Supplementary information

Graphene fiber-based asymmetric micro-supercapacitors

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Supplementary information: Text

Calculation

(1) The specific capacitance calculated by CV:

\[ C_A = \frac{A \times u \times \left( U_2 - U_1 \right)}{\int_{U_1}^{U_2} I dU + \int_{U_2}^{U_1} I dU} \]

Where \( C_A \) (mF/cm²), \( A \) (cm²), \( u \) (V/s), \( U_2 \) and \( U_1 \) (V), and \( I \) (A) are the area specific capacitance, the surface area of single electrode, scanning rate, high and low potential limit of CV tests, and the instant current of CV curves, respectively. Here \( C_A \) is the specific capacitance of single fiber. Relationship between specific capacitance of single fiber and the whole capacitor is:

\[ C_{tA} = \frac{1}{4} C_A \]

For F-asym-mSCs, specific capacitance of total device \( (C_{tA}) \) was calculated by the following equation:

\[ C_{tA} = \frac{A' \times u \times 2(U_2 - U_1)}{\int_{U_1}^{U_2} I dU + \int_{U_2}^{U_1} I dU} \]
Where $A'$ is the **total area** of two electrodes, the other symbols are the same as above. Here the $C_S$ is the specific capacitance of whole capacitor cell.

(2) The specific capacitance calculated by GCD:

$$C_A = \frac{I \times t}{\Delta U \times A}$$

Where $C_A$ (mF/cm$^2$), I (mA), $t$ (s), $\Delta U$ (V), and A (cm$^2$) are the area specific capacitance, the discharge current, the discharge time, the potential window and **surface area of single electrode**, respectively. Here $C_A$ is the specific capacitance of single fiber.

For F-asym-mSCs, $C_{tA}$ was calculated by the following equation:

$$C_{tA} = \frac{I \times t}{\Delta U \times A'}$$

Where $A'$ is the **total surface area of two electrodes**, the other symbols are the same as above. Here the $C_{tA}$ is the specific capacitance of whole capacitor cell.

(3) The volume capacitance and linear capacitance are calculated as followed:

$$C_V = \frac{C_A \times A}{V}$$

$$C_V = \frac{C_A \times A}{L}$$

Where A (cm$^2$), V (cm$^3$), and L (cm) are surface area of electrode, volume of electrode and length of electrode, respectively.

(4) The energy density ($E$) and power density ($P$) were calculated by the following equations:

$$E = \frac{C U^2}{2}$$

$$P = \frac{E}{t}$$

Where $C_A$ (mF/cm$^2$), U (V), t (s) are specific capacitance of whole capacitor cell, potential window, and discharge time, respectively.
**Figure S1.** (a) Mechanical properties of GMF1, GMF5, GMF9, and GMF12. (b) Mechanical properties of LGO fibers and GCF11.
**Figure S2.** (a)-(k) SEM images of neat GF surface (a and b); GMF5 (c and d); GMF 9 (e and f); GMF 24 (g, h and i); Cross-section of GMF24 (j and k). (l) TEM image of GMF9. (Scale bars of Figure a, c, e, g, j are 10 μm; Figure b, h, l are 500 nm; Figure d and i are 1 μm; Figure f is 2 μm; Figure k is 4 μm.)
Figure S3. Cycling stability test of GMF9.
Figure S4. (a) and (b) Cross-section images of GCF observed by SEM, scale bars: 5μm and 1μm. (c) CV curves of GCF at different scan rates. (d) GCD curves at different current densities.
Figure S5. (a) $C_A$ of GF, GMF1, GMF5, GMF9, GMF12, and GCF at different scanning rates. (b) $C_A$ of GF, GMF1, GMF5, GMF9, GMF12, GCF at different current density.
**Figure S6.** (a) GCD curves of parallel F-asym-mSCs at different potential windows (current density 0.5 mA/cm²). (b) GCD curves of parallel F-asym-mSCs at different current densities when operating voltage is 1.6 V.
Figure S7. (a) $C_t$ and $E_a$ of two-ply F-asym-mSCs at different scanning rates. (b) $C_t$ and $E_a$ of two-ply F-asym-mSCs at different current densities.
Supplementary information: Tables

Table S1. Summary of electrochemical performances of F-asym-mSCs obtained from the CV tests.

<table>
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<tr>
<th>Scan rate (mV/s)</th>
<th>$C_{id}$ (mF/cm$^2$)</th>
<th>$C_{il}$ (mF/cm)</th>
<th>$C_{ir}$ (F/cm$^3$)</th>
<th>$P_A$ (W/m$^2$)</th>
<th>$E_A$ (μWh/cm$^2$)</th>
<th>$P_V$ (W/cm$^3$)</th>
<th>$E_V$ (mWh/cm$^3$)</th>
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Table S2. Summary of electrochemical performances of F-asym-mSCs obtained from GCD tests.

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<th>Current density (mA/cm²)</th>
<th>C_{dA} (mF/cm²)</th>
<th>C_{dL} (mF/cm)</th>
<th>C_{dV} (F/cm³)</th>
<th>P_{d} (W/m²)</th>
<th>E_{d} (μWh/cm²)</th>
<th>P_{f} (W/cm³)</th>
<th>E_{f} (mWh/cm³)</th>
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