Supporting Information for

Dry-Spinning Approach to Continuous Graphene Fibers with High Toughness

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Materials: Graphite powder (35 mesh) was purchased from Qingdao Henglide Graphite Co., Ltd. HI (45%), H2SO4 (98%), KMnO4, P2O5, HCl (38%), H2O2 (30%) and ethanol (95%) were purchased from Sinopharm Chemical Reagent Co. Ltd.

Characterizations: The liquid crystals (LC) behavior of graphene oxide suspension and coagulating process of GO fiber were characterized by polarized optical microscope (POM) with ZEISS Axio Scope A1. Rheological behavior was characterized by a ARG2 rheometer. The surface morphology and cross section structure of fibers were characterized by a Hitachi S4800 field-emission scanning electron microscope (SEM) system. Mechanical property tests were performed on HS-3002C with a loading rate of 0.5mm per minute and the gauge distance was 5mm. All the electrical properties were measured by Keithley 2400 multiple-function source-meter. Raman spectra was acquired using a Reinshaw inVia-Reflex Raman microscopy at an excitation wavelength of 532 nm.

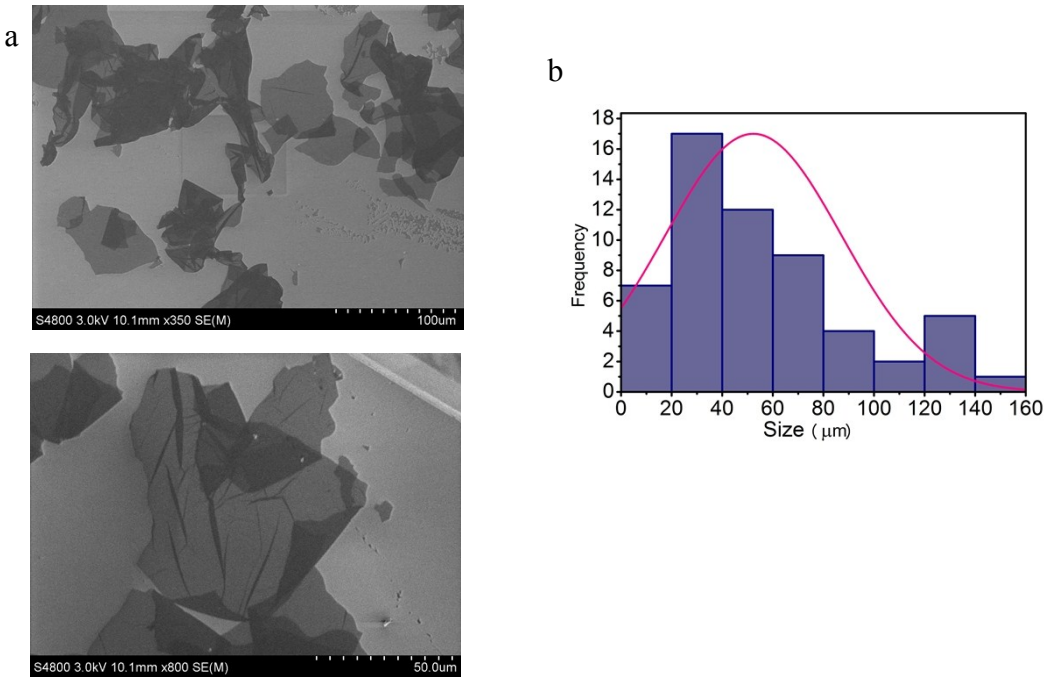

Figure S1. Size distribution of GO sheets. (a) SEM images of GO sheets on a silicon wafer. (b) The histogram of size distribution of GO sheets.

Figure S2. The shrink of GO gel fiber from radial direction. The POM images of GO gel fiber after 0s (a), 45s (b) and 90s (c).

