One-pot Approach to Prepare Stretchable and Conductive Regenerated Silk Fibroin/CNT Films as Multifunctional Sensors

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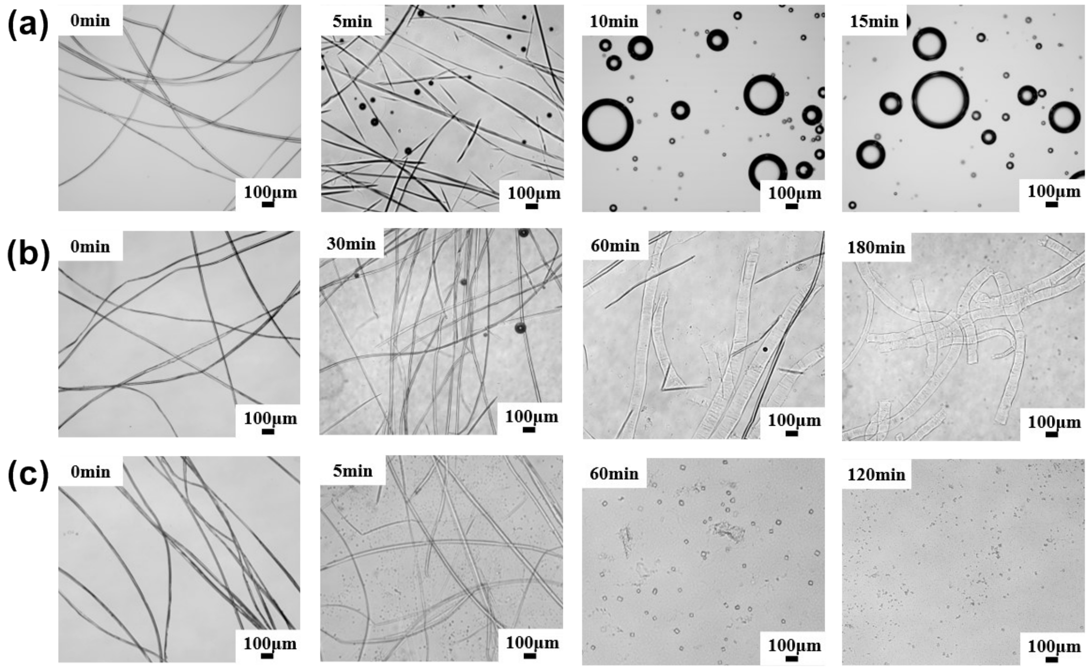

Figure S1. The dissolution process of silk fibroin. (a) aq. PA (85 wt%), 25°C. (b) aq. LiBr, 60°C. (c) aq. $CaCl_2/CH_3CH_2OH/H_2O$, 80°C.

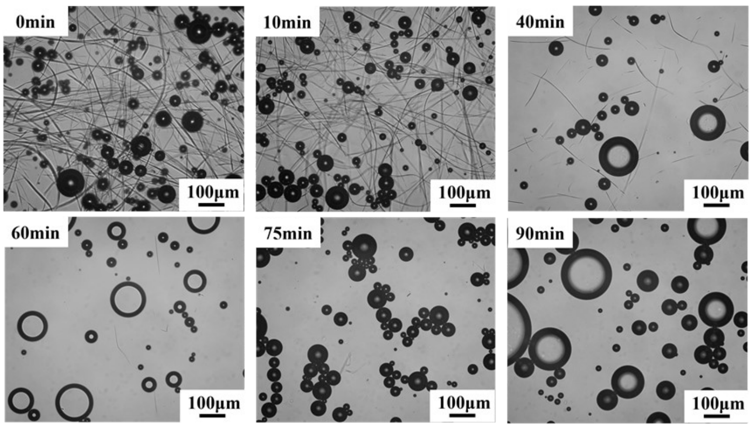

Figure S2. The dissolution process of silk fibroin in 75 wt% H_3PO_4, at 25°C.

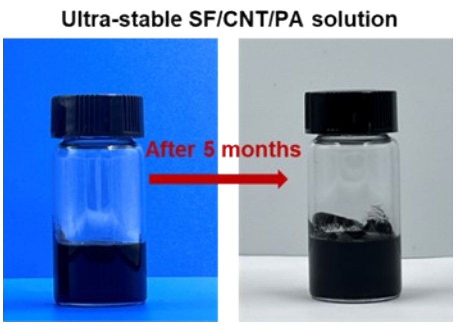

Figure S3. Optical micrographs of a H_3PO_4/CNT/SF solution after 5 months since the preparation.

