Electronic Supplementary Information

Layered Coating of Ultraflexible Graphene-Based Electrodes for High-Performance In-plane Quasi-Solid-State Micro-supercapacitor

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Calculation

The areal capacitance from CV curves can be calculated by Eq. 1:

$$C_A = \frac{\int I(v)dv}{AVS} \qquad (1)$$

Where C_A is the areal capacitance (mF cm^{-2}), I is the response current (A), A is the area of electrodes (cm^2), V is the voltage window (V), and S is the scan rate (mV s^{-1}). The specific capacitance from GCD curves can be calculated by Eq. 2:

$$C_A = \frac{I\Delta t}{AV} \qquad (2)$$

Where C_A is the areal capacitance (mF cm^{-2}), I is the discharge current (A), A is the area of electrode (cm^2), Δt is the discharge time (s), and V is the voltage window (V).

The energy density of the device can be calculated by the Eq. 3:

$$E=\frac{C*V^2}{2*h} \qquad (3)$$

and the power density is given by Eq. 4:

$$P=\frac{E}{t} \qquad (4)$$

Where E is the energy density (Wh cm^{-3}), C is the areal capacitance (mF cm^{-2}) calculated by GCD curves, V is the voltage window (V), h is the thickness of electrode (cm), P is the volumetric power density (W cm^{-3}), and t is the discharge time (s).

