

Supporting Information

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**Hierarchically Assembled Counter Electrode for Fiber
Solar Cell Showing Record Power Conversion Efficiency**

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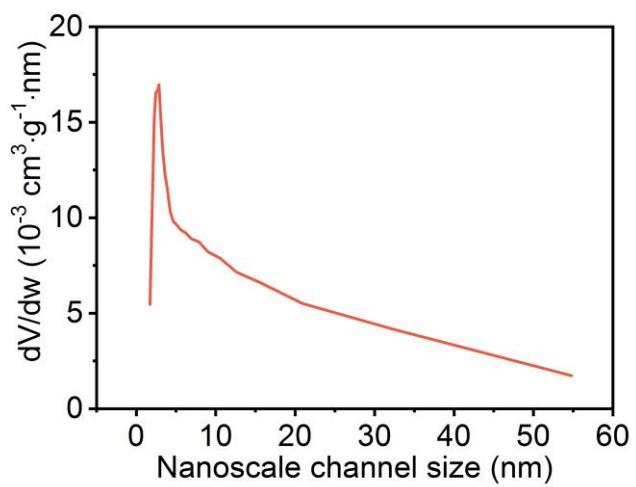


Figure S1. N₂ absorption-desorption isotherms of CNT.



Figure S2. Photograph of reels of primary CNT fibers.

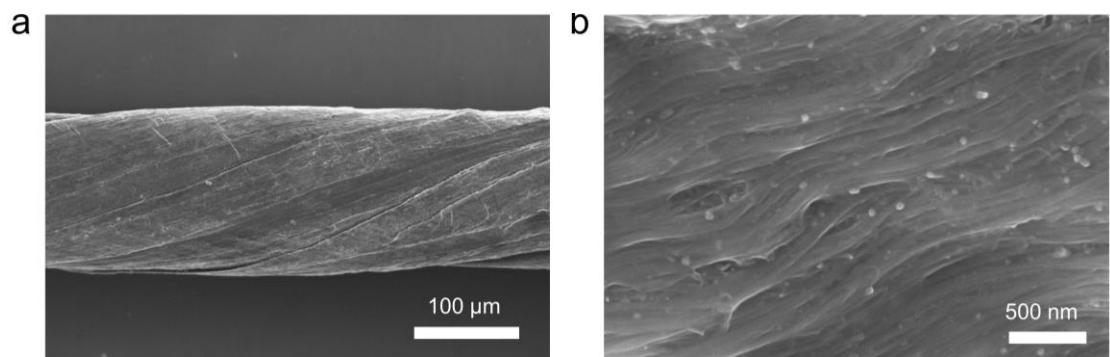


Figure S3. SEM image of CNT fiber (with diameter of $\sim 150 \mu\text{m}$) at low (a) and high (b) magnifications.

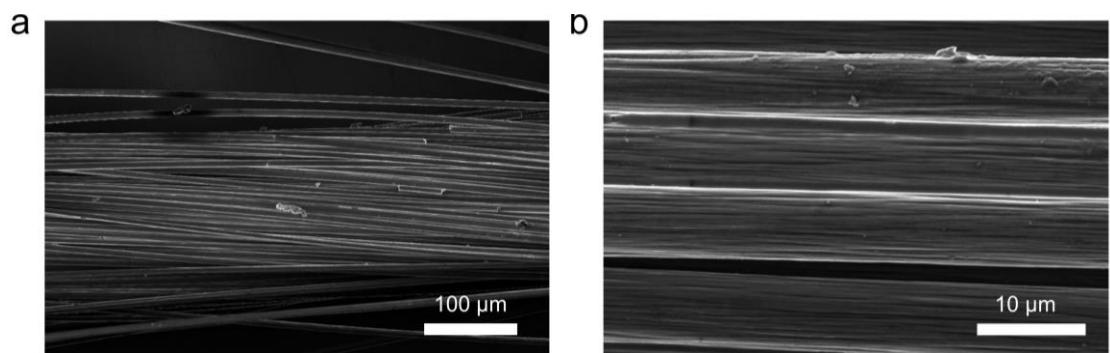


Figure S4. SEM image of carbon fiber (with diameter of $\sim 150 \mu\text{m}$) at low (a) and high (b) magnifications.

