Electronic Supplementary Information (ESI)

All-in-One GrapheneFiberSupercapacitor

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Experimental Section

GO suspension is prepared by modified Hummers method as reported.[s1] The GO fiber was produced by wet spinning method through a single capillary spinneret similar to our previous work.[s2] The region-confined reduction of GO was carried out as described in ref. s3. The argon ion lasers of 458nm were used to obtain the RGO layer on the GO fiber by a laser scanning confocal microscope (OLYMPUS FV1000). The ×10 objective lens was used to focus the laser (about 1.7μm of spot size) on the samples, 40μs exposure duration of each voxeland facial scan mode was chosen to reduce the exposed area in the vision. The two RGO parts were controlled to have almost same area by precisely controlling the mobile platform on the microscope to maintain the symmetry structure of the device. About 15 seconds were taken for reducing each of chosen regions (about 2mm long and 1/4 width of the fiber diameter). The laser spots were moved to the desired part by moving the mobile platform. The process can be easily repeated with a system that involved a laser source and a

moving-controllable mobile platform.

All electrochemical behaviours of the device were measured by a CHI660D electrochemical workstation. Two gold sheets were contacted with the RGO parts and connected with the external circuit. One of the gold sheet was wrapped by a thin plastic of PVDC insulating layer to prevent the short circuit of the device. Ionic liquid as electrolyte was 1-butyl-3-mthylimidazolium tetrafluoroborate and used as received. The capacitances calculated by the charge/discharge curves were based on the equation: $C=I/(dV/dt)A$, where I is the discharge current and A is the surface area of the RGO part and is equal to 1/4 of the total value of □π multiplied by the diameter of the fiber and the device length.[s4] The capacitances calculated by the CV curves were derived from the equation: $C = I/sA$, where I is the average cathode current, s is the scan rate and A represents the surface area of the electrode.

The morphologic characterization of the fiber was carried out by JEM-7001F SEM unit and the mechanical property of the fiber were obtained by a material testing system (SHIMADZU AGS-X 50N). The strain rate for a centimeter gauge length is 1 mm/min with a preload of 0.5N. Raman spectra were measured under ambient condition using a RenishawmicroRaman spectroscopy system with a 514.5nm argon-ion laser.

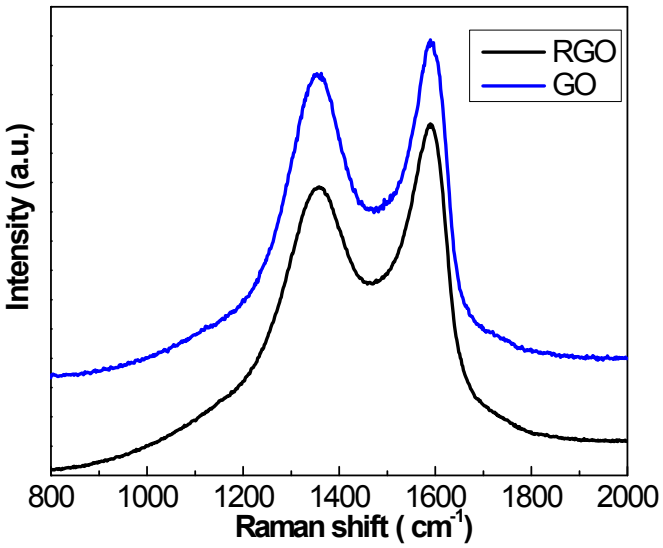

Fig.S1 Raman spectra of RGO and GO parts of the RGO/GO/RGO fiber.

Both RGO and GO regions have two remarkable bands around 1354 $cm^{-1}$and 1598 cm^{-1} assigned to the D- and G-bands of carbon, respectively. The G-band is related to graphitic carbon and the D-band isassociated with the structural defects or partially disordered structures of graphitic domains. The I_D/I_G value of GO was calculated to be 0.83, while that of RGO decreased to 0.78, indicating that GO sheets were reduced and their conjugated structures were partly restored.