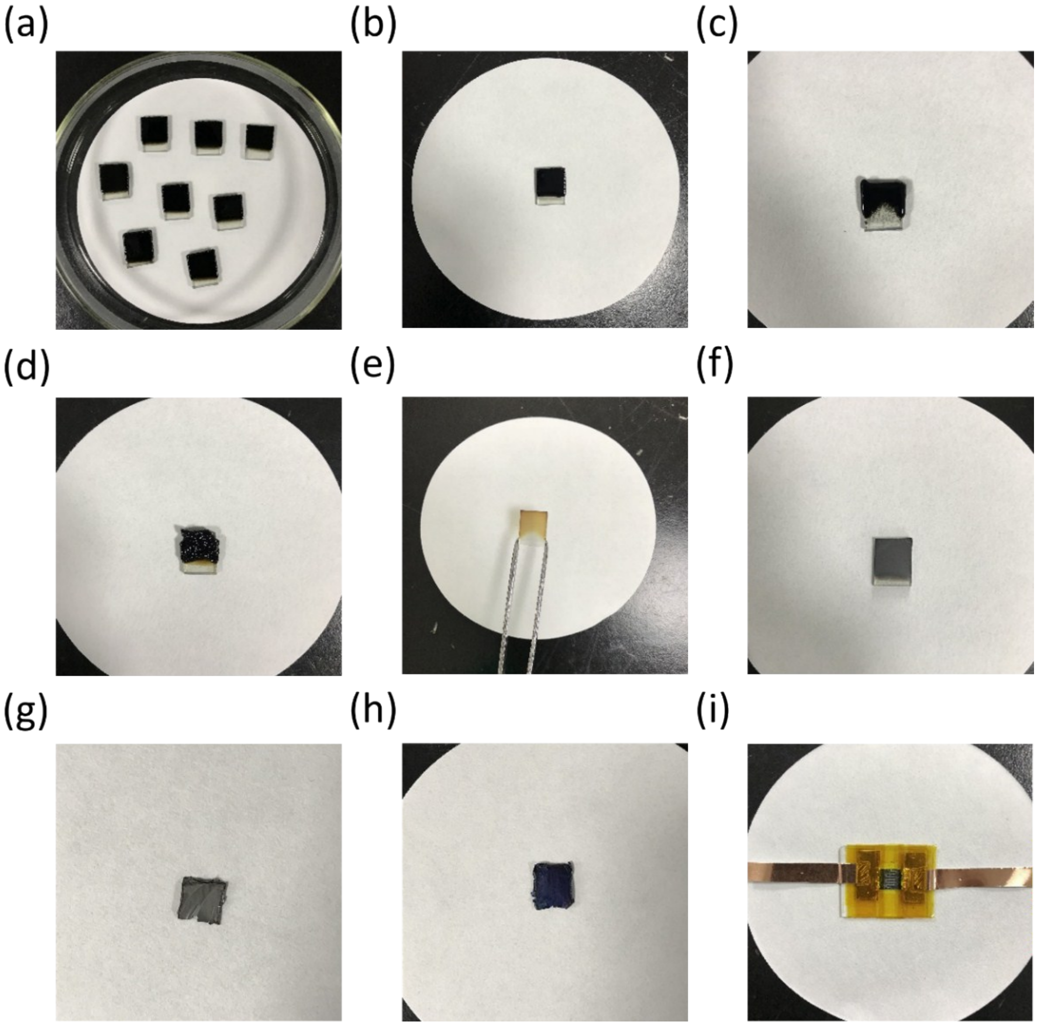

Fig. S1. Digital photos of devices at each fabrication step. (a) and (b) The images of pre-reduced GO (p-rGO) films taken immediately after EPD and the size of FTO glass is 1.2 cm * 1.6 cm. (c) p-rGO film which was partially reduced by L-AA. (d) P-rGO film which was reduced deeply. (e) Non-reduced GO film. (f) The dry p-rGO film. (g) The completely reduced rGO film (c-rGO). (h) The MGF. (i) The MSC based on MGFs.

