Supplementary Information

Fibrous Nanocomposites of Carbon Nanotube and Graphene-Oxide with Synergetic Mechanical and Actuative Performance

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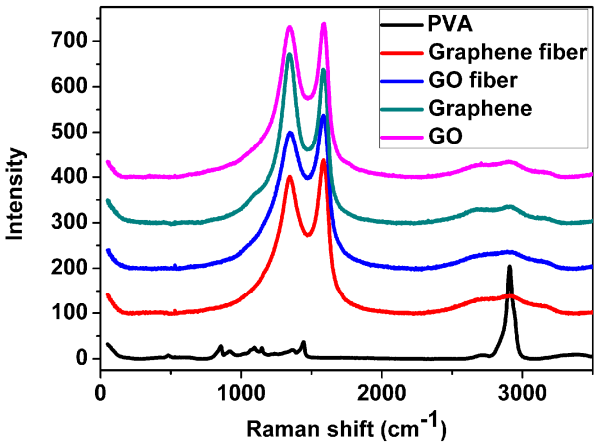
Experimental Methods

Chemicals: SWCNTs prepared by CVD method were purchased from Chengdu Organic Chemicals Co. Ltd., and were pretreated in 14 M nitric acid at 140 ^{o}C for 6 h before use. Graphite flake (bought from alfa-aesar Co., ltd) was exfoliated into GO by modified Hummers' method. Poly (vinyl alcohol) (PVA, MW ~120K, alcoholysis 93.0%) were provided by Sinopharm Chemical Reagent Co., Ltd.

***Preparation of fibers*:** 35 mg GO was added in 10 mL NaOH solution (pH~ 10.6) and bath sonicated for 2 h. The obtained homogeneous solution was used to prepare fibers by PVA-based coagulation spinning. The procedure was similar to our previous report,[1] except that concentrated HCl was added in PVA solution to adjust the pH to below 2. Graphene solution was obtained by

reducing GO in an aqueous solution of hydrazine hydrate at a concentration ratio of GO: hydrazine = 10 mg: 1mmol. SDS was added to prevent the restack of the sheets, and the weight ratio of SDS: graphene was 3:1. Graphene fibers were prepared following the same procedure to GO fibers and the raw solution concentration was also 3.5 mg/mL. SWCNT fibers were prepared via the procedure reported in our previous work.[1] Two different nanocarbon solutions (SWCNT, GO or graphene) with the same concentration (3.5 mg/mL) were mixed together at different volume ratio and homogenized, and then used to prepare composite fibers.

Characterization: The morphology of graphene fiber and SWCNT-graphene composite fiber was characterized by a field emission scanning electron microscope (FESEM, JEOL, JSM-6700F). Atomic force microscope (AFM) images of GO and graphene sheets were recorded on an SPI-3800 microscope with a SPA-400 scanner. For the sample preparation, 0.5 mL GO or graphene solution with the concentration of 3.5 mg/mL was diluted in 10 mL methanol. Dip a clean and dry silicon wafer into the solution for a few times, and then dried at 60 ^{o}C. Mechanical performance of the fibers was measured by XQ-1 fiber strength instrument. Each sample was tested for 5 times and the average value was calculated. FT-IR spectra were recorded using a Nicolet iN10 spectrometer (Thermo Scientific). The resolution of the spectrometer was ~4 cm^{-1}. The actuation test was referred to Hu's work,[2] and the distance between the two electrodes was ~2 cm. Raman spectra were recorded using a Thermofisher spectrometer (DXR) with an excitation length of 532 nm.



S1 Raman spectra of GO, Graphene and their fibers

The I_D/I_G ratio decreased after the reduction, indicating the removal of carboxyl and hydroxyl groups according to previous report.[3] When spun into fibers, the D peaks of both GO and graphene decreased, which may be owing to the interaction of the defective sites of GO or graphene with surrounding PVA molecules.

Table 1 Conductivity of SWCNT, graphene and their composite fibers

sample	GO fiber -HCl	graphene fiber-HCl	annealed graphene fiber-HCl	SWCNT fiber	SWCNT/GO (2:1)	SWCNT/GO (2:1)-HCl
conductivity (S/cm)	--	1.41E-05	1.31E-04	0.01	0.021	0.38
sample	SWCNT/GO (1:1)-HCl	SWCNT/GO (1:2)-HCl	SWCNT/G (2:1)	SWCNT/G (1:1)	SWCNT/G (1:2)-HCl	--
conductivity (S/cm)	0.028	7.6E-06	0.0037	0.0025	3.48E-04	--

References:

1. R. R. Wang, J. Sun and L. Gao, *J. Phys. Chem. C*, 2010, **114**, 4923-4928.
2. Y. Hu, W. Chen, L. H. Lu, J. H. Liu and C. R. Chang, *Acs Nano*, 2010, **4**, 3498-3502.
3. S. Stankovich, D. A. Dikin, R. D. Piner, K. A. Kohlhaas, A. Kleinhammes, Y. Jia, Y. Wu, S. T. Nguyen and R. S. Ruoff, *Carbon*, 2007, **45**, 1558-1565.