Electronic Supplementary Material (ESI) for Soft Matter

Supplementary Figure Legends

Fig. S1 Optical image of the capture silk shows six puffs along the two main-axis fibers, semi-transparent by observation of high-resolution optical microscopy.

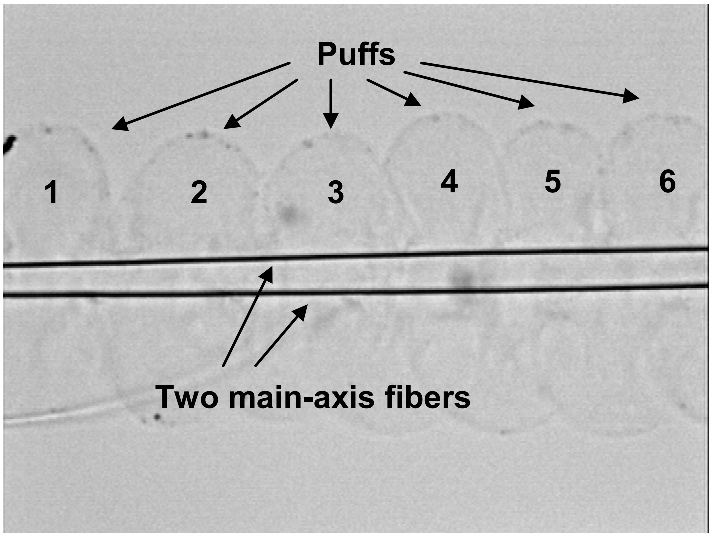

Fig. S1

Fig. S2 AFM image of rough feature by local observation on wetted spindle-knot. The rough surface can be composed of the nano-scale hump structures.

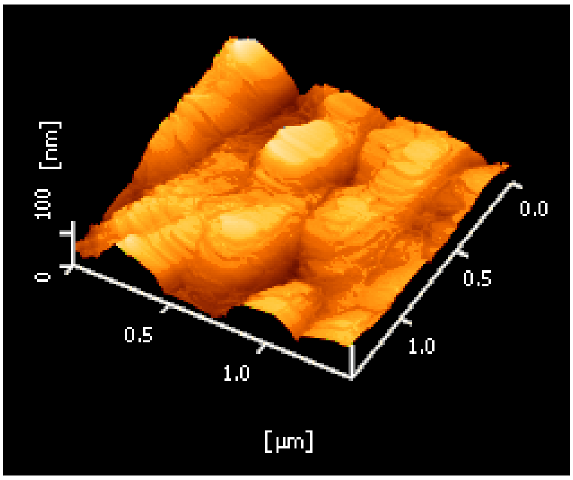

Fig. S2

Fig. S3 Optical image of artificial fiber with rough curved spindle-knots. It can be fabricated as following: The original fibers with about 6 ± 1.2 μm radii can be placed into polystyrene (PS) solution mixing with nanoscale silica sphere. Then a series of spindle-knots with silica sphere are distributed on the original fiber via the polymer-film break-up in Rayleigh instability[26] after rapidly drawing-out from above solution.

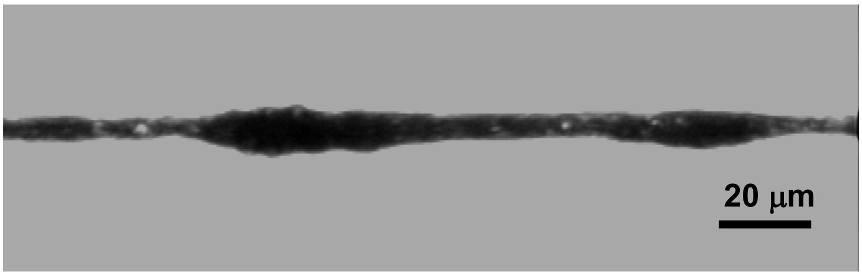

Fig. S3

Supplementary Analysis

Estimation on ability of adhesion-induced maximum hanging-drops

The water-capturing ability is valued by the maximum volume ratio ($V_{drop}/V_{silk,\ wet}$) of the hanging-drop to wet contact area on wetted spider capture silk. Based on optical image Figure S4a, the radii (R) of maximum hanging-drop is estimated as ~ 0.95 mm, so the maximum volume ($V_{drop}=4\pi R^3/3$) of drop is estimated as ~ 3.59 mm^3. The segment length (L) and average radii (R) of silk (that contacted by this maximum hanging-drop) is 0.519 and 0.0065 mm, respectively, and its volume ($V_{silk}=\pi R^2 L$) of contact area is estimated as ~ 6.89×10^{-5} mm^3. Figure S4b indicates the parameters of larger hanging-drop on horizontal silk. So, $V_{drop}/V_{silk,\ wet}$ is estimated roughly to up to ~ 52,000 in water-capturing ability.

