

## Change in the Mode of Spontaneous Motion of an Alcohol Droplet Caused by a Temperature Change

Ken NAGAI, Yutaka SUMINO, Hiroyuki KITAHATA and Kenichi YOSHIKAWA

*Department of Physics, Graduate School of Science, Kyoto University,  
Kyoto 606-8502, Japan*

(Received July 4, 2005)

An alcohol (pentanol) droplet exhibits spontaneous motion on an aqueous solution, driven by a solutal Marangoni effect. We found that the mode of such droplet motion changes depending on the temperature of the aqueous phase. When the temperature of the aqueous phase is 20 °C, a droplet with a volume of 1  $\mu\text{l}$  exhibits vectorial motion, whereas when the temperature is 25 °C, the droplet exhibits irregular motion. We discuss the mode change in relation to the solubility of pentanol in water.

### §1. Introduction

In general, at interface, there is the pressure which minimizes interfacial energy, e.g., Laplace pressure. When a spatial gradient of interfacial energy is present, spontaneous agitation is generated, and this is called the Marangoni effect. A droplet on a substrate with a nonuniform interfacial energy has been theoretically predicted to be driven by an interfacial-energy gradient, and this has been confirmed experimentally.<sup>1),2)</sup>

The interfacial energy is dependent on the chemical concentration. Thus, spontaneous motion due to the Marangoni effect can emerge when the distribution of the chemical concentration breaks symmetry around the periphery. For example, a camphor scraping with an asymmetric shape on water moves spontaneously because the camphor diffuses nonuniformly and there is a difference in the interfacial energy gradient around the scraping. The spontaneous motion of a scraping is translational or rotational depending on its shape.<sup>3)</sup> This result shows that the shape of interface determines the mode of the spontaneous motion due to the Marangoni effect.

In some systems, a droplet on a substrate spontaneously distorts and moves due to asymmetry of interfacial energy.<sup>4),5)</sup> In a system where the droplet moves spontaneously due to the Marangoni effect, the nature of the distortion of the interface and the mode of droplet motion can change depending on various parameters. We have previously shown that an alcohol droplet on water can spontaneously assume an asymmetric shape and move due to the Marangoni effect. The droplet selects a certain mode of motion depending on its volume. This mode selection can be interpreted in terms of competition between the droplet volume and the critical wave number in the instability due to the Marangoni effect.<sup>6)</sup> In the present report, we note that the mode of the motion of an alcohol droplet with a fixed volume can be changed by controlling the temperature of the aqueous phase.

## §2. Experiments

One hundred milliliters of an aqueous solution containing 2.3 vol% pentanol in a container was dipped into a temperature-controlled water bath (Taitec, Saitama; SM-05R) and then poured into a petri dish with a diameter of 180 mm. Next, 1  $\mu\text{l}$  of pentanol (Wako, Kyoto) stained with ink (Pilot Corporation, Tokyo; INKSP-55-B) to visualize the motion of the droplet was placed on the aqueous phase (Fig. 1). A high-speed video camera (RedLake MASD Inc., San Diego; Motion Scope PCI) was used to monitor the motion of the droplet at room temperature at 60 frames per second.

The observation on the droplet motion in the petri dish was performed at ambient temperature as a preliminary experiment. We have a future plan to extend our study on a system with more precise control of temperature.

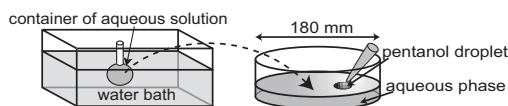


Fig. 1. Schematic illustration of the experimental setup.

## §3. Results

When the temperature of the aqueous phase was 20 °C, the droplet exhibited an asymmetric morphology, and the droplet motion was vectorial, as shown in Fig. 2(a). The speed of the motion was almost constant except when the droplet collided and reflected at the wall, as shown in Fig. 2(b). When the temperature of the aqueous phase was 25 °C, the droplet maintained a circular shape, and the droplet motion was irregular and intermittent, as shown in Fig. 3. These results show that the mode of droplet motion changed from vectorial motion to irregular motion with an increase in the temperature of the aqueous phase.

## §4. Discussion

The increase in the temperature induces the changes in the solubility of pentanol in water, the rate of pentanol evaporation, and interfacial tension: The solubility of pentanol in water decreases with the increase in temperature as shown in Table I. The higher temperature causes the higher rate of evaporation. The interfacial tension tends to decrease with the increase in temperature.

