Supplementary Material

Laser-Assisted Fabrication of Flexible Monofilament Supercapacitor

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Comparison of laser sintering and hotplate sintering

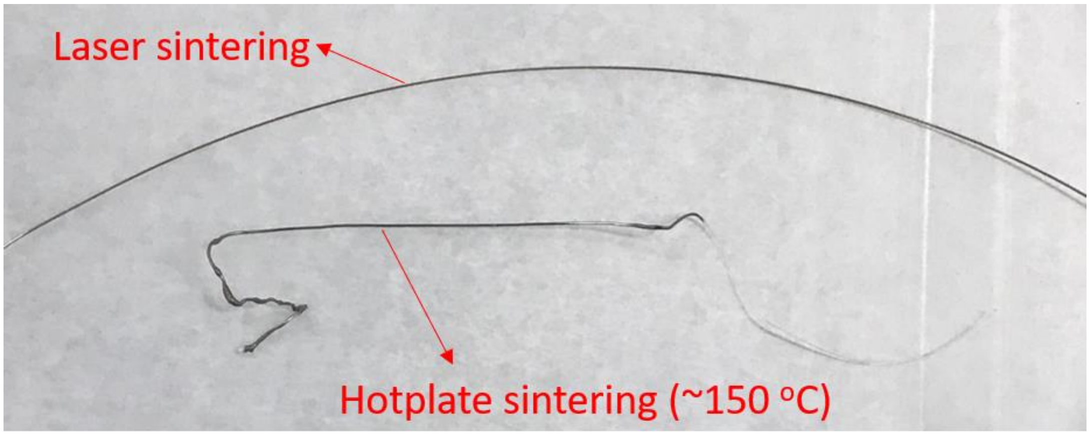

Figure S1. Digital images of AgNP-coated fibers sintered using a 532 nm laser and a hotplate at ~150 °C.

EDS elemental maps of the AgNP/AgNW/Au-coated fiber

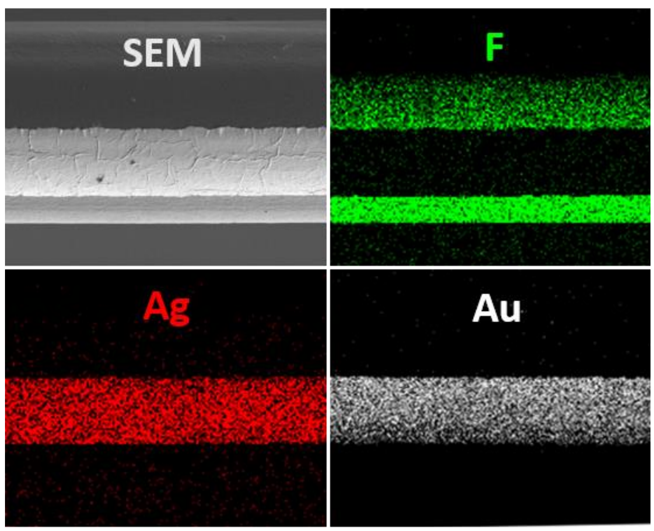

Figure S2. Top-view EDS elemental maps of the AgNP/AgNW/Au-coated fiber.

