Spontaneous Blebbing of Oil Droplet Caused by Generation /Annihilation of the Lamellar Structure

One of the important mechanisms for biological motility is the generation/collapse of an elastic aggregate of biopolymers. In order to mimic the essential features of the mechanism, we constructed a system where spontaneous deformation of an oil droplet is induced by the generation/collapse of a surfactant aggregate. Upon placing an oil droplet with a fatty acid on an aqueous phase with a cationic surfactant, the oil droplet spontaneously deformed intensively for more than 1 hour. Accompanied with the appearance of the deformation, the aggregate was generated at the oil-water interface. The structure of the aggregate was confirmed with small-angle X-ray scattering, indicating that the aggregate was highly-ordered lamellar with an interlayer distance of 405 Å.

Biological objects utilize various physical mechanisms to induce their motion. One of such mechanisms is the generation of elastic aggregates; for instance, generation of F-actin gel from G-actin. The construction of chemical systems that mimic some parts of biological motility is relevant for understanding physical mechanisms, especially the processes of spatio-temporal formation without the control of genes.

To construct such a system, a setup composed of an aqueous phase and an organic phase, such as an oil-water system, is useful. A far-from-equilibrium condition can be easily imposed by setting the initial condition for this system so that spontaneous pattern formation occurs.

In this study for the above-mentioned purpose, we constructed a physico-chemical model system composed of an aqueous phase with a cationic surfactant, trimethylstearylammonium chloride (STAC) and an organic phase, tetradecane, with a fatty acid, palmitic acid (PA).

STAC and PA were purchased from Tokyo Chemical Industry Co., Ltd. (S0087 and P1145; assay >97% and >99.5%, respectively). Tetradecane for an organic phase was obtained from Wako Pure Chemical Industries, Ltd. (207-10705; assay >99%). These chemicals were used without further purification. Water was purified by a Millipore milli-Q system.

As an aqueous phase, we prepared a STAC aqueous solution at concentrations of C_s. As an organic phase, palmitic acid was dissolved into tetradecane at concentrations of C_p. The critical micelle concentration of STAC is about 0.3 mM at room temperature.

To investigate the aggregate structure, we used a small-angle X-ray scattering (SAXS) apparatus installed at BL-15A. The incident X-rays were monochronized by Ge(111), and the wavelength was 1.5 Å. We used a cell made of acryl resin as shown in Fig. 1(a); 350 μL of the organic phase with C_p = 10 mM was placed on 700 μL of the aqueous phase with C_s = 25 mM.

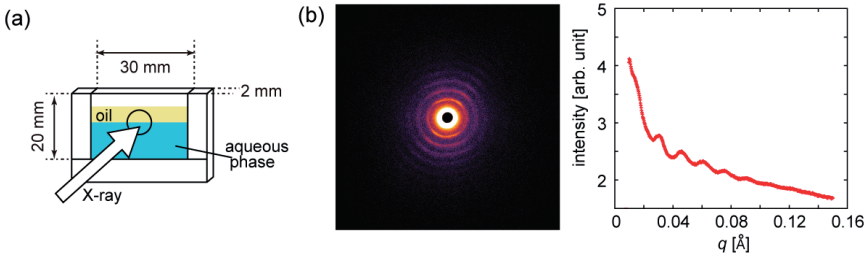

Figure 1
Inspection of aggregate generation: (a) Experimental setup. (b) SAXS image and its azimuthal average.

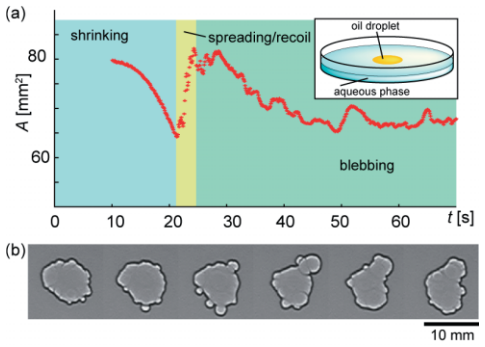

Figure 2
Manner of droplet deformation: (a) Change in apparent area, *A*, of an oil droplet. Inset: Schematic representation of experimental setup. (b) Blebbing motion observed in late stage of droplet deformation. Snapshots start from 48 s and were taken every 2 s.

To observe the droplet motion, the concentration of the aqueous phase was C_s = 0.67 mM. The diameter of the Petri dish was 145 mm, and the volume of the aqueous phase was 100 mL. A 100-μL oil droplet with C_p = 20 mM was placed on the aqueous surface. The deformation of the oil droplet was monitored by a digital video camera with the acquisition rate of 30 Hz. All measurements were carried out at room temperature (~22°C).

Figure 1(b) shows the SAXS image and its azimuthal average. The multi-peak profile corresponds to higher-order peaks of the first Bragg peak at 0.0155 $Å^{-1}$. The profile indicates that the aggregate is highly ordered lamellar. The interlayer distance, *d*, is calculated as *d* = 405 Å, which means that a large amount of water should be included in the structure [1]. Existence of such lamellar structure was confirmed with different combinations of C_s and C_p.

Figure 2 shows the manner of deformation after the oil droplet was placed on the aqueous surface. Initially, the oil droplet shrunk for 18 s, and then spread and recoiled in fractions of a second. After such a vivid change in apparent area, the oil droplet started blebbing at the interface, which continued for over 1 h. The aggregate started to appear after the oil droplet started to bleb.

From these results, we consider that the generated aggregate is a key factor of the oil droplet deformation, especially blebbing. Assuming from its structure that the aggregate is elastic and permeable, the blebbing can be explained with a mathematical model of the generation and collapse of the elastic aggregate [2, 3]. Continuous generation of the elastic aggregate at the oil droplet surface induces an increase in the internal pressure of the oil droplet. A bleb can appear upon breaking of the aggregate nearby triggered by an infinitesimal perturbation at the interface. We believe that the mathematical model can be further verified by inspecting the aggregate structure in the course of blebbing.

REFERENCES

[1] Y. Sumino, H. Kitahata, H. Seto, S. Nakata and K. Yoshikawa, *J. Phys. Chem. B* **113** (2009) 15709.
[2] Y. Sumino, H. Kitahata, H. Seto and K. Yoshikawa, *Phys. Rev. E* **76** (2007) 055201.
[3] Y. Sumino, H. Kitahata, H. Seto and K. Yoshikawa, in preparation.

BEAMLINE

15A

Y. Sumino[1], H. Kitahata[2], H. Seto[3] and K. Yoshikawa[4]
([1]The Univ. of Tokyo, [2]Chiba Univ, [3]KEK-IMSS, [4]Kyoto Univ. and ICORP-JST)