

Nematic Liquid-Crystal Necklace Structure Made by Microfluidics System

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1. Fabrication of a microfluidics device

For large droplets (diameter > 100 μm), a microfluidics device, which has a flow-focusing geometry, inspired by the previous report was used [28]. We should note here that in the reference [28] the device has three inlets, while in our experiment only two inlets are necessary. Therefore our device has two inlets, one for the dispersed phase and one for the continuous phase, as we show schematically in Fig. 2k in the main text.

For small droplets (diameter < 50 μm) we have used a device with single co-flow geometry shown in Fig. 2j of the main text. The squared capillary (Vitrocom, 8100-100) was modified to hydrophilic by a SC-1 solution. SC-1 solution was prepared by mixing water, hydrogen peroxide, and ammonium solution with the weight ratio of 5:1:1. The capillary was immersed into this solution and was annealed at 90 °C overnight. Then it was washed by pure water, dried, and used. This squared capillary was fixed on a slide glass by a two-component epoxy glue (UHU PLUS). A round capillary (Microcaps, Drummond) was pulled finely by the pullers of SUTTER Instrument (Model P-1000) and of NARISHIGE (PC-10). The processed capillary was used without modification. The round capillary was inserted into the squared capillary fixed on the slide glass. At one edge of the fixed squared capillary the needle whose tip was cut and grained smoothly was fixed by a two-component epoxy glue. Then the liquid which was poured from the grained needle can flow between the circle and the squared capillaries and represents the continuous phase of the device. After drying at 60 °C, liquids were flown into the microfluidics devices. Before experiments, the devices were washed with pure water.

2. Experimental setting of a microfluidics device

2-1. Large droplets (diameter > 100 μm)

For the dispersed phase, the setting was done as described below. A glass syringe (Micro syringe TLL, Hamilton) was filled with pure water and connected to a filter (Minisart 16534K, Sartorius), which was connected to the tube (Polyethylene Tubing 427421, Intramedic). The syringe was set to the micro-syringe pumps (World Precision Instruments-1002X, KF-Technology 1000). The syringe was pushed by the pump and the filter and the tube were filled with pure water.

Next, the tip of the tube was immersed into the dispersed phase. The dispersed phase (150 ~ 200 μl) was sucked from the tip of the tube by withdrawing the pump. Then the tip of the tube was connected to the inlet for the dispersed phase of a microfluidics device.

For the continuous phase, a glass syringe was filled with the continuous phase and connected to a filter. The needle whose tip was cut and grained smoothly, was connected to the filter and the tube was connected to the needle. The syringe was set to the micro-syringe pump. The syringe was pushed by the pump to fill the filter, the needle and the tube by the continuous phase. Then the tip of the tube was connected to the inlet for the continuous phase of a microfluidics device.

2-2. Small droplets (diameter < 50 μm)

In the microfluidics device for small droplets, the puddle made by the tip-cut needle was not used for the inlet of the dispersed phase, and therefore, we can reduce the amount of the dispersed phase compared to the case of large droplets. At first the round capillary was filled with the dispersed phase by thin needle and then the water-filled tube was connected to the round capillary. By pumping the water-filled tube, the dispersed phase was pushed into the system.

For the continuous phase, the procedure is the same as this for the large droplets.

3. Calculation of the Capillary and Weber numbers

The Capillary number (Ca) is defined as

$$Ca = \frac{\eta V_{PVA}}{\sigma},$$

where η is the viscosity of PVA aqueous solution, V_{PVA} is the typical speed of PVA aqueous solution and σ is the interfacial tension between 5CB and PVA solution. The Capillary number Ca represents the ratio between the viscous drag force and the surface tension. When Ca is smaller than unity, surface tension dominates and the system is in the dripping regime [S1].

The Weber number (We) is defined as

$$We = \frac{\rho V_{5CB}^2 L}{\sigma},$$

where ρ is the density of 5CB, V_{5CB} is the speed of 5CB at the nozzle, L is the diameter of a tip of the capillary. The Weber number We represents the ratio between the inertia and the surface tension. When We is smaller than unity, surface tension dominates, forcing the system to drip.

The Capillary and Weber numbers of the droplets shown in Fig. 2 are shown in Table 1. Capillary numbers are quite smaller than unity and Weber numbers are below unity for large droplets and close to unity for small droplets. As the result, our microfluidics system is in the dripping regime.