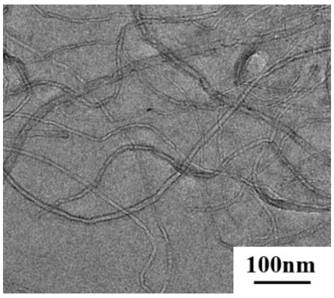

Figure S4. TEM image for SF/CNT showing the CNTs modified by SF was homogeneously dispersed.

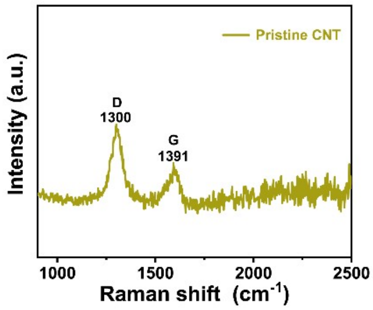

Figure S5. Raman spectrum of pristine CNT.

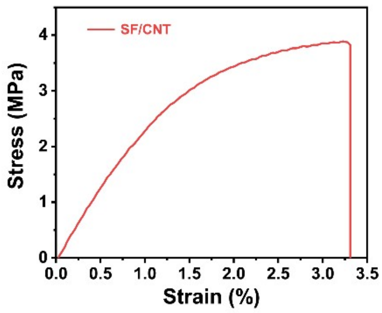

Figure S6. The tensile stress-train curves of the neat SF/CNT-1.

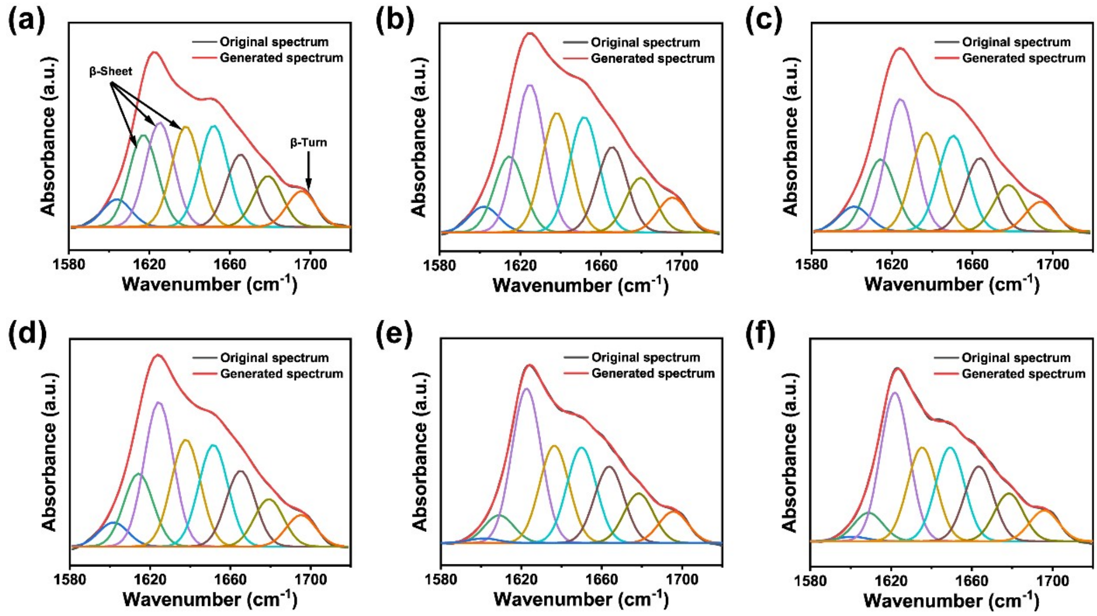

Figure S7. FTIR spectra of SF/GL films and SF/GL/CNT films. (a) Deconvolution of amide I band in FTIR spectra of SF/GL films. (b) Deconvolution of amide I band in FTIR spectra of SF/GL/CNT-1 films. (C) Deconvolution of amide I band in FTIR spectra of SF/GL/CNT-3 films. (d) Deconvolution of amide I band in FTIR spectra of SF/GL/CNT-5 films. (e) Deconvolution of amide I band in FTIR spectra of SF/GL/CNT-7 films. (f) Deconvolution of amide I band in FTIR spectra of SF/GL/CNT-9 films.