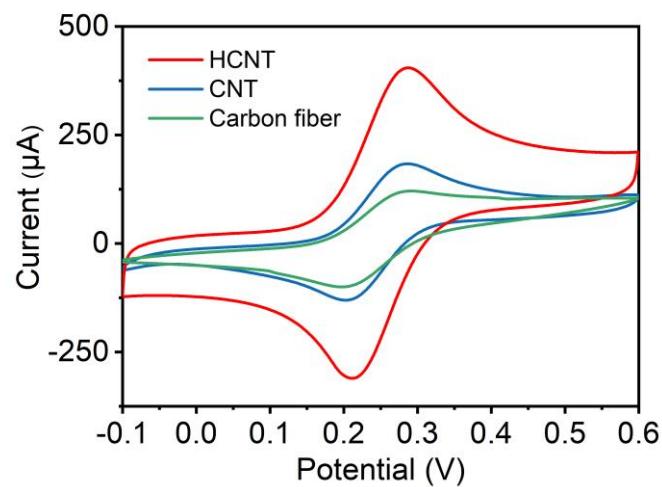


Figure S5. CV curves of electrochemical activity area measurement in the 20 mM $\text{K}_4\text{Fe}(\text{CN})_6$ and 0.2 M KCl at $20 \text{ mV}\cdot\text{s}^{-1}$.

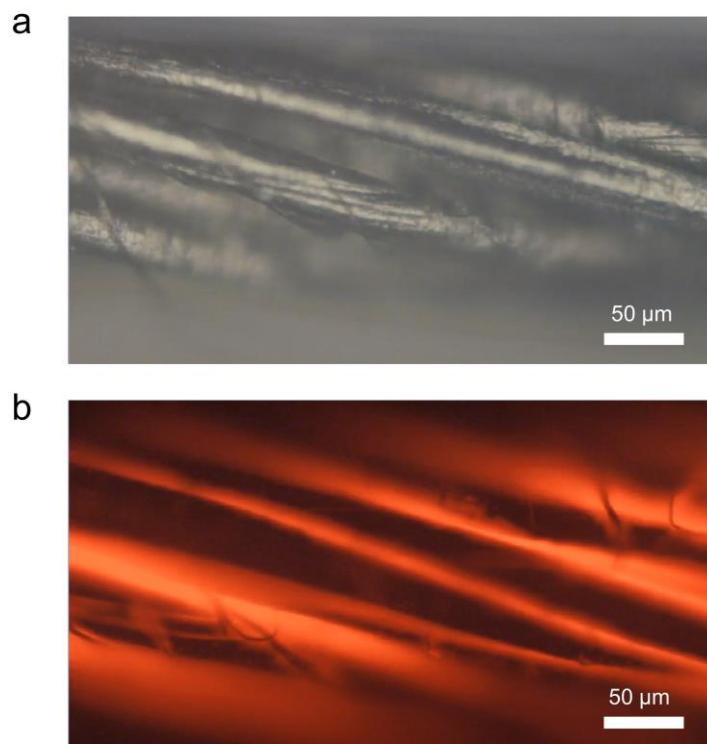


Figure S6. Optical (a) and fluorescence (b) micrographs of HCNT fiber with ethanol solution containing rhodamine indicate that the electrolyte infiltrated into microchannels.

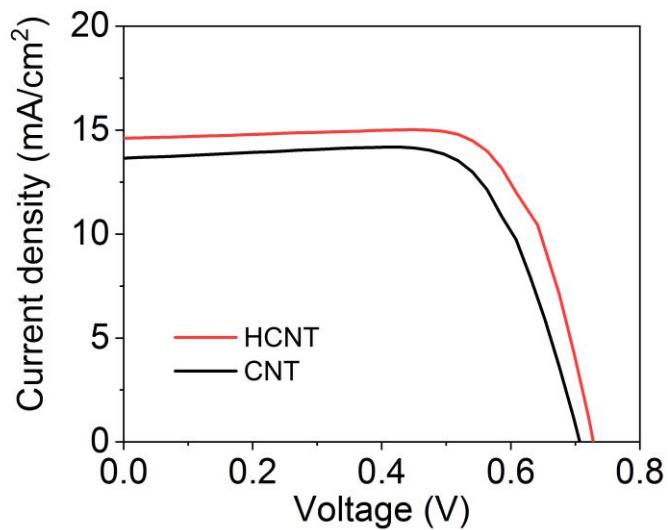


Figure S7. J - V curves of N719-sensitized fiber solar cells (with I^-/I_3^- electrolyte) using HCNT and CNT fibers as counter electrodes.

