



Relaxation dynamics of the Marangoni convection roll structure induced by camphor concentration gradient



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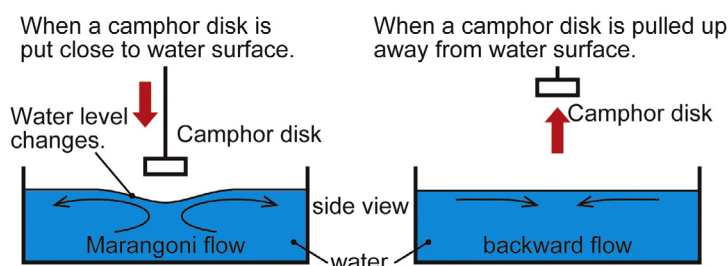
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HIGHLIGHTS

- When a camphor disk approaches water surface, Marangoni convection occurs.
- When a camphor disk moves away, water surface moves in opposite direction.
- Backward flow can be explained considering water surface deformation.

GRAPHICAL ABSTRACT



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ABSTRACT

When a camphor disk is placed close to a water surface, Marangoni convection occurs due to the surface tension gradient originating from the spatial distribution of camphor molecules at the water surface. We put plastic floats on the water surface to investigate the surface Marangoni flow, and observed that the plastic floats moved away from the camphor disk due to Marangoni convection. When the camphor disk was pulled up away from the water surface, the Marangoni convection weakened and finally disappeared. At that time, we observed that the floats approached the position just below the camphor disk. We discuss the mechanism of such float motion as related to the change in the structure of Marangoni convection and the change in the water level.

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1. Introduction

Marangoni convection occurs when there exists a surface tension gradient on liquid. Near the water surface, convective flow is induced from a region with lower surface tension to one with higher surface tension [1,2]. Marangoni convection is classified into

two types: solutal Marangoni convection and thermal Marangoni convection. That is, convection induced by any surface tension gradient originating from a concentration gradient is called solutal Marangoni convection [3,4], and that originating from a temperature gradient is called thermal Marangoni convection. Recently, pattern formation related to surface Marangoni flow was reported, and understanding the dynamics of such flow is an important issue in nonequilibrium physics [5,6].

Here, we consider the solutal Marangoni convection, especially that induced in a camphor-water system. When a camphor disk is placed on a water surface, camphor molecules dissolve from the

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disk and are distributed on the water surface around it, which causes a decrease in surface tension [7–13]. The camphor–water system has been investigated as a model of active matter [14,15].

It was reported that Marangoni convection occurs when a camphor disk is put on a water surface [16–18]. This convection can be understood based on a decrease in surface tension due to the distribution of camphor molecules at the water surface. Such a spatial distribution induces Marangoni stress at the water surface that drives Marangoni convection [19,20].

Without directly touching a camphor disk on a water surface, camphor molecules are sublimated from the camphor disk and adsorbed at the water surface, resulting in a spatial gradient of the surface tension. Thus, the Marangoni convection was observed by setting a camphor disk close to the water surface without touching it. In this situation, small plastic floats on the water surface moved escaping from the approaching camphor disk due to the effect of the Marangoni convection. We also observed the floats moved toward the hovering camphor disk when the camphor disk was pulled up away from the water surface. This is surprising since the reverse flow was generated even when the Marangoni convection was decaying.

In the present paper, we clarify the mechanism of this curious phenomenon based on the time change in the structure of the Marangoni convection and that of the water surface. At first, the motion of plastic floats at the water surface was monitored, when a camphor disk was put close to and then pulled up away from the water surface. The structure of the Marangoni convection in the aqueous phase was unveiled. The Marangoni convection can deform the shape of the water surface [21], and so the time change in the water level might clarify the mechanism of the reversed flows. We considered that the reverse flow may be related to changes in the water level, and performed numerical calculations to confirm our proposed mechanism.

2. Experiments

Camphor was purchased from Wako Pure Chemical Industries (Japan), and a camphor disk (diameter: 3 mm, thickness: 1 mm) was prepared using a pellet die set for Fourier transform infrared spectroscopy. Urea was purchased from Nacalai Tesque, (Japan). Purified water was made with a water purifying system (Autostill WG23, Yamato Scientific Co., Ltd., Japan).

