# Supporting Information

# Self-Propelled Oil Droplets and Their Morphological Change to Giant Vesicles Induced by a Surfactant Solution at Low pH

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## Figures

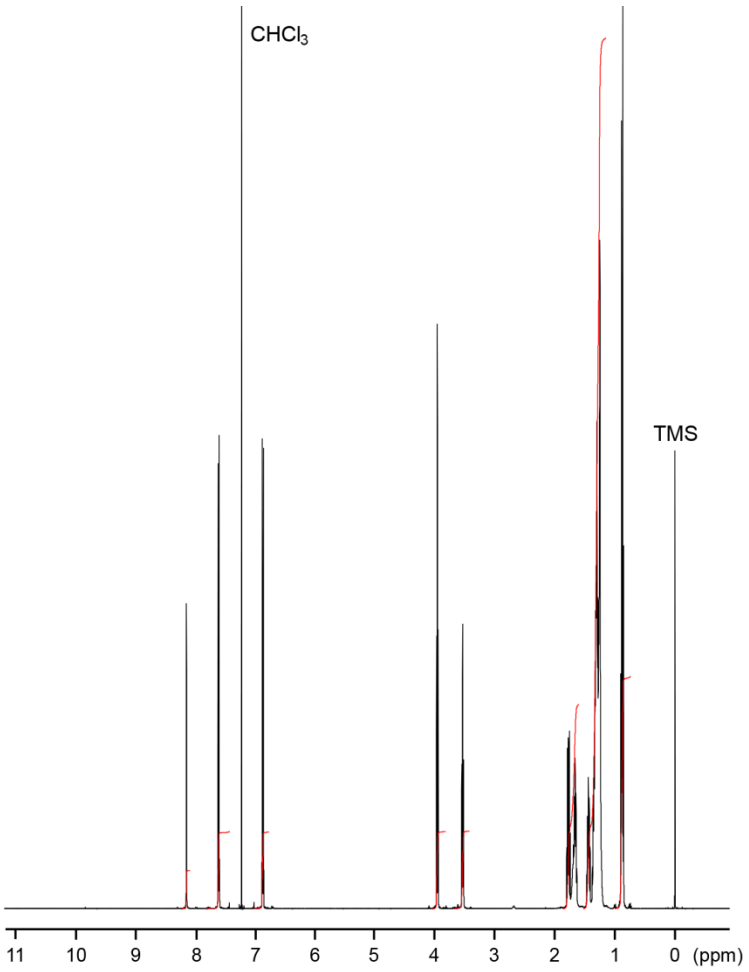


**Figure S1.** $^{1}$H NMR spectrum of HBDA dissolved in $CDCl_3$ (500 MHz).

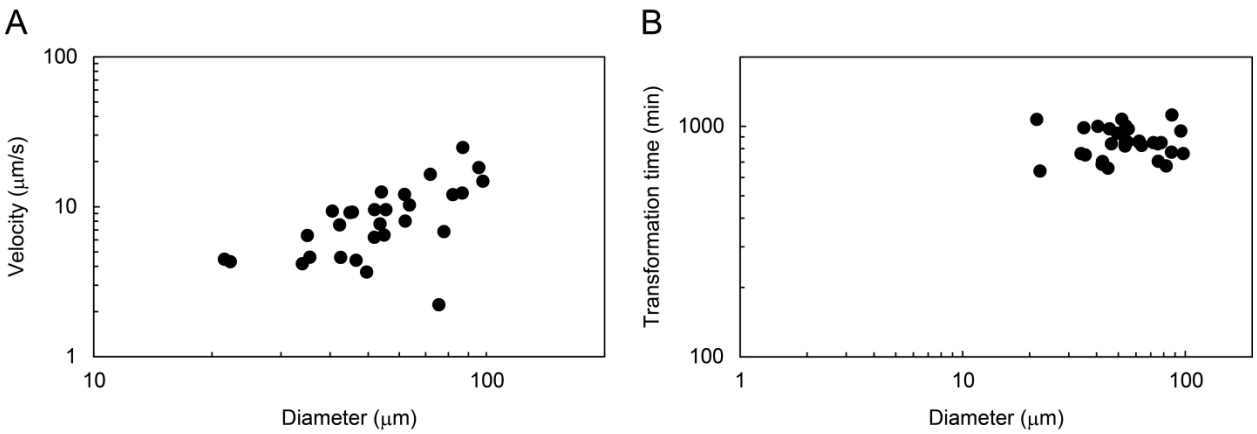


**Figure S2.** Relationship between the initial velocity and diameter of the self-propelled oil droplet. We calculated the initial velocities of the oil droplets as the average velocity within 10 seconds of commencement using microscopy (A). The flocculate particle transformation time is shown in (B).