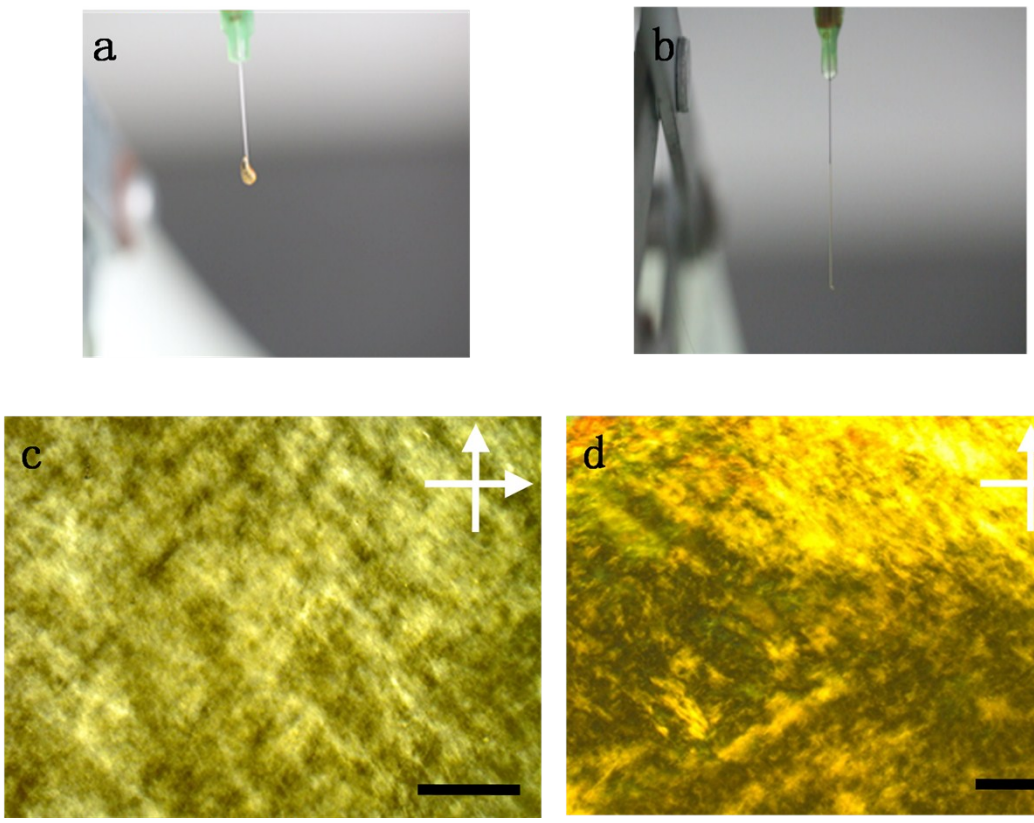

Figure S3. (a) A droplet formed from 4.5 mg g^{-1} GO/THF-H_2O dispersion. (b) A gel fiber formed from 8 mg g^{-1} GO/THF-H_2O dispersion. (c) (d) The POM images of 4.5 mg g^{-1} and 8 mg g^{-1} GO/THF-H_2O dispersion, respectively. Scale bar: 500μm.

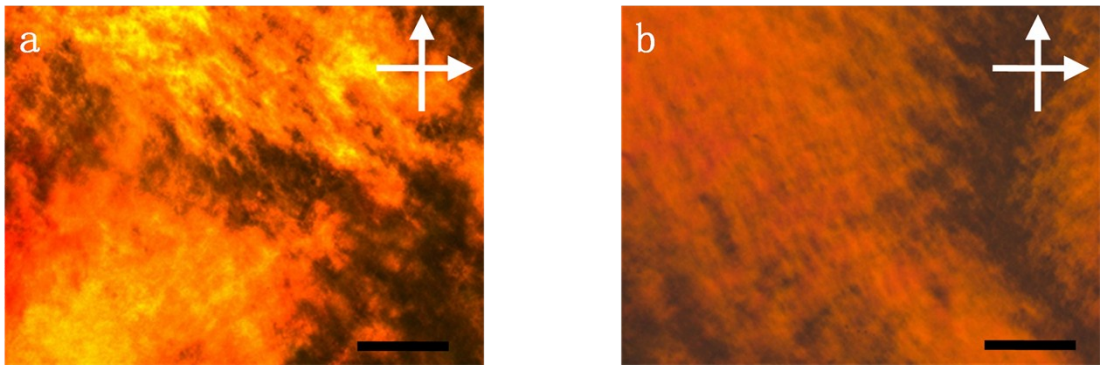

Figure S4. The POM images of GO/DMF-H_2O dispersion (a) and GO/ethanol-acetone dispersion (b). Scale bar: 500 μm.

