

1 Supporting information for

2 **In-situ Alignment of Graphene Nanoplatelets in Poly (vinyl alcohol)**
3 **Nanocomposite Fibers with Controlled Stepwise Interfacial Exfoliations**

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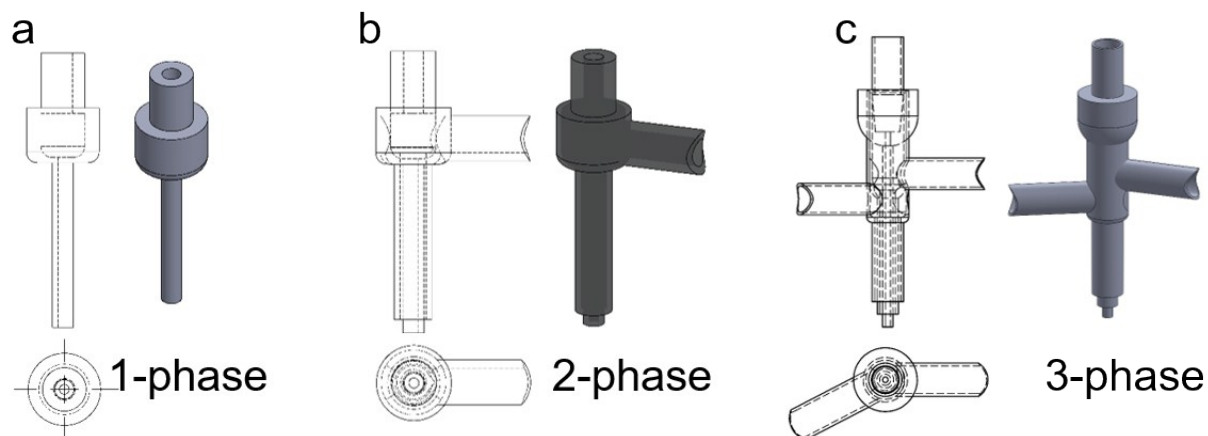
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21 **Figure S1.** In-house design using mechanical modeling software, Creo Direct Express, of
 22 spinnerets for (a) 1-phase/D-phase, (b) 2-phase, and (c) 3-phase fibers. All spinnerets were
 23 printed using 3D printing technics (i.e., fused deposition modeling (FDM), Dimension Elite,
 24 Stratasys). Acrylonitrile butadiene styrene (ABS) was used as the resin. After printing,
 25 spinnerets were soaked in dilute NaOH solution at 70 °C for 8 hours to eliminate the supporting
 26 structures. The minimum feature for the printer is 0.5 mm.

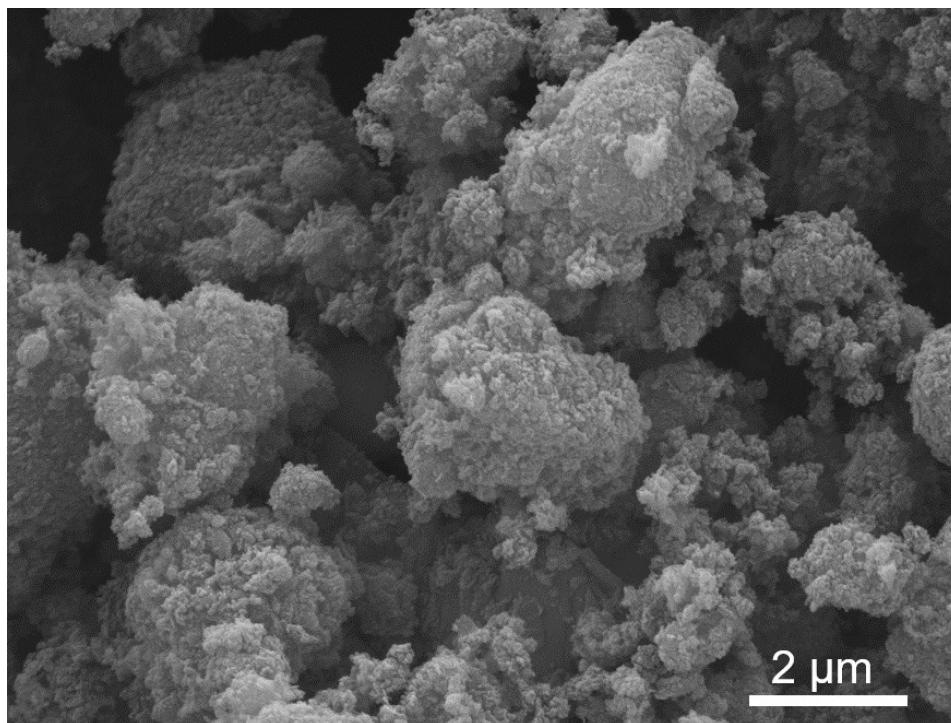
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Fiber Spinning Conditions				
Fiber	3-phase	2-phase	1-phase	D-phase
Spinning dope compositions	20 wt% PVA in DMSO, and 20 wt% GNPs in DMSO		20 wt% PVA in DMSO	3.5 wt% GNPs/PVA dissolved in DMSO with 20 wt% PVA/DMSO
Injection speed (m/min)	PVA: 3 GNPs: 1			
Air-gap distance (cm)	1-2 cm			
Take-up speed (m/min)	8			
Coagulation	24 hrs in methanol at room temperature 25 °C			
Fiber Drawing Conditions				
	3-phase	2-phase	1-phase	D-phase
DR ₋₁₀₀	8.47	7.74	7.55	6.87
DR ₋₁₅₀	1.59	1.44	1.30	1.26
DR ₋₂₀₀	1.21	1.33	1.30	1.16
DR_{-total}	16.29	14.82	12.76	10.04
D ₋₁₀₀ (μm)	111.2	124.2	138.1	124.8
D ₋₁₅₀ (μm)	88.9	103.7	103.8	110.9
D ₋₂₀₀ (μm)	80.3	89.8	91.1	102.9
Fiber structure	PVA/GNPs/PVA laminate	PVA/GNPs core-shell	PVA	GNPs dispersed in PVA

