Supporting information

Microstructural evolution of regenerated silk fibroin/graphene oxide hybrid fibers under tensile deformation

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Atomic Force Microscopy (AFM)
The thickness of graphene oxide was characterized by Multimode NanoScope IV system (Veeco, Santa Barbara, CA). The AFM images were performed in the tapping mode with a scanning range of 5 μm and a scanning frequency of 1.001 Hz. The samples were prepared by placing one droplet of the diluted GO solution on fresh mica and drying it at room temperature.

Particle size distribution
A Malvern Particle size & Zeta Potential Analyzer (Zetasizer Nano S90) was used to analyze the GO dispersion solution size distribution.

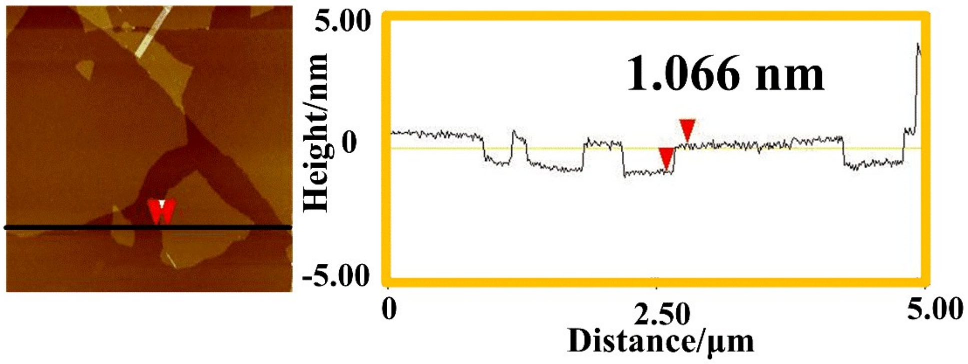

Fig. S1 AFM images of the GO nanosheets and corresponding height profile.

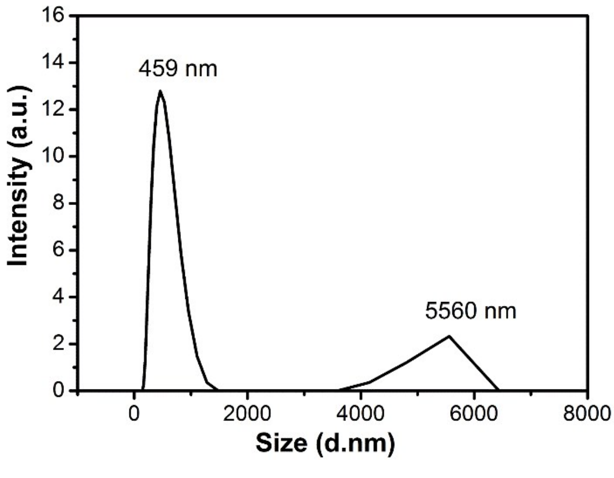

Fig. S2 The particle size and particle size distribution of GO dispersion solution tested by Malvern Particle size & Zeta Potential Analyzer.

Table S1. The parameters of SR-WAXD on RSF/GO fiber in elastic deformation zone

Code	m/kg	Load/N	σ^a/MPa	ε^b/%
A0	0	0	0	0
A1	1.512×10^{-4}	1.482×10^{-3}	15.84	0.4
A2	3.024×10^{-4}	2.964×10^{-3}	31.68	0.8
A3	4.536×10^{-4}	4.445×10^{-3}	47.50	1.2

[a] The diameter of fiber is about 1.092×10^{-5}m, [b] initial modulus of fibers is about 3.9 GPa, σ is stress and ε represents strain.

Table S2. The parameters of SR-WAXD on RSF fiber in elastic deformation zone

Code	m/kg	Load/N	σ^a/MPa	ε^b/%
C0	0	0	0	0
C1	1.512×10^{-4}	1.482×10^{-3}	16.04	0.4
C2	3.024×10^{-4}	2.964×10^{-3}	32.08	0.8
C3	4.536×10^{-4}	4.445×10^{-3}	48.11	1.1

[a] The diameter of fiber is about 1.085×10^{-5}m, [b] initial modulus of fibers is about 4.3 GPa, σ is stress and ε represents strain.

