

Multi-scale structure formation through spontaneous interface blebbing

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Introduction

We have reported the deformation of an interface between cationic surfactant aqueous solution and anionic surfactant organic solution [1-3]. The deformation is induced by the generation of elastic aggregates at the oil-water interface. The aggregation was also investigated with small angle X-ray scattering (SAXS) measurement, and revealed that it has lamellar structures with a large interlayer distance around 40 nm [2]. In this study, quasi-two dimensional condition was used to study the manner of blebbing without the geometrical effect due to the droplet shape. It was found out that the oil-water interface showed blebbing as in the case of the droplet system. During the blebbing motion, a regular array of aggregation pillars was formed spontaneously in the non-blebbing region of the interface. To investigate the detailed aggregation structure, micro-beam SAXS observation was performed to unveil microscopic structure of the aggregation. Surprisingly enough, such coherent structures were formed spontaneously from nanometer to millimeter scales, through the dynamical behavior of the interface without any external control.

Experiments

We prepared stearyltrimethylammoniumchloride (STAC) aqueous solution as an aqueous phase, whose concentration was 50 mM, and palmitic acid (PA) tetradecane solution as an organic phase, whose concentration was 20 mM. We used the cell schematically shown in Fig. 1(a) where sidewall was made up with Kapton film. A 600- μ l of aqueous phase was put into the cell, and then a 300- μ l of organic phase with was put on the aqueous phase.

For the measurement of spatial structure of aggregation in the pillars, we used microbeam SAXS apparatus installed at BL4A, Photon Factory, KEK. The X-ray beam radius was $\sim 5 \mu\text{m}$, and the wavelength was $\sim 1.1 \text{ \AA}$. We started the measurements 4800 s after the organic phase was set on the aqueous phase. We moved the system stepwisely in a horizontal direction at a rate of $\sim 2 \mu\text{m/s}$, and measured the local structures of aggregates. The measurements were conducted every 10 s ($\sim 20 \mu\text{m}$ step).

Results and Discussion

Figure 1 shows the microbeam SAXS results on a single pillar. As shown in Fig. 1(a), we performed microbeam SAXS measurement along a line about 1 mm

below the interface. The SAXS results suggest regular lamellar structures with an interlayer distance, d , around 29.1 nm, which obtained from $d = 2\pi/k_c$ where k_c was a peak-to-peak distance. The orientation of the lamellar structure depends on the measurement position in a single pillar. Figure 1(b) shows the spatial dependence of the total intensity, I_t , and the orientation of the lamellar structure, θ_p . The x axis is set horizontally, parallel to the oil/water interface. The total intensity, I_t , is defined as the radial and angular sum of scattering intensity, where direct beam intensity is eliminated. The orientation, θ_p , is defined as the angle of the maximum scattering intensity. I_t showed small flickering with respect to x indicating that the pillar is formed by grains of aggregates whose size should be comparable to the step size $\sim 20 \mu\text{m}$. The orientation is strongly correlated with the position, x , in the region with higher I_t . The experimental results show that the horizontal lamellar structure is formed at the center part of the pillar, while tilted one is formed at the edge as schematically shown in Fig. 1(c).

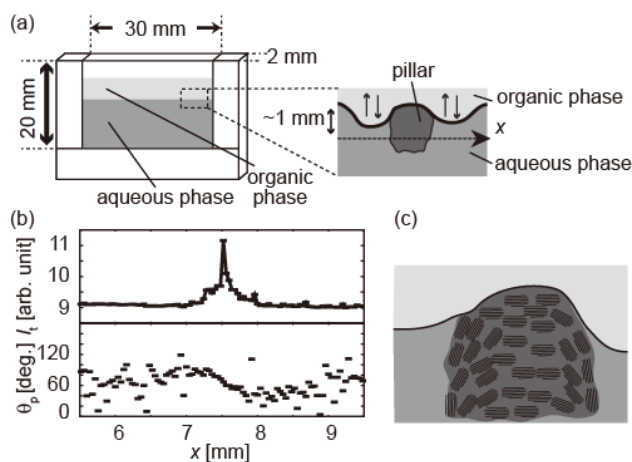


Figure 1: (a) Schematic illustration of experimental setup. (b) I_t and θ_p against the position x . The orientation of the lamellar structure has strong correlation with the position in a pillar structure. (c) Schematic illustration of microscopic structure in a pillar structure of aggregates.

References

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