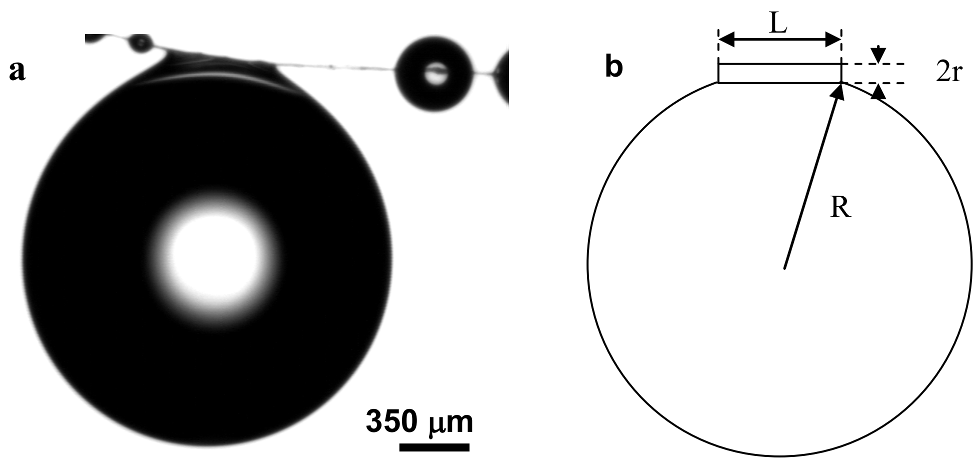

Fig. S4

Analysis on maximum radii of the hanging-drops on wetted spider silks

The water drops resting on silks, show that gravity could be balanced with adhesive force. For a horizontal silk, the water drop hanging was very similar to a drop pending from a cylindrical nozzle of radius (a), in which the weight of the drop $4\pi\rho g R^3/3$ could be balanced with the force of surface tension $2\pi a\gamma$.[22] According to the model of large drops hanging on the smooth thin fibers, the force resulting from the horizontal fiber was equivalent to the force generated by two similar fibers connected radially to the drop at the same location. Due to numerous humps with 200-500 nm diameters on the knot surface (Fig. 2b-c), the true area (A_{true}) was larger than the apparent area ($A_{apparent}$). The surface roughness r was ~ 1.32 that was estimated based on AFM image (Fig. S2). The adhesive force becomes $4\pi rb\gamma\cos(\pi/2-\alpha)$, was balanced by gravity, *i.e.*,

$$4\pi rb\gamma\cos(\pi/2-\alpha) = 4\pi\rho g R^3/3 \tag{1}$$

Thus radius of drop is:

$$R'_H \sim [3rb\kappa^{-2}\sin\alpha]^{1/3} \tag{2}$$

Where, α is the off-axis angle; the κ^{-1} was the capillary length $\kappa^{-1}=(\gamma/\rho g)^{1/2}$ (denoting ρ and γ as the liquid density and surface tension; κ^{-1} is 2.7 mm for water); b could be averaged by the number of drop covering the knots and the spacing of knots, *i.e.* $b\sim(kb_r+nb_a)/(k+n)$, where k and n were the number of 'knots' and of spacing between 'knots' covered by drop, respectively; b_r and b_a indicated the maximum and minimum radii of silk with periodic knots (Fig. 2c), respectively. According to the equation (2), we estimated R'_H ~ 0.74 mm (in Fig. 3c), at α ~ 71°, b ~ 0.015 mm.

Furthermore, considered into the effect on Laplace pressure in difference, the maximum radius of hanging-drop should be improved. Here, we consider the b and R are independent variable, so an equation can be obtained by differential coefficient to Eq. (1):

$$\Delta R = \Delta b \cdot r\kappa^{-2} \sin\alpha / R^2 \tag{3}$$

Where, given $\Delta b = b_r - b_a = 0.015$ mm, $R = 0.74$ mm, $\Delta R = 0.249$ mm. So we estimated the maximum radius of hanging-drop on horizontal silk can be $R'_{H,max} = R'_H + \Delta R = 0.989$ mm, which is in well agreement with the testing value (0.95 mm).

As for the vertical silk, the adhesive force acting on a drop was given by

$$b_r \cos\theta_r - b_a \cos\theta_a = \frac{2R^3}{3r\kappa^{-2}} \tag{4}$$

$$R'_V \sim [3/2 \cdot r\kappa^{-2}(b_r \cos\theta_r - b_a \cos\theta_a)]^{1/3} \tag{5}$$

From Eq.(5), $R'_V \sim 0.598$ mm, which is in basic agreement with the testing value ($R_V \sim 0.54$ mm).

Table 1 the testing data of off-axis angles α related to the upper (θ_u) /below (θ_b) contact angles of hanging-drops on the horizontal silk.