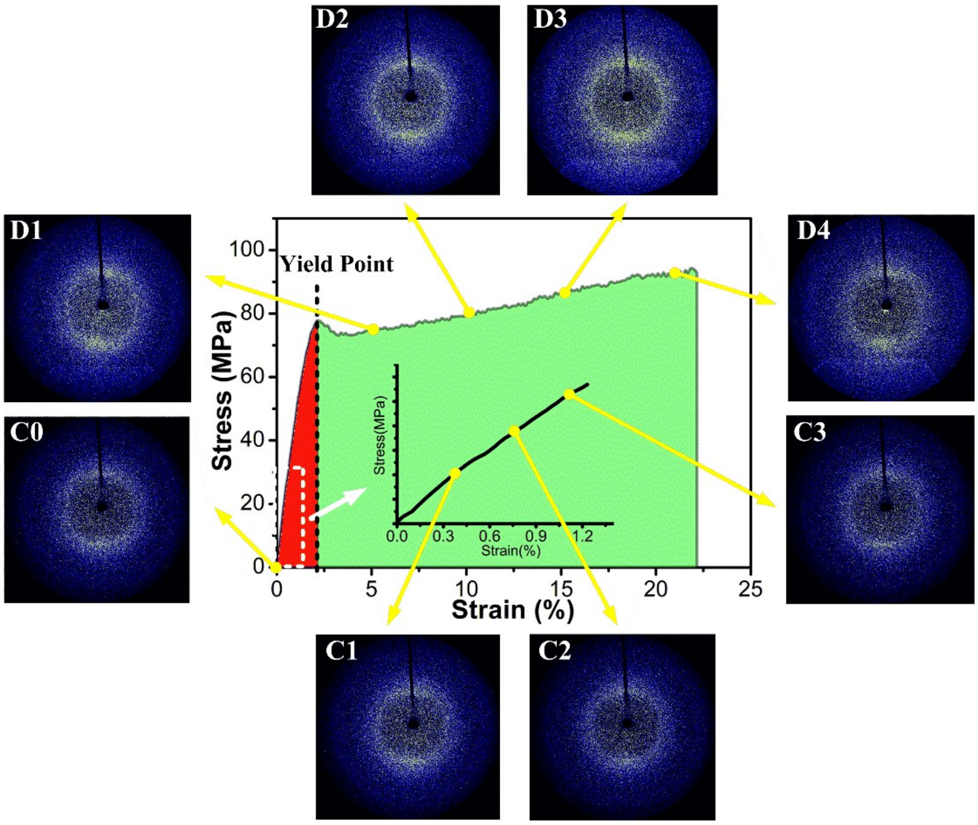

Fig. S3 The stress-strain curve of the RSF hybrid fibers at a 2X draw ratio and the corresponding 2D-WAXD patterns at different strains.

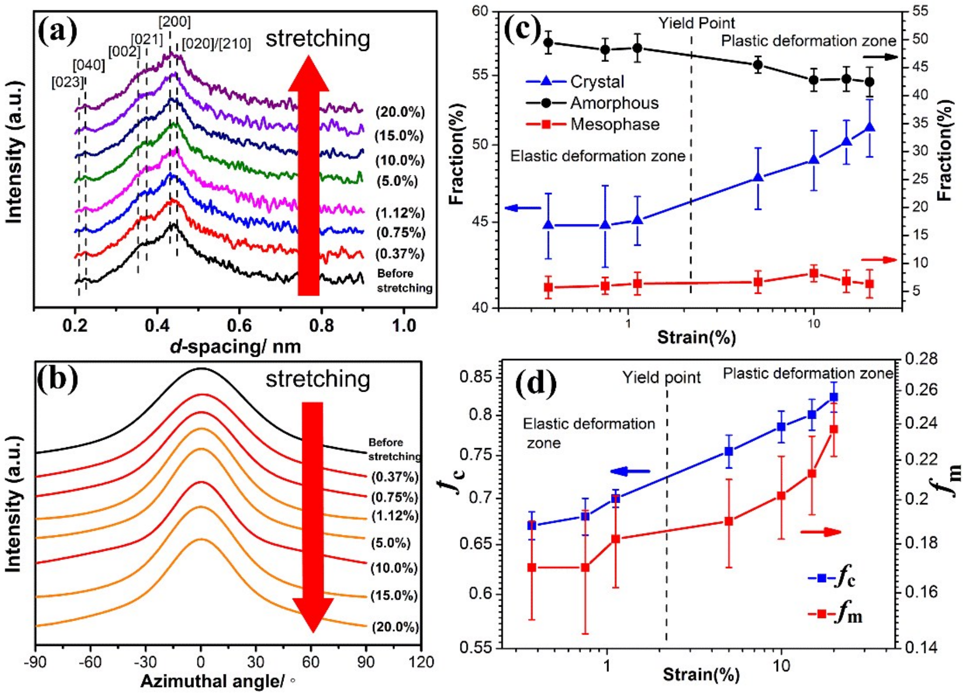

Fig. S4 Analysis of (a) the 1D-WAXD diffractograms of RSF fibers in different stretching states: (b) azimuthal profiles at d-spacing of 0.45 nm, (c) the variation of fraction of crystallinity, amorphous and mesophase, and (d) the variation of Herman's orientation factor of crystalline region and mesophase.

For RSF fibers after the yield point, the fraction of mesophase increased firstly but decreased at higher strain. Since the fraction of mesophase in the RSF fibers was lower than that in RSF/GO fibers, we considered that in the RSF fibers, it was amorphous phase that was converted to crystalline structure and mesophase during the tensile deformation, and the mesophase was also converted to crystalline structure with the increase of the strain.

Both f_c and f_m of pure RSF silk fibers also had slight change in the elastic deformation zone, but increased steadily in the plastic deformation. Finally they reached 0.84 and 0.24, respectively, at the strain of 20%.