Development of a numerical model for Marangoni convection in the micro-scale environment

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Abstract

Marangoni convection is induced in liquids by surface tension gradient along a free surface. Such flows also develop in nano- and micro-scale systems and play important roles. To have a better understanding for the phenomena occurring in processing of such micro-scale systems, generally two kinds of numerical simulation approaches have been considered: continuum and discrete (molecular dynamics). While the continuum-based techniques cannot capture the intermolecular effects, the molecular dynamics approach requires huge computational cost. To address the adverse futures of these techniques, a new numerical method has been developed by combining the computational fluid dynamics (CFD) from the continuum side and Langevin dynamics from the discrete approach. The present simulation results have shown that this new numerical technique can successfully study and predict the phenomena occurring in macro-scale process applications.

Key words: Marangoni convection, Computational fluid dynamics, Langevin equation, Micro-scale environment, Multi-phase flow, S-CLSVOF method, Numerical modeling, Multi-scale modeling

1. Introduction

Temperature or concentration differences along a free surface give rise to a surface tension gradient, and the unbalanced force along the free surface develops Marangoni convection in the liquid (Scriven and Sternling, 1960). Although this surface tension driven flow is generally weaker than natural convection, it may become significant in some environments such as the microgravity environment where the buoyancy driven flows becomes weaker (e.g. Lappa, 2004; Kawamura et al., 2012) and micro-scale (e.g. Darhuber and Troian, 2005; Karbalaei, et al., 2016). For example, the surface tension driven flow has attracted attention following the interesting phenomena observed by Pettit in his microgravity experiments on the International Space Station (e.g. Pettit, 2003; Pettit, 2011; Ueno et al., 2010; Limsukhawat et al., 2013; Yamamoto et al., 2013; Yamamoto et al., 2016; Shiratori et al., 2016). However, it was also shown that such a convective flow may also occur in the micro-scale environment of the applications of manufacturing micro- and nano-devices. The works of Murata and Mochizuki, 2006; Maier et al., 2012; Das et al., 2015a; Das, 2015b have shown that Marangoni convection develops even in the nano-meter scale.

Although the driving force of the Marangoni convection (which is the surface tension gradient) is the same, the physical modeling in macroscopic and microscopic systems differs. Macroscopic phenomena are modeled on the basis of continuum approximation while microscopic phenomena are modeled using discrete methods that incorporate intermolecular forces. For certain applications it is also necessary to develop models that are between macro- and micro-scales: meso-scale. Examples of the meso-scopic numerical models are the Dissipative Particle Dynamics (DPD) method (Hoogerbrugge and Koelman, 1992) and the coarse-grained molecular dynamics (CGMD) method (Kremer and Grest, 1990). These methods are bottom-up modeling based on the molecular dynamics approach.

However, in the present article we adopted the top-down modeling. That is, the present work introduced a new numerical model built upon the continuum approach. The objective was to develop a numerical model to express multiphase...
flows with considering the momentum, energy and overall mass balance (continuum), and the species mass transport with Marangoni convection at the micro scale. To achieve this goal, the discrete and continuum numerical techniques are combined into a single numerical model. The final goal of this study is to develop the numerical method to solve the momentum, energy and mass balances in the domains with a gas-liquid interface in the micro- or meso-scale systems.

2. Numerical Analysis

As mentioned earlier, the continuum and discrete methods are coupled, and the micro-scale phenomena are introduced by considering the interactions between particles in the discrete method. In order to express the movement of molecules (such as surfactants) and the interactions between them in the domain, we used the discrete method for species transfer. For the momentum, overall mass and energy balances, we adopted the continuum approach with incorporating thermal fluctuation terms.