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29 **Table S1.** Experimental details of fiber spinning and drawing conditions. Since the fibers were
30 not perfectly circular after drawing, their diameter values were calculated based on the weight
31 method (i.e., mass and length of the fiber were used to obtain the cross-section areas and
32 equivalent diameters). PVA and GNPs densities were 1.19 g/cm³ and 2.2 g/cm³, respectively.
33 DR: draw ratio; D: diameter (μm).

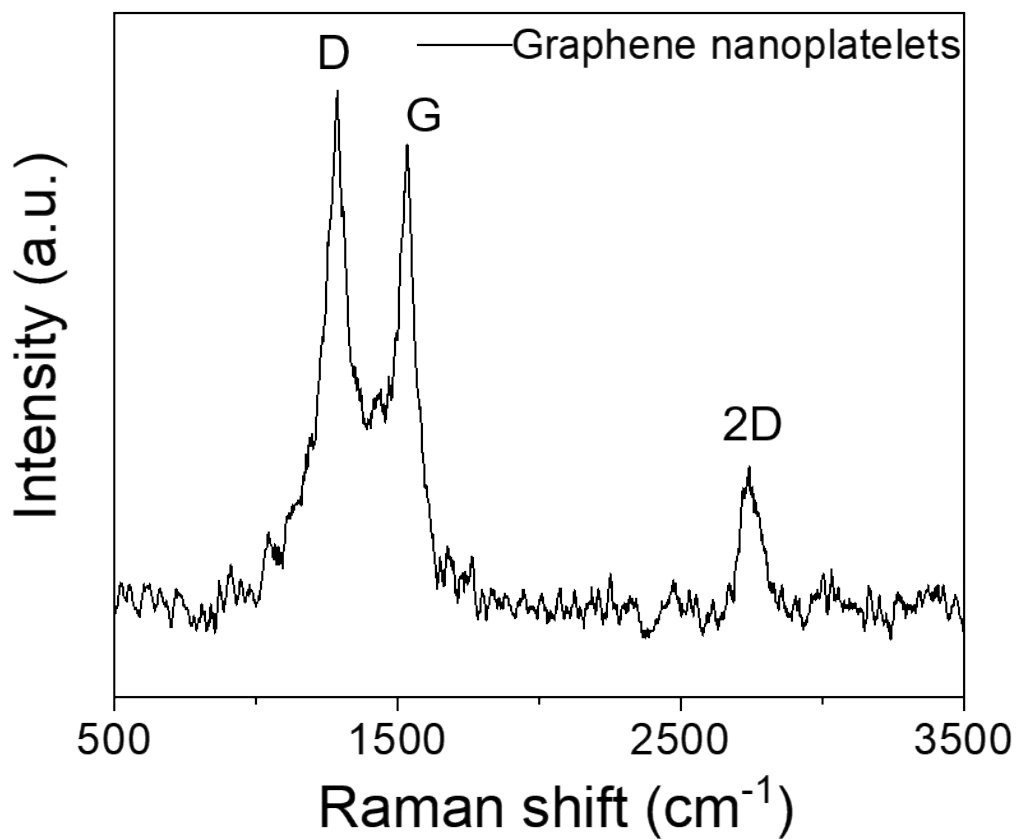
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36 **Figure S2.** SEM image of the GNPs as received shows a particle size smaller than 2 μm. The size
37 is not very uniformly distributed.