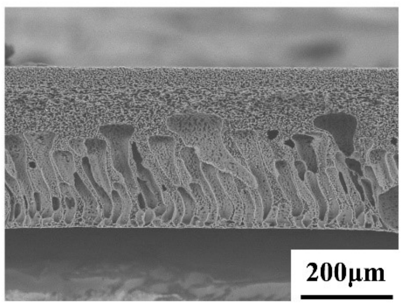

Figure S8. The SEM images of SF/GL films.

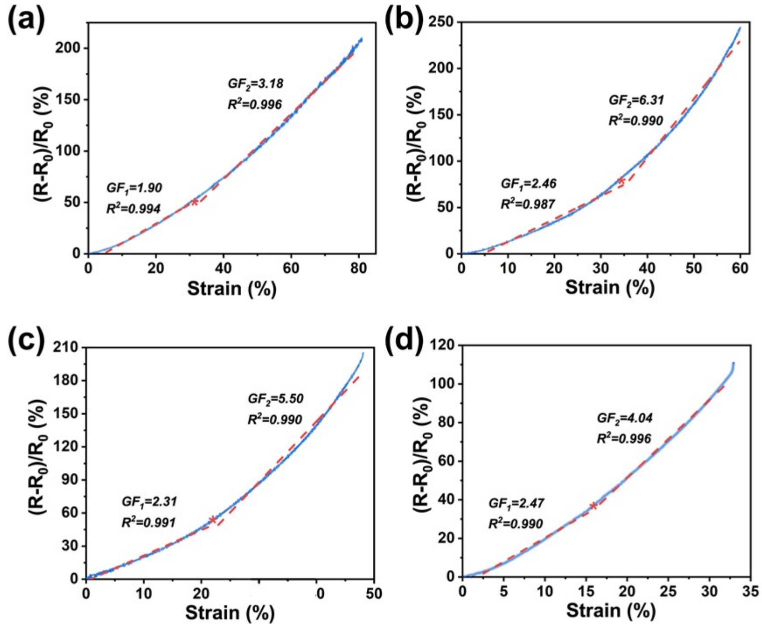

Figure S9. Typical relative resistance versus strain curve of SF/GL/CNT films with varying CNT content at a stretching rate of 10 mm min^{-1}. (a) SF/GL/CNT-1. (b) SF/GL/CNT-5. (c) SF/GL/CNT-7. (d) SF/GL/CNT-9.

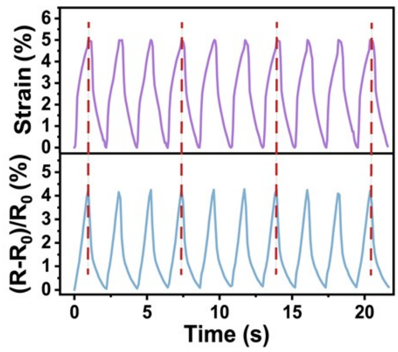

Figure S10. Dynamic response of the sensor to ten loading and unloading cycles at 5 % strain, indicating the negligible signal hysteresis.

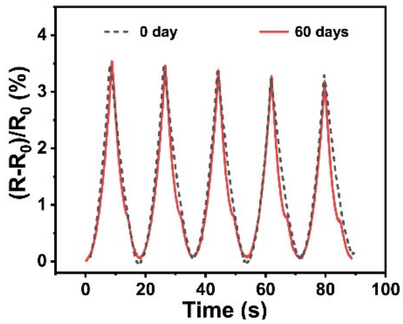

Figure S11. The relative resistance change vs time during the loading-unloading cycles at a strain of 3% for the initial SF/GL/CNT-3 sensor and the sensor after placing 60 days.

