Experimental Study on Marangoni Convection Instability Induced by Evaporation of Silicone Oil Droplets on a Heated Substrate (Postprint)

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Abstract

Experimental observations were made of the Marangoni convection instability phenomenon and its evolution during the evaporation of 0.65cSt silicone oil droplets on a heated substrate. The critical conditions for Marangoni convection instability were measured, and the effects of wetting radius and Ma number on the stability of Marangoni convection within droplets were analyzed. The results show that Bénard-Marangoni convection cells only emerge within the droplet when the contact angle decreases below a certain critical value. The cells exhibit a "petal" morphology—pointed and slender at the droplet apex, arc-shaped near the three-phase contact line, and linear where cells compress each other. As evaporation proceeds, the cells become shorter and thicker. The number of cells increases with increasing wetting radius and Ma number. The critical contact angle for the formation of Bénard-Marangoni convection cells increases with Ma number. The droplet edge consistently exhibits thermocapillary convection without Bénard-Marangoni convection cells.

Full Text

Experimental Investigation on Marangoni Convection Instability Induced by Evaporation in a Sessile Silicone Oil Droplet on a Heated Substrate

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Abstract

This study experimentally investigates the Marangoni convection instability and its evolution in evaporating 0.65 cSt silicone oil droplets on a heated substrate. The critical conditions for Marangoni convection instability were measured, and the effects of wetting radius and Ma number on the stability of Marangoni convection within the droplet were analyzed. Results reveal that Bénard-Marangoni (BM) convection cells emerge only when the contact angle decreases below a critical threshold. These cells exhibit a distinctive "flower-like" morphology: sharp and slender at the droplet apex, circular-arc shaped near the three-phase contact line, and straight where adjacent cells compress each other. As evaporation proceeds, the cells become shorter and thicker. The number of cells increases with both wetting radius and Ma number. The critical contact angle c for BM convection onset increases with increasing Ma number. Thermocapillary convection persists at the droplet edge without forming BM convection cells.

Keywords: Droplets, evaporation, thermocapillary convection, Bénard-Marangoni convection, stability

Introduction

Droplet evaporation is a ubiquitous phenomenon in nature with significant implications for practical applications including spray cooling, atomization combustion, inkjet printing, and DNA molecular imaging [?]. Recent studies have shown that during the main stage of droplet evaporation, the evaporation rate is faster at the droplet edge than at the apex, creating non-uniform surface temperature distribution and surface tension gradients that induce Marangoni convection within the droplet [?]. Early experimental work by Zhang and Xu [?] at Tsinghua University investigated evaporating volatile organic droplets on heated surfaces, identifying distinct flow regions including convection zones, Bénard convection zones, stagnation zones, and edge zones. Ren et al. [?] subsequently suggested through numerical simulation that Marangoni convection could become unstable during rapid droplet evaporation.

Deegan et al. [?] proposed that Marangoni convection is responsible for the formation of coffee-ring deposits from dried liquid drops containing solid particles. Hu and Larson [?] discovered that thermocapillary flow direction reverses when the contact angle falls below 14°, transitioning from edge-to-apex flow at larger angles to apex-to-edge flow at smaller angles. Xu and Luo [?] identified a stagnation point on the droplet surface where flow directions diverge on either side. Jin and Hu [?] observed that evaporating droplets develop a pair of vortices that persist longer at lower substrate temperatures but gradually disappear as temperature increases. Girard et al. [?] found that when the heating radius exceeds the droplet wetting radius, the three-phase contact line temperature is higher than the droplet apex temperature, and vice versa. Ghasemi and Ward [?] demonstrated that thermocapillary convection plays a crucial role in energy transport during droplet evaporation, particularly near the three-phase contact line.

More recently, Sefiane, Brutin, and colleagues [?] employed infrared thermography to observe hydrothermal waves resulting from unstable Marangoni convection in evaporating methanol, ethanol, and FC-72 droplets with wetting radii smaller than the capillary length, analyzing the variation of wave number during the evaporation process.

Existing research has primarily focused on highly volatile droplets, attributing Marangoni convection instability to thermocapillary flow driven by tangential temperature gradients at the droplet surface—characterized as hydrothermal waves. However, the curved free surface of a droplet inevitably generates both radial normal temperature gradients and surface tangential temperature gradients that are continuously coupled throughout the evaporation process. Whether normal temperature gradients can induce Bénard-Marangoni (BM) convection within droplets remains poorly understood. This study addresses this knowledge gap by investigating Marangoni convection in less volatile silicone oil droplets evaporating on heated substrates, with particular emphasis on experimental observation of BM convection instability characteristics and analysis of the influences of wetting radius and substrate temperature.

Experimental Setup

A schematic of the experimental apparatus is shown in Figure 1 [Figure 1: see original paper]. The circular substrate, machined from copper with a polished surface coated with superhydrophobic material, was mounted on a cylindrical acrylic chamber connected to a constant-temperature oil bath (PP07R-20-A12Y, PolyScience, temperature control accuracy ±0.005°C) for substrate temperature regulation. The actual substrate temperature was measured using four K-type thermocouples. Three leveling screws ensured horizontal alignment of the platform.