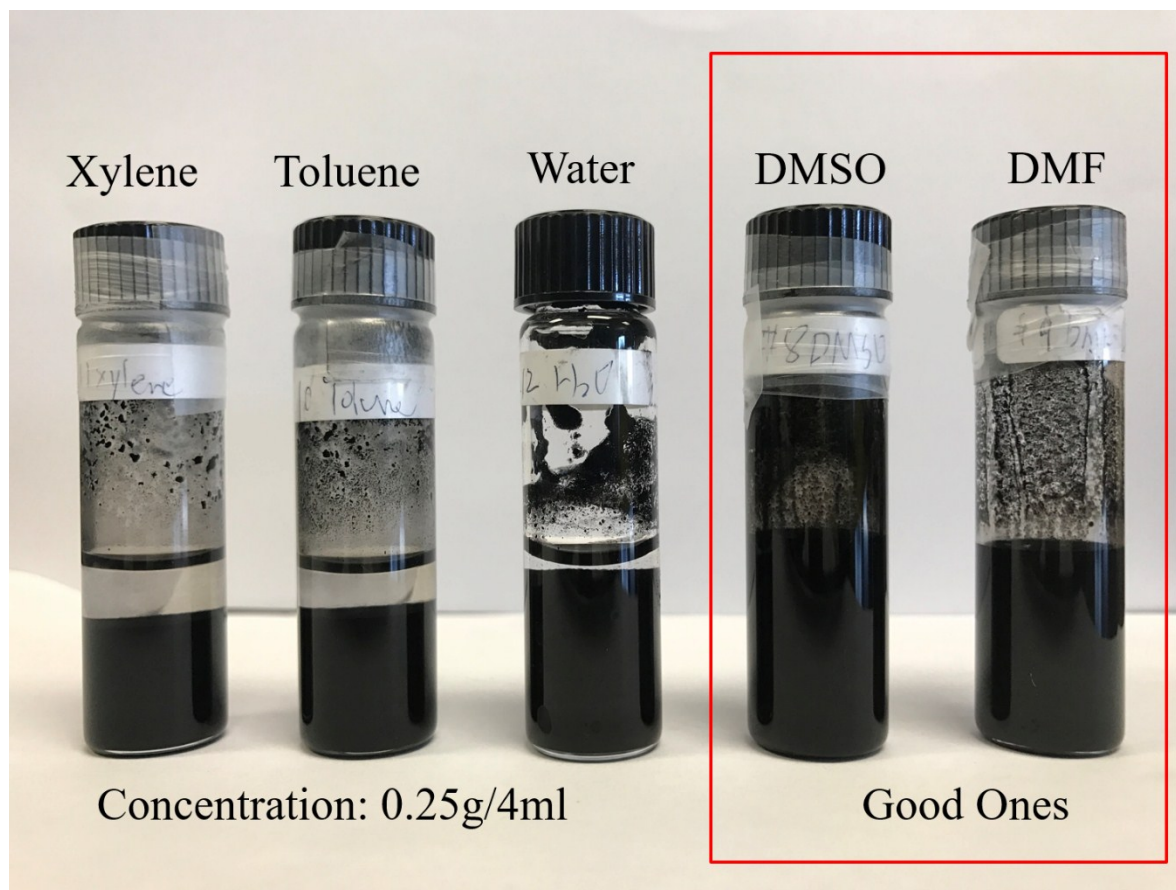
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40 **Figure S3.** Raman spectroscopy of the obtained GNPs. The D band, G band, and 2D band are at
41 1350 cm⁻¹, 1530 cm⁻¹, and 2750 cm⁻¹, respectively. The intensity ratio of the D and G band is ~1,
42 which is relatively high, indicating the presence of some defects and disorders.