2.1. Numerical Model for Momentum and Heat Transfer (Continuum Approach)

The momentum, overall mass, and energy balances were expressed within the continuum approach in this study; leading to the following governing equations of the momentum balance, continuity, and energy balance with thermal fluctuation (Landau and Lifshitz, 1959; Landau and Lifshitz, 1979):

\[
\frac{\partial(\rho u)}{\partial t} + \nabla \cdot (\rho uu) = -\nabla p + \nabla \cdot \mu(\nabla u + \nabla u^T) + \rho g + F_\sigma + F_F
\]
\[
\nabla \cdot u = 0
\]
\[
\frac{\partial T}{\partial t} + u \cdot \nabla T = \nabla \cdot \kappa \nabla T + Q_F
\]

where \( u \) is the flow velocity, \( p \) pressure, \( \rho \) the liquid density, \( \mu \) the liquid viscosity, \( g \) the gravitational acceleration, \( T \) temperature, \( \kappa \) the fluid thermal diffusivity. The source term \( F_\sigma \) is the surface tension term and \( F_F, Q_F \) are the thermal fluctuation terms which are expressed as:

\[
F_F = \nabla \cdot S
\]
\[
Q_F = \nabla \cdot (Q + u \cdot S)
\]

where \( S \) is the fluctuation tensor and \( Q \) is the fluctuation vector. Their average and variance are as follows:

\[
< S_{ij} > = 0
\]
\[
< Q_i > = 0
\]
\[
< S_{ij} S_{kl} >= \frac{2k_BT}{\Delta tV_c}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})
\]
\[
< Q_i Q_j >= \frac{2k_BT^2}{\Delta tV_c}\delta_{ij}
\]

where \( k_B \) is the Boltzmann constant, \( \Delta t \) the time step width, \( V_c \) the discrete volume of the numerical grid cell, \( k \) the thermal conductivity, and \( \delta \) Kronecker’s delta. The surface tension term \( F_\sigma \) is given in subsection 2.4.

2.2. Numerical Model for Species Transfer (Discrete Approach)

To express molecular interactions in a microscopic phenomenon, we adopted a discrete approach that makes use of the Langevin equation. In this study, the particles expressed in the Langevin equation are considered as clusters of molecules. Hence, the particle size is quite larger than molecules sizes. The Langevin equation is given as follows

\[
m \frac{d^2 x}{dt^2} = -\xi \frac{dx}{dt} + R(t) + F_{add}
\]

where \( m \) is the particle mass, \( x \) the position vector, \( \xi \) the coefficient of the drag term, \( R(t) \) the random vector and \( F_{add} \) the additional force term. The first term on the right hand side in the Langevin equation is the drag force term, the second
term represents the random fluctuating term, and the third term is the conservative additional term which is taken as zero in this study. That is, we do not consider molecular interactions since this article’s objective is to determine whether the present numerical method can predict the macroscopic phenomena under consideration. The average and variance of the random vector $\mathbf{R}(t)$ are:

\[
\langle \mathbf{R} \rangle = 0 \quad (11)
\]

\[
\langle \mathbf{R}_i \mathbf{R}_j \rangle = \frac{2D\xi^2}{\Delta t} \delta_{ij} \quad (12)
\]

where $D$ is the diffusion coefficient used in the microscopic phenomena. To satisfy the macroscopic diffusion, we use Einstein’s relation for the fluctuation strength. The coefficient of the drag term $\xi$ is:

\[
\xi = C_D A_p \frac{\rho |\mathbf{u}_l - \mathbf{u}_s|}{2} \quad (13)
\]

where $C_D$ is the drag coefficient, $A_p$ the projected area and subscripts $l$ and $s$ indicate the liquid and solid phases, respectively. For the drag model, we adopted Stokes’ drag law:

\[
C_D = \frac{24}{Re_p} \quad (14)
\]

where $Re_p$ is the particle Reynolds number defined as:

\[
Re_p = \frac{\rho |\mathbf{u}_l - \mathbf{u}_s| d_s}{\mu_l} \quad (15)
\]

where $d_s$ is the particle diameter. By substituting Eqs. (14) and (15) into Eq. (13), the coefficient of drag term $\xi$ can be transformed into

\[
\xi = 3\pi \mu_l d_s \quad (16)
\]

By substituting Eq. (16) into Eq. (12), one can express the variance of the fluctuation term as:

\[
\langle \mathbf{R}_i \mathbf{R}_j \rangle = \frac{2D(3\pi \mu_l d_s)^2}{\Delta t} \delta_{ij} \quad (17)
\]

The fluctuation strength is introduced into the particles to satisfy this variance.