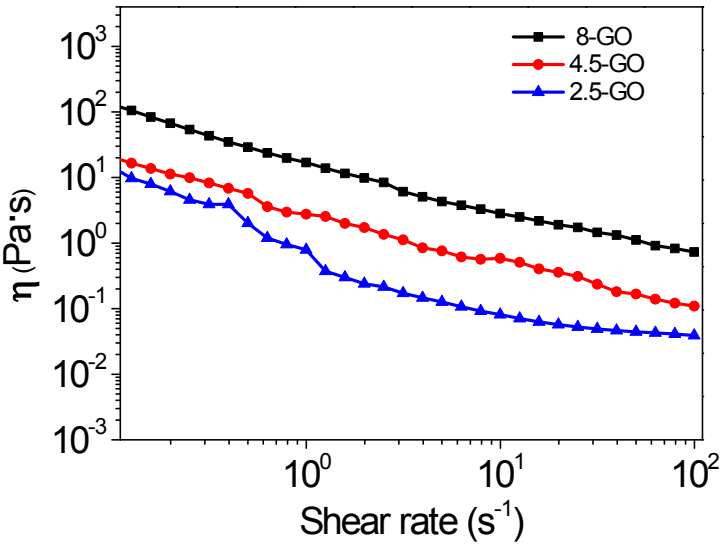

Figure S5. The viscosity of GO dispersion at 2.5 mg g^{-1}, 4.5 mg g^{-1} and 8 mg g^{-1} with different shear rate.

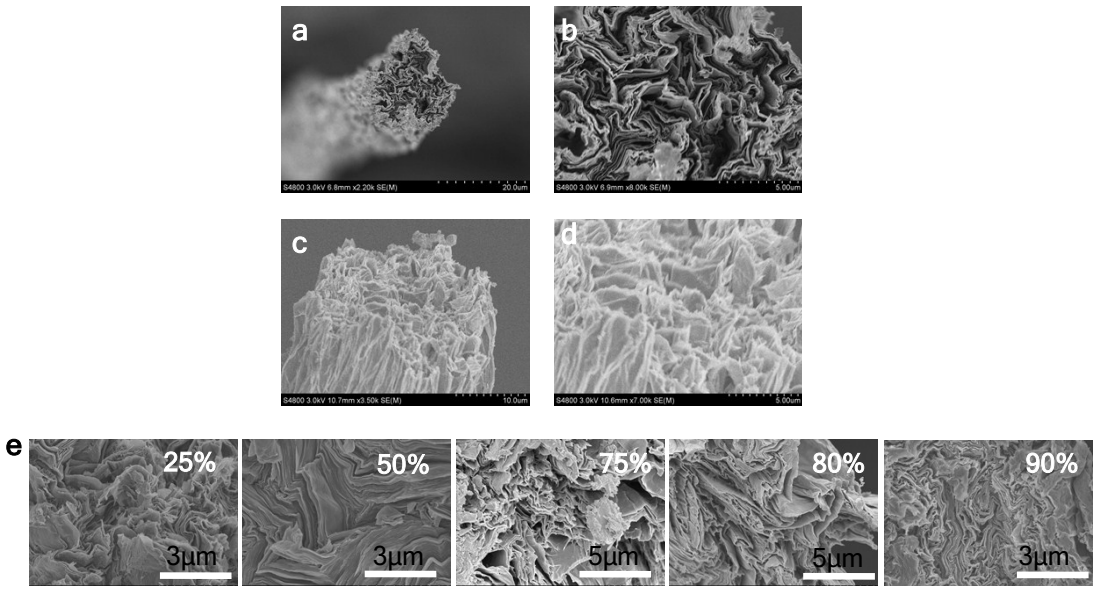

Figure S6. The SEM images showing the cross section of rGO fiber and GO fiber. (a) (c) The cross section images from front and side direction, respectively. (b) (d) The magnified images of (a) (c), respectively. (e) SEM images of transverse section of GO fibers with different volume ratio of THF.