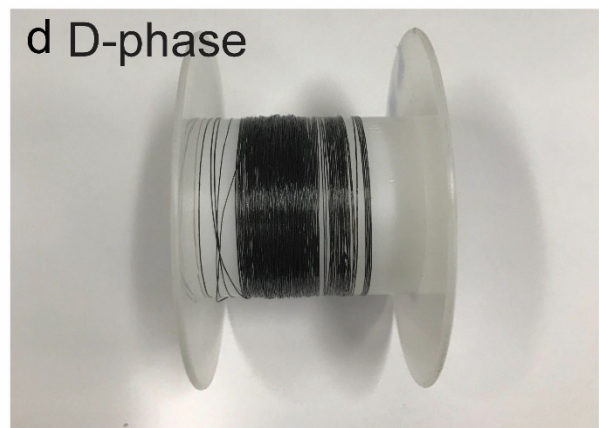
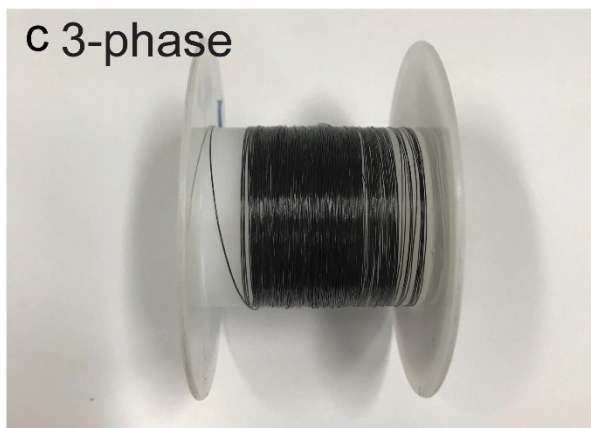
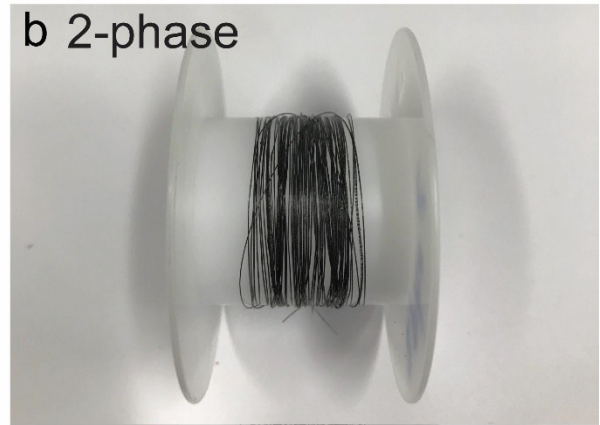
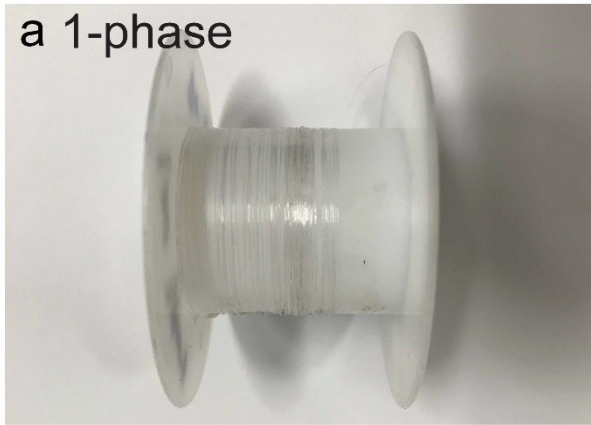
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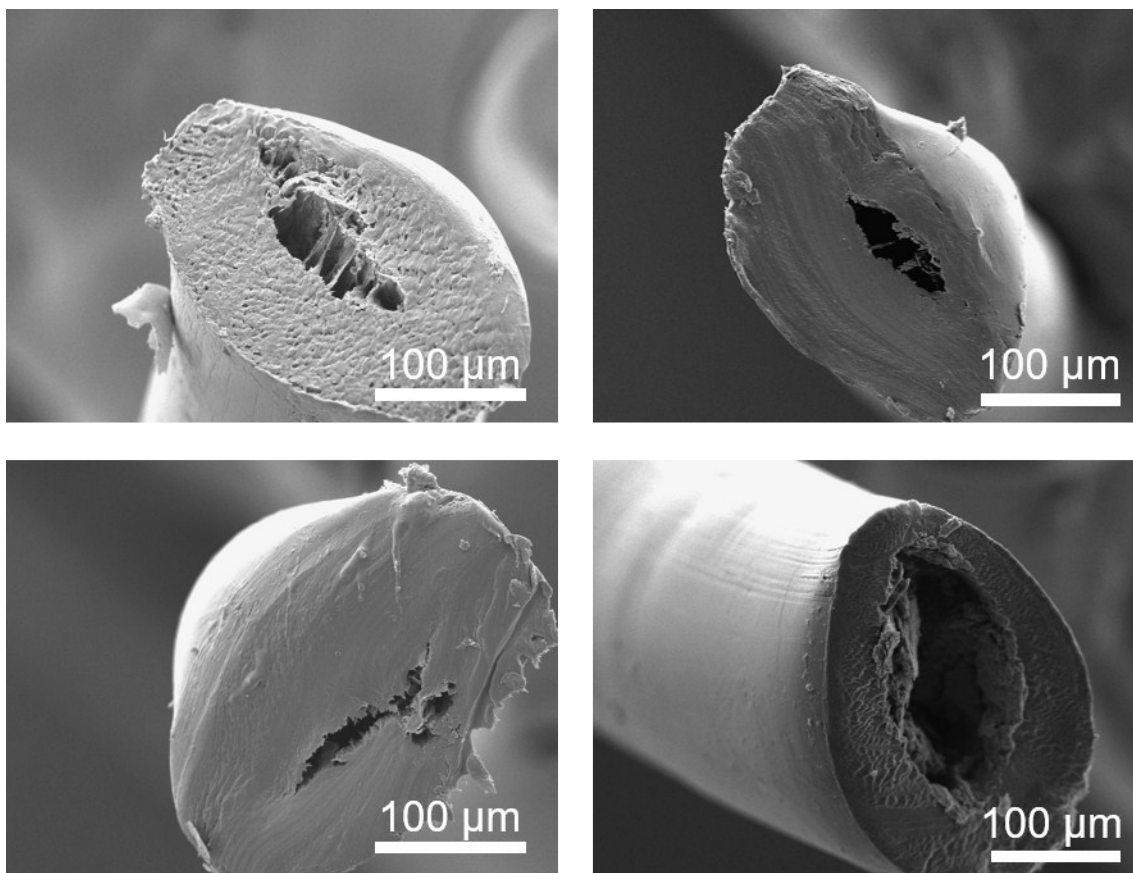
46 **Figure S4.** Dispersion quality of GNPs in 5 solvents: xylene, toluene, water, dimethyl sulfoxide
47 (DMSO), and dimethylformamide (DMF). All five samples were stirred using a vortex shaker and
48 were tip sonicated for 10 minutes. The above image was taken after 2 hours of rest. DMSO and
49 DMF had similar dispersion qualities, whereas xylene, toluene, and water had considerable
50 GNPs sedimentation.