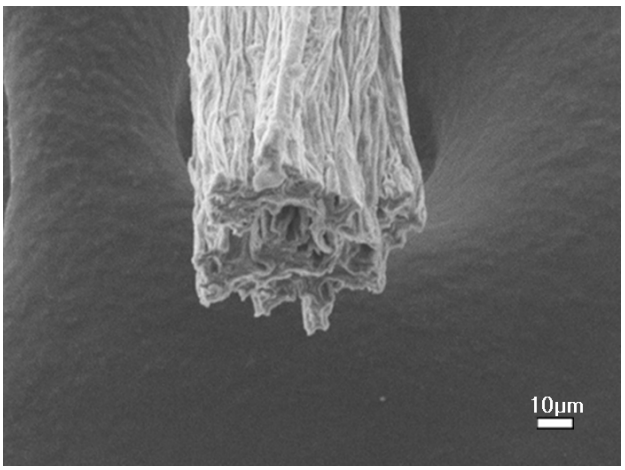

Fig. S2 SEM image of the cross-section of RGO/GO/RGO fiber.

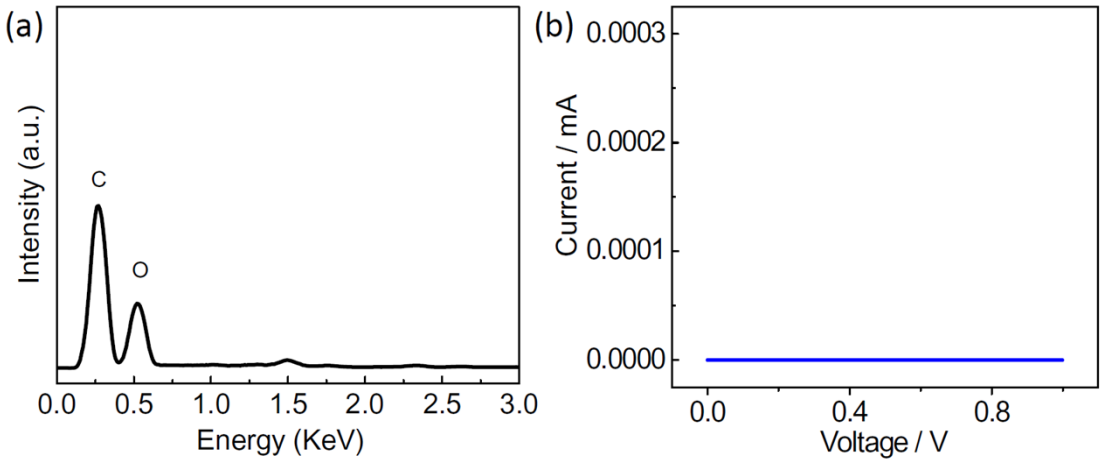

Fig.S3 The typical EDS (a) and I-V curve of the GO fiber after standing in the ambient condition for one day.

The GO fiber is generally stable under the experimental condition. Both its chemical composition and nonconductive nature remain almost unchanged.

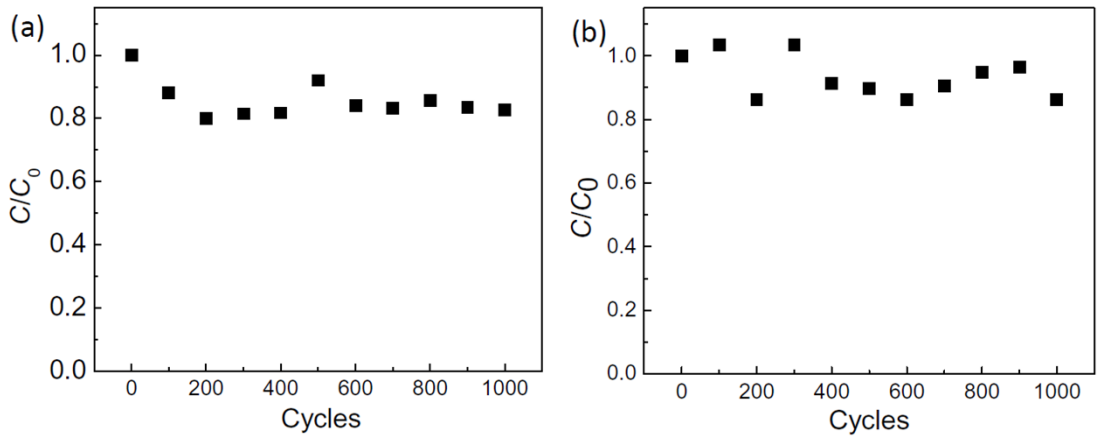

Fig. S4 Capacitance C related to the initial capacitance C_0 of the RGO/GO/RGO fiber for different bending cycles. (a) is measured at straight state, and (b) is measured at the bent state.