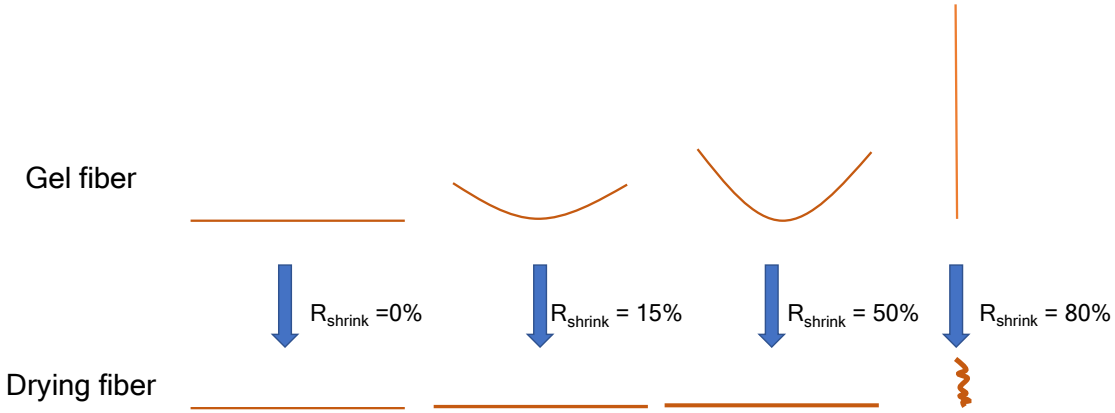

Figure S7. A scheme of the drying process of GO gel fiber hanging a support with different shrinkage ratio.

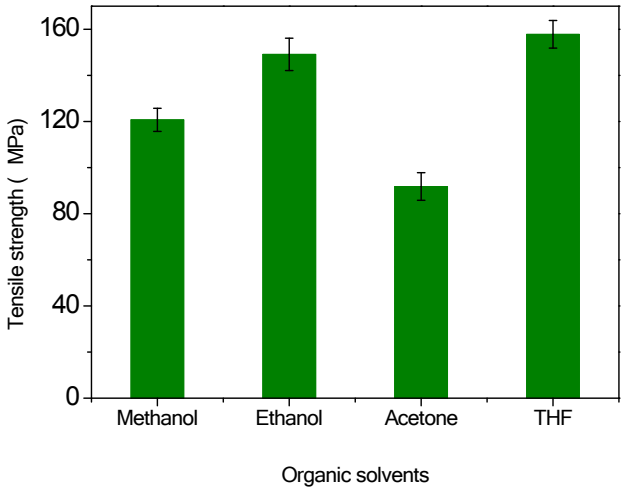

Figure S8. Comparison of the mechanical properties among the fibers with different organic solvents including methanol, ethanol, acetone and THF. (The volume ratio between organic solvent and water is 1:1. The GFs fabricated from THF/H_2O dispersion possessed the best mechanical property.)

