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Terrestrial and microgravity experiments on onset of oscillatory thermocapillary-driven convection in hanging droplets

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ABSTRACT

Thermocapillary-driven convection in a hanging droplet is experimentally investigated under normal- and micro-gravity conditions. A droplet is hung on a heated cylindrical rod facing downward, and another rod cooled is placed just beneath the droplet to create the designated temperature difference between both ends of the droplet. A transition of the flow field from a two-dimensional axisymmetric 'steady' flow to three-dimensional time-dependent 'oscillatory' ones by increasing temperature difference is realized. The oscillatory flow is accompanied with thermal waves due to so-called hydrothermal wave (HTW) instability, which propagates over the free surface at a constant frequency. The present study aims to understand the transition conditions of the flow from the steady to the oscillatory ones, and to characterize the convective field inside the droplet with the HTW by imposing a range of temperature differences. We discuss the transition condition and the flow fields in the oscillatory regimes after the transition obtained in the terrestrial and on-orbit experiments in 'Kibo,' the Japanese Experiment Module aboard the International Space Station.

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1. Introduction

A thermocapillary effect is realized when there is a surface tension gradient along a liquid-gas interface due to the temperature dependence of the surface tension $\partial\gamma/\partial T$, where γ is the surface tension and T the temperature. Along the liquid-gas interface with a non-uniform temperature field, the surface tension difference drives the fluid from the higher temperature region toward the lower temperature one if $\partial\gamma/\partial T$ has a negative value. The resulting convection is called thermocapillary-driven convection or Marangoni convection. Such convection is observed in numerous technological applications such as in the processes of crystal growth [1]. Among various geometries of the crystal growth methods, we focus on the thermocapillary-driven flow inside a hanging droplet formed on a heated rod (Fig. 1). Such a geometry has been employed especially in protein crystal growth processes known as a hanging drop method [2–6]. It has been indicated that the induced flow inside the hanging droplet does affect the quality of the crystal.

The intensity of the flow induced by the thermocapillary effect is generally described by using a non-dimensional number of Marangoni number $\mathcal{M}$, defined as

$$\mathcal{M} = \frac{|\gamma_T|\Delta TL}{\rho\nu\kappa} = \mathcal{RP}, \tag{1}$$

where ρ, ν and κ are the density, kinematic viscosity and thermal diffusivity of the test fluid, respectively, $\gamma_T = \partial\gamma/\partial T$ is a temperature coefficient of surface tension, ΔT is the temperature difference to which the droplet is exposed, L is the characteristic length of the target geometry, $\mathcal{R}$ is the Reynolds number defined as $(|\gamma_T|\Delta TL)/(\rho\nu^2)$ and $\mathcal{P}$ is the Prandtl number defined as ν/κ.

The behaviors of the suspended particles inside the hanging droplet due to the thermocapillary-driven flow are discussed [7] to indicate unique particle behaviors known as the particle accumulation structure, PAS, after Schwabe et al. [8]. Takakusagi and Ueno [7] indicated the flow patterns in terms of the behaviors of suspended particles in the hanging droplet by changing the

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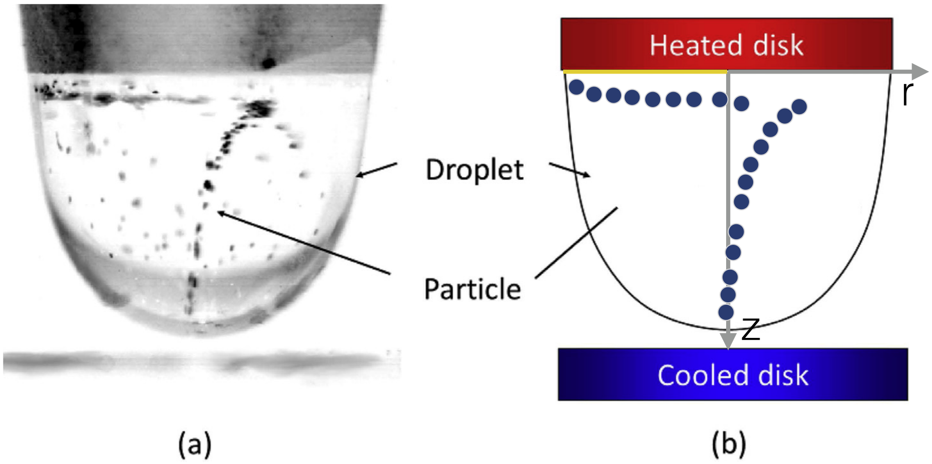

Fig. 1. Target geometry of a hanging droplet: (a) snapshots and (b) their schematic views observed from side.

