $\varepsilon = 0.025L$ is $Pe_c \approx 33.25$. In this case the droplet
350 diameter is $2R_d = 103 \mu m$ and is larger than the width of micro-channel system $L = 70 \mu m$. However, since the disperse phase is completely de-wetting, this phase can not contact the wall surface. Therefore the droplet is not a sphere but is squeezed in the width direction so that the interface of droplet does not touch the wall surface and is parallel to the wall (See Fig.3(c)).

355 The whole emulsification process is demonstrated in Fig. 4. This comparison shows excellent match between numerical and experimental results, including the size of droplet and the movement of droplet in the downstream. The value of mobility is experimentally justified as $M_c = \chi\epsilon^2$, where $\chi = 820m \cdot s/kg$ and $\epsilon = 0.025L$. The whole duration, from the disperse phase entering into the throat of micro-channel to the exiting of droplet near the computational outlet is 360 about 70 ms. It is found that the breakup location of disperse phase is near the entrance of throat and the breakup process experiences a very short time (less than 1ms). The breakup process indicates the dominant capillary effect over the shear stress in squeezing regime. The velocity of droplet in the downstream is 365 constant, which is confirmed in both the experiments and numerical simulations.

Fig. 5 shows the comparison of droplet volume in initial shape and in downstream respectively. Four different values of mobility are used to investigate the effect of interfacial diffusion on the droplet movement process in the micro-channel. In phase field method, the interfacial diffusion(the Gibbs-Thomson effect) is inevitable, since the Cahn-Hilliard equation utilizes the diffusion term 370 in the right hand side of the equation (8). The Péclet number $Pe = \frac{\epsilon Q_c}{ML\sigma}$ represents the ratio of advection to diffusion, which means that large mobility will cause strong interfacial diffusion. The droplet volume nearly keeps constant in these four different values of mobility. In Fig. 5, the largest mobility is 375 $\chi = 1280(\text{unit: } m \cdot s/kg)$ where $\epsilon = 0.025L$. The diffusion effect in our model is so small that the distance between the leading edge and the rear edge of droplet decreases less than 1%.

4.3. *The effect of interfacial thickness on droplet formation process and breakup of the disperse phase*

380 The interfacial thickness is an artificial diffuse layer introduced by phase field method and plays a very important role in two-phase flow models. In the Navier-Stokes equation, the force term $G\nabla\phi$ represents the surface tension force acting on the binary fluids as a body force. This force is not fixed because it is a function of the chemical potential G , which in turn is a function of the

Figure 5: The comparison between initial droplets and droplets in downstream with different values of mobility (where $\varepsilon = 0.025L$). The tuning mobility parameter are $\chi = 12.8(a), 204.8(b), 820(c), 1280(d)$ (unit: $\text{m} \cdot \text{s} / \text{kg}$) respectively. The left column shows the initial droplets and the right column shows the droplets in downstream. The 2D cross section is in the middle of the channel. The flow rates of the disperse phase and the continuous phase are $0.5 \mu\text{L}/\text{min}$ and $1 \mu\text{L}/\text{min}$ respectively.

interfacial thickness. Thus the surface tension body force varies with the inter-
 385 facial thickness. Besides, the phenomenological mobility is also a function of
 interfacial thickness $M = \chi \varepsilon^2$. Therefore, we can see that the interfacial thick-
 ness both affect the surface tension force and the relaxation time of interface.
 In our numerical simulations, we define the thickness as four different values:
 390 $Cn = 0.025, Cn = 0.03, Cn = 0.035, Cn = 0.04$ to investigate the effect of ε on
 the emulsification process.

In Fig. 6, it can be seen that the size of droplet varies with four different
 interfacial thicknesses when the tuning parameter of mobility is fixed as $\chi =$
 $820 \text{ m} \cdot \text{s} / \text{kg}$. The size of droplets increases with the value of ε and the shape

Figure 6: The 2D cross section comparison of droplet using four different interfacial thicknesses. The 2D cross section is in the middle of the channel. The value of mobility is $M = \chi\varepsilon^2$, where $\chi = 820m \cdot s/kg$. (a) $\varepsilon = 0.025L$; (b) $\varepsilon = 0.03L$; (c) $\varepsilon = 0.035L$; (d) $\varepsilon = 0.04L$. The flow rates of the disperse phase and the continuous phase are $0.5\mu L/min$ and $1\mu L/min$ respectively.

