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On the motion of droplets driven by solutal Marangoni convection in alloy systems with a miscibility gap

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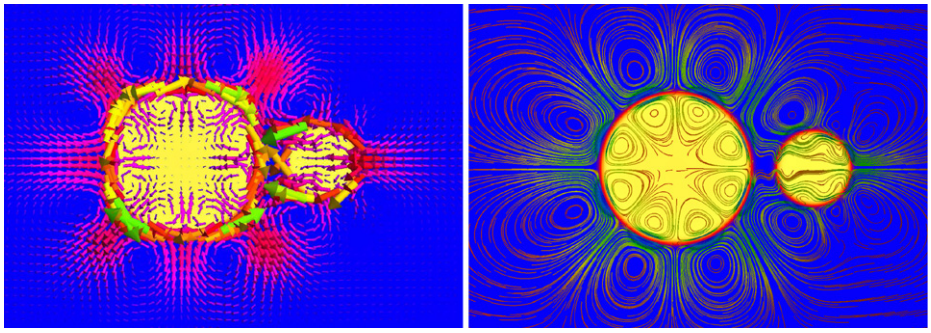
HIGHLIGHTS

- Analysis of solutal Marangoni convection in the bipolar coordinate.
- A combined Cahn–Hilliard and Navier–Stokes model with capillary tensor.
- Repulsion or attraction is affected by droplet radius and the Marangoni number.

GRAPHICAL ABSTRACT



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ABSTRACT

In the first part of this work, we analytically study the motion of two droplets driven by solutal Marangoni convection in a bipolar coordinate. Particular solutions for the Laplace and Stokes equations are found by applying Robin type boundary conditions for mass transfer and by utilizing continuity of stream function and impenetrability at the surface of droplets. The solutions for the Laplace and Stokes equations are connected by the tangential stress balance between the viscosity stress and the Marangoni stress caused by concentration gradients. In the second part, we numerically investigate the motion of two droplets in an immiscible fluid by solving the combined convective Cahn–Hilliard and Navier–Stokes equations, where the capillary tensor is used to account for the Marangoni force. A significant outcome of the present work is that the attraction or repulsion of droplets is determined by droplet radius and the Marangoni number. In both cases, we obtain the stream lines affected by the spacing between droplets and the ratio of the radius of the droplet.

1. Introduction

The motion of droplets driven by solutal Marangoni convection in an immiscible fluid is a commonly observed phenomenon in nature [1–4], e.g. the swimming of oil in water, and is coupled in some phase transformation processes [5,6], e.g. the motion of the minority liquid droplet in the continuous phase during spinodal decomposition. The mass transport during the motion of the droplets is induced by the surface tension gradient that is caused by concentration gradients along the surface of the droplet. A powerful method that is used to study this effect is a combination of the convective Cahn–Hilliard equation with the Navier–Stokes equation, where the mass transport is governed

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by the Cahn–Hilliard equation and the convection follows the Navier–Stokes equation.

For low Reynolds number fluids with small Péclet number, the Navier–Stokes equation reduces to the Stokes equation by defining a stream function and the diffusion equation becomes the Laplace equation. The general solutions of the Stokes and Laplace equations in the bipolar coordinate have been found by Stimson and Jeffery [7,8]. Based on the work of Stimson and Jeffery and applying appropriate boundary conditions, the motion of two droplets driven by Marangoni convection has been analytically studied by Golovin et al. [9].

The Stokes equation is for steady state fluids where temporal evolution of the velocity is not involved. In the context of hydrostatic equilibrium, the pressure difference across a fluid–fluid interface is given by the Young–Laplace equation. For non-uniform surface tension, the system is not in hydrostatic equilibrium any more and the pressure distribution around the droplet evolves with time and, in general, does not follow the Young–Laplace equation. This effect has not been considered in the analysis of Golovin et al. and will be treated in our work. Moreover, a stress tensor (often called capillary tensor) derived from Noether's theorem

$$\underline{\underline{\mathbf{\Theta}}} \propto (\nabla c)^2 \mathbf{I} - \nabla c \otimes \nabla c,$$

where $\mathbf{I}$ is the identity tensor and c is the molar concentration, is proposed in Ref. [10] to be included in the Navier–Stokes equation to compute the Marangoni convection. This stress tensor or its equivalent formats have been widely used for different setups [11–14]. In the analysis of Golovin et al., the stress balance condition at a fluid–fluid interface was applied to solve the Laplace and Stokes equations. When the fluid–fluid interface evolves with time, it would be high intricate to apply the stress balance condition at each time step. This problem can, however, be solved by including the capillary tensor in the Cahn–Hilliard–Navier–Stokes model.

In the first part of our investigation, we analytically study the motion of two droplets driven by solutal Marangoni convection in a bipolar coordinate by referring to the work of Golovin et al. [9]. The particular solutions for the Laplace and Stokes equations are found by choosing appropriate matching conditions at the surface of the droplets. The bridge for the Laplace and Stokes equations is the tangential force balance between the viscosity stress and the Marangoni force at the surface of each droplet. The isolines for the concentration and the stream lines affected by the spacing between droplets and by the radius of the droplets have been investigated.