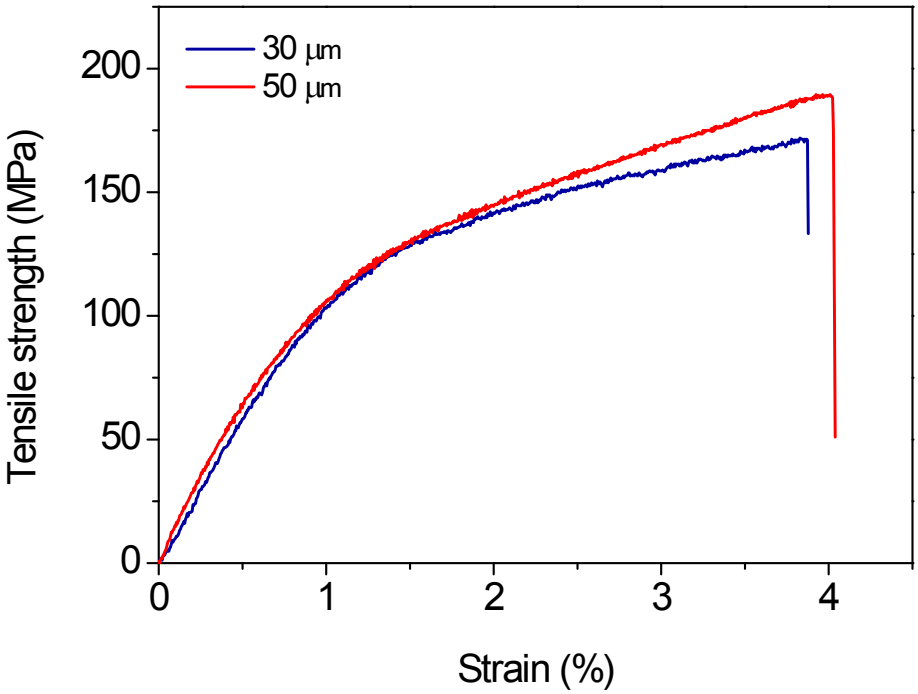

Figure S9. The mechanical properties of GFs with medium (~30 μm) and large (~50 μm) GO sheets. For the small (5 μm) size GO, droplets only formed after the nozzle but not continuous gel fiber, and we can not get continuous GO fibers by dry-spinning. For the medium and large GO sheets, the mechanical behavior of their wet-spun fibers is quiet similar, implying that the medium size is beyond the critical size, above which the mechanical strength of fibers scale up with the GO size.

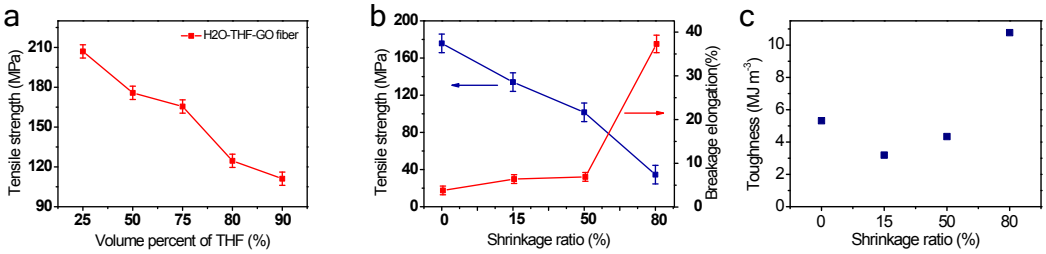

Figure S10. The mechanical property of GO fiber. (a) The fiber fabricated from GO dispersion with different volume percent of THF. (b) The fiber fabricated with different shrinkage ratio.

