



## Counter-current motion of a droplet levitated on a liquid film undergoing Marangoni convection



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### ABSTRACT

This paper experimentally and analytically investigates the motion of a levitated droplet against the Marangoni flow in an immiscible outer fluid. Based on our earlier experiments, when a droplet is released from a height  $\sim 1.5$ –4 times its diameter from the liquid surface, it can overcome the impact and stay levitated at the liquid–air interface due to the existence of an air gap between the droplet and the liquid film. Surprisingly, such a levitated droplet, moves toward the heating source against the Marangoni convection. In order to explain this behavior, we propose a simple approach: first, the Marangoni convection inside the thin film is considered without the droplet floating on the surface. By using a level-set method and solving the Navier–Stokes equation, the free surface velocity and deformation are calculated. Then, these quantities are used to solve for droplet velocity and drag coefficient simultaneously using a force balance. In order to compare the simulation results, experiments with levitated water droplets on an immiscible carrier liquid, FC-43, are conducted for various temperature gradients and droplet velocities are measured at different locations using high-speed imaging. The experimental results are in good agreement with the developed theoretical model. For a Reynolds number range of 2–32, it is shown that the drag coefficients are up to 66% higher than those for the fully immersed sphere at the same Reynolds numbers. Finally, a correlation is proposed to calculate the drag coefficient of levitated droplets for various temperature drops across the channel.

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### 1. Introduction

Surface tension gradient caused by a temperature gradient on a liquid surface leads to an imbalance of tangential stresses at the fluid interface. Due to this surface tension gradient in the tangential direction, an unbalanced force distribution at the interface is created, which starts the fluid motion. This phenomenon is called the Marangoni effect. Several studies have numerically modeled the Marangoni convection during heating and evaporation of droplets [1,2]. The main focus of these studies were the effect of thermo-physical properties, phase change and contact line dynamics on droplet evaporation. However, the current study focuses on the surface distortion created by local heating of a thin sheet of liquid, which creates Marangoni convection. This distortion of the thin liquid sheet is due to the higher surface tension of colder liquid which pulls the warmer surrounding fluid with lower surface tension. There have been several investigations that take advantage of thermocapillary drift to actuate droplets on solid surfaces, however in all these works even at high temperature

gradients the droplet velocity was low, i.e. microns per second, and the motion was in the direction of decreasing temperatures [3,4]. The non-uniformity of the spreading coefficient ( $S = \sigma_{\text{carrier}} - \sigma_{\text{carrier-drop}} - \sigma_{\text{drop}}$ , where  $\sigma$  indicates interfacial tension) could be due to chemical agents on the solid surface, thermal gradients or electrical charge, and any of these could result in droplet motion. Issues such as droplet pinning or droplet evaporation has motivated researchers to use liquid platforms instead of solid substrates where droplets would be dispensed on a layer of immiscible liquid and transported by means of thermal gradients [5].

The thermocapillary drift of bubbles and droplets in reduced gravity was investigated thoroughly [6–9]. Zhang et al. [7] found the crucial role of inertia in determining an asymptotic solution for temperature field when a drop moves in a vertical temperature gradient at small values of thermal Péclet number. Marangoni flotation of liquid droplets was studied experimentally and numerically by Dell'Aversana et al. [8]. Their results suggested that surface tension gradient helps to form a stable air film in between the droplet and pool surface which prevents sinking of the droplet. Greco and Grigoriev [10] investigated thermocapillary driven motion of interfacial droplets and found that in some cases, the droplet suspended at an interface of two liquid layers migrated

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in the direction opposite to the temperature gradient. Samareh et al. [11] used the volume of fluid method to simulate thermocapillary migration of a deformable drop in immiscible fluids with variable surface tension. Wegener [12,13] numerically studied the effect of droplet diameter, kinematic viscosity ratio and partition coefficient on the mass transfer of a solute in a Marangoni dominated liquid–liquid system. Wu [14] under the quasi-steady state assumption modeled the thermocapillary droplet migration in a uniform temperature gradient at large Reynolds and Marangoni numbers.

Our preliminary experimental observations [15–17] showed for the first time that droplets can move in either direction of a thermal gradient, depending on its shape at the air–liquid interface. Infra-red thermal imaging was used to determine the surface temperature of the carrier fluid. It was demonstrated that a spherical drop moved towards the heat source, and a sessile drop away from it. By using a simple analysis, it was conjectured that spherical drops would roll down the free surface slope due to gravity.

In the current study, using detailed modeling supported by experiments, it will be shown that the liquid surface deforms due to a temperature gradient, and that a levitated drop dispensed on the liquid surface moves down the slope with an air gap underneath, and does not roll down. The drops that do not levitate are submerged, revealing only a lens shaped surface above the fluid surface. Since the sessile drop has been investigated extensively in the literature, in this paper, we focus on levitated droplets. First, we provide a theoretical foundation to solve the Marangoni convection in the absence of droplet and obtain solutions for deformation at the free surface and free surface velocity. Using Marangoni solution for the slope of the liquid layer height and surface velocity, we then provide a model based on force balance to iteratively solve for drag coefficient and velocity of the droplet. The droplet velocity profile is then compared with the experimental profiles for four temperature gradients.

