Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information

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

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Supplementary Information contains:

Supplementary Figures S1-8.

Supplementary table S1.

Supplementary Figures and Legends

Figure S1:

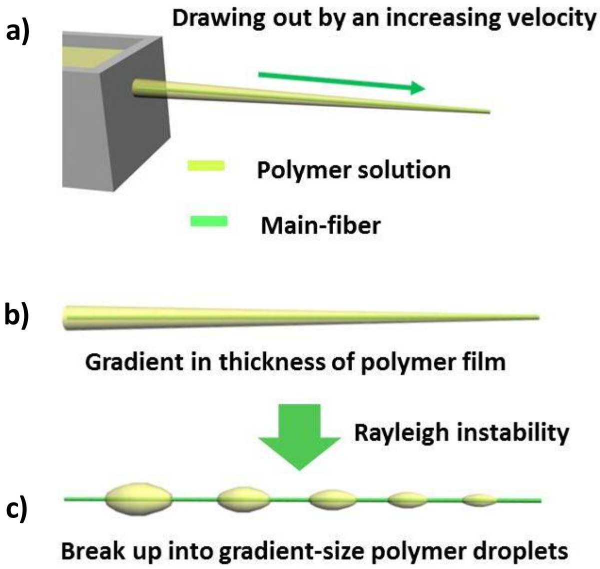

Fig. S1. Illustration of uni-direction gradient-step spindle-knots formed on fiber. a) A main-fiber is drawn out from polymer solution via an increasing velocity. **b)** A gradient thick polymer solution film forming along main-fiber. **c)** The cone-gradient thickness film would break up into the gradient-step 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.

Figure S2:

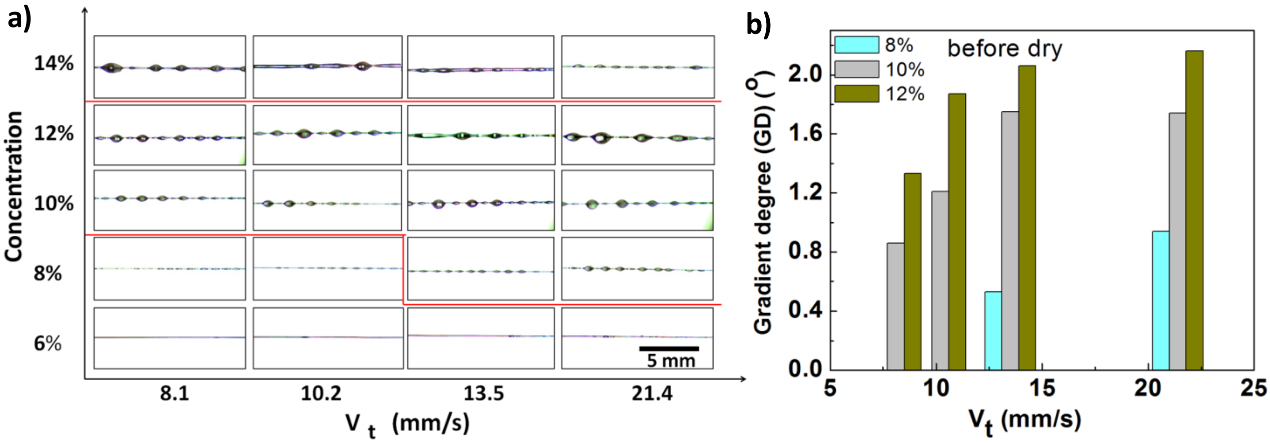

Fig. S2. Optical images of Rayleigh instability break-up into gradient size droplet via low and high concentration film at changing-controlled drawing velcoity of 8.1–21.4 mm/s. a) At low concentration (≤ 6 wt.%) the film is too thin, little gradient-step spindle-knot forms on fiber. When the concentration of solution (weight percent) is between 8%–12 wt.%, we can get excellent graidnet-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.%.