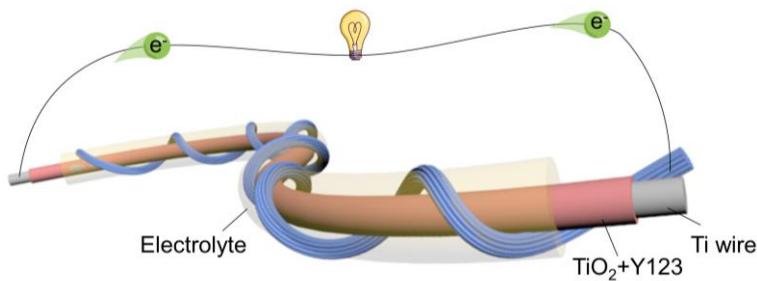


Figure S8. Structure of FDSSC and flow direction of electrons in external circuit.

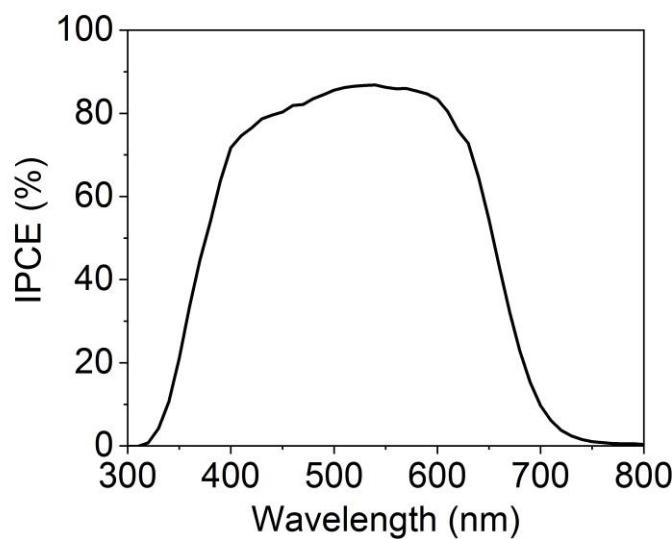


Figure S9. IPCE spectra of HCNT@Pt-based FDSSC.

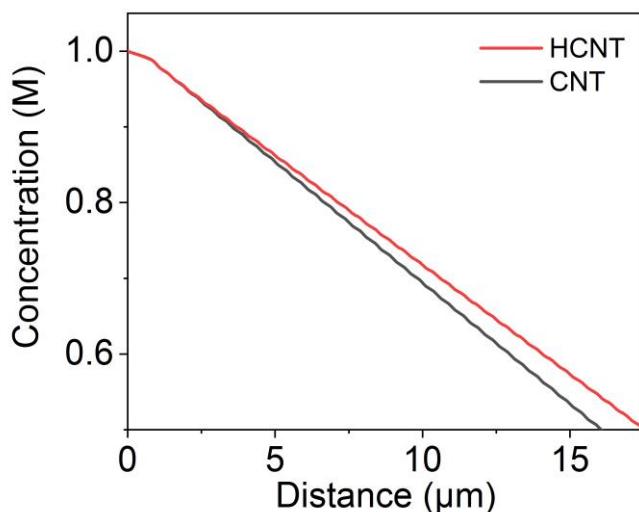


Figure S10. The curves of simulated ion concentration inside the fiber electrodes along the distance from electrodes surface. The simulation was performed by COMSOL Multiphysics 6.0. The initial concentration of Co^{3+} was 1 M. The diffusion coefficient was $1 \times 10^{-10} \text{ m}^2/\text{s}$.