Comparison of AgNP and AgNP/AgNW layers

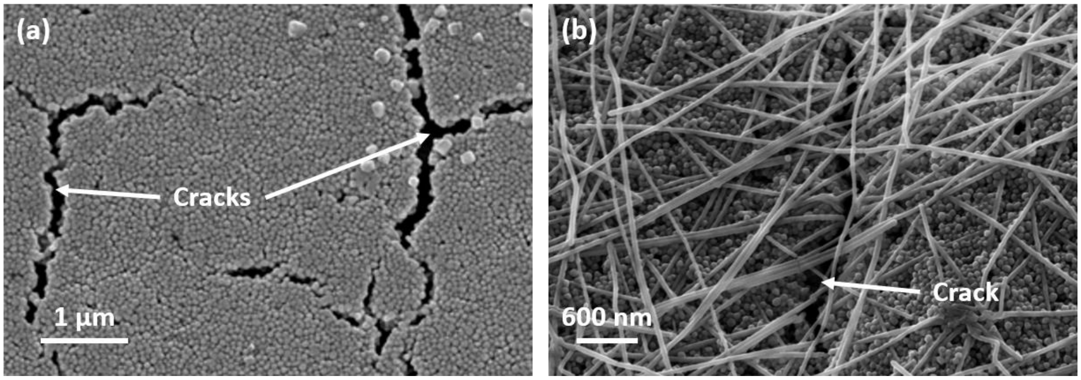

Figure S3. Top-view SEM images of (a) a AgNP layer and (b) a AgNP/AgNW layer near cracks.

Electrochemical characteristics of G-SCs and MG-SCs

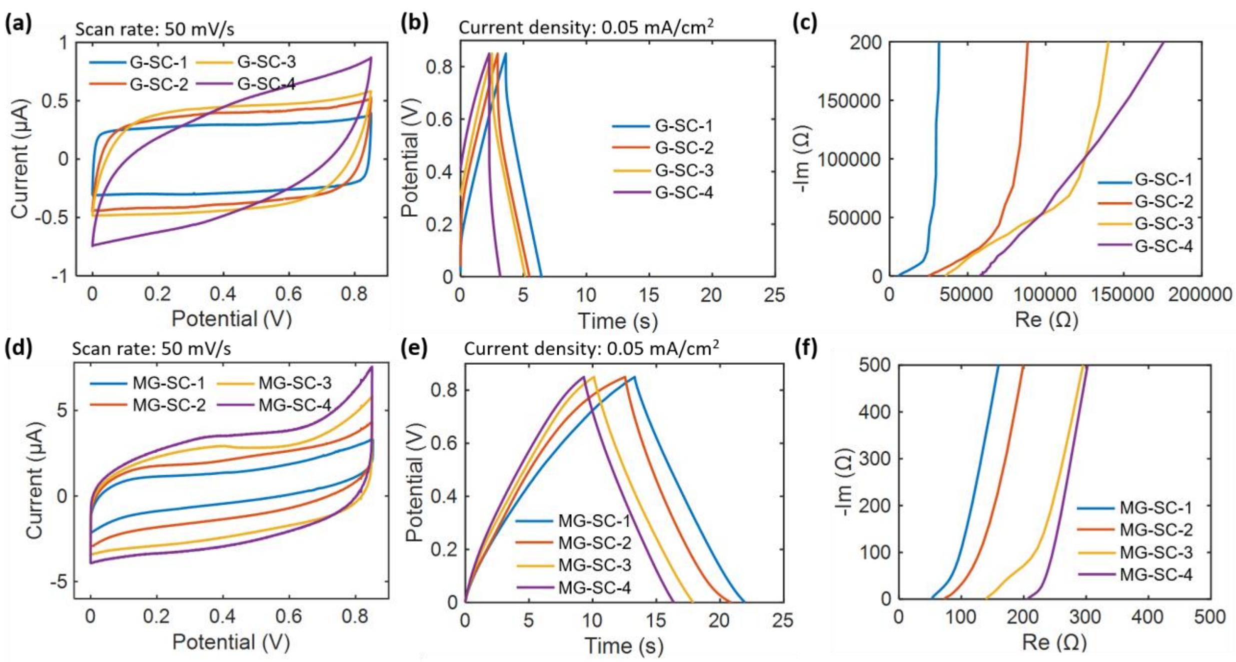

Figure S4. Cyclic voltammetry (CV) , galvanostatic charge/discharge (CC) curve, and electrochemical impedance spectroscopy (EIS) for (a, b, c) G-SCs and (c, d, e) MG-SCs, respectively.