395 of droplet changes from sphere to plug as the volume becomes larger and the droplet formation process is delayed. This indicates that the original energy and force balance is broken and the value of mobility $M = \chi\varepsilon^2$, $\chi = 820m \cdot s/kg$ is not the characteristic mobility if $\varepsilon \neq 0.025L$. The relaxation time changes with the mobility, thereby greatly affects the droplet formation process. Thus
400 we should find a relationship between the tuning mobility parameter χ and interfacial thickness ε to keep the relaxation time and the diffusion term as reasonable values so that the phase field model can match the experimental phenomena.

The relaxation time of interface should be determined as constant to keep the
405 energy stable and the Péclet number would vary with the control of mobility and interfacial thickness so that interfacial diffusion is calculated to be reasonable. The Péclet number corresponding to the characteristic mobility is the exact ratio between advection and diffusion to reflect the real physical process. In Fig. 7 we use four different thicknesses and the corresponding reasonable tuning
410 mobilities to examine the breakup process of disperse phase and the dynamic shape of droplet. In all of the four groups (a,b,c,d), the relation between tuning mobility and interfacial thickness is fixed to keep the phenomenological mobility constant $M = \chi\varepsilon^2 = constant$, which means the relaxation time of interface is

Figure 7: The comparison of breakup process using four different interfacial thicknesses. The 2D cross section is in the middle of the channel. The characteristic mobility is $M_c = \chi \varepsilon^2$: (a1-a2) $\varepsilon = 0.025L$, $\chi = 820m \cdot s/kg$; (b1-b2) $\varepsilon = 0.03L$, $\chi = 570m \cdot s/kg$; (c1-c2) $\varepsilon = 0.035L$, $\chi = 418m \cdot s/kg$; (d1-d2) $\varepsilon = 0.04L$, $\chi = 320m \cdot s/kg$. The flow rate of the disperse phase and the continuous phase are $0.5\mu L/min$ and $1\mu L/min$ respectively.

also kept to be constant. For $\varepsilon = 0.025L$ the tuning mobility is $\chi = 820m \cdot s/kg$
 415 as the characteristic value, then the corresponding characteristic values for $\varepsilon =$
 $0.03L$, $\varepsilon = 0.035L$ and $\varepsilon = 0.04L$ are $\chi = 570m \cdot s/kg$, $\chi = 418m \cdot s/kg$ and
 $\chi = 320m \cdot s/kg$ respectively. The Péclet number $Pe = \frac{\varepsilon Q_c}{ML\sigma}$ in these four
 different thickness are $Pe = 33.25$, $Pe = 39.9$, $Pe = 46.55$ and $Pe = 53.2$
 correspondingly. It means that the Péclet number is linearly proportional to
 420 the interfacial thickness since the mobility and other parameters are kept to be
 constant. The numerical results of the four groups show excellent agreements
 in the breakup process and the size of droplet.

The interfacial body force and the interfacial energy dominate the breakup
 process. The time span of breakup process is very short (less than 1ms) and
 425 there is an instability of the two-phase flow which causes the minimization of
 interfacial energy after the breakup. From the enlarged picture at the entrance of
 throat, we can see that there are symmetric circular flows in the bulk with bullet
 shape, which indicates the instability of interfacial energy and the momentum
 equilibrium in horizontal direction.

Figure 8: The comparison of droplet size in four different disperse flow rates (the left column shows the 3D results, the middle column shows the 2D cross section results and the right column shows the experimental results.). The 2D cross section is in the middle of the channel. The characteristic mobility is $M_c = \chi \varepsilon^2$, where $\varepsilon = 0.025L$, $\chi = 820m \cdot s/kg$. The flow rate of disperse phase varies from $0.1\mu L/min$ to $1\mu L/min$. (a) $Q_d = 0.1\mu L/min$; (b) $Q_d = 0.2\mu L/min$; (c) $Q_d = 0.5\mu L/min$; (d) $Q_d = 1\mu L/min$. The flow rate of continuous phase is fixed as $1\mu L/min$.

4.4. The effect of flow rate on the size of droplet

The capillary number is calculated using the physical properties and flow rate of the continuous phase $Ca = \mu_c Q_c / L^2 \sigma$. In our numerical study, however, the value of Ca is fixed to 0.016 since the physical properties and the flow rate of the continuous phase are constant. Therefore we only investigate the effect of flow rate of the disperse phase on the size of droplet.