Figure S3:

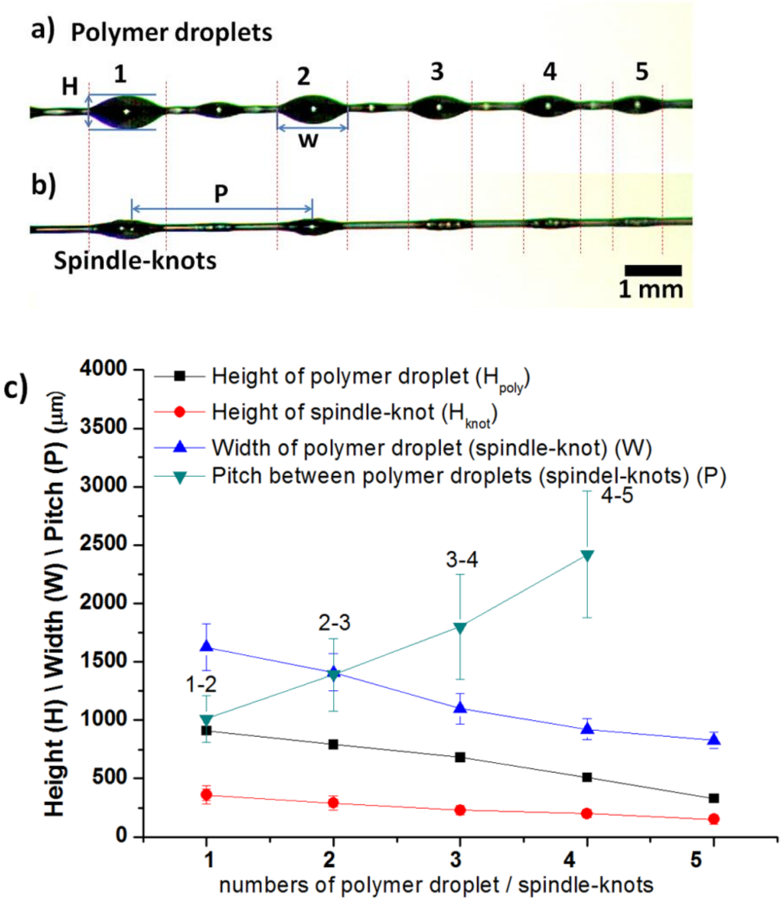

Fig. S3. a) The fiber with 5 differented-sized polymer droplets numbered 1,2,3,4,5. **b)** GSF with 5 different-sized spindle-knots corresponding to the polymer droplets 1-5. The heights of polymer droplets are marked with H, widths are marked with W. The pitchs of spindel-knots are indicated with P. **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 adfter dry about 7 min. The W and P can be kepted mostly in size after dry.

Figure S4:

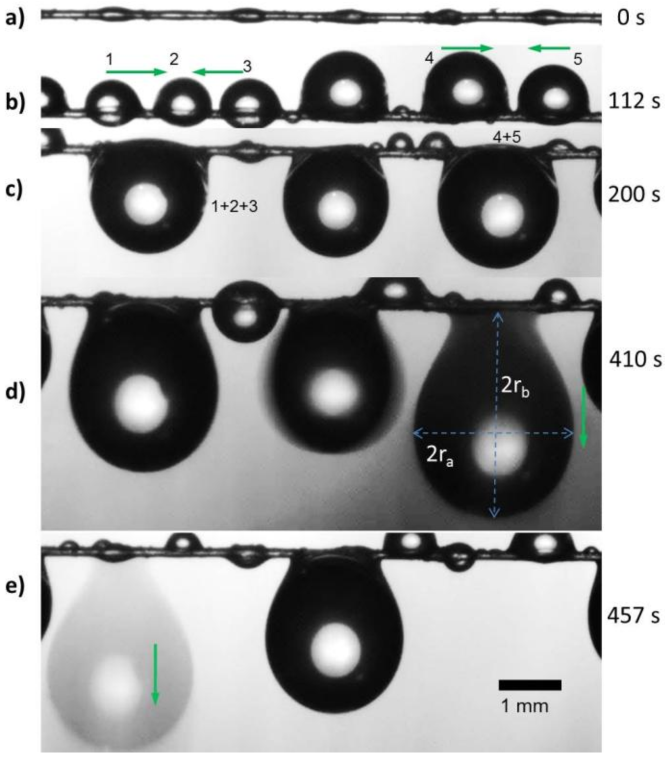

Fig. S4. Optical images of water collection on fiber with uniform size spindle-knots. a) A fiber with uniform size spindle-knots. **b)** At ~ 112 s, the condensed droplet 1, 2, 3, 4, 5 grow, and tend to be coalesced each other. **c)** The coalesced droplet (1+2+3), and droplet (4+5) appear at 200 s. **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:

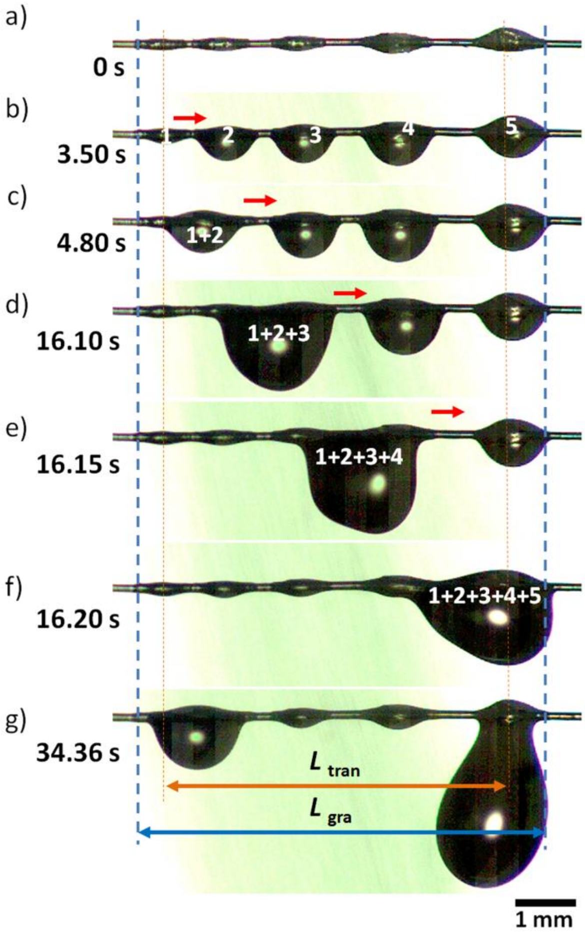

Fig. S5. **In-situ observation on directional coalescence of a hydrophobic isooctane ($(CH_3)_2CHCH_2C(CH_3)_3$) condensed droplet along the GSF**. **a)** GSF with 5 spindle-knots with gradient steps (L_{gra} ~ 6.8 mm). **b)** The droplets 1,2,3,4,5 form on every knots, and droplet 1 tends to move at ~ 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 direcitonal coalescence of droplets realizes the transport of droplet in L_{tran}=5.6 mm along GSF with L_{gra}=6.8 mm at ~ 34.36 s.

Figure S6:

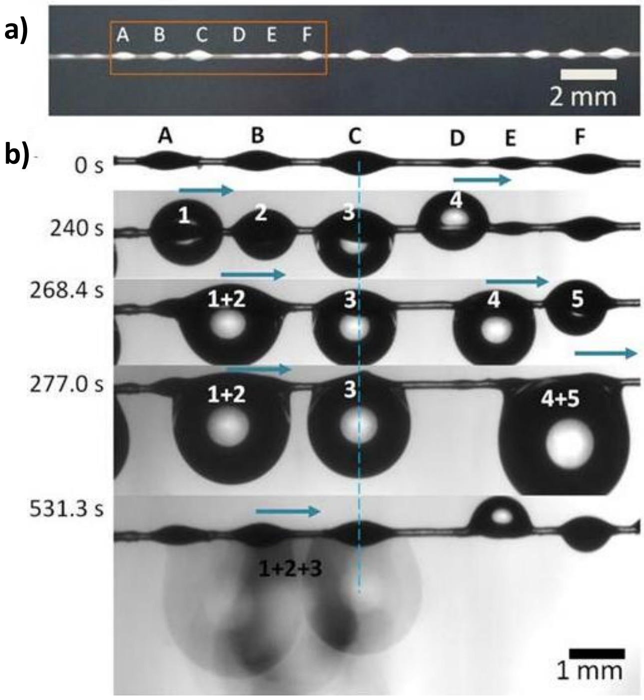

Fig. S6. Droplet transport stability between periodic gradients of GSF. a) Optical images of periodical GSF. There are three periodicities of continuous uni-direction gradient mode. **b)** Droplet transport property on the connecting segment (marked with red rectangle) between both gradient spindle-knots 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.