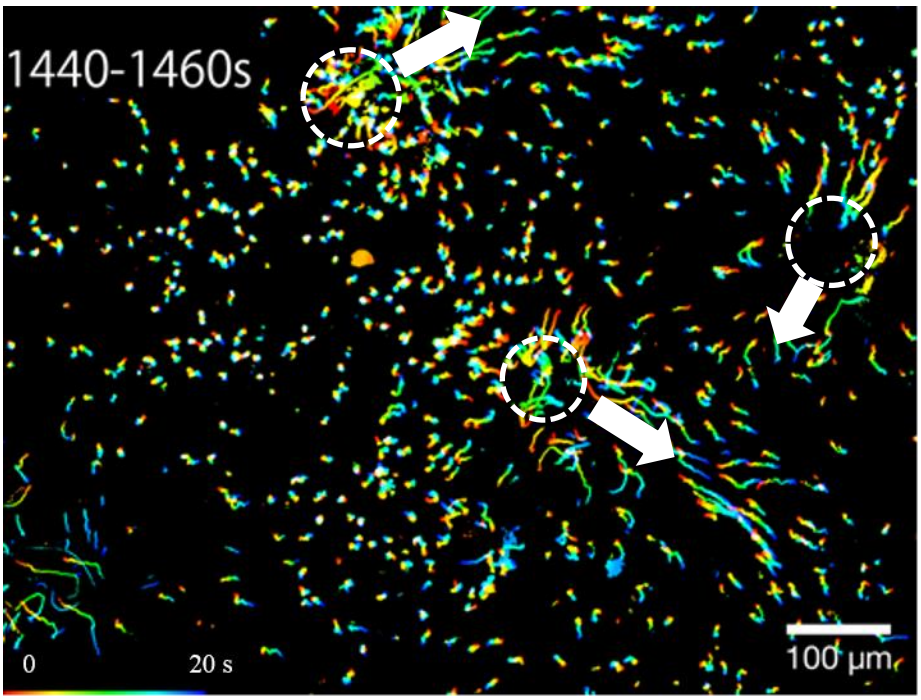


**Figure S3.** Trajectories of the flow fields of spherical droplets exhibiting self-propelled motion using hydrophilic fluorescent microspheres dispersed in an aqueous phase of the HBDA emulsion system (exposure time: 66.7 ms, 15 frames/sec, analyzed by Image J macro given from http://www.jaist.ac.jp/ms/labs/hiratsuka/index.php/). The dot lines and arrows represent the position of the droplets and the direction of self-propelled motion, respectively. The microspheres flowed according to the oil droplet locomotion, indicating that it was associated with the surrounding water advection to the opposite direction at its rear side.