In the second part of our work, we study the motion of the droplets by employing the combined Cahn–Hilliard and Navier–Stokes (CHNS) equations, regarding that the analytical solution is, however, for the steady state where there is no temporal evolution for the concentration, velocity and pressure. In contrast to the analytical model in the first part, the advantages of the CHNS model are that (i) it captures a diffusive-physical interfacial width in the scale of nanometer, (ii) by writing the capillary tensor as one force term in the Navier–Stokes equation it can avoid the explicit-application of the matching boundary conditions, e.g. stress balance, at the fluid–fluid interface, and (iii) it is more realistic since the temporal evolution in the diffusive path is evolved. As the analytical solution for the CHNS model is non-trivial, we solve the CHNS model numerically on a staggered mesh. The most important result we find is that whether the droplets approach or repel each other depends on the radius of the droplet and the Marangoni number, which has not been found in the analytical solution. This phenomenon is explained in terms of dynamic pressure affected by Marangoni number and the radius of the droplet. We further study the motion of two unequally-sized droplets affected by the ratio of their radii.

The paper is structured as follows: In Section 2, we present the analysis. The numerical model is described in Section 3. Section 4 concerns the simulation results and discussion. The conclusion and remarks are given in Section 5.

2. Analysis

In the analysis, we assume undeformable interface and the droplet is in the form of a sphere or a nearly sphere. The capillary number $C := \nu U_c/\sigma$, where ν is the viscosity, U_c is the characteristic velocity and σ is the interfacial tension, measures a ratio of the viscosity force to the interfacial tension force. It is an indicative parameter for the degree of deformation. For $C \ll 1$, the droplet stays in the form of a sphere or a nearly sphere [15]. The characteristic velocity U_c is evaluated by D/d_0 where D is the diffusivity and d_0 is the capillary length. With $D \sim 1\times10^{-9}$ m^2/s, $d_0 \sim 1\times10^{-9}$ m, $\sigma \sim 0.1$ J/m^2 and $\eta \sim 1\times10^{-3}$ Pa s, the capillary number C is about 10^{-4}, which is much less than 1. Based on this, we study the motion of two nearly spherical droplets in the bipolar coordinate.

2.1. Bipolar coordinate

The relation between the bipolar coordinate (ϱ, φ) and the cylindrical coordinate (z, x) is defined by [16]

$$z + ix = iq \cot\left[\frac{1}{2}(\varphi + i\varrho)\right], \quad \varphi \in [-\pi, \pi],\ \varrho \in (-\infty, \infty), \quad (1)$$

where q is a positive constant. Eq. (1) yields

$$z = \frac{q \sinh\varrho}{\cosh\varrho - \cos\varphi}, \qquad x = \frac{q \sin\varphi}{\cosh\varrho - \cos\varphi}. \quad (2)$$

By eliminating φ in Eq. (2), we have

$$(z - q\coth\varrho)^2 + x^2 = \left(\frac{q}{\sinh\varrho}\right)^2,$$

which defines a circle with center at $(q\coth\varrho,\ 0)$ and radius $q/\sinh\varrho$. For the left droplet with radius b and the right droplet with radius a, we have

$$a = \frac{q}{\sinh\alpha}, \qquad b = \frac{q}{\sinh\beta},$$

where α and β denote the surface of the right and left droplets in the bipolar coordinate, as shown in Fig. 1. The separation distance between the two droplets is given by

$$\begin{aligned} d &= \left(q\coth\alpha - \frac{q}{\sinh\alpha}\right) - \left(-q\coth\beta + \frac{q}{\sinh\beta}\right) \\ &= a(\cosh\alpha - 1) + b(\cosh\beta - 1). \end{aligned}$$

Choosing a as the dimensionless factor for space, we get the new relation between the bipolar coordinate and the cylindrical coordinate

$$z = \frac{\sinh\alpha \sinh\varrho}{\cosh\varrho - \cos\varphi}, \qquad x = \frac{\sinh\alpha \sin\varphi}{\cosh\varrho - \cos\varphi}.$$

The separation distance is now expressed as

$$d = (\cosh\alpha - 1) + \frac{b}{a}(\cosh\beta - 1).$$

Realizing that $\cosh^2\alpha = 1 + \sinh^2\alpha$, we get the following relation between α in the bipolar coordinate and the separation distance and the ratio of the radii

$$\cosh\alpha = \frac{(d+1+r)^2 + 1 - r^2}{2(d+1+r)},$$

$$\cosh\beta = \frac{(d+1+r)^2 - 1 + r^2}{2r(d+1+r)},$$

where $r = b/a$. In our analysis, without loss of generality, we assume that the bigger droplet is the one with radius b, thus $r \geq 1$.