### 2.3. Coupling between the Methods of Continuum and the Discrete Approaches

The methods of continuum and discrete approaches (which will be called continuum method and discrete method, respectively) must be properly coupled. The continuous concentration distribution is reconstructed by the particle distribution calculated by the Langevin equation as:

\[
C = C_{\text{max}} \sum \mathbf{V}_{p,i} \frac{V_{p,i}}{V_c} \quad (18)
\]

where $V_p$ is the particle volume. The summation of particle volume occupied in a numerical grid cell is used for the calculation of the concentration distribution. In this study, we used volume fraction in representing concentration. The concentration profile calculated by Eq. (18) is not smooth. To obtain a smooth concentration profile, Laplacian filter (Lafaurie et al., 1994; Hoang et al. 2013) is used after solving Eq. (18).

\[
C = \frac{\sum_i C_i |\mathbf{S}_i|}{\sum_i |\mathbf{S}_i|} \quad (19)
\]

where $\mathbf{S}_i$ is the numerical cell area vector and subscript $f$ indicates the numerical cell face.
2.4. Multiphase Flow Modeling

We adopted a simple coupled Volume of Fluid with the Level Set (S-CLSVOF) method for the multiphase flow simulation. This method uses the algebraic Volume of Fluid (VOF) method, and the curvature and the normal vector to the interface are calculated by the Level-Set function. This method was first developed by Albadawi et al. (2013). The VOF method used in this study is its original form that was implemented in an open source software, OpenFOAM.

\[
\frac{\partial \alpha}{\partial t} + \nabla \cdot (\alpha \mathbf{u}) + \nabla \cdot ((1 - \alpha) \alpha \mathbf{u}_r) = 0 \tag{20}
\]

where \( \alpha \) is the volume fraction of Fluid 1, and \( \mathbf{u}_r \) is the relative velocity between Fluid 1 and Fluid 2. The volume fraction \( \alpha \) is defined as:

\[
\alpha = \begin{cases} 
0 & \text{Fluid2} \\
1 & \text{Fluid1} \\
0 < \alpha < 1 & \text{Interface}
\end{cases} \tag{21}
\]

Furthermore, the relative velocity \( \mathbf{u}_r \) is expressed as:

\[
\mathbf{u}_r = \mathbf{u}_1 - \mathbf{u}_2 \tag{22}
\]

where the subscripts 1 and 2 are used to describe Fluid 1 and Fluid 2, respectively. The third term in Eq. (20) compresses the interface thickness in this algebraic VOF method. For details the reader is referred as Rusche (2002), Weller (2008), Deshpande et al. (2012) and Albadawi et al. (2013). After solving Eq. (20), the following re-initialization equation is solved

\[
\frac{\partial \psi}{\partial t} = \text{Sign}(\psi_0)(1 - |\nabla \psi|) \tag{23}
\]

with

\[
\psi_0 = (2\alpha - 1)\Gamma \tag{24}
\]

\[
\Gamma = 0.75\Delta x \tag{25}
\]

where \( \psi \) is the Level-Set function, \( \text{Sign} \) is the sign function, \( \psi_0 \) the initial Level-Set function calculated by volume fraction \( \alpha \), \( \Gamma \) the non-dimensional number related to the initial Level-Set function and \( \Delta x \) the width of the numerical grid cell.

By the use of the Level-Set function, the curvature and the interface normal vector are calculated by

\[
k = - \int_{\Omega} \nabla \cdot \mathbf{n} d\Omega = - \int_{S} \mathbf{n} \cdot dS = \sum_{i} n_i \cdot S_i \tag{26}
\]

\[
\mathbf{n}_t = \frac{(\nabla \psi)_t}{|(|\nabla \psi|_t + \delta_1)} \tag{27}
\]