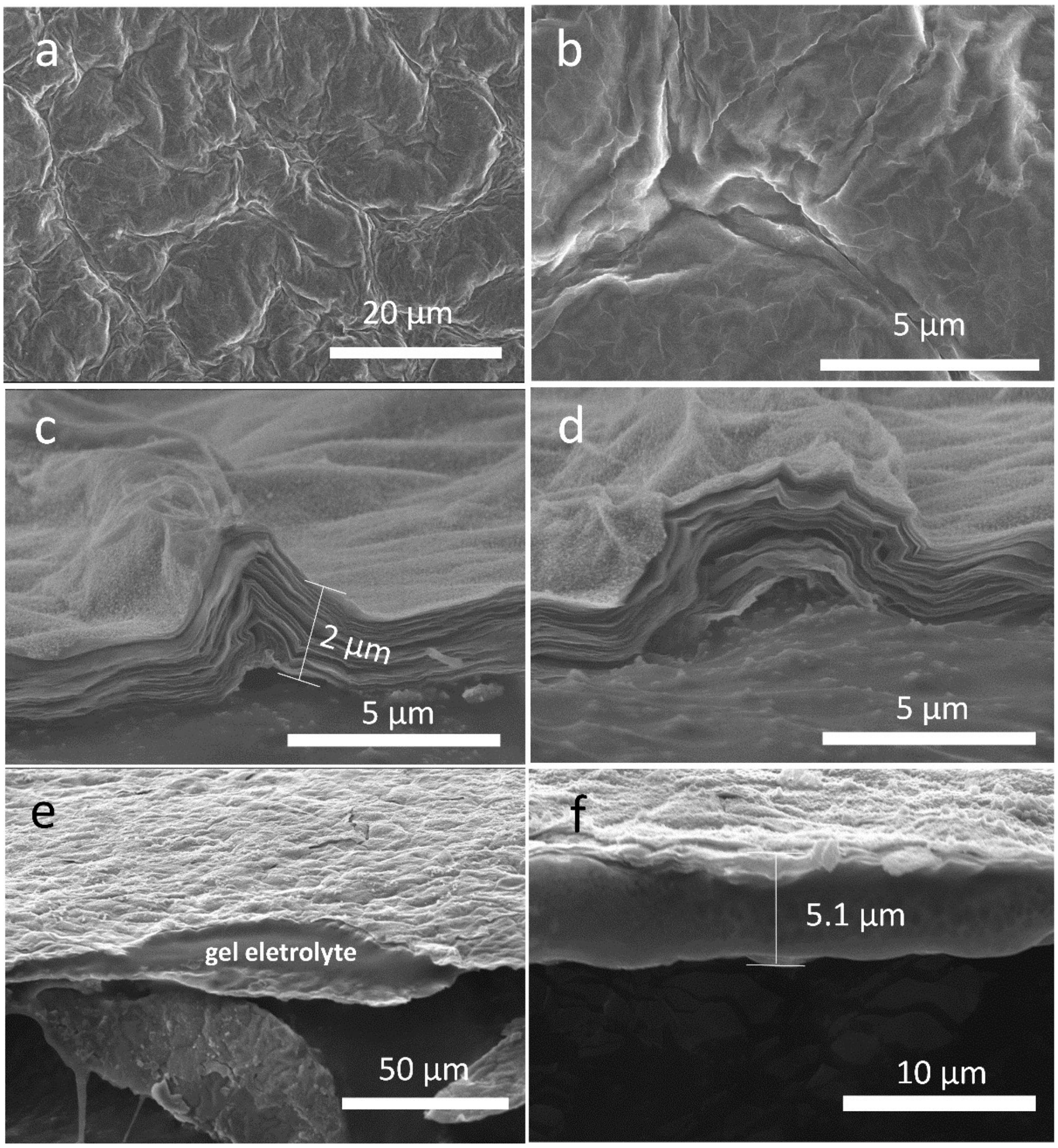

Fig. S2. (a) and (b) SEM images of c-rGO. (c) and (d) Cross-section image of MGF-60 showing the thickness of MGF-60 is about 2 μm. (e) and (f) Cross-section image of MGF-60-2.

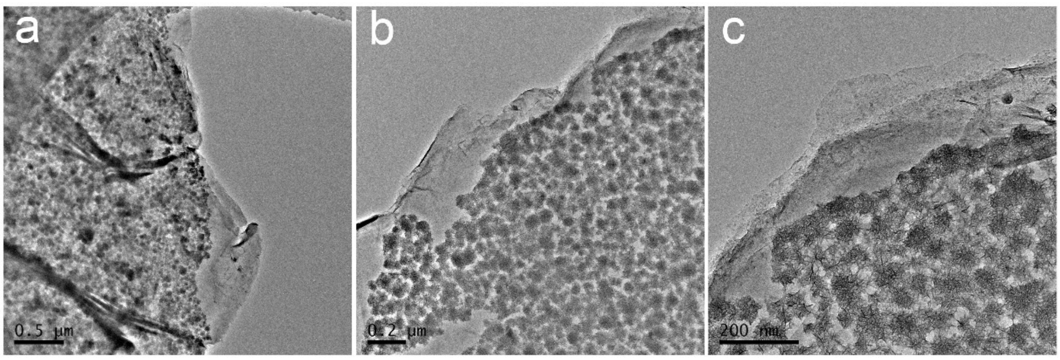

Fig. S3. (a), (b) and (c) TEM images of MGF-60.

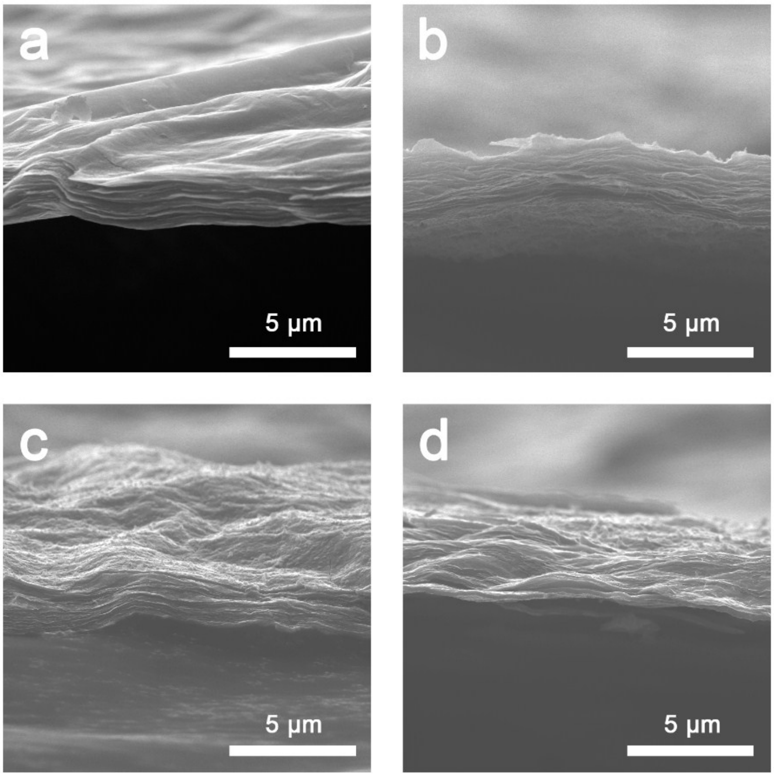

Fig. S4. Cross-section images of (a) MGF-0, (b) MGF-30, (c) MGF-90 and (d) MGF-120.