Fig. 8 clearly shows the comparison of droplet volume between experimental and numerical results. The experimental and numerical results match very well. The size of droplet increases with the flow rate of disperse phase. This phenomenon satisfies the mass conservation law. However, the droplet volume is not linearly proportional to the flow rate of disperse phase because the droplet formation process is also affected by the surface tension force and the dynamic energy equilibrium (including both interfacial energy $\int_{\Omega} \frac{1}{2} \varepsilon |\nabla \phi|^2 d\Omega$ and bulk free energy $\int_{\Omega} \frac{(\phi^2 - 1)^2}{4\varepsilon} d\Omega$) in the two phase fluids system. Experimental results

Figure 9: The comparison of droplet velocity between experimental and numerical results in four different flow rates of disperse phase. The characteristic mobility is $M_c = \chi \varepsilon^2$, where $\varepsilon = 0.025L$, $\chi = 820 \text{ m} \cdot \text{s}/\text{kg}$. The flow rate of continuous phase is fixed as $1 \mu\text{L}/\text{min}$.

of droplet radius in the x - y plane are $40 \mu\text{m}$, $42.2 \mu\text{m}$ and $51.5 \mu\text{m}$ for the flow rate
of disperse phase at $0.1 \mu\text{L}/\text{min}$, $0.2 \mu\text{L}/\text{min}$ and $0.5 \mu\text{L}/\text{min}$ respectively. The
shape of droplet at $1 \mu\text{L}/\text{min}$ is not a sphere but a plug and the 3D the numerical
results show that the interface of this dispersed plug does not touch the wall
surface. The length of this plug flow is about $128 \mu\text{m}$ in the x direction. It can
be noticed that the droplet volume obtained from the experiments fluctuates
because the unsteady flow due to the micro-pumps. The numerical results of
droplet radius are $41.5 \mu\text{m}$, $45.2 \mu\text{m}$ and $51.2 \mu\text{m}$ correspondingly and the length
of plug is $140.2 \mu\text{m}$. The discrepancy between the experimental and numerical
results is less than 10% and is caused by the relatively large instability induced
by the micro-pump when operating at a low flow rate.

4.5. The velocity of droplet and period of droplet formation process

The velocity of droplet in the downstream channel after emulsification process is studied in both experiments and numerical simulations. From the experiments we can see that the droplet or plug moves along the micro-channel at a constant speed. The velocity of droplet in P-F model is also acquired by analyzing the numerical results.

Different from the experimental measurements, the computational cases only include 0.5 second in time and only 2 to 4 droplets forms in the whole process. Therefore we cannot use the time average method to measure the velocity of

around 100 droplets. Instead, we use the position of leading edge of interface in
465 disperse droplet to measure the droplet velocity. The measurement starts from
the moment when the droplet is completely formed and ends when the leading
interface gets close to the outlet. The velocity we measured in numerical cases
is also not stable for each frame (per 0.002 to 0.005 second), thus we used
the average velocity for the whole frames to express the droplet velocity in the
470 downstream.

Numerical results of the velocity are higher than the corresponding exper-
imental values when $Q_d = 0.1\mu\text{L}/\text{min}$, $0.2\mu\text{L}/\text{min}$ and $0.5\mu\text{L}/\text{min}$, but lower
than the experimental results when $Q_d = 1\mu\text{L}/\text{min}$. However, the discrepancy
is less than 15%. Besides the instability caused by the micro-pumps, there are
475 other possible reasons which can contribute to this discrepancy. These factors
include the variation of physical properties caused by temperature change and
the wall surface roughness which can lead to the increase of resistance at wall
surface and change the boundary condition at solid wall. From Fig. 9, it clearly
shows that the velocity of droplet increases with the flow rate of disperse phase
480 and this tendency can be found in both experimental and numerical results. It
indicates that the numerical results are reasonably accurate in describing the
transitional movement process in the downstream channel.

The periods of droplet formation process are obtained in different flow rates
of the disperse phase both in experiments and numerical models. To account
485 the effect of unsteady flow due to the micro-pump, we analyze a very long
time span (over 10s) to acquire the variation of period. We carefully observe
the emulsification process in the experiments and acquire the periods in four
different flow rates ($0.1\mu\text{L}/\text{min}$, $0.2\mu\text{L}/\text{min}$, $0.5\mu\text{L}/\text{min}$, $1\mu\text{L}/\text{min}$). According
to the experimental data, the periods of emulsification process are 116~170 ms
490 for $Q_d = 0.1\mu\text{L}/\text{min}$, 81~89 ms for $Q_d = 0.2\mu\text{L}/\text{min}$, 54~63 ms and $Q_d =$
 $0.5\mu\text{L}/\text{min}$ and 38~46 ms for $Q_d = 1\mu\text{L}/\text{min}$. It can be seen that the span of
period is very large when the flow rate of disperse phase is small, which indicates
the unsteadiness effect is obvious at smaller flow rates.