EDX and XPS of MnO_2

Figure S5a shows the energy-dispersive X-ray spectroscopy (EDX; Emax Energy EX-250, Horiba) result of an MGM fiber electrode. The EDX spectrum confirms the existence of Mn and O in the electrode. Figure S5b shows the X-ray photoelectron spectroscopy (XPS; K-alpha Plus, Thermo Fisher Scientific) spectrum collected by the survey scan of a MnO2 layer that was prepared on a large-area graphene film by using the same electrodeposition method. Figure S5c shows the XPS spectrum of Mn 2p that displays a peak separation of 11.6 eV.

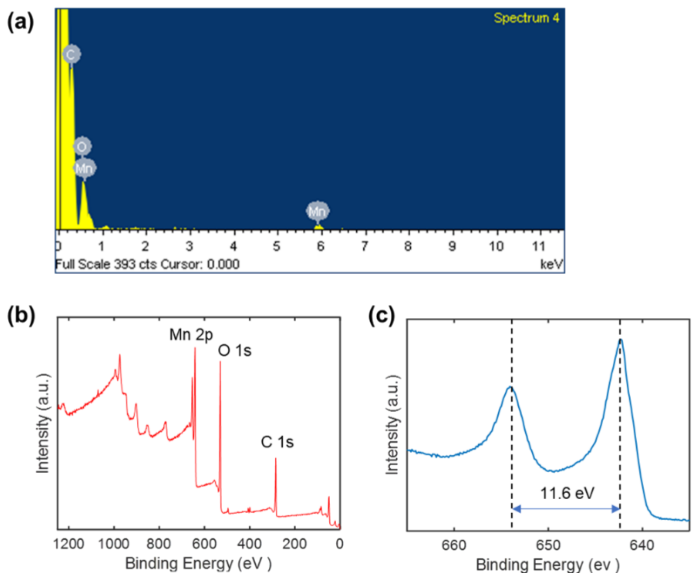

Figure S5. (a) EDX spectrum of an MGM electrode fiber. (b) Survey XPS spectrum and (c) XPS spectrum of Mn 2p that were obtained from a MnO_2 layer that was electrodeposited on a graphene film.

***Electrochemical characteristics of MGM-SC obtained via* 3-electrode measurements**

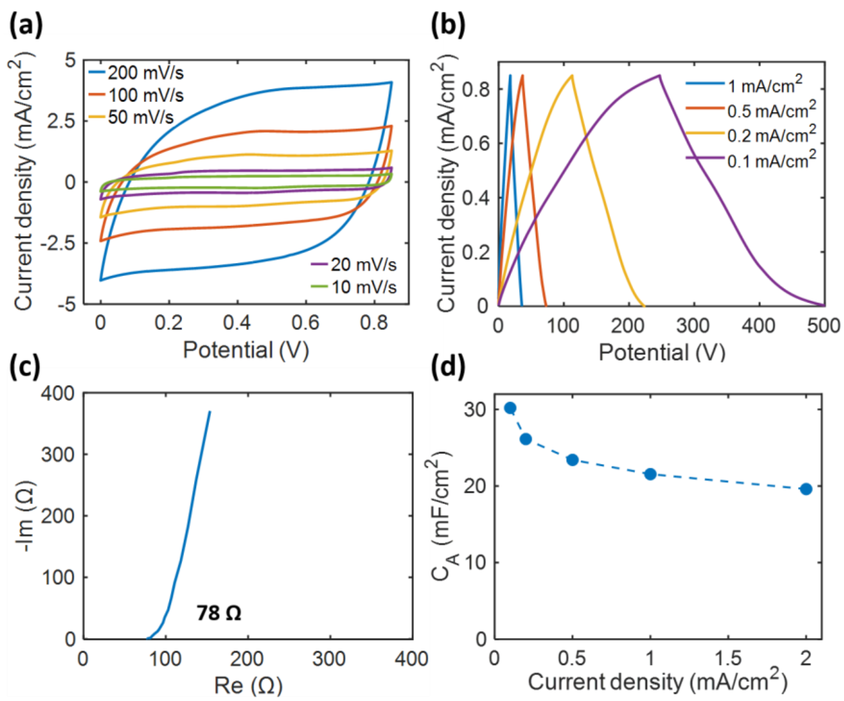

Figure S6. (a) CV , (b) CC , (c) EIS , and (d) areal capacitances at a range of current densities for a single MGM fiber (length: 3.5 cm).