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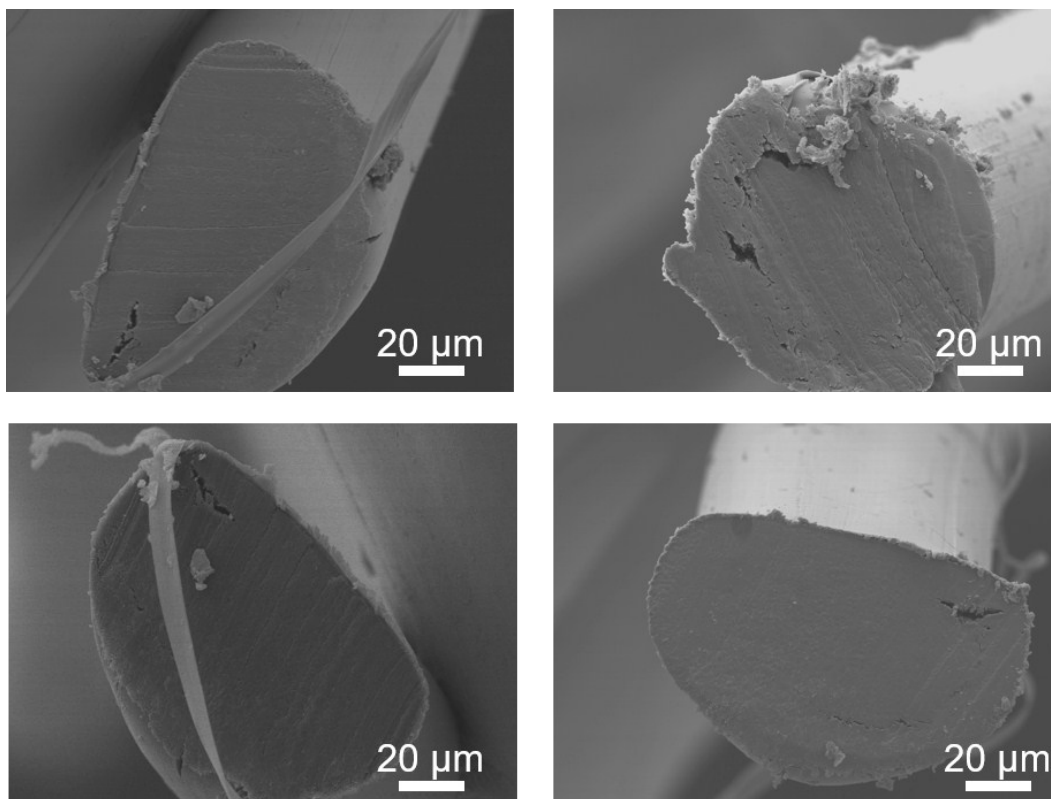
53 **Fig S5.** Post-drawn fibers of (a). 1-phase, (b) 2-phase, (c) 3-phase, and (d) D-phase.



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55 **Figure S6.** SEM of 2-phase fibers at four different cross section points. The variation in void
56 diameter and GNPs distribution indicate poor interactions between GNPs and polymer matrix in
57 the 2-phase fibers.

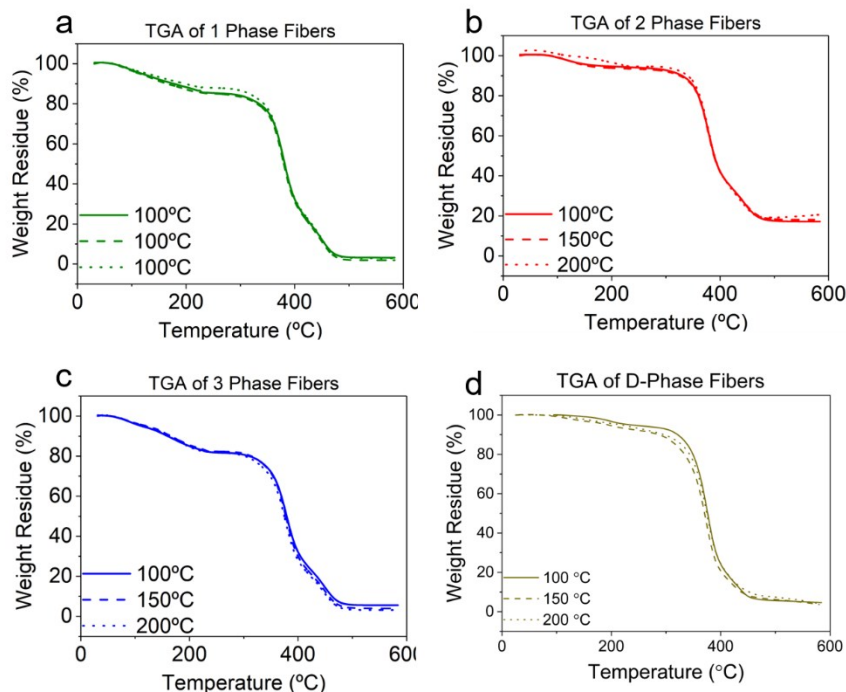
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60 **Figure S7.** SEM of 3-phase fibers at four different cross section points. Compared to SEM
61 images of undrawn fibers, the area of the middle layer is significantly reduced. Note that
62 although liquid nitrogen was used before cutting the fiber, PVA polymer was still sheared and
63 covered part of the GNPs channel.

