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

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Abstract

The experiments observed the Marangoni convection instability phenomena and their evolution during the evaporation of 0.65cSt silicone oil droplets on a heated substrate, measured the critical conditions for Marangoni convection instability, and analyzed the effects of wetting radius and Ma number on the stability of Marangoni convection within droplets. The results revealed 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-like" morphology, being relatively sharp and slender at the droplet apex, arc-shaped in the vicinity of the three-phase contact line, and straight where adjacent cells compress each other. As evaporation progresses, the cells become shorter and thicker. The number of cells increases with increasing wetting radius and Ma number. The critical contact angle c for the generation of Bénard-Marangoni convection cells increases with increasing Ma number. At the droplet edge, thermocapillary convection persists without the formation of Bénard-Marangoni convection cells.

Full Text

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

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Abstract

This study experimentally investigates 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 the droplet were analyzed. Results show that Bénard-Marangoni convection cells only emerge when the contact angle decreases below a critical value. These cells exhibit a "flower-like" morphology: sharp and slender at the droplet apex, circular-arc shaped near the three-phase contact line, and straight where cells compress against each other. As evaporation progresses, the cells become shorter and thicker. The number of cells increases with both wetting radius and Ma number. The critical contact angle c for Bénard-Marangoni convection onset increases with increasing Ma number. Thermocapillary convection persists at the droplet edge without forming Bénard-Marangoni cells.

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

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0 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 [1]. Recent studies have revealed that during the main stage of droplet evaporation, the evaporation rate is faster at the edge than at the apex, leading to non-uniform surface temperature distribution and consequently surface tension gradients that induce Marangoni convection within the droplet [2]. Early experimental work by Zhang Nengli and Xu Youren at Tsinghua University [3] investigated evaporation of volatile organic droplets on heated surfaces, identifying distinct flow regions within the droplet: convection zones, Bénard convection zones, stagnation zones, and edge zones. Ren Zepei et al. [4] subsequently proposed through numerical simulation that Marangoni convection may become unstable during rapid droplet evaporation. Deegan et al. [5] suggested that Marangoni convection is responsible for the coffee-ring deposition pattern formed by dried liquid drops containing solid particles. Hu and Larson [6] discovered that thermocapillary flow direction reverses when the contact angle falls below 14°, flowing from the edge to the apex at larger angles and in the opposite direction at smaller angles. Xu and Luo [7] identified a stagnation point on the droplet surface where flow directions diverge. Jin Zheyan and Hu Hui [8] observed that a pair of vortices forms during droplet evaporation, persisting longer at lower substrate temperatures but gradually disappearing as temperature increases. Girard et al. [9] found that when the heating radius exceeds the droplet wetting radius, the three-phase contact line temperature is higher than the droplet apex temperature, with the reverse occurring when the heating radius is smaller. Ghasemi and Ward [10] 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 [11-16] used infrared thermography to observe hydrothermal waves resulting from Marangoni convection instability in evaporating methanol, ethanol, and FC-72 droplets with wetting radii smaller than the capillary length, analyzing the variation of wave numbers during evaporation.

Review of existing literature reveals that current research primarily focuses on volatile droplets, attributing Marangoni convection instability mainly to thermocapillary convection driven by tangential temperature gradients at the droplet surface—characterized as hydrothermal waves. However, droplets possess curved free surfaces where radial normal temperature gradients and surface tangential temperature gradients always coexist and couple during evaporation. Whether Bénard-Marangoni convection (hereinafter referred to as BM convection) induced by normal temperature gradients can occur in droplets remains poorly understood. To address this gap, the present study investigates Marangoni convection induced by evaporation of low-volatility silicone oil droplets 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.

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1 Experimental Apparatus and Methods

The experimental apparatus is schematically illustrated in Figure 1 [Figure 1: see original paper]. The circular substrate, machined from copper with a polished surface coated with superhydrophobic material, is 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 is measured by four K-type thermocouples. Three leveling screws ensure horizontal alignment of the platform. Aluminum powder tracer method is employed to visualize Marangoni convection instability within the droplet.

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

For contact angle measurements, four readings are taken and averaged in each experiment, with the maximum deviation from the mean defined as measurement error. Thermocouples are calibrated against the constant-temperature oil bath across the range of 10–50°C before experiments, reducing maximum error to 0.027%.

2 Experimental Results and Analysis

2.1 Evolution of Bénard-Marangoni Convection Cells

For a droplet with wetting radius r = 2.14 mm and initial contact angle = 41.15°, at Ma = 8.146 × 10 (Ma = _TΔTr/ , where ΔT = T_w - T_a), ambient temperature T_a = 295.95 K, relative humidity RH = 56%, and substrate temperature T_w = 298.95 K, the Marangoni convection phenomenon is shown in Figure 2 [Figure 2: see original paper]. Initially, tracer particles reveal symmetric flow from the three-phase contact line toward the droplet apex and then downward to the bottom—stable thermocapillary convection without instability. At t = 18 s, "flower-like" vortex cells emerge within the droplet (Figure 2.b). For each "petal," fluid flows upward from the edges to the center and then downward into the droplet interior, consistent with Bénard-Marangoni convection characteristics. Careful observation reveals morphological differences from hexagonal cells in flat liquid layers: the cells are sharp and slender at the droplet apex, circular-arc shaped near the three-phase contact line, and straight where adjacent cells compress against each other. As evaporation proceeds, the number of BM cells gradually increases, reaching 13 uniformly distributed cells extending from the apex to near the contact line at t = 26 s (Figure 2.c). However, no cells form at the droplet edge, where thermocapillary convection persists. Droplet thinning during evaporation alters cell morphology: by t = 32 s (Figure 2.d), cells become shorter and thicker, and at t = 45 s (Figure 2.f), they resemble hexagonal cells in flat layers before gradually disappearing as evaporation completes.

Figures 2(a) and 2(b) demonstrate that 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 different Ma numbers, experiments control droplet volume V, wetting radius r, and ambient temperature T_a while varying substrate temperature T_w to change Ma, measuring _c when BM convection initiates. Results shown in Figure 3 [Figure 3: see original paper] reveal that _c increases monotonically with Ma. This occurs because both normal and tangential temperature gradients coexist in the droplet. Tangential gradients drive thermocapillary convection that transports heat from the 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 gradients—particularly near the apex—allowing weaker thermocapillary flow and easier BM convection onset. Therefore, BM convection only emerges when the contact angle falls below a threshold value at constant Ma. As Ma increases, the normal temperature gradient strengthens, favoring BM convection formation and enabling it to dominate over thermocapillary convection even at higher contact angles, thus increasing _c.

2.2 Effects of Wetting Radius and Ma Number

For a fixed wetting radius, the maximum number of cells n varies with Ma, as shown in Figure 5 [Figure 5: see original paper]. The maximum cell count increases with Ma because larger normal temperature gradients promote BM convection generation. For droplets with different wetting radii, cell morphology shows no significant differences, but the maximum cell number varies. At T_w = 303.15 K, T_a = 295.15 K, and RH = 60%, the maximum cell numbers are 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.

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3 Conclusions

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

1. Bénard-Marangoni cells only form when the contact angle decreases below a critical value. The cells exhibit a "flower-like" morphology distinct from hexagonal cells in flat layers: 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 shorter and thicker. Thermocapillary convection persists at the droplet edge without cell formation.
2. The critical contact angle _c for Bénard-Marangoni convection onset increases with Ma number. Under identical conditions, droplets with larger wetting radii contain more cells due to greater accommodation capacity. Cell number also increases with Ma number.

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Note: Figure translations are in progress. See original paper for figures.