Cyclic charge/discharge testing for MGM-SC

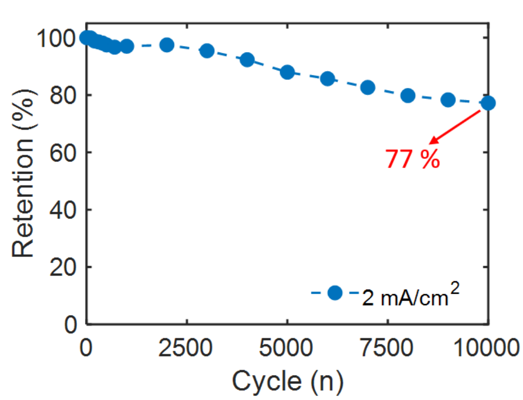

Figure S7. Capacitance retention for 10000 cycles of charge/discharge for MGM-SC.

Ragone plot

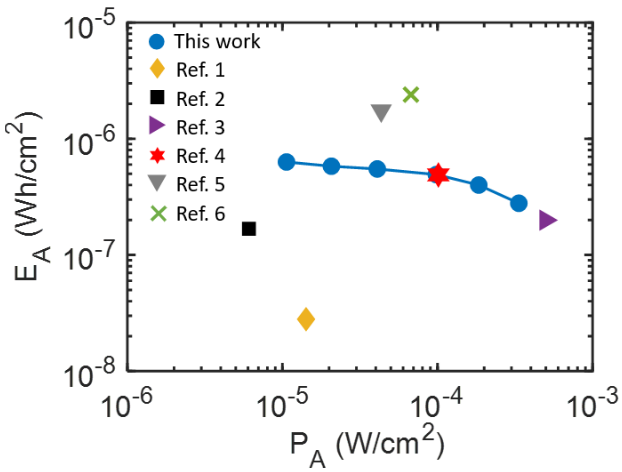

Figure S8. Area-specific Ragone plot of the MGM-SC and other reported fiber SCs. For comparison the data from ref. 1, 2, 3, 4, 5, 6 were reproduced by using Origin® software.

The areal energy density (E_A) and power density (P_A) in this work and ref. 4 and 6 were calculated by:

$$C_{cell} = \frac{I \times t}{(V - IR_{drop})} \qquad \text{(F)} \qquad (1)$$

$$E_A = \frac{1}{2} \times \frac{C_{cell}}{2A} \times \frac{V^2}{3600} \qquad \text{(Wh/cm}^2\text{)} \qquad (2)$$

$$P_A = \frac{E_A \times 3600}{t} \qquad \text{(W/cm}^2\text{)} \qquad (3)$$

, where I is the discharge current (A), t is the discharge time (s), V is the operation voltage (V), IRdrop is the voltage drop at the beginning of the discharge curve (V), and A is the surface area of a single fiber electrode (cm^2).[7, 8] It should be noted that for ref. 2, the C_A and E_A values were calculated by

