Supplementary Materials A Large-Scale Spinning Approach to Engineering Knittable Hydrogel Fiber for Soft Robots

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- Movie S2. Unfold of compressed cage.

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Figure S1. Photograph of the self-lubricate phenomenon. (**A**) Before the removal of the hydrogel pillar, one end of pillar slipped out due to gravity. (**B**) Beginning of removal of the hydrogel pillar, the lubricant layer began to separate from the hydrogel pillar. (**C**) During removal, lubricant droplets formed after the hydrogel pillar. (**D**) After removal, lubricant left in the PTFE tube.



Figure S2. FTIR spectrum of solid AMPS, pregel solution, and collected lubrication layer.



Figure S3. Distance-Intensity curve of UV lamp used.



Figure S4. The diameter fiber produced with different PTFE tubes.



Figure S5. ESEM image of single network fiber. Cured statically, demolded by peeling the PTFE tube open.



Figure S6. SEM image of hydrogel fiber. (A) Lyophilized fiber. (B) Desiccated fiber.



Figure S7. Cycle performance of electro actuation under field strength of 0.5 V mm⁻¹.



Figure S8. Changes of fiber during swelling. (A) Swell curve of dry fiber. **(B)** ESEM image of fractured fiber, no cracks were found on the surface, indicating the cracks were formed from the inside.



Figure S9. Swell curve of water swelled fiber in TEG.



Figure S10. Comparison of mechanical strength of hydrogel fiber actuators. ¹⁻¹⁰



Figure S11. Cyclic tensile test of T-fiber. The T-fiber was continuously stretched and released at a set stress of 80% of its maximum tensile strength with a constant rate of 50 mm min⁻¹. After 30 cycles, the T-fiber exhibited a slight increase strain (below 2%), probably attributed to the translocation between polymer chains in fiber structure. The strain of the T-fiber reached to 7% after 79 cycles due to plastic deformation.



Figure S12. FTIR spectrum of TEG, TEG treated gel and pristine gel.



Figure S13. Illustration of welding of hydrogel fibers.



Figure S14. ESEM and SEM images of T-fiber knots. (A) ESEM image of water swelled T-

fiber knot. (B) SEM image of lyophilized water swelled T-fiber knot.



Figure S15. Comparison of mechanical strength. (A) Comparisons of tensile fracture strength for swelled fiber, T-fiber, and activated T-fiber. Cyclic tensile test of (B) as synthesized water swelled fiber, and (C) activated T-fiber. The fiber was stretched to 80% of its maximum tensile strength at a constant rate of 50 mm min⁻¹ and back continuously. The changes of the curves were caused by the gradually shrinking of fiber due to evaporation of water.



Figure S16. ESEM images of reswelled T-fibers. (A) ESEM image of water swelled T-fiber.

(B) ESEM image of activated T-fiber. (C) ESEM image of activated T-fiber's cross-section.



Figure S17. Electro-actuation performance of water swelled fiber and activated T-fiber.



Figure S18. Electro-actuation performance of single fiber under different electric field strength.



Figure S19. Illustration of the electric field in test tank. (A) Illustration of the method to connect the entire test water tank and make it into one electrode. (**B**) Side view of the electric potential distribution of such design. (**C**) Front view of the electric potential distribution of such design.



Figure S20. Illustration of the electric field around hand-shaped actuator. (A) Electric potential distribution of two electrodes in such design. (B) Illustration of supporting frame and electrode used in hydrogel hand.



Figure S21. UV-Vis Spectrum of the solution during the polymerization of AMPS (Diluted 200 times with deionized water). The UV-Vis spectra of the AMPS solution under different curing time were investigated. With the increase of curing time, the adsorption curve changed with the decreasing concentration of the initiator 2-Hydroxy-4'-(2-hydroxyethoxy)-2-methylpropiophenone. After UV curing for 20 min, the absorption curves gradually stabilized, indicating the complete reaction. To ensure the complete polymerization, we chose the UV curing time as 30 min.



Figure S22. Setup of the SLS equipment.



Figure S23. Photograph of surfaced cracked fiber.



Figure S24. ¹H NMR of partially polymerized pregel solution after the first polymerization without the crosslinker. The rate of conversion was derived from the peak area by $3b/(c+d+3b) \times 100\%$.



Figure S25. SEM image of the lyophilized core-sheath fiber



Figure S26. Equipment and definition of the electro-actuation test. (A) Schematic of water tank used for the electro-actuation test. **(B)** Measurement of Bending angle.



Figure S27. Comparison of camera raw filter. (**A**) Raw ESEM image. (**B**) Camera Raw filtered ESEM image.

Method	Major composite	Diameter (mm)	Feeding rate (uL min ⁻¹)	Post polymerization	Tensile strength (MPa)	Ref.
Self- lubricate spinning	Acrylamide Poly(2- Acrylamido- 2-	0.59 to 2.43	30 to 250	None	0.114 to 5.6 (T-fiber)	This work
	methylpropa nesulfonic acid)					
Solution spinning	Acrylic acid Polyvinyl Alcohol	0.57 to 1.57	N/A	20 h	0.485 to 0.738	2
Solution spinning	Sodium alginate	0.24 to 0.6	50 to 400	30 min	0.06 to 0.25	10
Microfluid spinning	Poly(N- isopropylacryl amide) Sodium alginate Graphite oxide	0.4 to 0.98	300 to 800	20 h	0.087 to 0.194	11
Solution spinning	Chitosan Poly(ethylene	0.9 to 1.8	100 to 400	4 h	0.07 To 0.25	3
Microfluid spinning	N- isopropylacryl amide Poly(N- isopropylacryl amide-co- hydroxyethyl methacrylate)	0.04 to 0.21	1300 to 3000	24 h	N/A	12
Electro- spinning	Poly(N - isopropyl acrylamide)	N/A	N/A	4 h	N/A	13
Microfluid spinning	Poly(N - isopropyl acrylamide)	0.05 to 0.72	50 to 400	None	0.17 to 0.42	4

Table S1. Comparison of different spinning methods.

	Sodium alginate Graphite oxide					
Microfluid spinning	Acrylamide Sodium alginate	0.2 to 0.6	N/A	20 h	0.143 to 0.393	1
Dynamic crosslinki ng spinning	Poly(ethylene glycol) diacrylate	0.041 to 0.118	150 to 450	None	N/A	14
Electrospi nning	Acrylamide Poly(acrylami de)	0.01	N/A	4 h	N/A	15

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