No.	sinα=1-h/*R*	α	h/*R*	Below CA (θ_b)	Upper CA (θ_u)	Error in θ_b	Error in θ_u	(θ_b−θ_u)
1	0.053	3.038	0.947	43.5	40	0.5	5	3.5
2	0.096	5.509	0.904	43	38	0.5	3.5	5
3	0.119	6.834	0.881	51	40	2.5	0.5	11
4	0.13	7.47	0.87	49	48	0.5	3	1
5	0.152	8.743	0.848	47	45	1.5	3.5	2
6	0.228	13.179	0.772	55	41	4	0.5	14
7	0.29	16.858	0.71	51	38	4	6	13
8	0.356	20.855	0.644	50.5	40	3.5	5	10.5
9	0.41	24.205	0.59	51	36	2	5.5	15
10	0.436	25.849	0.564	58.5	34.5	1	3	24
11	0.495	29.67	0.505	51	33.5	1	7	17.5
12	0.527	31.803	0.473	52	33	2.5	5.5	19
13	0.561	34.125	0.439	51.5	33	1.5	5.5	18.5
14	0.605	37.229	0.395	54	30	1.5	6.5	24
15	0.629	38.976	0.371	47	26	0.5	7	21
16	0.639	39.717	0.361	45	27.5	0.5	6.5	17.5
17	0.672	42.222	0.328	55	27.5	0.5	2	27.5
18	0.717	45.807	0.293	47	28	1	3	19
19	0.716	45.725	0.284	51	28	1.5	2.5	23
20	0.722	46.22	0.278	43	24.5	1	3	18.5
21	0.727	46.635	0.273	42	23.5	0.5	1.2	18.5
22	0.748	48.147	0.252	44	20.5	0.5	1	23.5

Note: h is the height of drop above horizontal silk; *R* is the radii of drop.

Table 2 the testing data of a hanging-drop on horizontal silk

No	R(mm)	α (^{o})	$R^3/b\kappa^{-2}$	sinα
1	0.122	4.4	0.0471	0.0771
2	0.148	6.1	0.08312	0.1065
3	0.172	6.3	0.1316	0.1093
4	0.201	8	0.2106	0.1388
5	0.222	11.6	0.2813	0.2005
6	0.238	15.0	0.3498	0.2594
7	0.275	19.1	0.5368	0.3279
8	0.315	23	0.8062	0.3908
9	0.327	24.3	0.906	0.411
10	0.360	31.9	1.2073	0.5278
11	0.367	32.3	1.2821	0.5345
12	0.389	37.3	1.5224	0.6054
13	0.413	41.4	1.8203	0.6613
14	0.441	45.2	2.2206	0.7095
15	0.465	48.4	2.6099	0.7476
16	0.487	50.4	2.9825	0.7703
17	0.492	53.5	3.0912	0.8035
18	0.58	75.5	5.05	0.968

Note: κ^{-1} is the capillary number, 2.7 mm for pure water; R is the radii of drop; α is the off-axis angle of drop; b is the effective radii of silk.

Table 3 the testing data of a hanging-drop on vertical silk

No	R (μm)	b_r	b_a	θ_r	θ_a	$R^3/b\kappa^{-2}$	$b_r\cos\theta_r$-$b_a\cos\theta_a$ (μm)
1	5.61	5.2	4.4	15	19	0.0242	0.886
2	7.07	5.2	4.2	14	20	0.0485	1.099
3	12.98	5.3	4.2	13	20	0.300	1.2172
4	15.39	5.4	4.2	12	21	0.500	1.3632
5	17.41	5.5	4.1	12	27	0.7242	1.7357
6	20.47	5.4	4.7	14	23	1.1758	0.9251
7	22.33	5.6	4.1	12	28	1.5273	1.8579
8	23.60	5.6	4.2	14	27	1.8030	1.6918
9	29.46	5.8	4.1	16	35	3.5061	2.2256
10	29.94	5.8	4.2	17	34	3.6818	2.0650
11	33.40	6.0	4.0	19	36	5.1091	2.4365
12	36.66	6.7	3.9	23	42	6.7606	3.2690
13	37.25	6.9	3.6	24	52	7.0879	4.0871
14	39.75	7.4	3.5	26	62	8.6151	5.0078
15	40.58	6.8	3.9	22	55	9.1667	4.068
16	41.02	7.7	3.5	23	64	9.4667	5.5536
17	41.80	7.8	3.5	23	65	10.2939	5.5754
18	42.18	7.6	3.7	25	61	10.2939	5.0941
19	44.51	7.8	3.5	23	65	12.0970	5.7563
20	436	11.8	4.2	21	78	11.35×10^3	10.146

Note: κ^{-1} is the capillary number, 2.7 mm for pure water; b_r, b_a are the top and bottom radius of the drop-coated region; θ_r, θ_a are the top and bottom semi-angle of drop.