The mode change observed in the present study indicates that the rise of temperature increases the critical volume where the mode changes. In the previous study, we have shown that the increase in

Table I. Solubility of pentanol in water. The error of temperature is  $\pm 0.5$  °C and that of solubility is  $\pm 0.05$  ml/100 ml water.

temperature of water (°C)	solubility (ml/100 ml water)
21.0	2.70
21.8	2.66
22.9	2.61
24.6	2.54
27.4	2.50
31.0	2.46

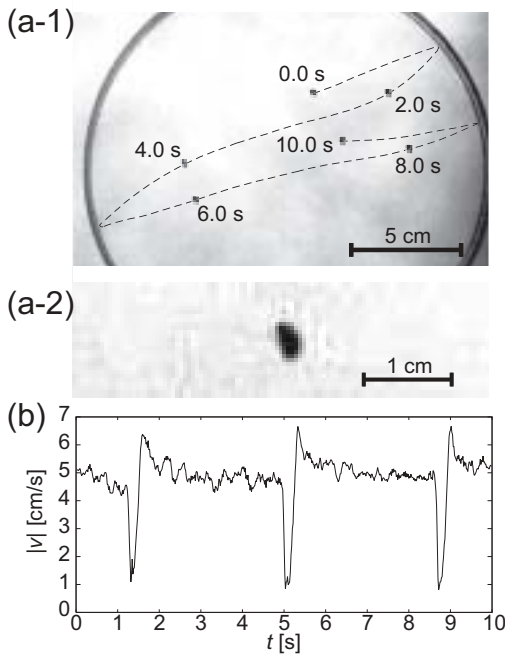


Fig. 2. Spontaneous motion of an alcohol droplet when the temperature of the aqueous phase was 20 °C. (a-1) Trace of the center of mass of the droplet for 10 s. The droplet moved vectorially. (a-2) Morphology of the droplet at 6.0 s. (b) Time trace of the speed of translational motion. The speed of the motion was almost constant except when the droplet collided and reflected at the wall.

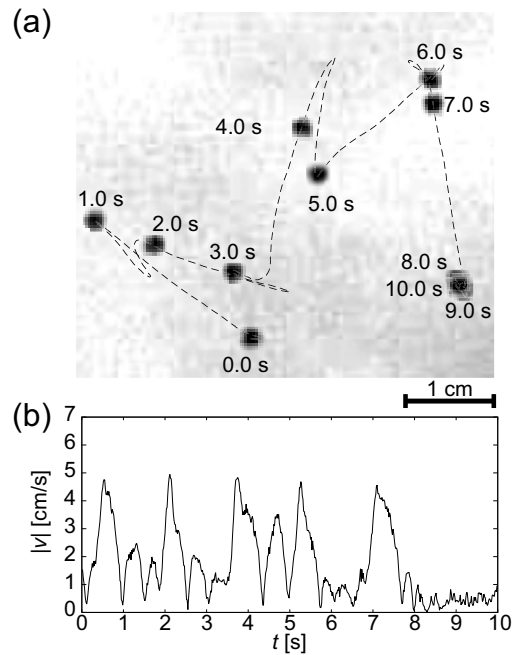


Fig. 3. Motion of an alcohol droplet when the temperature of the aqueous phase was 25 °C. (a) Trace of the center of mass of the droplet for 10 s. (b) Time trace of the speed of translational motion. The droplet moved irregularly and intermittently.

such critical volume is induced by the decrease in solubility, or by the decrease in the rate of evaporation, or by the decrease in the ratio of air-aqueous-phase interfacial tension to pentanol-aqueous-phase interfacial tension.<sup>6)</sup> The decrease in solubility induced by the increase in temperature can explain the experimental trend. Whereas, the increase in the rate of evaporation is against it. The effect of the change in interfacial tension may be negligible, according to the result of our preliminary experiment (data not shown). From these consideration, it is suggested that the mode change by change in temperature is mainly induced by the change in pentanol solubility in water, though there remains the possibility that the change in interfacial tension could have some contribution.

## §5. Conclusion

We have shown that the mode of the motion of an alcohol droplet on an aqueous phase changes with a change in the temperature of the aqueous phase. This mode

change is thought to be mainly caused by a change in the solubility of pentanol in water, although further experiments are needed to clarify the effect of the change in interfacial tension.

### Acknowledgements

This work was supported by a Grant-in-Aid for the 21st Century COE (Center for Diversity and Universality in Physics) from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

### References

- 1) F. Brochard, *Langmuir* **5** (1989), 432.
- 2) M. K. Chaudhury and G. M. Whitesides, *Science* **256** (1992), 1539.
- 3) S. Nakata, Y. Iguchi, S. Ose, M. Kuboyama, T. Ishii and K. Yoshikawa, *Langmuir* **13** (1997), 4454.
- 4) F. D. D. Santos and T. Ondarçuhu, *Phys. Rev. Lett.* **75** (1995), 2972.
- 5) Y. Sumino, N. Magome, T. Hamada and K. Yoshikawa, *Phys. Rev. Lett.* **94** (2005), 068301.
- 6) K. Nagai, Y. Sumino, H. Kitahata and K. Yoshikawa, *Phys. Rev. E* **71** (2005), 065301.