Supplementary Information

A fiber-shaped solar cell showing record power conversion efficiency of 10%

Xuemei Fu, [†] Hao Sun, [†] Songlin Xie, Jing Zhang, Zhiyong Pan, Meng Liao, Limin Xu, Zhuoer Li, Bingjie Wang, Xuemei Sun and Huisheng Peng*

State Key Laboratory of Molecular Engineering of Polymers, Department of Macromolecular Science, Laboratory of Advanced Materials, Fudan University, Shanghai 200438, China. *E-mail: penghs@fudan.edu.cn

[†]*These authors contributed equally to this work.*

Experimental Section

Calculation of power conversion efficiency. For the fiber-shaped DSSC, the power conversion efficiency (η) was determined by the equation of $\eta = V_{OC} \times J_{SC} \times$ FF, where V_{OC} , J_{SC} and FF referred to the open-circuit voltage, short-circuit current density and fill factor, respectively. The effective area used for calculation of the short-circuit current density was the projected area of working electrode derived from the product of diameter and length of working electrode according to the generally recognized method¹⁻³.

Characterization. The morphology and structure were characterized by fieldemission scanning electron microscope (Ultra 55, Zeiss) operated at 5 kV equipped with energy dispersive spectroscopy. The J-V characteristic curves were measured via a Keithley 2420 Source Meter under illumination (100 mW cm⁻²) of simulated AM1.5 solar light coming from a solar simulator (Oriel-Sol3A 94023A equipped with a 450 W Xe lamp and an AM1.5 filter). The light intensity was calibrated using a reference Si solar cell (Oriel-91150). The mechanical properties of the fibers were measured by a HY0350 Table-Top universal testing instrument with a gauge length of 6 mm. Electrical conductivities were tested by a Keithley 2400 Source Meter. Cyclic voltammogram and electrochemical deposition were completed on an electrochemical workstation (CHI 660E). The cross sections were obtained by cutting the core-sheath fibers with a focused Ga ion beam using 30 kV voltage and 20 nA beam current (Zeiss Auriga), followed by polishing using ion beam with currents ranged from 1 to 4 nA. Non-destructive 3D imaging of the core-sheath fiber was achieved from 3D X-ray microscope (ZEISS Xradia 510 Versa). The hydrophilic CNT sheets were characterized via Fourier Transformation Infrared Spectrometer (NEXUS 470, Thermofisher), X-ray photoelectron spectroscopy (Axis Ultra DLD, Kratos) equipped with a monochromatic $Al_{K\alpha}$ X-ray source and contact angle testing instrument (OCA40, Dataphysics).

References

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- S2 D. Y. Liu, M. Y. Zhao, Y. Li, Z. Q. Bian, L. H. Zhang, Y. Y. Shang, X. Y. Xia, S. Zhang, D. Q. Yun, Z. W. Liu, A. Y. Cao and C. H. Huang, *ACS Nano*, 2012, 6, 11027-11034.
- S3 Y. P. Fu, H. W. Wu, S. Y. Ye, X. Cai, X. Yu, S. C. Hou, H. Kafafy and D. C. Zou, *Energy Environ. Sci.*, 2013, 6, 805-812.

Supplementary Figures



Fig. S1 (a and b) SEM images of a pristine CNT fiber at low and high magnifications, respectively.



Fig. S2 Infrared spectra of bare CNT and O-CNT.



Fig. S3 X-ray photoelectron spectroscopy characterization of bare CNT and O-CNT.



Fig. S4 Stress-strain curves of O-CNT fibers with increasing O₂ plasma processing time.



Fig. S5 Dependence of electrical conductivity of O-CNT fibers on O_2 plasma processing time.



Fig. S6 (a) SEM image of a Pt/CS-CNT fiber wound around a Ti/TiO_2 wire photoanode. (b) Higher magnification of TiO_2 at (a).



Fig. S7 Comparison of *J-V* curves of fiber-shaped DSSCs using 50% Pt/CS-CNT fiber and bare CNT fiber as the counter electrodes.



Fig. S8 Comparison of *J-V* curves of fiber-shaped DSSCs based on electrodeposited Pt/CS-CNT fiber and Pt-coated CS-CNT fiber by pyrolysis. The Pt content was determined to be 14%.

Counter electrode	$V_{oc}\left(\mathrm{V} ight)$	J_{sc} (mA cm ⁻²)	FF	η (%)
70% Pt/CS-CNT	0.731	18.52	0.70	9.48
50% Pt/CS-CNT	0.725	19.43	0.71	10.00
30% Pt/CS-CNT	0.722	18.00	0.67	8.72
10% Pt/CS-CNT	0.739	16.08	0.63	7.49
0% Pt/CS-CNT	0.740	13.14	0.60	5.83
50% Pt/CNT	0.736	17.49	0.65	8.37
Pt wire	0.743	14.59	0.56	6.11

Table S1. Photovoltaic parameters of the fiber-shaped DSSCs from Fig. 3g.