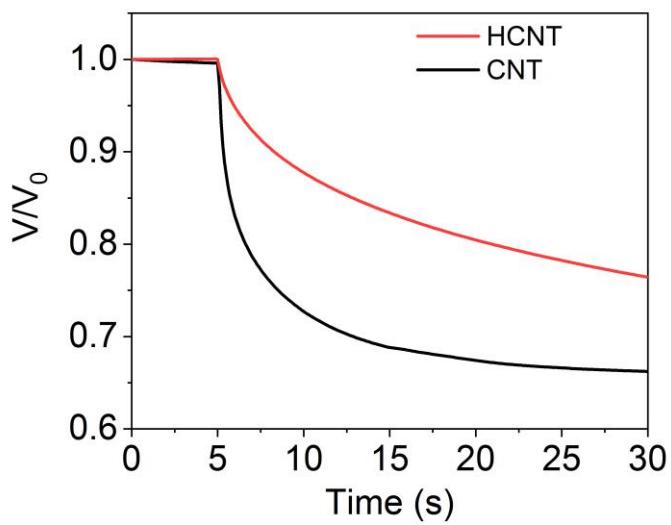


Figure S11. The open-circuit voltage decay curves of HCNT-based and CNT-based FDSSCs.

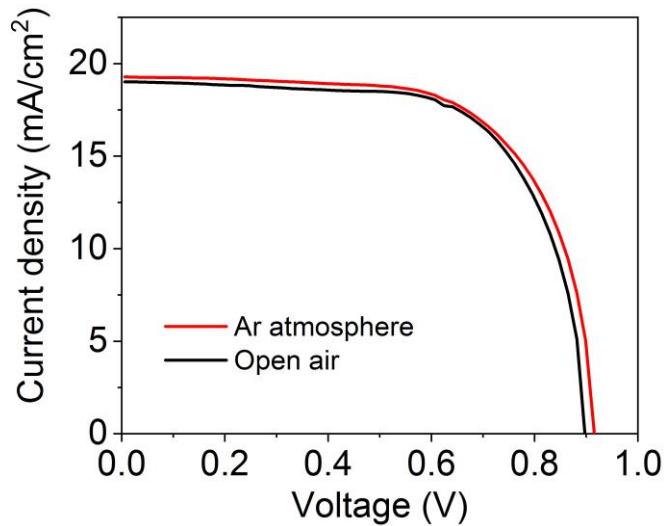


Figure S12. J - V curves of the FDSSCs measured under Ar atmosphere and open air conditions.

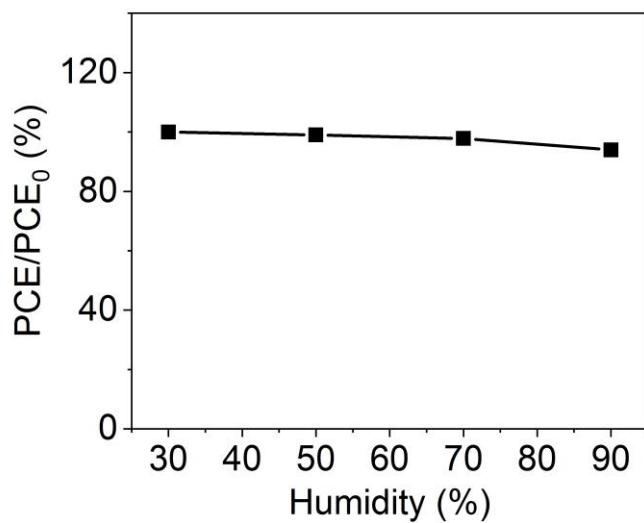


Figure S13. Variations of PCEs for FDSSCs under different humidity levels.

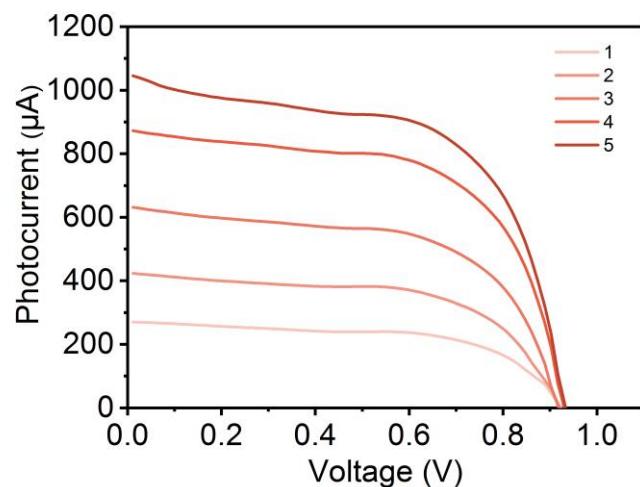


Figure S14. *I-V* curves of the fabric modules with increasing numbers of FDSSCs connected in parallel.