intensity of the thermocapillary effect (or by changing $\mathcal{M}$) in a series of terrestrial experiments. They categorized the induced flows into the state of the two-dimensional (2D) axisymmetric steady flow and the states of the three-dimensional (3D) oscillatory flow states, and indicated the thresholds between those flow states. The 3D oscillatory flows can be further categorised into 'Standing wave' and 'Travelling wave' judging from the behavior of suspended particles in the droplet and the variation of its surface temperature. Such a thermal wave traveling over the free surface is induced by so-called hydrothermal wave (HTW, hereafter) instability, a phenomenon shown by Smith and Davis [9] in a thin liquid film of high-$\mathcal{P}$ fluid exposed to a temperature gradient over the free surface.

It has been also known that the evaporation of the fluid from the droplet induces convective motion inside the droplet as well in drying process of the sessile droplets settled on a substrate [10]. In such a case, heat transfer via latent heat as well as sensitive heat results in more complex convection due to the thermocapillary and evaporation effects [11–19]. Even when one pays attention to pure thermocapillary-driven convection, comprehensive mechanism of the induced convection and resultant suspended particles motions has not fully understood. Recently the thermocapillary and buoyant flow in sessile and hanging droplets for a wide range of liquid materials, droplet shapes, levels of gravity, temperature differences as well as heat-transfer coefficients between the liquid and the ambient atmosphere was numerically investigated by Masoudi and Kuhlmann [20]. The flow field and the accompanying particles' behaviors induced by the thermocapillary effect, however, have not been characterized in this geometry. We aim to characterize the flow field and the accompanying particles' behaviors in order to apply this method not only to the high-quality protein crystal growth, but also to techniques of mixing and stirring particles without using an external force in a closed system in a hanging droplet.

We have carried out on-orbit experiments on the thermocapillary-driven flow in the half-zone liquid bridge of higher $\mathcal{P}$ fluid and of larger size than those employed in the terrestrial experiments [21–27]. These series of microgravity experiments, called Marangoni Experiment in Space (MEIS), had been conducted as the first scientific experiments on the Japanese Experiment Module 'Kibo' aboard the International Space Station (ISS) since 2008. The main objectives of the project MEIS are

1. to determine the threshold of the flow transition from 2D steady to 3D oscillatory flows due to the thermocapillary effect [21,22,26],
2. to measure 3D flow structures of oscillatory flows [21,25,28], and
3. to illustrate the transition route to chaotic flows [23,24],

in a geometry of large-scale half-zone liquid bridge of high-$\mathcal{P}$ fluids. It is emphasized that stable and long-duration microgravity conditions bring us a great benefit to conduct a series of experiments with a geometry of larger characteristic length than that in the terrestrial experiment; In the terrestrial experiment, we have to limit ourselves to handling a droplet with smaller characteristic length in order to prevent a significant deformation due to the gravity force. In order to realize a high-$\mathcal{M}$ condition under a small L, we have to add larger temperature difference ΔT as indicated in Eq. (1), which would result in significant evaporation of the test fluid. If one employs higher $\mathcal{P}$ fluid under smaller L, one needs to add a larger ΔT in order to realize the same value of $\mathcal{M}$. That is why we restrict ourselves to using rather small $\mathcal{P}$ fluid under normal gravity. In the space experiment, on the other hand, one may employ larger L without static deformation, so that ΔT maintains relatively small even with higher $\mathcal{P}$ fluid. This is one of the substantial benefits of the on-orbit experiments in the International Space Station. In the MEIS project, five series of experiments, MEIS-1, MEIS-2, MEIS-3, MEIS-4 and MEIS-5 were carried out in the Japan fiscal year of 2008, 2009, 2011, 2010 and 2012, respectively. During the experiments in the MEIS-1, a hanging droplet on a heated rod was realized and the suspended particles in the heated droplet accumulate on a single line like a whip as observed in a small-scale droplet [7]. We carried out another experiment of hanging droplet in MEIS-3, and observed similar flow regimes in the droplet to those under normal gravity conditions.

In the present study, we investigate the flow transition condition from the 2-D steady flow to 3-D oscillatory one, and of the fundamental frequency of the oscillatory flows through the terrestrial and on-orbit experiments.

2. Experiment

A hanging droplet as shown in Fig. 1 is the target geometry in the present study. We prepare a droplet sustained on the heated rod, whose end surface has a radius R. And the co-axial rod of the same R is placed facing the tip of the droplet (see Appendix A). The experimental apparatus for the terrestrial experiments is described in Fig. 2. The basic structure of the terrestrial system was described by Takakusagi and Ueno [7]. Two sets of optical systems were employed in the present study to observe temperature

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