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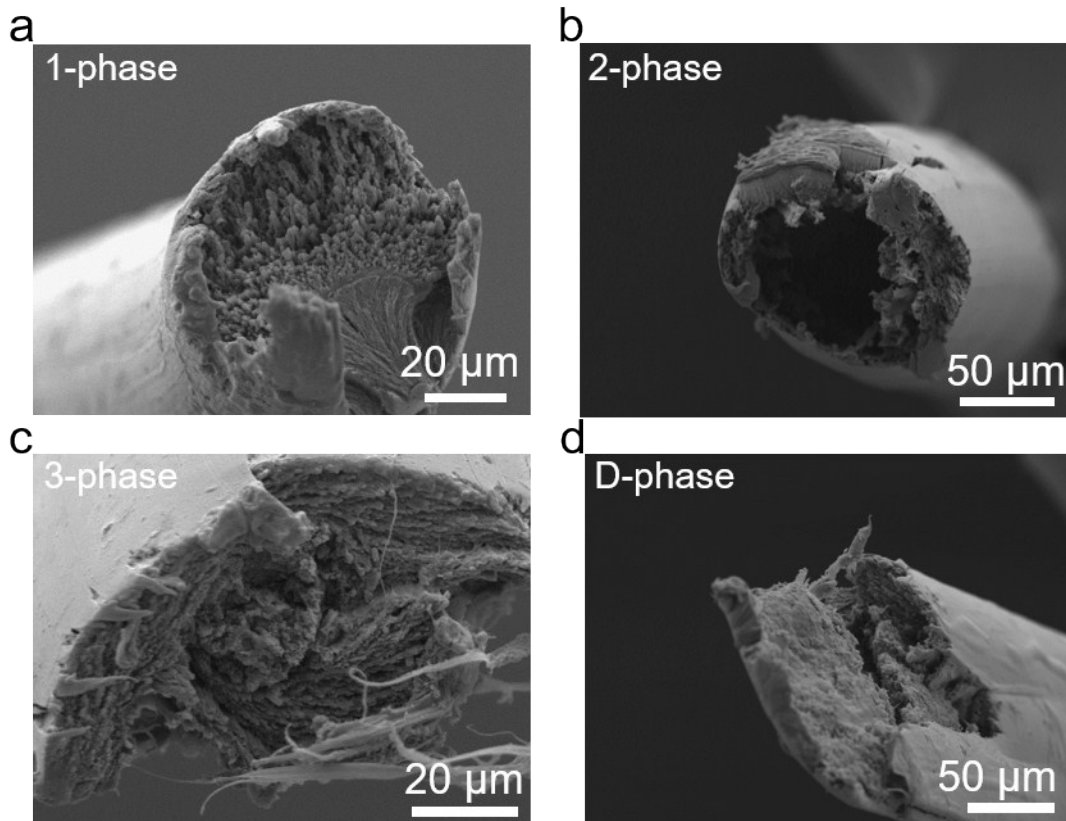
66 **Figure S8.** Thermogravimetric analysis on (a) 1-phase, (b) 2-phase, and (c) 3-phase (d) D-phase

Fiber samples	Draw ratio	Weight residue (%)
2-phase	7.74	16.2
	11.15	18.2
	14.82	19.0
3-phase	8.47	6.0
	13.4	4.4
	16.29	3.3
D-phase	6.9	4.3
	8.7	3.7
	10.4	3.8

67 fibers from room temperature to 600 °C under nitrogen.