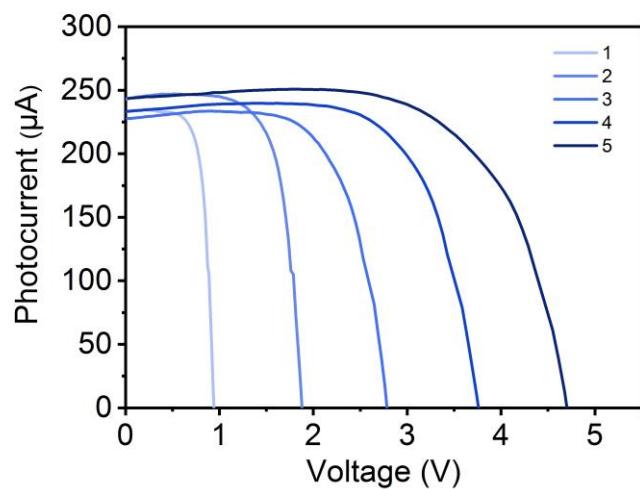


Figure S15. *I-V* curves of the fabric modules with increasing numbers of FDSSCs connected in series.

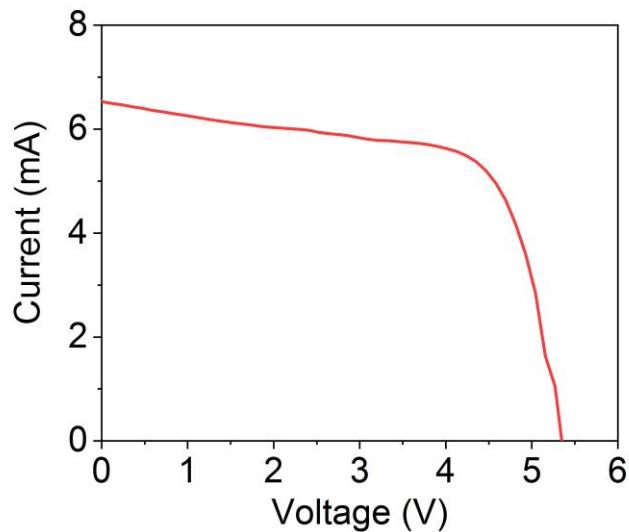


Figure S16. The *J-V* curve of the photovoltaic textile measured under the illumination of AM 1.5G.

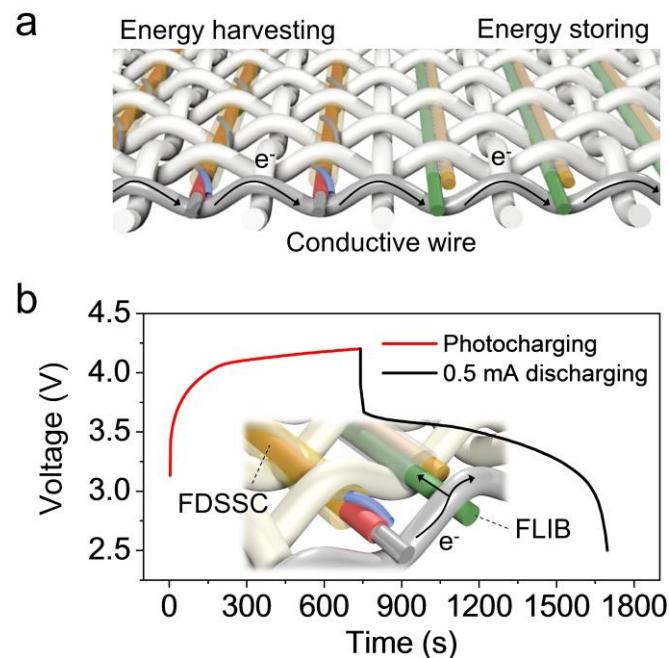


Figure S17. (a) Scheme to a textile woven by FDSSCs and FLIBs. (b) Photocharging curve of FDSSCs under AM 1.5 (red line) and discharging curve at 0.5 mA (black line) of the FLIB. Inset: operating mechanism of electron flow along conducting wire from the photoanode to the negative electrode.