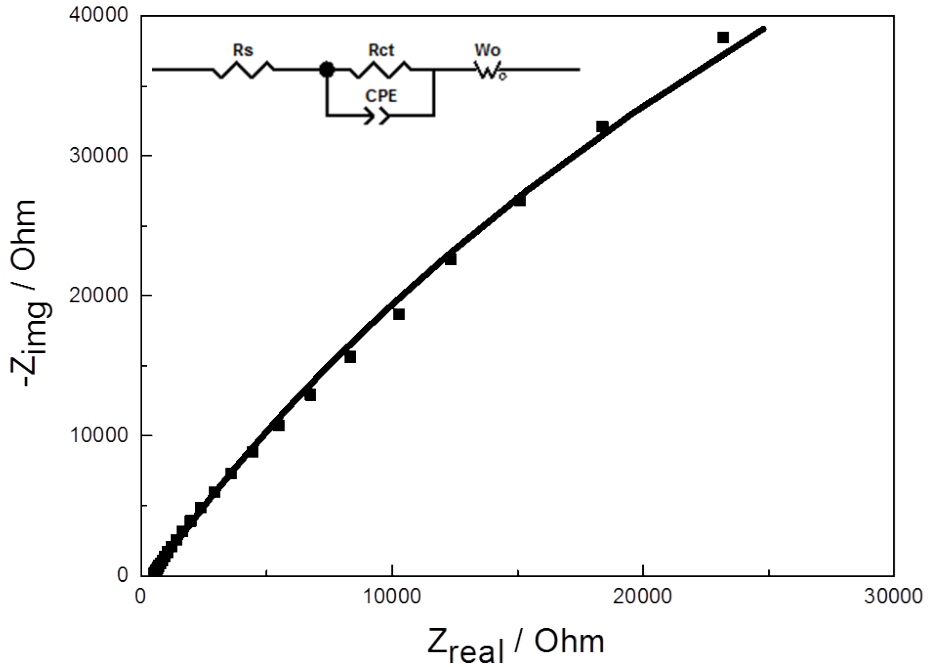

Fig. S5 The EIS spectrum of the fiber microcapacitor (scattered points) and the fitting curve (solid curve). Inset of the picture: the equivalent circuit used to fit the EIS spectrum.

Fig. S5 shows the EIS spectrum of the fiber microcapacitor (scattered points) and the fitting curve of the spectrum (solid curve). The raw data are fitted by the following equivalent circuit, where Rs is the serial resistance of the capacitor, Rct stands for the

charge transfer resistance at the GO/G interface, CPE represents a constant phase element used to simulate the capacitive component of the capacitor and Wo is the Warburg element contributed from the diffusion of the electrolyte, respectively. Based on the current equivalent circuit, the serial resistance of the capacitor is estimated to be 455 Ohm.

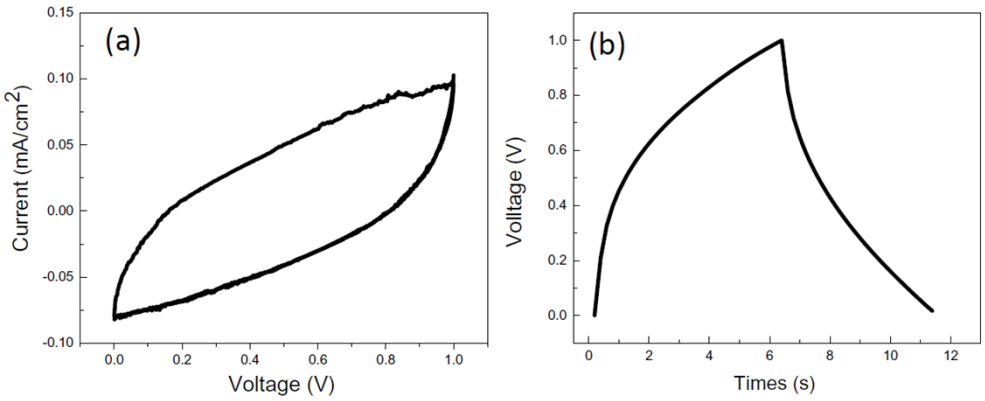

Fig. S6 (a) The CV curve at scan rate of 0.05V/s and (b) The charge/discharge curve at current density of 100μA/cm^2 with ionic liquid electrolyte for the RGO/GO/RGO fiber with an approximate reduced width of 1/5 fiber perimeter. The calculated capacitance is only 0.5mF/cm^2.