2.1. Experiment I

To evaluate the surface Marangoni flow induced by a camphor disk, a water phase (depth: 5 mm) was prepared by pouring purified water (volume: 157 mL) into a glass Petri dish (diameter: 200 mm). Then, three plastic floats (diameter: 3 mm, thickness: 0.1 mm, mass: 1 mg) were put onto the water surface. The camphor disk was connected to a height adjusting z-stage (OSMS20-35, resolution: 2 μm , Sigma Koki, Japan) using a platinum wire (diameter: 0.5 mm, length: 30 mm) to control the vertical position of the disk. The schematic illustration of the experimental system is shown in Fig. 1a. At $t = 0$, the camphor disk was rapidly (scan rate: 15 mm s^{-1}) moved from $d = 25$ to $d \sim 0^+$ mm, where d is the distance from the water surface to the bottom surface of the camphor disk. The location of the camphor disk was kept at $d \sim 0^+$ mm without touching the water for 300 s, and then it was rapidly moved away from the water surface to $d = 25$ mm. Finally, it was fixed at $d = 25$ mm. Here, $d \sim 0^+$ mm means that we could not measure the precise distance between the camphor disk and the water surface, but it was approximately 0.5 mm. The motion of the plastic floats was monitored from above with a digital video camera (HDR-CX590, Sony, Japan; time resolution, 1/30 s). In order to evaluate the surface flow near

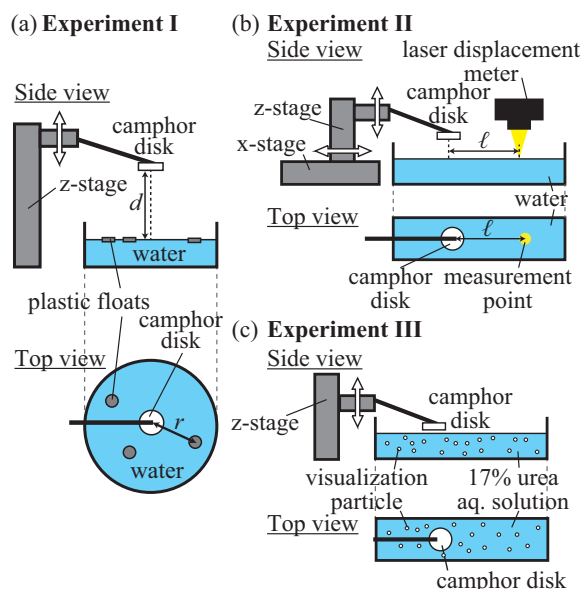


Fig. 1. Schematic illustration of the three experimental equipments. (a) Experiment I for the observation of the plastic floats. (b) Experiment II for the measurement of water level. (c) Experiment III for the observation of the visualization particles. r is the distance of a plastic float from the center of the camphor disk. ℓ is the distance between projection of camphor disk center on water surface and the measurement point. d is the distance from the water surface to the bottom surface of the camphor disk.

the camphor disk, some floats were placed near the camphor disk and released just before the camphor disk was pulled up away from the water surface. The motion of the plastic floats was analyzed by image processing software.

2.2. Experiment II

To evaluate the change in water level in the water chamber due to the Marangoni flow, we prepared a rectangular water chamber (width: 9 mm, height: 20 mm, length: 145 mm). A water phase was prepared by pouring 6.5 mL of purified water (depth: 5 mm) into this chamber. The vertical position of the camphor disk was controlled with the z-stage. At first, the camphor disk was moved close to the water surface from $d = 25$ to $d \sim 0^+$ mm without touching the water surface. The location of the camphor disk was kept at $d \sim 0^+$ mm for 60 s, and then it was rapidly moved away from the water surface to $d = 25$ mm. The water level was measured with a laser confocal displacement meter (LT9010M, Keyence, Japan; minimal resolution 0.01 μm , working distance 6 mm, spot diameter 2 μm), when the camphor disk was fixed close to the water surface. To change the distance, ℓ , between projection of camphor disk center on water surface and the measurement point, the lateral position of the z-stage was controlled by an x-stage device (XLSG80, Misumi, Japan), as shown in Fig. 1b. Time series of water level was measured successively, and we obtained the change in the water level between with and without a camphor disk, by eliminating the effect of the water evaporation for each camphor position.

2.3. Experiment III

To visualize the convective flow around the camphor disk, visualization particles (DIAION, HP20S, Mitsubishi Chemical, Japan; particle size 100–200 μm , particle density 1.01 g mL^{-1}) were added to the aqueous phase (volume: 6.5 mL, depth: 5 mm) in the rectangular water chamber (width: 9 mm, height: 20 mm, length: 145 mm). The schematic illustration is shown in Fig. 1c. Here, 17 wt/v% urea was dissolved in the aqueous phase to produce a

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