The surface tension term has two components: normal and tangential. The tangential component represents Marangoni convection. The surface tension term is expressed as:

\[
F_{\sigma} = \sigma_{T} \delta \nabla \psi + \sigma_{C}(\nabla C - \mathbf{n}(\mathbf{n} \cdot \nabla C))\delta' \tag{28}
\]

where \( \sigma \) is the surface tension, \( \delta \) the delta function, \( \sigma_{T} \) the surface tension coefficient of temperature and \( \sigma_{C} \) the surface tension coefficient of concentration. The delta function \( \delta' \) is calculated by the Level-Set function as

\[
\delta' = \begin{cases} 
\frac{1}{\pi \epsilon} \left(1 + \cos \left(\frac{\pi \epsilon}{\epsilon}\right)\right) & |\psi| < \epsilon \\
0 & \text{else.}
\end{cases} \tag{29}
\]

where \( \epsilon \) is the interface thickness (\( \epsilon = 1.5\Delta x \)). The physical properties are updated by the volume fraction \( \alpha \) as

\[
\mu = \mu_1 \alpha + \mu_2 (1 - \alpha) \quad \rho = \rho_1 \alpha + \rho_2 (1 - \alpha) \quad \kappa = \kappa_1 \alpha + \kappa_2 (1 - \alpha) \quad D = D_1 \alpha + D_2 (1 - \alpha) \tag{30}
\]

The related details and validations for this multi-phase flow method can be found in Yamamoto et al. (in press).
2.5. Discretization
The governing equations are discretized by the finite volume method, and the numerical methods of both approaches are introduced into an open source software, OpenFOAM. For time derivative, the implicit Euler method is used for the continuum method and the explicit Euler method for the discrete method. The spatial discretization for the continuum method is done by linear interpolation. The coupling between velocity and pressure fields is the Pressure-Implicit with Splitting of Operators (PISO) method (Issa, 1985).

3. Results and Discussions

Validation of the numerical methods used is carried out in the following manner:
(1) Comparing the Langevin equation with the diffusion equation in equilibrium
(2) Comparing the Langevin equation with the diffusion equation with flow, and
(3) Comparing the multiphase flow with Marangoni convection with the discrete method.

3.1. Comparing Langevin equation with diffusion equation in equilibrium
In the case of no flow, the numerical solution obtained from the Langevin equation without additional effects must correspond to that obtained from the diffusion equation. Thus, we validated the Langevin equation with respect to the following diffusion equation

\[ \frac{\partial C}{\partial t} + u \cdot \nabla C = D \nabla^2 C \]  

which is also solved using the same discretization schemes as those described in subsection 2.5.

![Fig. 1](image)

Fig. 1 (a) Schematics of the 2-D numerical simulation domain, (b) initial concentration distribution obtained from the diffusion equation and (c) the initial particle distribution from the Langevin equation.

The computational domain here is a two-dimensional square with the sides of 100 μm. The boundaries represent the impermeable walls through which we impose the zero concentration flux condition in the solution of the diffusion equation, and a perfect collision condition in the solution of the Langevin equation. A schematic description of the computational domain is shown in Fig. 1 (a). The sides of this area are set to 12 μm. The numerical grid has uniform spacing with 50 × 50 grid resolution in each direction. The initial conditions for diffusion and Langevin equations are shown in Fig. 1 (b) and (c), respectively. The number of particles is 2,500 and two values of diffusion coefficient are utilized: \( D = 1.0 \times 10^{-8} \) and \( 1.0 \times 10^{-9} \) m²/s. These values are smaller than that of usually used the diffusion coefficient value. The time increment has to be enough small in solving the Langevin equation. The time increments used in this calculation were \( 1 \times 10^{-6} \) s and \( 1 \times 10^{-8} \) s for \( D = 1.0 \times 10^{-8} \) and \( 1.0 \times 10^{-9} \) m²/s. Ensemble average was taken by using 10-run calculations to achieve the quantitative comparison.

The simulation results are shown in Fig. 2. Comparing Fig. 2 (a) with Fig. 2 (c), one can be seen that the concentration profiles are in good agreement with each other. Quantitative comparisons are also given in Fig. 2 (d). Quantitatively, we also obtain a good agreement between the predictions from the Langevin and diffusion equations. With these results, we can state that macroscopic diffusion can be studied directly by the discrete method.

3.2. Comparing Langevin equation with diffusion equation with flow
Next we compare the Langevin equation with the diffusion equation in the presence of flow. The calculation conditions are similar to that presented in subsection 3.1 with the condition that the upper wall can slide in the horizontal
direction to the right in Fig. 1. In this case, the physical properties are transformed into the dimensionless numbers of Reynolds number \((Re = \frac{\rho_{\text{wall}} \cdot u_{\text{wall}} \cdot d}{\mu} = 100)\) and Schmidt number \((Sc = \frac{\nu}{D} = 0.2, 10)\). The representative length \(d\) is taken as the side length of the numerical domain.