The aluminum particle tracer method was employed to visualize Marangoni convection instability within droplets. Prior to experiments, a small amount of aluminum powder (0.1 wt%) was mixed into the silicone oil and stirred uniformly. Several microliters of this mixture were carefully deposited onto the substrate using a micro-syringe to form spherical cap-shaped droplets. A microscope positioned directly above the droplet enabled real-time observation and recording of Marangoni convection instability phenomena. Contact angles were measured using a contact angle goniometer (JC2000DM, accuracy ±0.1°), while wetting radii were determined from microscope images. The working fluid was 0.65 cSt silicone oil with properties listed in Table 1 . The minimal aluminum powder concentration ensured negligible effects on fluid properties. In all experiments, the droplet wetting radius remained smaller than the capillary length, rendering buoyancy effects negligible. Ambient temperature was controlled by air conditioning, and relative humidity was measured using a wet-and-dry bulb thermometer.

Contact angle measurements were repeated four times per experiment and averaged, with maximum deviation from the mean defined as measurement error. Thermocouples were calibrated against the constant-temperature oil bath across the range 10–50°C prior to experiments, reducing maximum error to 0.027%.

Experimental Results and Analysis

Evolution of Bénard-Marangoni Convection Cells

For a droplet with wetting radius r = 2.14 mm and initial contact angle = 41.15° evaporating under conditions of Ma = 8.146 × 10 (Ma = _TΔTr/ , where ΔT = T_w - T_a), ambient temperature T_a = 295.95 K, and relative humidity RH = 56%, the Marangoni convection at substrate temperature T_w = 298.95 K is illustrated in Figure 2 [Figure 2: see original paper]. Initially, tracer particles revealed symmetric fluid flow from the three-phase contact line toward the droplet apex and then downward to the droplet base, representing stable thermocapillary convection without instability (Figure 2.a). At t = 18 s, "flower-like" vortex cells emerged within the droplet (Figure 2.b). For each "petal," fluid flowed upward from the edges toward the center and then descended into the droplet interior—characteristic of Bénard-Marangoni (BM) convection. The cell morphology differed from the hexagonal cells in flat liquid layers: cells were sharp and slender at the droplet apex, circular-arc shaped near the three-phase contact line, and straight where adjacent cells compressed each other.

As evaporation progressed, the number of BM cells gradually increased. By t = 26 s (Figure 2.c), 13 cells were uniformly distributed across the droplet surface, extending from the apex to near the three-phase contact line. However, no cells formed at the droplet edge, where thermocapillary convection persisted. Droplet thinning during evaporation caused progressive morphological changes: by t = 32 s (Figure 2.d), cells had become shorter and thicker. At t = 45 s (Figure 2.f), the droplet was sufficiently thin that the cells resembled the hexagonal pattern of flat liquid layers before gradually disappearing as evaporation completed.

Critical Contact Angle for BM Convection Onset

As evident from Figures 2(a) and 2(b), BM convection cells do not appear immediately but only when the contact angle decreases below a critical value. To determine this critical contact angle c under various Ma numbers, experiments were conducted with controlled droplet volume V, wetting radius r, and ambient temperature T_a, while varying substrate temperature T_w to modify the Ma number. The measured critical contact angles are presented in Figure 3 [Figure 3: see original paper], showing that c increases with increasing Ma number.

This relationship arises because both normal and tangential temperature gradients coexist within evaporating droplets. Tangential gradients drive thermocapillary convection that transports heat from the three-phase contact line to the apex, reducing the normal temperature difference and suppressing BM convection. At small contact angles, the droplet becomes flatter, resembling a thin liquid layer with reduced tangential temperature gradients, particularly near the apex, where thermocapillary convection weakens and BM convection can more readily develop. Therefore, BM convection emerges only when the contact angle falls below a threshold value for a given Ma number. As Ma number increases, the enhanced normal temperature gradient favors BM convection formation, enabling it to dominate over thermocapillary convection even at higher contact angles, thus raising the critical contact angle.

Effects of Wetting Radius and Ma Number

For droplets with different wetting radii, cell morphology showed no significant differences, but the maximum cell number varied substantially. Under conditions of T__w = 303.15 K, T__a = 295.15 K, and RH = 60%, the maximum cell numbers were 19 for r = 1.81 mm (Figure 4 [Figure 4: see original paper].a), 23 for r = 2.73 mm (Figure 4.b), and 26 for r = 3.23 mm (Figure 4.c). Evidently, larger wetting radii accommodate more cells due to greater droplet capacity.

At constant wetting radius, the maximum cell number n also varies with Ma number, as shown in Figure 5 [Figure 5: see original paper]. The maximum cell number increases with Ma number because higher Ma numbers produce larger normal temperature gradients that promote BM convection formation, thereby generating more cells.

Conclusions

Through experimental investigation of Marangoni convection instability in 0.65 cSt silicone oil droplets on heated substrates, this study observed the evolution of Bénard-Marangoni convection cells, measured their instability onset conditions, and examined the effects of wetting radius and Ma number on convection stability. The key findings are:

(1) Bénard-Marangoni cells emerge only when the contact angle decreases below a critical threshold. These cells exhibit a distinctive “flower-like” morphology—sharp and slender at the droplet apex, circular-arc shaped near the three-phase contact line, and straight where cells compress each other. During evaporation, cells become progressively shorter and thicker. Thermocapillary convection persists at the droplet edge without forming BM cells.

(2) The critical contact angle c for BM convection onset increases with Ma number. Under identical conditions, droplets with larger wetting radii develop more cells due to greater accommodation capacity. Cell number also increases with Ma number.

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