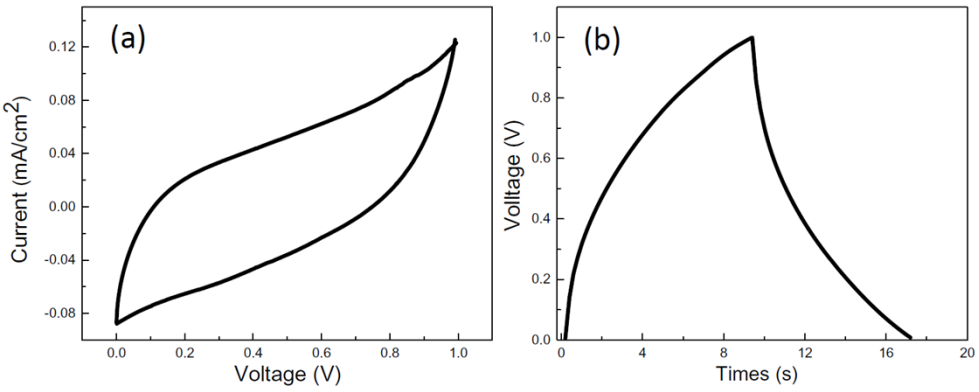

Fig. S7 (a) The CV curve at scan rate of 0.05V/s and (b) The charge/discharge curve at current density of 120μA/cm^2 with ionic liquid electrolyte for the RGO/GO/RGO fiber with an approximate reduced width of 1/2 fiber perimeter. The calculated capacitance by charge/discharge curve is about 0.9 mF/cm^2 at a current density of 120μA/cm^2.

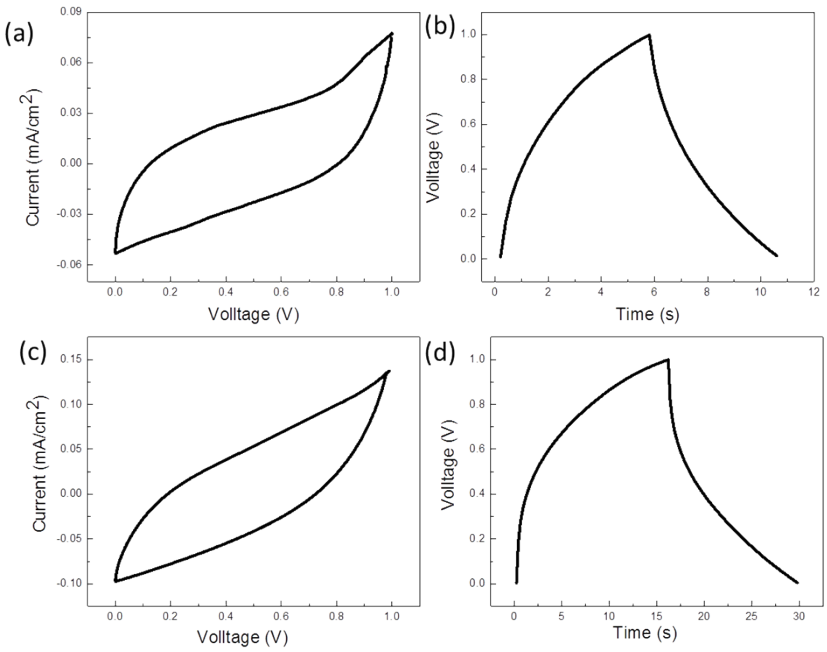

Fig. S8 (a,b) The CV curve at scan rate of 0.05V/s and the charge/discharge curve at current density of 80μA/cm^2 with ionic liquid electrolyte for RGO/GO/RGO fiber with 20μs exposure duration of each voxel. (c,d) The CV curve at scan rate of 0.05V/s and the charge/discharge curve at current density of 80μA/cm^2 with ionic liquid electrolyte for RGO/GO/RGO fiber with 100μs exposure duration of each voxel.