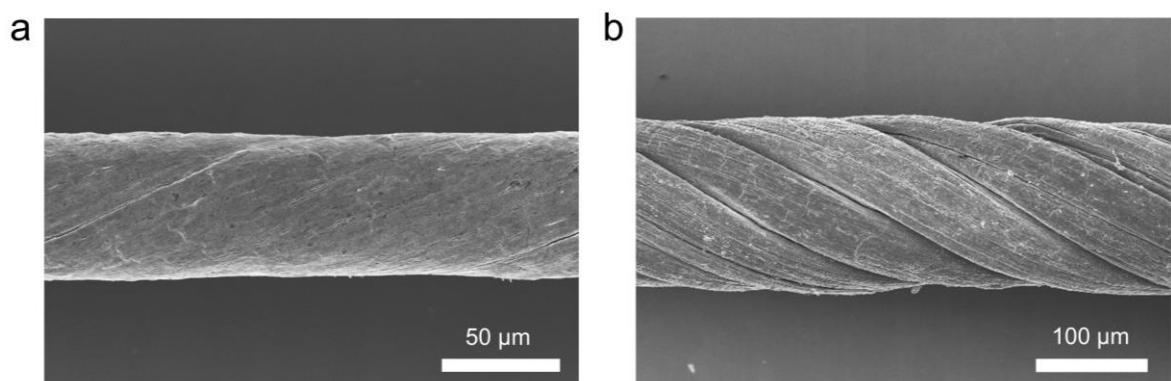


Figure S18. (a) SEM image of primary CNT fiber with diameter of 60 μm . (b) SEM image of HCNT-1 fiber with diameter of 150 μm .

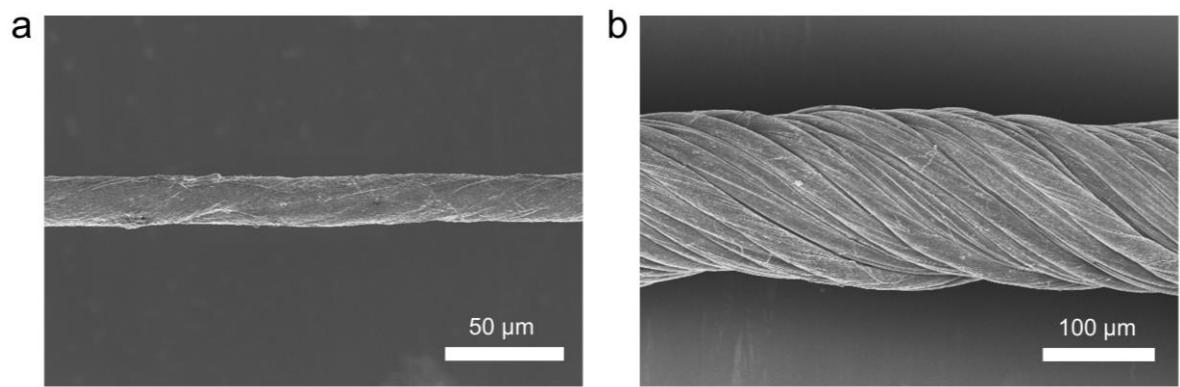


Figure S19. (a) SEM image of primary CNT fiber with diameter of 20 μm . (b) SEM image of HCNT-3 fiber with diameter of 150 μm .