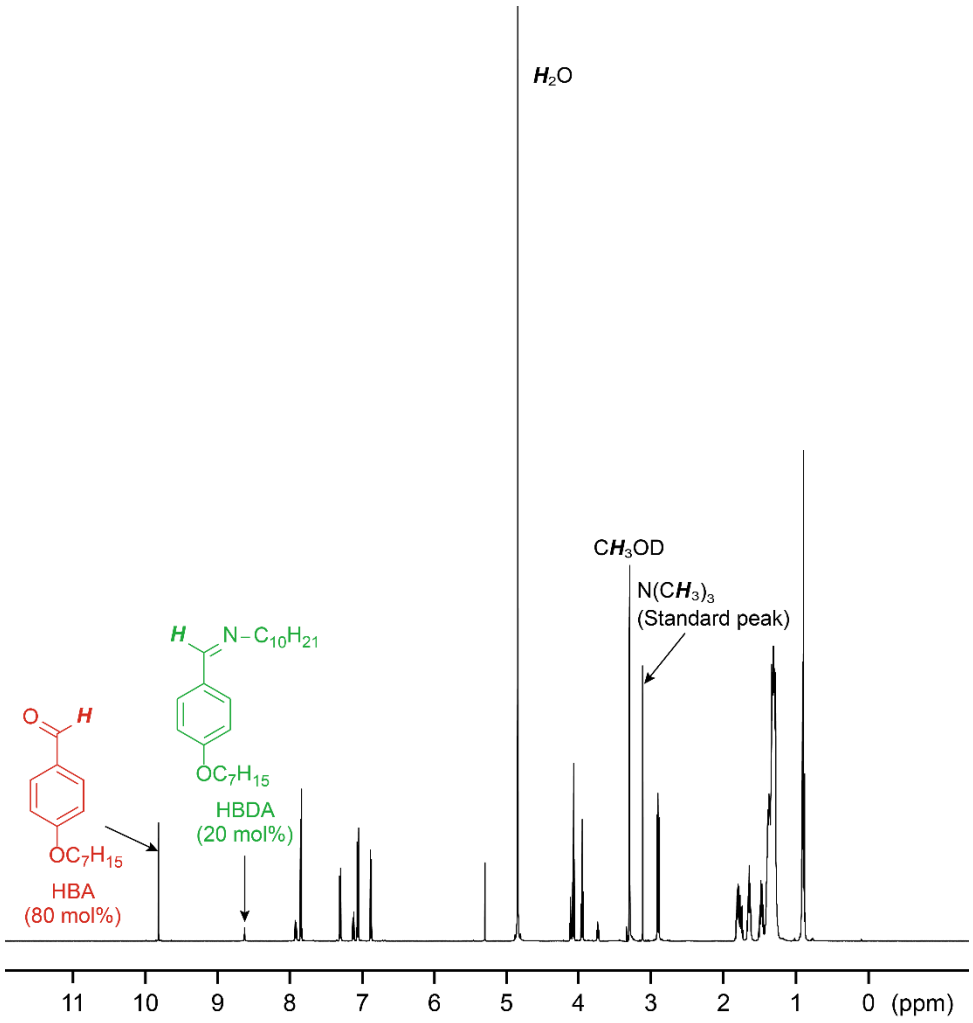


**Figure S4.** $^{1}$H NMR spectrum (MeOD-$d_4$) of the hydrolyzed products of HBDA (125 mM) in 10 mM C16TAB containing 0.1 M HCl (200 μL) at room temperature after 60 min. The percentage hydrolysis of HBDA was calculated by integrating the decrease in the signals for the imine methine protons at δ 8.22 ppm using the signal for trimethylammonium from C16TAB protons at δ 3.18 ppm as an internal standard peak.

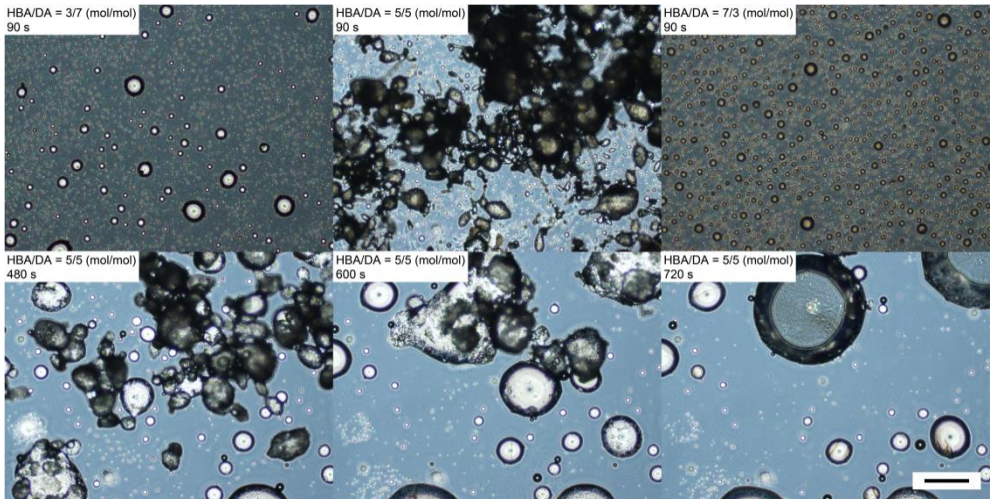


**Figure S5.** Phase contrast microscopy images of a HBA and DA mixture at 3/7, 5/5, and 7/3 (mol/mol) in 10 mM C16TAB solution containing 0.1 M HCl at room temperature. Bar: 50 μm.