The depth of the RGO part cannot be measured precisely. However, we could change the exposure time to adjust it. As for the area of RGO was fixed, the longer it was exposed, the deeper it supposed to be. At the exposed time of 20μs duration of each voxel, the capacitance is only 0.4mF/cm^2. As the exposed time increased to 100μs, the capacitance reached 1.1mF/cm^2, which is almost the same level of 40μs exposure duration of each voxel. That is to say, 40μs exposure duration of each voxel is sufficient for fiber capacitor, the longer exposed time and deeper depth have no significant influence on the capacitance performance.

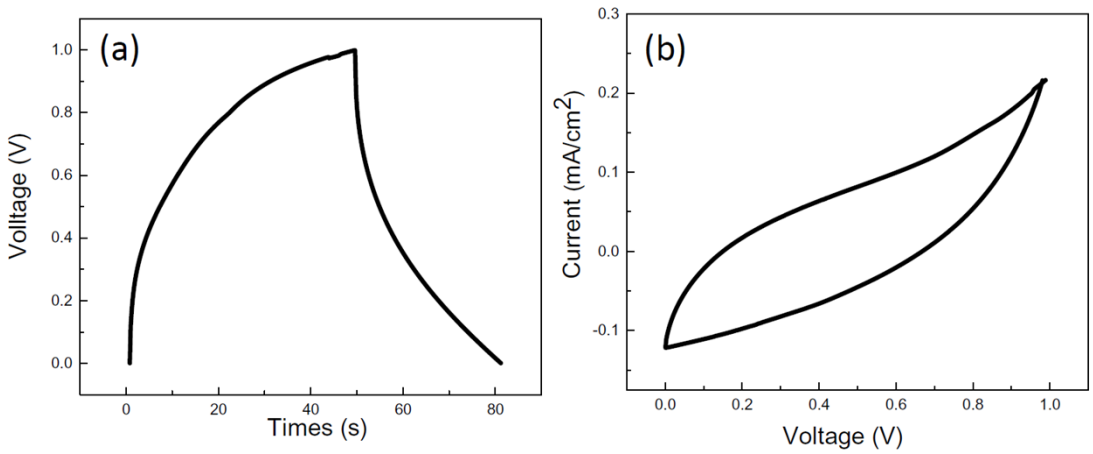

Fig. S9 (a) The CV curve at scan rate of 0.05V/s and (b) The charge/discharge curve at current density of 80μA/cm^2 with ionic liquid electrolyte for the RGO/GO/RGO fiber with a diameter of about 70μm and 1/4 fiber perimeter for RGO part. The calculated capacitance by charge/discharge curve is about 2.4 mF/cm^2.

Table S1 The capacitance and energy density of the fiber-based capacitors.

	Capacitance	**Energy density**	**Electrolyte**
MnO_2–ZnO nanowires[1]	2 mF/cm^2	2.7×10^{-8}Wh/cm^2	PVA/H_3PO_4 gel electrolyte
CNT fiber and Ti wire [2]	0.6 mF/cm^2	1.5×10^{-7}Wh/cm^2	NH_4F and H_2O in ethylene glycol
MWCNT/MnO_2 fiber [3]	3.57 mF/cm^2	1.7mWh/cm^3	PVA/H_3PO_4
RGO/GO films [4]	0.6 mF/cm^2	4.3×10^{-4}Wh/cm^2	None
Current work	1.2 mF/cm^2	5.4×10^{-4}Wh/cm^2	Ionic liquid
Current work	2 mF/cm^2	6.4×10^{-4}Wh/cm^2	0.1M $NaClO_4$ in acetonitrile

References

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s2 Y. Zhao, C.C. Jiang, C.G. Hu, Z.L. Dong, J.L. Xue, Y.N. Meng, N. Zheng, P.W. Chen, L.T. Qu, *ACS nano*, 2013, **7**, 2406–2412.

s3 H.H. Cheng, J. Liu, Y. Zhao, C.G. Hu, Z.P. Zhang, N. Chen, L. Jiang, L.T. Qu, *Angew. Chem. Int. Ed.*, 2013, **52**, 40, 10482–10486.

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