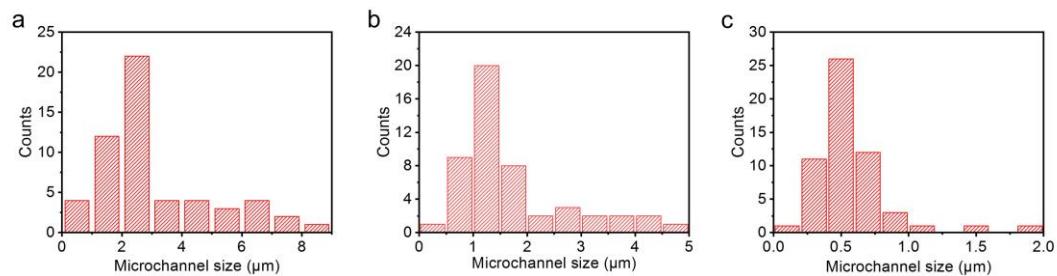


Figure S20. Statistical distribution of the channel size for microchannels in HCNT-1 fiber (average size of $3 \pm 1.97 \mu\text{m}$) (a), HCNT-2 fiber (average size of $1.7 \pm 1.01 \mu\text{m}$) (b), and HCNT-3 fiber (average size of $0.5 \pm 0.29 \mu\text{m}$) (c). The sample number was 50 for each statistical analysis.

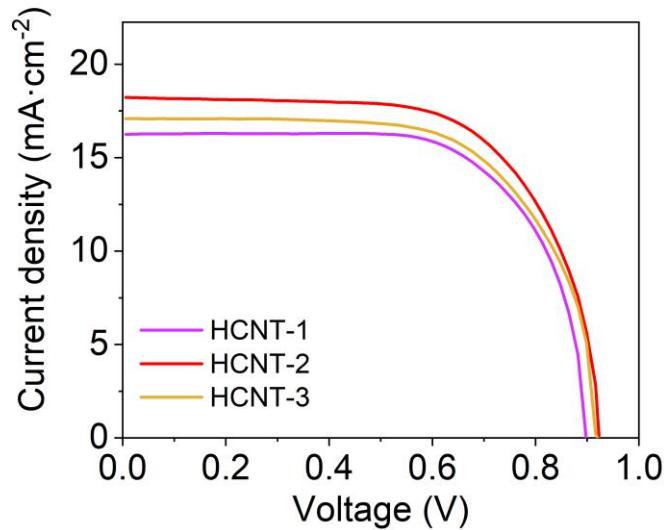


Figure S21. J - V curves of the FDSSCs using different counter electrodes.

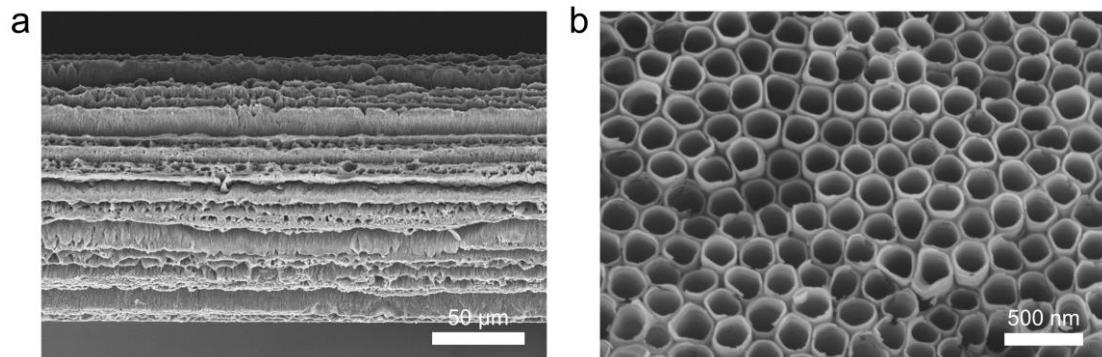


Figure S22. SEM images of fiber photoanode at (a) low and (b) high resolution.