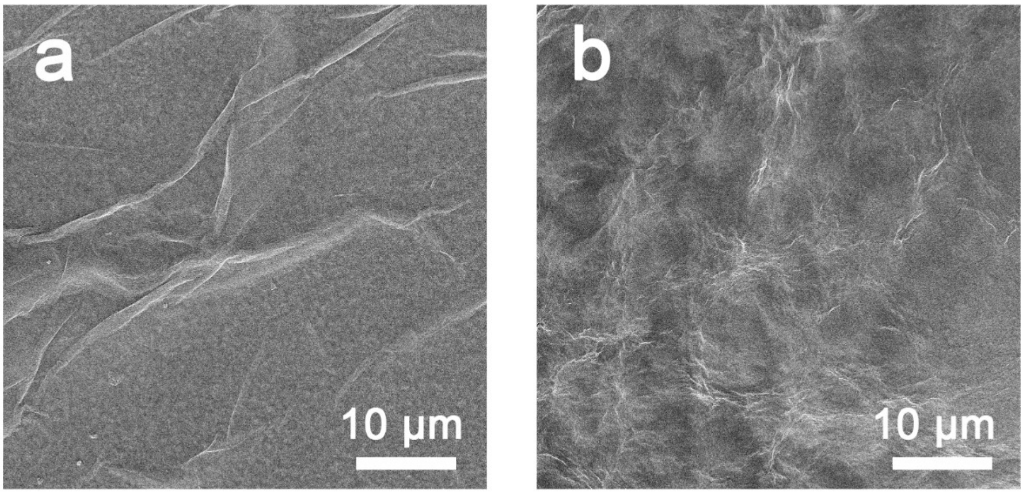

Fig. S5. SEM images of (a) GO and (b) p-rGO.

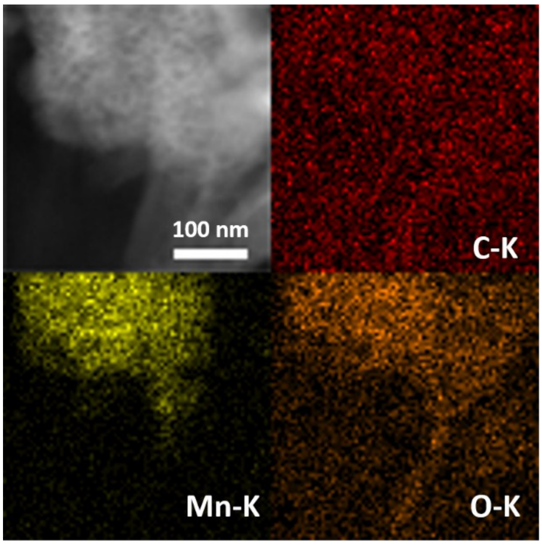

Fig. S6. Element mapping images of MGF.