$$C_A = \frac{I}{A} \times \frac{t}{(V - IR_{drop})} \qquad (4)$$

$$E_A = \frac{1}{2} \times C_A \times \frac{V^2}{3600}\ . \qquad (5)$$

Stress-strain curve of the PVDF fiber

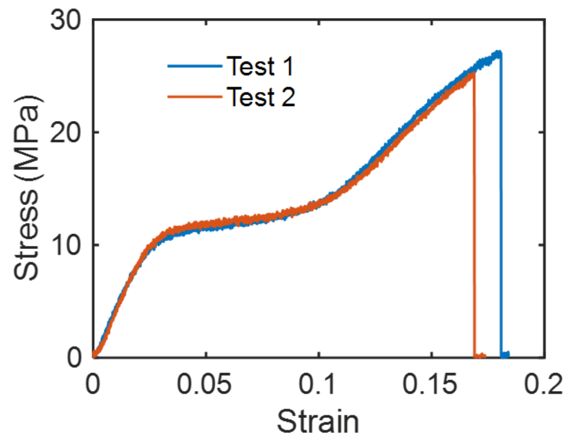

Figure S9. Stress-strain curve of the PVDF fiber.

Droplet-coating method

The AgNPs/AgNWs coated fiber was prepared by using the droplet-coating method. A Ag ink droplet, which was suspended to the syringe needle end, wetted part of the PVDF fiber, enveloping its entire cross-section. The syringe was then translated at a speed of 5 mm/s using a motorized linear stage, so that the droplet swept the entire surface of the fiber. This sweeping was repeated 10 times.

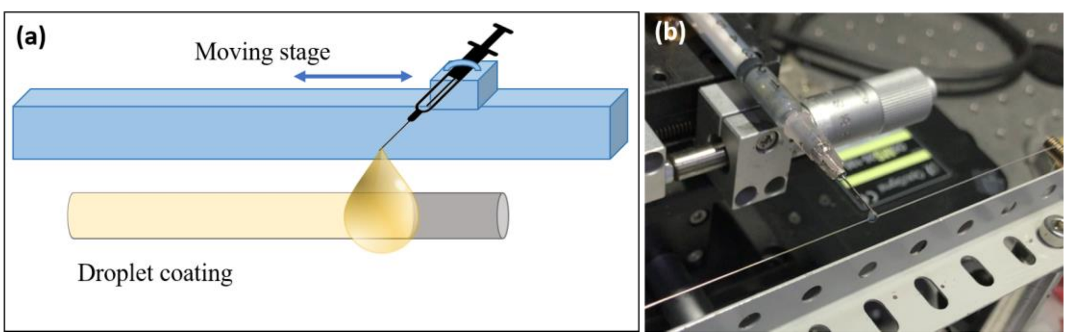

Figure S10. (a) Schematic of the droplet-coating method. (b) Digital image of the droplet-coating setup.

Bendability comparison

Table S1. Bendability comparison of fiber SCs.

Type	Material	Diameter (μm)	Bending angle (º)	Bending radius (cm)	Bending times (n)	Retention (%)	ref
All-in-one Monofilament	PVDF fiber, thin metal layer, Gr, MnO_2	SC device fiber: 320–340	180	1.5–0.25	-	92.5	This work
			150	0.75	3000	94	
	GO fiber, rGO	SC device fiber: 50	-	0.75	160	~80	9
Parallel	Au coated polymer fiber + ink	Single electrode fiber: 200	180, 360	-	-	95	10
	Gr based functional fiber	Single electrode fiber: ~50	0–180	-	-	120	8
			180	-	1000	90	
	Cu wire /rGO/ MnO_2	Single electrode fiber: 150	60	-	5000	80	11
	CNT fiber/CNTs/ PANI	Single electrode fiber: 55	0–180	-	-	95	12
			180	-	500	99.8	
	rGO on Au wire	Single electrode fiber: 130	0-120, S shape	-	-	99	13
			90	-	1000	90	
	Hollow Gr/ conducting polymer fiber	Single electrode fiber: 112	0–180	-	-	105	14
			180	-	500	95	
	MnO_2/porous Ni wire	Single electrode fiber: 300	0-180	-	-	90	15
	All-carbon hybrid fibers	Single electrode fiber: 30	120	-	1000	96	16
		Single electrode fiber: 236	120	-	1000	89	
Twisted	MWCNT/ OMC fiber	Single electrode fiber: 150	180	-	1000	90	5
	rGO fiber	SC device fiber: 500	0–360, intertwined	-	-	90	17
			180	-	1000	90	
	SWCNTs/ PAniNW/gel yarn	Single electrode fiber: ~50	0–180	-	-	95	4
	GF@PEDOT	Single electrode fiber: 90	0–250	-	-	90	18
			180	-	300	80	
Coaxial	GF/Gr sheath	SC device fiber: ~190	180	-	100	92	19