We also obtain the periods in the numerical simulations. We choose the time

495 step size to be 0.0002s and perform 3000 steps in time evolution. The period of droplet formation process can be accurately determined from the evolution of pressure. The evolution of pressure in the central point at the entrance of throat is shown in Fig. 10, with $Q_d = 0.2\mu\text{L}/\text{min}$.

The numerical solutions of pressure at the entrance of throat vary periodically with time in Fig. 10 and there are about 3 fluctuations from 0.2s to 0.5s. The pressure increases when the disperse phase is squeezed by the continuous phase to form a droplet. When the droplet breaks up, the pressure suddenly drops to a very low value. It clearly indicates that there are three sudden drops of pressure at $T \approx 0.26\text{s}$, 0.31s and 0.41s . We obtain the periods for four given flow rates: the values of period are 179 ms for $0.1\mu\text{L}/\text{min}$, 99 ms for $0.2\mu\text{L}/\text{min}$, 61 ms for $0.5\mu\text{L}/\text{min}$ and 41 ms for $1\mu\text{L}/\text{min}$. By comparing the numerical solutions with the experimental periods we can see that the numerical periods are within the experimental spans when the flow rate is $0.5\mu\text{L}/\text{min}$ or $1\mu\text{L}/\text{min}$. It indicates the numerical and experimental periods match very well when the disperse flow rate is relatively high. The numerical periods are slightly higher than the experimental periods when the flow rate is small ($0.1\mu\text{L}/\text{min}$ and $0.2\mu\text{L}/\text{min}$). The unsteady flow may lead to this mismatch, and it is difficult to keep flow steady in the small flow rates.

5. Conclusions

515 A 3D phase field model, the Cahn-Hilliard-Navier-Stokes model with generalized Navier boundary condition, was used with finite element method to simulate the droplet formation process in a flow-focusing device. To justify this phase field numerical model and investigate the selection criterion of several critical parameters, experiments in the same configuration was set up. The fluid flow was dominated by interfacial force since the capillary number was fixed as a small value ($Ca \approx 0.016$). We found the phenomenological mobility in the Cahn-Hilliard equation is a critical parameter for the emulsification process. This parameter determines the diffusion term in the Cahn-Hilliard equation

Figure 10: (a) The evolution of pressure in central point at the entrance of throat and (b) the location of fluid pressure in 2D cross section. The characteristic mobility is $M_c = \chi \varepsilon^2$, where $\varepsilon = 0.025L$, $\chi = 820 m \cdot s/kg$. The flow rates are $0.2 \mu L/min$ and $1 \mu L/min$ for disperse and continuous phase respectively.

and the relaxation time of the interface in order to control the droplet volume
 525 and shape. We defined the characteristic mobility by a comparison between
 numerical and experimental results. This characteristic mobility is associated
 with the chemical energy in the diffusion term of the Cahn-Hilliard equation,
 and thus the mobility should be affected by the interfacial tension between given
 specific fluids. In this paper the two-phase fluids consisted of water for the dis-
 530 perse phase and mineral oil for the continuous phase. Therefore the mobility
 in this water-mineral oil system was found by comparing with the experimental
 results. The mobility of other different two-phase fluid systems should also be
 investigated in our work in future. In our model the numerical convergence
 of sharp interface limit was guaranteed with the properly selected interfacial
 535 thickness. The interfacial thickness was also found to play a key role in the
 phase field model. The variation of thickness could lead to the change of energy
 dynamic equilibrium in the two-phase system and also cause the variation of
 characteristic mobility. To ensure the numerical results capturing the physical
 process correctly, i.e., the experimental data, we found that the multiplication

540 between the tuning mobility and the square of interfacial thickness needed to
 be kept constant ($\chi\varepsilon^2 = \text{Constant}$) to guarantee the mobility ($M = \chi\varepsilon^2$) as
 a constant. This criterion also guarantees that Péclet number ($Pe = \frac{\varepsilon Q_c}{ML\sigma}$)
 represents the reasonable ratio between advection and diffusion. The Péclet
 number Pe is proportional to the interfacial thickness when the mobility and
 545 other physical properties are fixed. The effect of flow rate of the disperse phase
 was also studied and a good agreement between numerical results with the char-
 acteristic mobility and experimental results was achieved. The droplet velocity
 in the downstream and the period of droplet formation process obtained in the
 numerical model generally matched the experimental results. By comparing the
 550 numerical results with experiments, we justified the correctness of our phase
 field model, thus this model can be used for our further study to investigate
 more complex two-phase flow systems.

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