Supplementary Information

Directional size-triggered microdroplet target transport on gradient-step fibers

Yan Xue, Yuan Chen, Ting Wang, Lei Jiang, and Yongmei Zheng *

Key Laboratory of Bio-Inspired Smart Interfacial Science and Technology of Ministry of Education, Department of Chemistry and Environment, Beihang University, Beijing, 100191 (P. R. China)

*Correspondence and requests for materials should be addressed to Y. Z. (zhengym@buaa.edu.cn)

Supplementary Information contains:
Supplementary Figures S1-8.
Supplementary table S1.
Fig. S1. Illustration of uni-direction gradient-step spindle-knots formed on fiber. a) A main-fiber is drawn out from polymer solution via an increasing velocity. b) A gradient thick polymer solution film forming along main-fiber. c) The cone-gradient thickness film would break up into the gradient-step droplets due to gradient Rayleigh instability. After dryness, the gradient-step droplets form the gradient-step spindle-knots from the big one to the small one.
Fig. S2. Optical images of Rayleigh instability break-up into gradient size droplet via low and high concentration film at changing-controlled drawing velocity of 8.1–21.4 mm/s. a) At low concentration (≤ 6 wt.%) the film is too thin, little gradient-step spindle-knot forms on fiber. When the concentration of solution (weight percent) is between 8%–12 wt.%, we can get excellent gradient-step spindle-knot fiber (the red line area). At high concentration (>12 wt.%), e.g., 14 wt.%, the gradient-step spindle-knots would not be excellent as the solution is too sticky. b) The gradient degree (GD) of well-formed structure on GSF (before dry) versus drawing velocity $V_t$ (ultimate velocity) by using solution concentration of 8 wt.%, 10 wt.%, 15 wt.%. 

Fig. S2: 

![Optical images of Rayleigh instability](image_url)
Fig. S3. a) The fiber with 5 different-sized polymer droplets numbered 1,2,3,4,5. b) GSF with 5 different-sized spindle-knots corresponding to the polymer droplets 1-5. The heights of polymer droplets are marked with H, widths are marked with W. The pitchs of spindle-knots are indicated with P. c) The relationship of H, W, P. The H is decreased in size of ~ 33.8–45% from polymer droplet (■ line) to spindle-knot (● line) for every ones after dry about 7 min. The W and P can be kepted mostly in size after dry.
**Figure S4:**

**Fig. S4. Optical images of water collection on fiber with uniform size spindle-knots.** 

a) A fiber with uniform size spindle-knots.  

b) At ~ 112 s, the condensed droplet 1, 2, 3, 4, 5 grow, and tend to be coalesced each other.  
c) The coalesced droplet (1+2+3), and droplet (4+5) appear at 200 s.  
d) Droplet (1+2+3) and droplet (4+5) become bigger. Droplet (4+5) falls off with the critical volume of ~ 11.2 μl at ~ 410 s.  
e) Droplet (1+2+3) falls off with the critical volume of ~ 8.7 μl at ~ 457 s without transportation along fiber. The scale bar is 1 mm. Green arrows represent the direction droplet transport and falling. The critical volume of droplet is estimated by using $V=(4\pi r_a^2 r_b)/3$, where $V$ is critical volume of droplet, $r_a$ and $r_b$ represent the radii of droplet as shown in graph, respectively.
Figure S5:

**Fig. S5.** In-situ observation on directional coalescence of a hydrophobic isooctane \(((\text{CH}_3)_2\text{CHCH}_2\text{C(\text{CH}_3)}_3)\) condensed droplet along the GSF. a) GSF with 5 spindle-knots with gradient steps \(L_{\text{gra}} \approx 6.8\) mm. b) The droplets 1,2,3,4,5 form on every knots, and droplet 1 tends to move at ~ 3.5 s. c) Droplet 1 coalesces with droplet 2, forms droplet (1+2) at ~ 4.8 s. d) Droplet (1+2) coalesces with droplet 3 at ~ 16.10 s. e) Continuous to coalesce with droplet 4, forms droplet (1+2+3+4) at ~ 16.15 s. f) Finally forming droplet (1+2+3+4+5) at ~ 16.2 s. g) The directional coalescence of droplets realizes the transport of droplet in \(L_{\text{tran}}=5.6\) mm along GSF with \(L_{\text{gra}}=6.8\) mm at ~ 34.36 s.
Fig. S6. Droplet transport stability between periodic gradients of GSF. a) Optical images of periodical GSF. There are three periodicities of continuous uni-direction gradient mode. b) Droplet transport property on the connecting segment (marked with red rectangle) between both gradient spindle-knots A→B→C and D→E→F (including the biggest spindle-knot adjacent to the smallest spindle-knot). Droplet 1, 2, 3 coalesce directionally into big droplet (1+2+3) at point C at ~ 531.3 s. Droplets finally move toward the biggest spindle-knot as target and meanwhile droplet 4, 5 coalesce directionally into big droplet (4+5) at point F, at ~ 277 s moving away from point D to F.
Fig. S7. Droplet coalescence with different modes. a) The normal coalescence. Droplets 1,2 coalesce into droplet (1+2), then coalesce with droplet 3 into droplet 3+(1+2), finally, coalesce into a bigger droplet 4+[3+(1+2)] at ~ 28.9 s. Another droplet 5 moves directionally at ~ 28.9–29.6 s. b-c) the “disorder” coalescence, as for (a-b) realising long-range transport, but for (c) the distance between two adjacent droplets is 2.8 mm > 2.2 mm, droplets can not transport continuously.
Fig. S8. Optical images of GSF with different gradient modes: a) Unidirection decreasing size, b) Unidirection increasing size; c) Middle symmetric size; d) Two-side symmetric size, which are fabricated by velocity-changable drawing-out coating technique. The different parameters are as following table S1.

Table S1: fiber fabricated parameters for different modes

<table>
<thead>
<tr>
<th>Gradient mode</th>
<th>$V_0$ (mm/s)</th>
<th>$V_t$ (mm/s)</th>
<th>$V_{t1}$ (mm/s)</th>
<th>$V_{t2}$ (mm/s)</th>
<th>$a$ (mm/s$^2$)</th>
<th>$a_1$ (mm/s$^2$)</th>
<th>$a_2$ (mm/s$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a) Uni-direction decreasing</td>
<td>0</td>
<td>4.8</td>
<td>--</td>
<td>--</td>
<td>7</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>b) Uni-direction increasing</td>
<td>6.3</td>
<td>0</td>
<td>--</td>
<td>--</td>
<td>--6</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>c) Middle-symmetric size</td>
<td>0</td>
<td>--</td>
<td>8.3</td>
<td>0</td>
<td>--</td>
<td>8</td>
<td>-9</td>
</tr>
<tr>
<td>d) Two-side symmetric size</td>
<td>9.3</td>
<td>--</td>
<td>0</td>
<td>7</td>
<td>--</td>
<td>-6</td>
<td>8</td>
</tr>
</tbody>
</table>

Note: the parameter can be defined: original velocity $V_0$, ultimate velocity $V_t$, the first stage ultimate velocity $V_{t1}$, the second stage ultimate velocity $V_{t2}$, accelerated speed $a$, the first stage accelerated speed $a_1$, the second stage accelerated $a_2$. 