## 2. Experimental setup

A thin layer of Perfluorotributylamine (FC-43, 3 M) is formed in a rectangular acrylic mini-channel. The dimensions of the experimental channel are  $D = 2$  mm deep,  $W = 8.5$  mm wide and  $L = 55$  mm long. Thermal gradients were created by passing electric current through Nichrome wire heater (28 Gauge, 4.15  $\Omega$ /ft) as heating element (Fig. 1). In order to obtain temperature field across the platform a high-resolution, high-speed infrared thermal imaging system was used. Due to thermal capacitance, the time scale for the droplets to acquire the temperature of the surrounding fluid is larger than the time scale over which droplets move along the length of the platform; hence they are essentially not at the same temperature as the surrounding liquid.

Infrared thermal imaging measurement technique provides spatially and temporally resolved temperature of surfaces

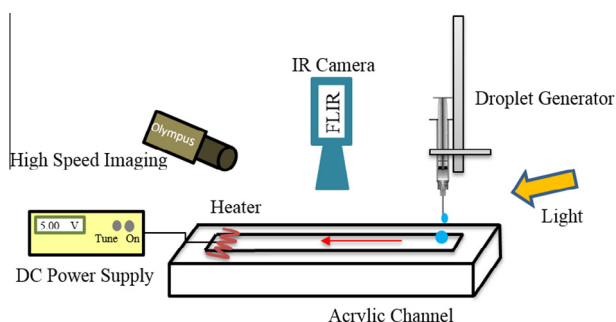


Fig. 1. Schematic of experiment set-up.

non-invasively. The infrared camera (SC 5000, FLIR) used in this study works based on InSb (Indium Antimonide – narrow band gap semiconductor sensor) with a spectral response range of 2.5–5  $\mu\text{m}$  in wavelength. The detector array provides a  $640 \times 512$  pixel digital image where each pixel is  $15 \mu\text{m} \times 15 \mu\text{m}$ . The temperature range used in the current experiment is 25–60  $^{\circ}\text{C}$  and this uses an integration time of 1.17 ms. The IR camera was operated at 100 fps with measurement accuracy of  $\pm 1$   $^{\circ}\text{C}$ . The IR camera captures surface temperature of the liquid depending on the intensity of thermal radiation recorded by the infrared sensor within the camera. The emissivity of the surface can be input into the software used for data processing. The emissivity used was 1.0. For a change of emissivity of  $\pm 0.03$ , temperature changes by 0.3  $^{\circ}\text{C}$ . Through Fourier Transform Infrared Spectroscopy (FTIR) it was confirmed that the absorption coefficient does not change by more than 1% for the fluids used in this study. A set of precision dispensing needles with various tip size (36G–23G) were used to release pendant droplets of deionized water manually on the surface of FC-43. The physical properties of the fluids used in the experiments are given in Table 1.

## 3. Results and discussion

### 3.1. Experimental levitation of spherical droplets

Some preliminary experiments were done to levitate a spherical droplet on a liquid surface and preserve its shape [17]. When the water droplet is released from a certain critical height, it is able to retain its spherical shape, while the liquid sheet below the droplet stretches. In order to create spherical droplets at the air–liquid interface, they were released as free falling droplets on to the liquid surface from different heights. Upon impact, an air gap develops between the drop and the surface that sustains the weight of the droplet. The entrainment and drainage of air in the thin gap between liquids is due to the relative motion of the fluid interfaces, which itself is generated due to surface tension gradients across the interfaces. It has been shown that a droplet can be kept floating on the surface of a liquid pool by maintaining a temperature difference between the droplet and the liquid pool [18]. Bearing action drags air in between the drop and liquid surface and the resultant pressure build-up supports the drop. Generally, non-coalescence of droplets is attributed to the existence of an air gap [19–21]. Through thermocapillary effects or vibration effects [22], a droplet can be kept levitating on the surface of a liquid pool. The fact that an active external source is required to prevent the droplet from collapsing into the underlying pool suggests that the air gap is continuously drained and replenished.

In the current work, not only is the drop levitated above the liquid pool without collapsing or immersing but also is propelled by the imposed thermal gradients in the desired path. Thus, the pressure buildup due to entrainment of flow and the relative motion of fluidic interfaces explains the longevity of the levitated droplets.

Based on simple solutions to energy equation which show exponential decay in temperature, the current temperature field was normalized and curve-fitted to take the form of an exponentially decaying function:  $\theta = a \cdot \exp(bx)$ ; typical values for coefficients from the current experimental data are  $a = 1$  and  $b = -1.7$ . As shown in Fig. 2, the thermal gradient drops gradually away from the heater.

Experiments were carried out for migration of spherical droplets 3 mm (volume  $\sim 14 \mu\text{L}$ ) in transient and steady thermal gradient conditions. The temperature differences along the channel were 25 $^{\circ}$ , 15 $^{\circ}$ , 10 $^{\circ}$  and 5  $^{\circ}\text{C}$ . In transient condition, the platform was initially at room temperature ( $\sim 19$   $^{\circ}\text{C}$ ) and the heater was turned on and the thermal gradient experienced by the droplet was recorded by evaluating the curve fitted to temperature

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