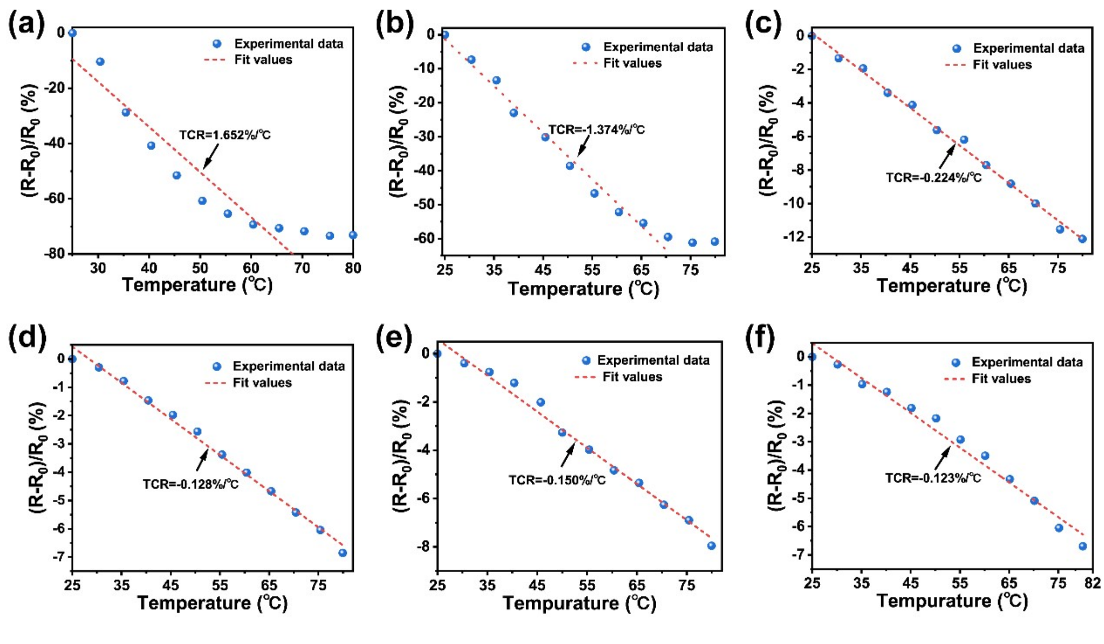

Figure S12. Normalized relative resistance changes of SF/GL/CNT sensosrs upon increasing temperature from 25 to 80 °C. (a) SF/GL/CNT-0.1 films. (b) SF/GL/CNT-1 films. (c) SF/GL/CNT-3 films. (d) SF/GL/CNT-5 films. (e) SF/GL/CNT-7 films. (f) SF/GL/CNT-9 films.

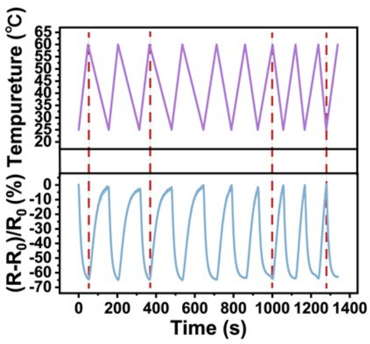

Figure S13. Dynamic response of the sensor to hot (60 °C) and cold (25 °C) environment, indicating the negligible signal hysteresis.

Table S1. The D, G, and I_D/I_G of the SF/GL/CNT sensors with varying CNTs content.

Sample	D (cm^{-1})	G (cm^{-1})	I_D/I_G
SF/GL/CNT-1	1306	1597	1.82
SF/GL/CNT-3	1310	1605	2.07
SF/GL/CNT-5	1311	1606	2.18
SF/GL/CNT-7	1314	1608	2.31
SF/GL/CNT-9	1315	1610	2.37

Table S2. Characterization of the β-Sheet content and crystallinity of SF/GL films and SF/GL/CNT films with FTIR spectra and XRD.

Sample	β-Sheet (%)	β-crystallinity (%)
SF/GL	35.3	14.06
SF/GL/CNT-1	37.1	15.37
SF/GL/CNT-3	39.6	17.80
SF/GL/CNT-5	43.4	20.66
SF/GL/CNT-7	46.1	22.67
SF/GL/CNT-9	33.7	13.61

Table S3. Summary of thermosensation capacities of temperature sensors

Sensing materials	TCR (%/℃)	Sensing ranges (℃)	Ref.
Pt NFs into silk fibroin membranes	0.205	20-60	51
Silk CFM	0.81	20-80	61
$Gr/SF/Ca^{2+}$	2.09	20-50	16
SS-PEDOT:PSS/rGO	0.99	20-50	59
Graphene-doped PU	0.815	20-100	58
PEDOT-TPU	0.95	20-40	60
BP/LEG on SEBS	0.1736	25-50	57
GO/polyurethane	0.80	30-80	56
SF/GL/CNT	1.6	25-80	**This work**