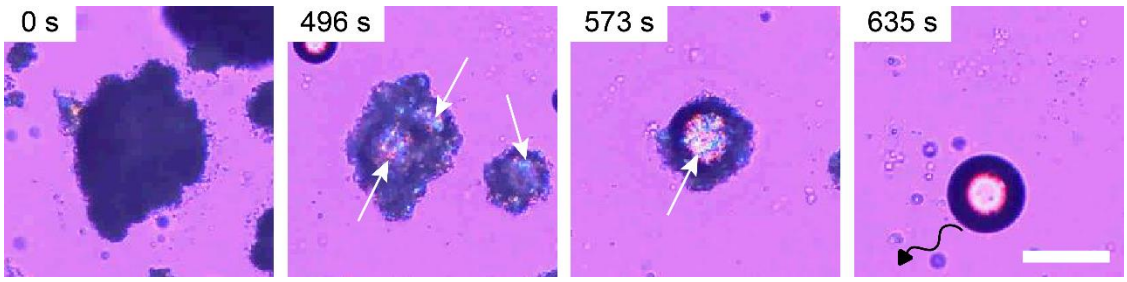


**Figure S6.** Typical micrographs of a mixture composed of HBA and DA (5/5 molar ratio) in 10 mM C16TAB solution containing 0.1 M HCl at room temperature under a polarized microscope. Morphological changes and locomotion of the droplets that had brilliant texture in a part of the aggregate were observed (indicated by white arrows). The black arrow represents the direction of self-propelled motion of droplet. Bar: 100 μm.

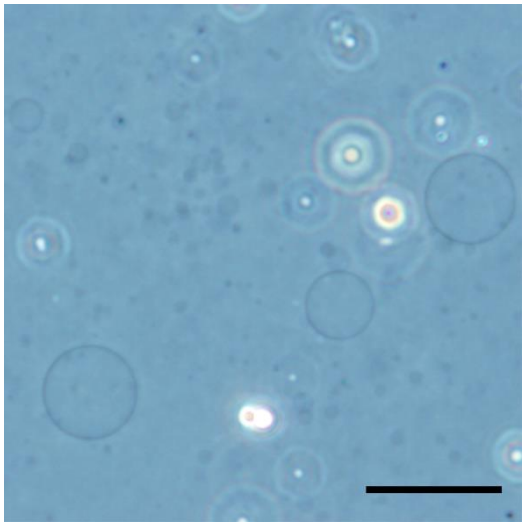

**Figure S7.** Phase contrast micrograph of giant vesicles forming in the dispersion composed of HBA (125 mM), DA (125 mM), C16TAB (10 mM), and HCl (100 mM) at room temperature 24 h after preparing the sample for observation. Bar: 40 μm.

## Tables

**Table S1** Effects of the surfactant concentration on the dynamics in a C16TAB solution at room temperature.

| Entry | C16TAB (mM) | Dispersion | Morphology | Motion time (min) |
|---|---|---|---|---|
| 1 | 10 | 1 M HCl | Undefined aggregates | n. o. |
| 2 | 10 | 0.1 M HCl | Change from undefined aggregates to spherical droplets | 30 |
| 3 | 10 | 0.01 M HCl | Change from undefined droplets to spherical droplets | n. o. |
| 4 | 10 | Water | Spherical droplets | n. o. |
| 5 | 10 | 0.1 M NaCl | Spherical droplets | n. o. |
| 6 | 50 | 0.1 M HCl | Change from undefined aggregates to spherical droplets | n. o. |

n. o. = not observed.

## Video clip

One video clip has been attached to show the dynamics of the HBDA emulsion, which was prepared in an acidic solution (0.1 M HCl) of C16TAB (0.01 M), and observed by phase contrast microscopy. The sequential dynamics, which consisted of the transformation of flocculate particles to spherical droplets and locomotion of droplet, were observed under low pH conditions using an oil compound HBDA containing an imine bond. This movie was 6 times faster than the real time observations.