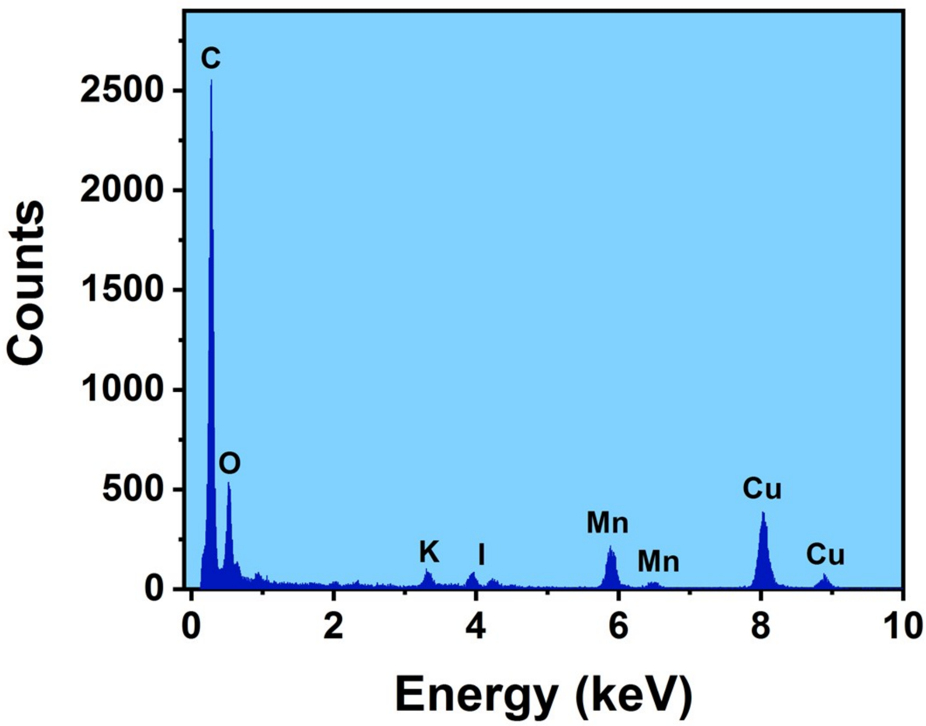

Fig. S7. EDX spectrum of MGF-60.

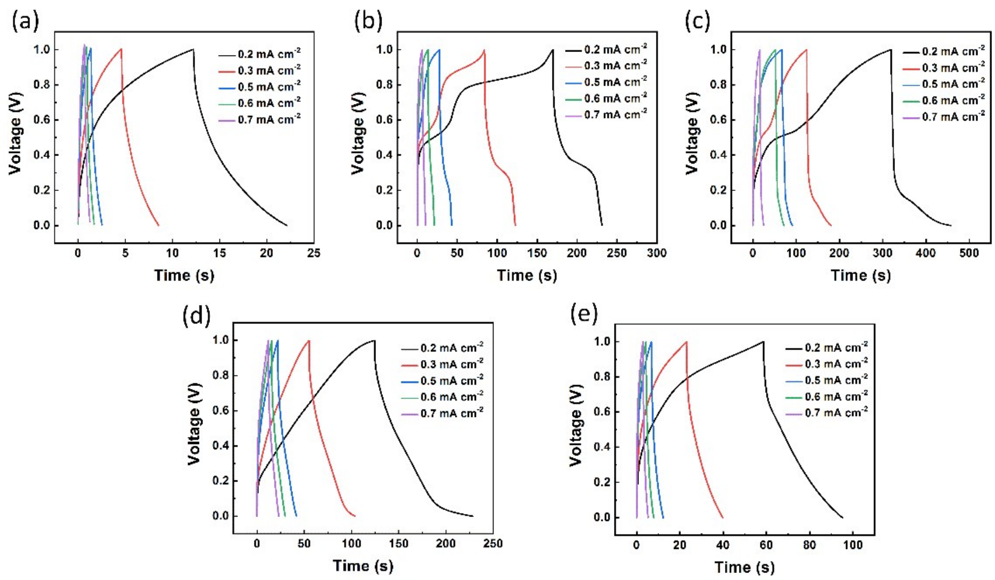

Fig. S8. GCD curves of each device with different hydrothermal growth time at current density ranging from 0.2 mA cm^{-2} to 0.7 mA cm^{-2}. (a) MGF-0-2. (b) MGF-30-2. (c) MGF-60-2. (d) MGF-

90-2. (e) MGF-120-2.

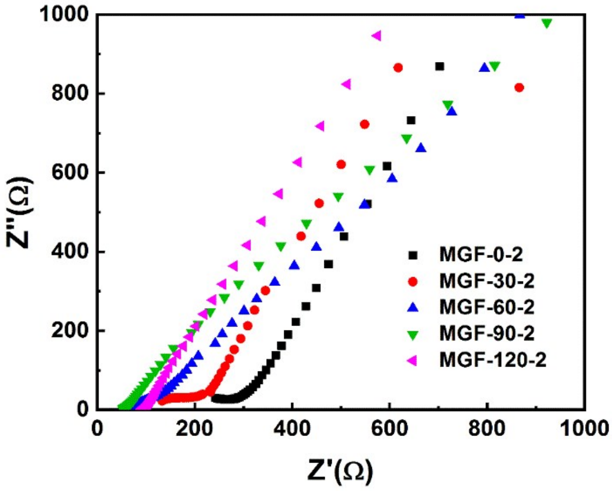

Fig. S9. Magnified Nyquist plots in the high-frequency region of samples.