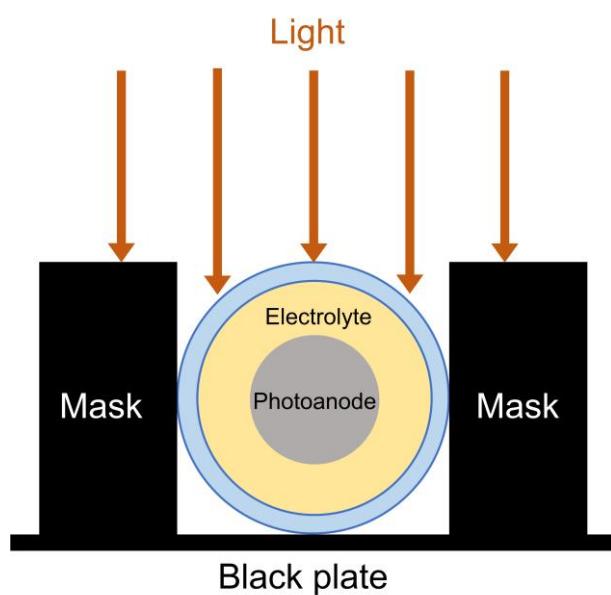


Figure S23. The schematic illustration of J - V measurement.

Table S1. EIS data in Figure 3c.

	$R_s/(\Omega \cdot \text{cm}^2)$	$R_{ct}/(\Omega \cdot \text{cm}^2)$	$Z_N/(\Omega \cdot \text{cm}^2)$
Carbon fiber	43.3	6.57	150.9
CNT fiber	7.9	0.44	141.4
HCNT fiber	6.7	0.38	84.3

Table S2. Photovoltaic parameters of the FDSSCs in Figure 3d.

	$V_{oc}/(\text{V})$	$J_{sc}/(\text{mA} \cdot \text{cm}^{-2})$	FF	PCE/(\%)
Carbon fiber	0.773	11.14	0.63	5.43
CNT fiber	0.889	15.11	0.66	8.89
HCNT fiber	0.923	18.26	0.66	11.14
HCNT@Pt fiber	0.907	19.32	0.68	11.94

Table S3. Photovoltaic parameters of the FDSSCs with different Pt loadings.

Pt loadings/(mg/cm ²)	$V_{oc}/(\text{V})$	$J_{sc}/(\text{mA}/\text{cm}^2)$	FF	PCE/(\%)
2.12	0.870	19.09	0.68	11.28
4.24	0.907	19.32	0.68	11.94
8.48	0.907	19.05	0.68	11.82

Table S4. Comparison of PCEs for fiber solar cells to date.

PVs Type	Cathode	Anode	PCE (%)	Reference
DSSC	HS-CNT fiber	Ti-TiO ₂ NT	11.94	This work
	Graphene fiber/Pt	Ti-TiO ₂ NT	8.45	[1]
	RGO/CNT fiber/Pt	Ti-TiO ₂ NT	8.50	[2]
	Pt wire	Multiple Ti-TiO ₂ NT	9.1	[3]
	Pt wire	Ti-TiO ₂ film-TiO ₂ NP	7.41	[4]
	CF@TiO ₂ @MoS ₂	Ti-TiO ₂ NT	9.5	[5]
	Pt/CS-CNT fiber	Ti-TiO ₂ NT	10.00	[6]
OSC	CF@PANI@CoSe	Ti-TiO ₂ NT	10.28	[7]
	Pt wire	Ti-TiO ₂ @PEOx	11.22	[8]
	PEDOT:PSS	SS-ZnO	2.3	[9]
	PEDOT:PSS	SS-ZnO	2.53	[10]
	PEDOT:PSS	Ti-TiO ₂ NP	1.78	[11]
	PEDOT:PSS	Ti-TiO ₂ NT	1.23	[12]
	PEDOT:PSS	Ti-ZnO	1.62	[13]
PSC	PEDOT:PSS	Graphene-ZnO	2.13	[14]
	OMeTAD	SS- TiO ₂ NP	3.3	[15]
	Spiro-OMeTAD	Ti-TiO ₂ NT	5.22	[16]
	CNT	PEN-TiO ₂	9.49	[17]
	Spiro-OMeTAD	Ti-TiO ₂ NP	6.58	[18]
	Spiro-OMeTAD	Ti-TiO ₂ NP	7.53	[19]
	Spiro-OMeTAD	Ti-TiO ₂ NP	10.79	[20]
	P3HT	ETL free	7.49	[21]

Table S5. The acetonitrile infiltration time for different fiber electrodes.

Number of sample	Infiltration time for CNT fibers (s)	Infiltration time for HCNT fibers (s)
1	45	19
2	35	21
3	48	27
Average	42.7	22.3

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