The computed concentration distributions obtained from the diffusion and Langevin equations are presented in Fig. 6.
3. In this case, as seen we also obtain a good agreement between the solutions. This shows that even in the presence of flow the Langevin equation can be used in lieu of the diffusion equation.

3.3. Multiphase flow with Marangoni convection with the discrete method

In this case the continuum and discrete methods are fully combined. The numerical simulation domain is presented in Fig. 4 (a) and the initial particle distribution is shown in Fig. 4 (b). The number of the particles is 10,000. In this case, the simulation conditions are the same as Sen and Davis (1982), except that used for particles (concentration distribution). Numerical accuracy was validated by comparing the present simulation results with those of Sen and Davis (1982). Aspect ratio $A = \frac{d}{W}$, Marangoni number $Ma = \frac{\sqrt{\gamma_{ij} \Delta T d}}{\rho_0 a}$ and Prandtl number $Pr = \frac{c}{\rho_0 a}$ have the values of 0.2, 0.2 and 0.2, respectively. The surface tension coefficient of concentration $\sigma_C$ is firstly assumed to be zero to compare the results with Sen and Davis (1982). That is, the passive concentration (particles) is introduced into the case of Sen and Davis (1982). The Schmidt number ($Sc = \frac{\sigma}{D}$) was set to be $1 \times 10^5$ and the diffusion is very small in this case. This method can simulate such a high-Sc-number fluid, although the conventional method using a diffusion equation is difficult to calculate such a high-Sc-number fluid.

![Fig. 4 Multiphase flow with Marangoni convection: (a) schematics of the numerical domain, and (b) initial particle distribution.](image)

![Fig. 5 Simulations results of the multiphase flow with the discrete method: (a) temperature distribution, (b) particle distribution and (c) concentration distribution reconstructed by particle distribution.](image)

The computed temperature, particle and concentration distributions are shown in Fig. 5. In this figure, the middle horizontal line indicates the gas/liquid interface. The interface is expressed by the contour of $\alpha = 0.5$. As seen, thermo-
capillary flow develops along the free surface towards the cold wall. The flow changes the interface shape slightly and affects the particles and the concentration distributions. The computed concentration profiles show clearly the "return flow" usually observed in Marangoni convection in thin films (Sen and Davis, 1982). Furthermore, the concentration distribution expands along the interface in accordance with the interface deformation.

Next, the present study investigated whether the concentration difference alone can produce Marangoni convection without temperature variation or not. Generally speaking, the contaminant reduces the surface tension. Therefore we adopted a negative surface tension coefficient of concentration to accommodate this effect. The concentration lowers the surface tension in this test. The initial solutal Marangoni number is defined as $Ma_C = \frac{\mu \xi A C_{\text{max}}}{\rho K}$ and the value is 0.2. The computed results are shown in Fig. 6. The results show the solutal Marangoni convection develops along the free surface towards the low concentration area. This means that the present numerical algorithm can predict the "return flow" induced by solutal Marangoni convection.

![Fig. 6 Computed concentration distribution induced by solutal Marangoni convection alone: (a) Particles distribution and (b) concentration distribution reconstructed by particles distribution.](image)

This newly developed method has two merits and one demerit. The merits are; (1) this model can handle the high-Sc-number fluid and (2) this model can introduce the intermolecular effect by introducing particle-particle pair potential. On the other hand, the method requires a larger computational cost compared with conventional method using macroscopic diffusion equation.

4. Conclusion

The present study introduced a new numerical method by combining the computational fluid dynamics (CFD) from the continuum side and Langevin dynamics from the discrete approach. The method is validated in the cases of no flow, with flow, and with multiphase flow. However, the validation was done only at the macroscopic scale. In future, we plan to introduce additional forces into the Langevin equation to model adsorption to the interface and the molecular interactions. In addition, the relationship between the Langevin and diffusion equations used in this study was at equilibrium. We also plan to introduce non-equilibrium relationship between these equations to obtain more accurate predictions.

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