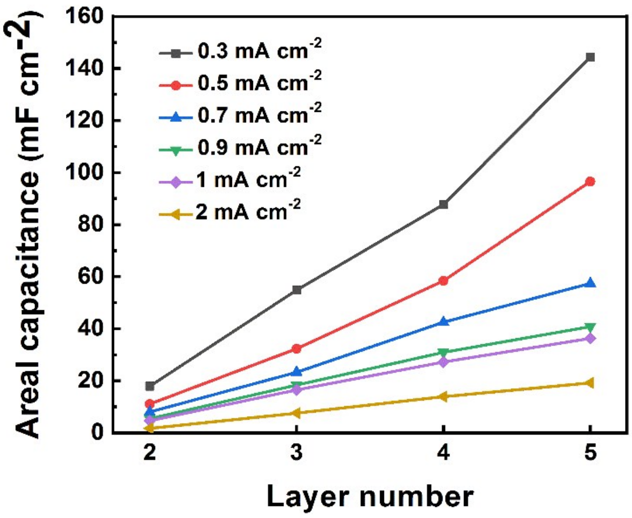

Fig. S10. The areal capacitance of MSCs based on MGF-60 with different number of layers at different current density.

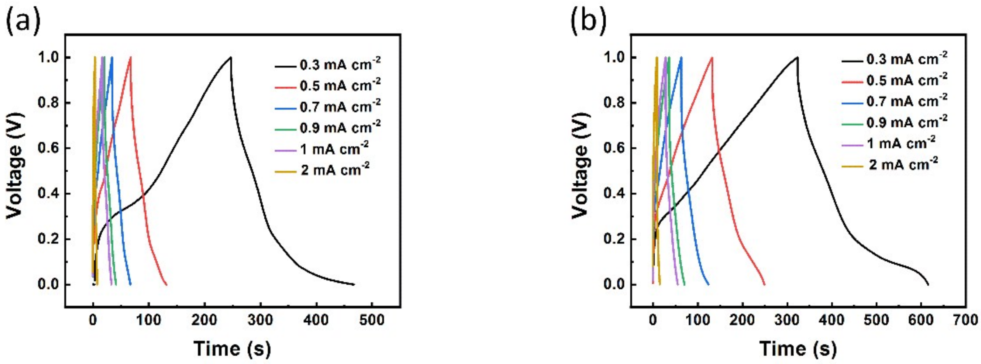

Fig. S11. (a) GCD curves of MGF-60-3. (b) GCD curves of MGF-60-4.

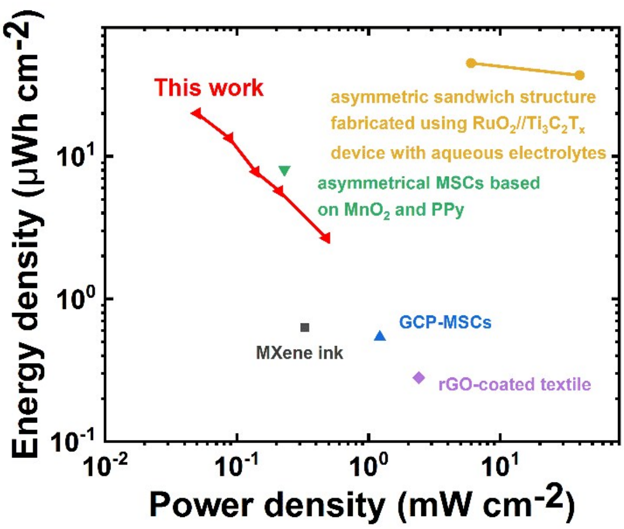

Fig. S12. Ragone plots show the areal energy density and power density of the MGF-60-5 MSC in comparison with other MSCs based on Mxene-RuO_2, Mxene ink, GCP-MSCs, rGO-coated textile,

MnO_2 and PPy.