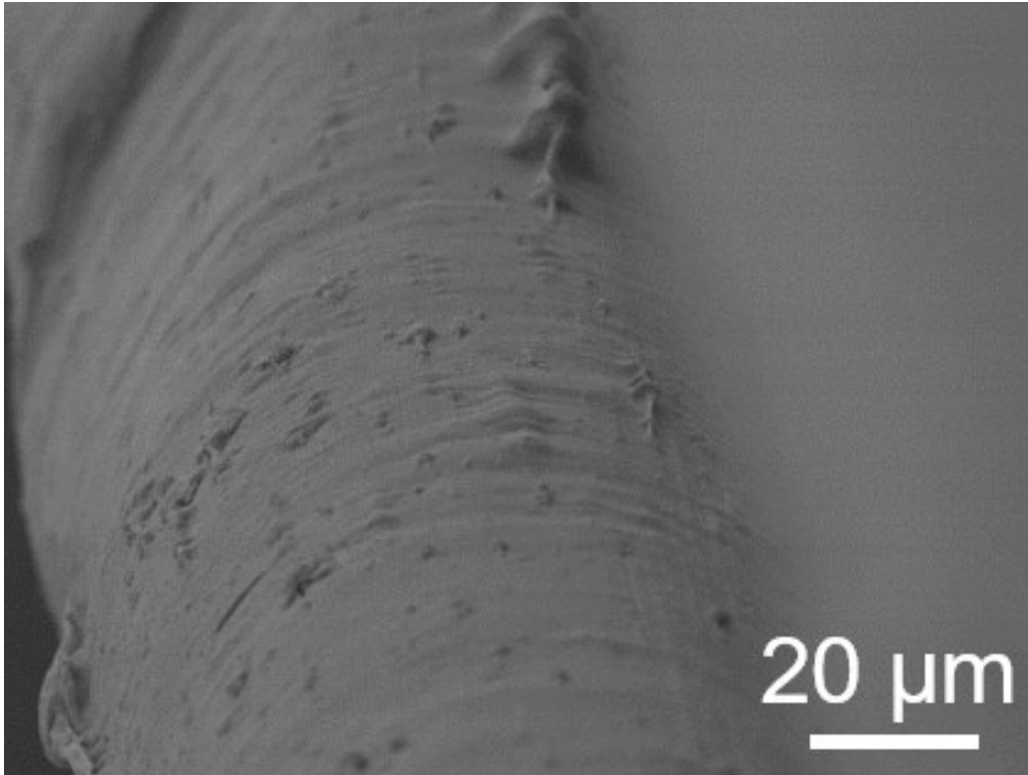
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69 **Table S2.** Weight residue changes as a function of draw ratio. Weight residue of the 1-phase
70 fiber was used to calibrate the weight residue contributed from potential leftover carbonized
71 PVA polymer. For 2-phase fibers, weight residue percentage increased as the draw ratio
72 increased. For 3-phase fibers, weight residue percentage decreased as the draw ratio increased.
73 For D-phase fibers, weight residue maintained the same.



74 **Figure S9.** SEM images of cross-sections after the tensile test for (a) 1-phase, (b) 2-phase, (c) 3-
75 phase, and (d) D-phase fibers. For 2-phase fibers, the fracture point was close to the void by the
76 aggregated GNPs. The clear fracture area of the 1-phase and 3-phase fibers suggests a better
77 extension of the polymer chains, which was reflected from their smaller strain values. For D-
78 phase, the fiber end diameters were reduced, indicating that the polymer chains were drawn to
79 a lesser degree.

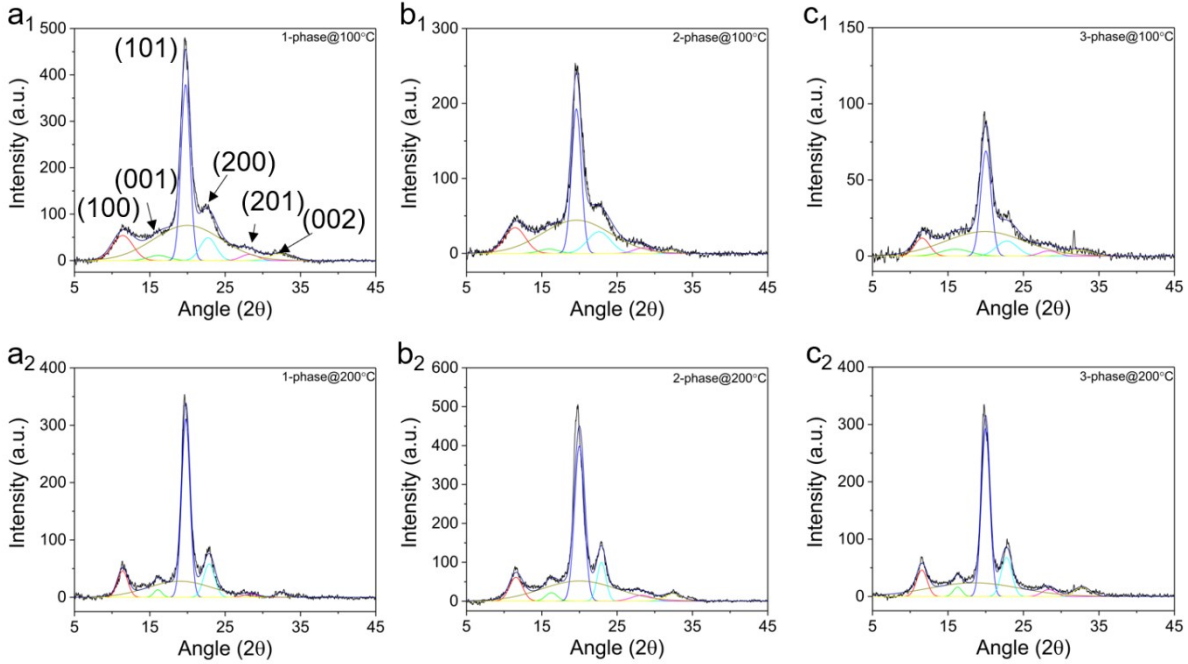
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82 **Figure S10.** SEM image of the surface of the D-phase fiber after drawing.

