Supplementary information

Graphene fiber-based asymmetric micro-supercapacitors

Bingna Zheng, Tieqi Huang, Liang Kou, Xiaoli Zhao, Karthikeyan Gopalsamy, Chao Gao*.

MOE Key Laboratory of Macromolecular Synthesis and Functionalization, Department of Polymer Science and Engineering, Zhejiang University, 38 Zheda Road, Hangzhou, 310027, P. R. China

Supplementary information: Text

Calculation

(1) The specific capacitance calculated by CV:

$$C_A = \frac{\int_{U_1}^{U_2} IdU + \int_{U_2}^{U_1} IdU}{A \times u \times (U_2 - U_1)}$$

Where C_A (mF/cm²), A (cm²), u (V/s), U₂ and U₁ (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, sepcific capacitance of total device (C_{tA}) was calculated by the following equation:

$$C_{tA} = \frac{\int_{U_1}^{U_2} IdU + \int_{U_2}^{U_1} IdU}{A' \times u \times 2(U_2 - U_1)}$$

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/cm²), I (mA), t (s), ΔU (V), and A (cm²) 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 (cm²), V (cm³), 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:

$$E = \frac{CU^2}{2}$$
$$P = \frac{E}{t}$$

Where C_A (mF/cm²), U (V), t (s) are specific capacitance of whole capacitor cell, potential window, and discharge time, respectively.

Supplementary information: Figures



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 μ m; Figure b, h, l are 500 nm; Figure d and i are 1 μ m; Figure f is 2 μ m; Figure k is 4 μ m.)



Figure S3. Cycling stability test of GMF9.



Figure S4. (a) and (b) Cross-section images of GCF observed by SEM, scale bars: 5μ m and 1μ m. (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/cm^2). (b) GCD curves of parallel F-*asym*-mSCs at different current densities when operating voltage is 1.6 V.



Figure S7. (a) C_{tA} and E_A of two-ply F-*asym*-mSCs at different scanning rates. (b) C_{tA} 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.

Scan rate (mV/s)	C_{tA} (mF/cm ²)	C_{tL} (mF/cm)	C_{tV} (F/cm ³)	P_A (W/m ²)	E_A (µWh/cm ²)	P_V (W/cm ³)	E_V (mWh/cm ³)
100	17.1	0.21	17.1	13.6	6.1	1.4	6.1
80	19.2	0.24	19.2	12.3	6.8	1.2	6.8
50	23.2	0.29	23.2	9.3	8.2	0.9	8.2
20	29.3	0.37	29.3	4.7	10.4	0.5	10.4
10	33.6	0.42	35.2	2.7	11.9	0.3	11.9

Current density (mA/cm ²)	C_{tA} (mF/cm ²)	C _{tL} (mF/cm)	C_{tV} (F/cm ³)	P_A (W/m ²)	E_A (μ Wh/cm ²)	P_V (W/cm ³)	E_V (mWh/cm ³)
0.1	19.1	0.24	19.1	0.4	6.7	0.40	6.7
0.2	18.3	0.23	18.3	0.8	6.3	0.79	6.3
0.5	16.8	0.21	16.8	1.9	5.5	0.19	5.5
0.8	15.8	0.20	15.8	3.0	4.9	0.30	4.9
1	15.4	0.19	15.4	3.7	4.6	0.37	4.6
2	13.4	0.17	13.4	6.7	3.3	0.67	3.3

Table S2. Summary of electrochemical performances of F-*asym*-mSCs obtained from GCD tests.