Gr: graphene, GF: graphene fiber, rGO: reduced graphene oxide, CNT: carbon nanotube, PANI: polyaniline, MWCNT: multi-walled carbon nanotube, SWCNT: single-walled carbon nanotube, PaniNW: polyaniline nanowires, OMC: ordered mesoporous carbon, PEDOT: Poly(3,4-ethylenedioxythiophene).

Specific capacitance comparison

Table S2. Comparison of specific capacitances of fiber SCs.

Type	Material	Electrolyte	C_L (mF/cm)	C_A (mF/cm^2)	Ref
Monofilament	PVDF fiber, Ag/Au/Gr/MnO_2	PVA-Na_2SO_4	**0.51** at 0.1 mA/cm^2	**12.3** at 0.1 mA/cm^2 * **9.5** at 2 mA/cm^2 *	This work
	GO fiber, rGO	$BMIMBF_4$	-	**1.2** at 80 μA/cm^2	9
		0.1 M $NaClO_4$ in CH3CN	-	**0.24** at 200 μA/cm^2	
Parallel	Gr based functional fiber	PVA-H_2SO_4	-	**74.25** at 40 mV/s *	8
	Cu/rGO/MnO_2	PVA-KOH	-	**140** at 0.1 mA/cm^2	11
	CNT fiber/CNT/PANI	PVA-H_3PO_4	-	**67.31** at 0.5 mA/cm^2	12
	MnO_2/porous Ni wire	PVA-KOH	-	**847.22** at 0.41 mA/cm^2	15
Twisted	GF/MnO_2	PVA-H_2SO_4	**0.143** at 100 mV/s	**9.1–9.6** at 2 μA	20
	Gr/conducting polymer microfibers	PVA-H_2SO_4	**0.58** at 0.53 mA/cm^2	**15.39** at 0.53 mA/cm^2	18
	SWCNTs/ PAniNW/gel yarn	PVA-H_2SO_4	-	**3.065** at 0.2 A/g *	4
Coaxial	Stainless steel, ink, AC	PVA-H_3PO_4	**0.1** at 40 μA	**3.18** 40 μA	21
	Nanoporous Au wire, MnO2	PVA-LiCl	-	**12** at 0.3 mA/cm2 **6** at 2 mA/cm2	22

Gr: graphene, GF: graphene fiber, GO: graphene oxide, rGO: reduced graphene oxide, CNT: carbon nanotube, SWCNT: single-walled carbon nanotube, PaniNW: polyaniline nanowires, NW: nanowire.

* The specific capacitance values reported in this paper, ref. 8 and ref. 4 were calculated from the equation:

$$C_X = 2\frac{I}{X} \times \frac{t}{(V - IR_{drop})} \tag{6}$$

, where I is the discharge current (A), t is the discharge time (s), V is the operation voltage (V), and IR_{drop} is the voltage drop at the beginning of the discharge curve (V). X can be the effective surface area (A) or length (L) of a single electrode. However, other studies in the table reported the specific capacitance value from the following equation:

$$C_X = \frac{I}{X} \times \frac{t}{(V - IR_{drop})}\,. \tag{7}$$

Therefore, the specific capacitance value reported in the marked papers has been divided by a factor of 2 in this table for fair comparison.

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