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85 **Figure S11.** XRD analysis of three types of fibers. (a₁), (b₁), and (c₁) are XRD patterns for 1-
 86 phase, 2-phase, and 3-phase fibers at 100 °C. (a₂), (b₂), and (c₂) are XRD patterns at 200 °C. The
 87 (1 0 1) planes are located at ~19.7209°, 19.81006°, 19.76602°, respectively, for 200 degrees.

88 Crystallite size was calculated for 1-phase, 2-phase, and 3-phase fibers according to Scherrer's
 89 equation:

$$90 \quad \tau = \frac{K\lambda}{\beta \cos\theta}$$

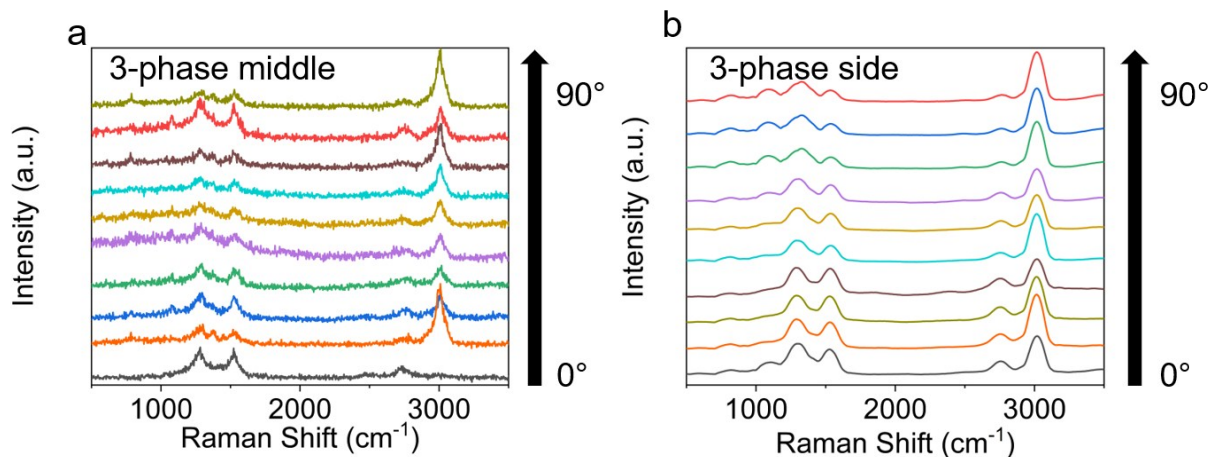
91 where τ , K , λ , β , and θ are the mean crystallite size, shape factor (0.9), X-ray wavelength (1.54),
 92 line broadening at FWHM, and Bragg angle.

93 Crystallinity degree was calculated based the following equation:

$$94 \quad \text{Crystallinity} = \frac{A_{\text{crystalline}}}{A_{\text{crystalline}} + A_{\text{amorphous}}} \times 100$$

Fiber type	Draw temperature (°C)	Crystallite size of (101) plane	Crystallinity (%)
1phase	100	6.7	54.4159
1phase	200	7.3	68.9205
2phase	100	5.1	54.5652
2phase	200	6.2	62.502
3phase	100	5.9	52.138
3phase	200	7.2	66.5621

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97 **Figure S12.** Raman spectra for an incident laser while changing the incident angle. The 3-phase
 98 fiber incident light was focused on the (a) middle and (b) side sections. The signature peaks all
 99 match previous Raman spectra for raw GNPs. The peak at $\sim 3000\text{ cm}^{-1}$ arises from the stretching
 100 vibrations of CH_2 in the PVA fiber. The I_{2D} band intensity at $\sim 2750^\circ$ was fitted and normalized as
 101 the incident angle increases from 0° to 90° .

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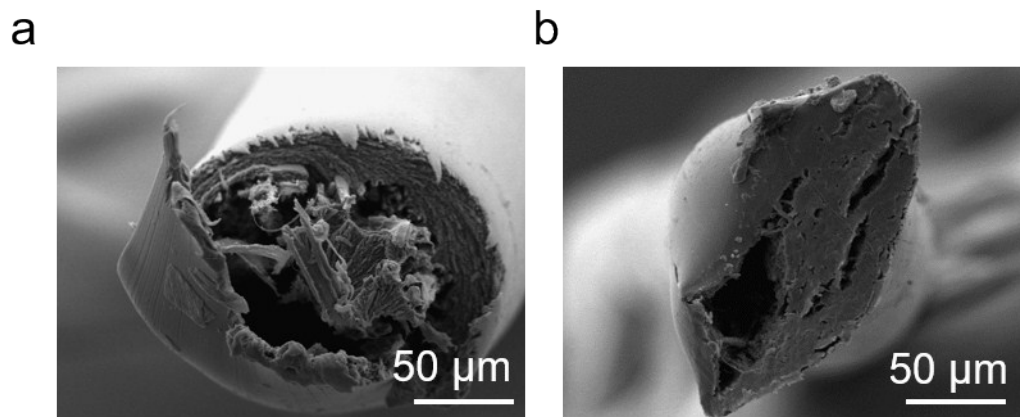


Figure S13. SEM images of the 3-phase fiber with large channel thickness. (a) fracture point. (b) after drawing at 200 °C. scale bar equals 50 μm.