Table 1 The Capillary and Weber numbers of the droplets shown in Fig. 2.

droplet diameter *10 ⁻⁶ [m]	tip diameter L *10 ⁻⁶ [m]	typical speed of PVA V_{PVA} *10 ⁻⁴ [m/s]	typical speed of 5CB V_{5CB} [m/s]	We	Ca *10 ⁻³
17	3	6	1	0.25	3
46	5	15	1.3	0.71	7.4
135	70	2.5	0.017	0.0017	1.2

The parameters used in the calculation are follows; the viscosity of the PVA solution η is 59 mPa·s as shown in the section 5 of the supporting information, the density of 5CB ρ is 1.01×10^3 kg/m³, and the interfacial tension between 5CB and PVA solution σ is approximately 12×10^{-3} N/m [S2, S3].

4. Segregation of a liquid crystal in a tether

We have observed in experiments that in some necklaces LC and PVA in a tether segregated after few days (Fig. S1).

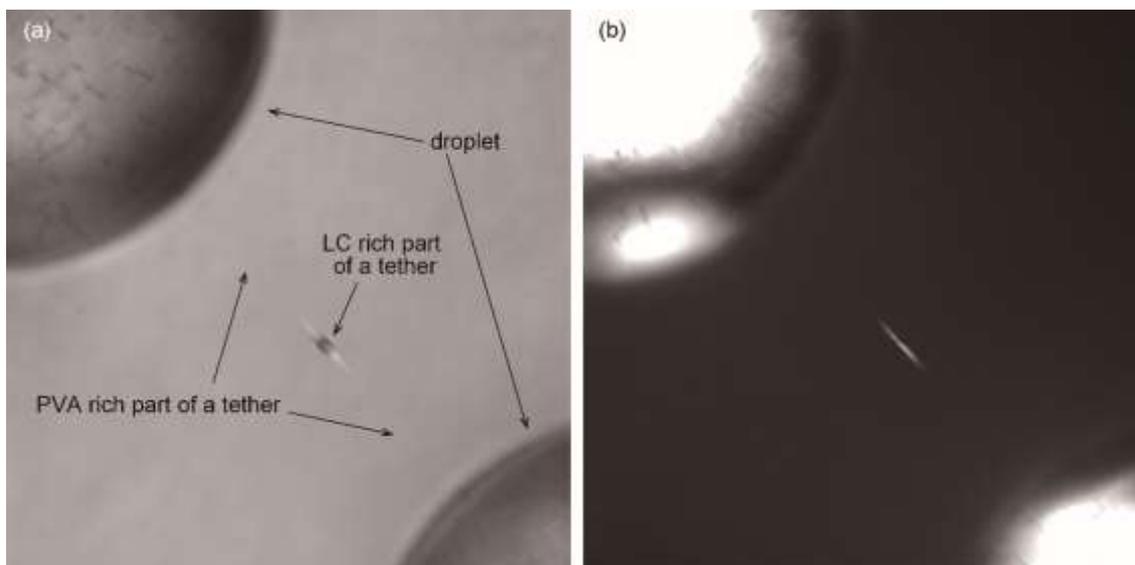


Fig. S1 Microscopic observation of the liquid crystal segregation in a tether. The droplet diameter is $210\ \mu\text{m}$. (a) The concentration of LC in the tether is not homogenous and LC is accumulated in the center of a tether, what can be also observed in the crossed polarized image (b). The tether is still connecting the droplets, but it consists of PVA rich part at both sides and LC rich part in the middle.

5. Measurement of the viscosity of a PVA solution

The viscosity of PVA 5 wt% aqueous solution η was measured by rheometer (Anton Paar, Physica MCR 301) with cylindrical cell. By scanning the shear rate $\dot{\gamma}$ from 0.01 to 10 s⁻¹, it is found that the viscosity is stable around $\dot{\gamma} = 0.1$ s⁻¹. We measured the viscosity $\eta = 59$ mPa·s at $\dot{\gamma} = 1$ s⁻¹ and $\eta = 60$ mPa·s at $\dot{\gamma} = 0.1$ s⁻¹. To calculate the spring constant of a tether and the Capillary number we used $\eta = 59$ mPa·s.

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