Figure S7:

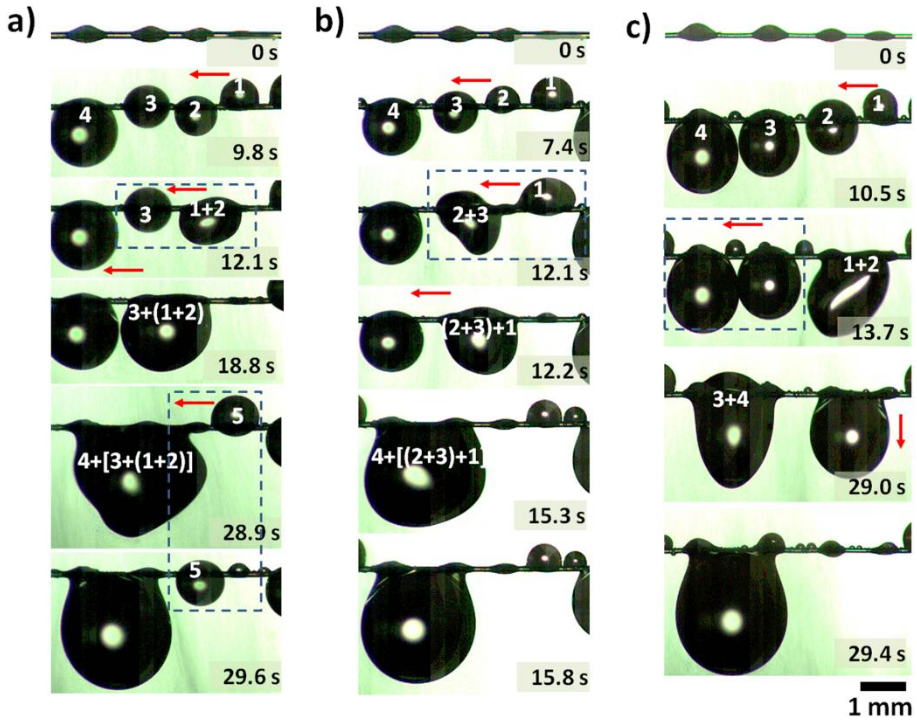

Fig. S7. **Droplet coalescence with different modes**. **a)** The normal coalescence. Droplets 1,2 coalesce into droplet (1+2), then coalesce with droplet 3 into droplet 3+(1+2), finally, coalesce into a bigger droplet 4+[3+(1+2)] at ~ 28.9 s. Another droplet 5 moves directionally at ~ 28.9–29.6 s. **b-c)** 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.

Figure S8:

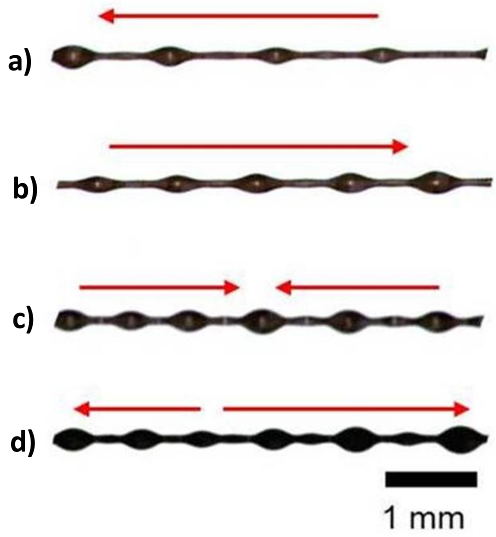

Fig. S8. Optical images of GSF with different gradient modes: **a)** Unidirection decreasing size, **b)** Unidirection increasing size; **c)** Middle symmetric size; **d)** Two-side symmetric size, which are fabricated by velocity-changable drawing-out coating technique. The diffferent parameters are as following table S1.

Table S1: **fiber fabricated parameters for different modes**

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

Note: the parameter can be defined: original velocity V_0, ultimate velocity V_t, the first stage ultimate velocity $V_{t\,1}$, the second stage ultimate velocity $V_{t\,2}$, accelerated speed a, the first stage accelerated speed a_1, the second stage accelerated a_2.