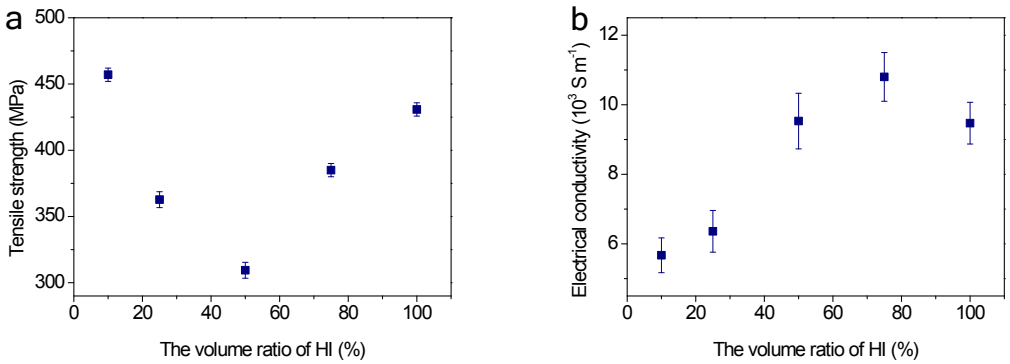

Figure S11. Comparison of mechanical and electrical properties among the GFs reduced with different volume between HI and ethanol including 4.5%, 11.25%, 22.5%, 33.75% and 45%. As the HI concentration increased, the trend of tensile strength had a valley at the HI concentration of 22.5%. This valley possibly caused by the competition between the weakening effect by ethanol (fast evaporation can give voids inside) and the enhancement effect by HI solution (it can bring dense structure of fibers). With increasing the concentration of HI, the electrical conductivity increased, however, when the concentration was up to 33.75%, the electrical conductivity became the highest value, which implies that the degree of reduction of GOFs is saturated.

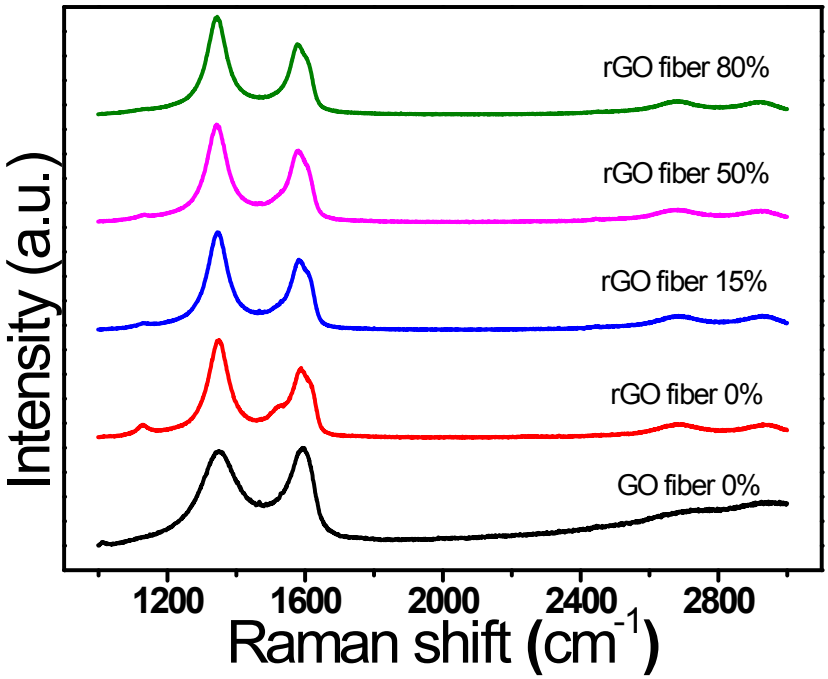

Figure S12. The Raman spectra of GO fiber with different shrinkage ratio and rGO fiber without shrink.

Table S1. Physical properties of solvent.

Property / Reagent	B.P. (°C)	S. V. P. (KPa)	S. T. (mN/m)(25°C)
DMF	152.8	3.46 (60°C)	32.5
H_2O	100	2.20 (19°C)	72
Ethanol	78.3	5.33 (19°C)	22.39
Acetone	56.5	53.32 (39.5°C)	18.8
THF	65.4	15.2 (15.2°C)	26.4
Methanol	64.7	12.3 (20°C)	18.8

Note: B. P., S. V. P. and S. T. are the abbreviation of boiling point, saturate vapor pressure and surface tension, respectively.

Table S2. Hansen solubility parameters of solvents.

Solvent	δ_d	δ_p	δ_h	$\delta_p + \delta_h$
Acetone	15.5	10.4	7.0	17.4
THF	16.8	5.7	8.0	13.7
DMF	17.4	13.7	11.3	25.0
Ethanol	15.8	8.8	19.4	28.2
Methanol	15.1	12.3	22.3	34.6
Water	15.5	16.0	42.3	58.3