# Electronic Supplementary Information for

# A graphene/carbon nanotube@π-conjugated polymer nanocomposite for high-performance organic supercapacitor electrode

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## Experimental section

### Data analysis of electrochemical measurements

The specific capacitance of the three-electrode system ($C_s$) was calculated by using the following equation:

$$C_s = I \, \mathrm{d}t \, / \, (m \, \mathrm{d}V) \tag{1}$$

in which $Cs$ (F g$^{-1}$) is the specific capacitance, $I$ (A) is the discharge current, $\mathrm{d}t$ (s) is the discharge time, $m$ (g) is the mass of the active material loaded in working electrode, and $\mathrm{d}V$ (V) is the potential window after IR drop.

The specific capacitance of the two-electrode cell configuration ($C_{\mathrm{cell}}$) was calculated by using the following equation:

$$C_{\mathrm{cell}} = I \, \mathrm{d}t \, / \, (m \, \mathrm{d}V) \tag{2}$$

in which $C_{\mathrm{cell}}$ (F g$^{-1}$) is the specific capacitance, $I$ (A) is the discharge current, $\mathrm{d}t$ (s) is the discharge time, $m$ (g) is the total mass of negative and positive electrodes, and $\mathrm{d}V$ (V) is the potential window after IR drop. The energy density ($E$, Wh kg$^{-1}$) and the power density ($P$, W kg$^{-1}$) for the two-electrode cell can be obtained using the following equations:

$$E = 0.5 \, C_{\mathrm{cell}} \, V^2 \, / \, 3.6 \tag{3}$$

$$P = 3600 \, E / \mathrm{d}t \tag{4}$$

in which $C_{\mathrm{cell}}$ (F g$^{-1}$) is the specific capacitance of the two-electrode cell, $V$ (V) is the potential window after IR drop, and d$t$ (s) is the discharge time.
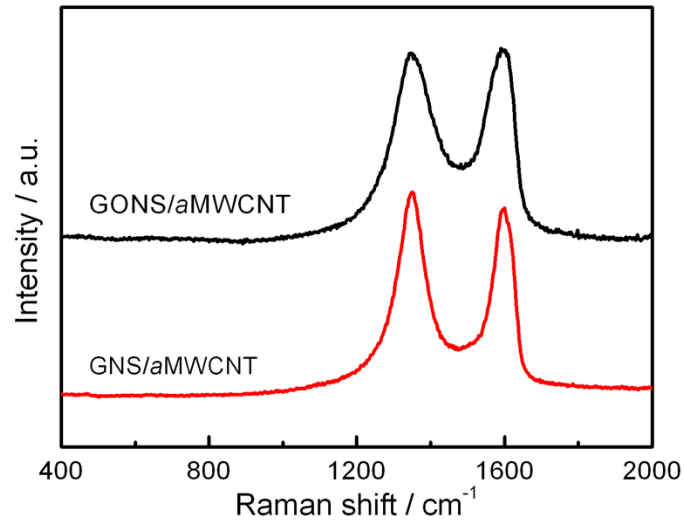
## Results and discussion



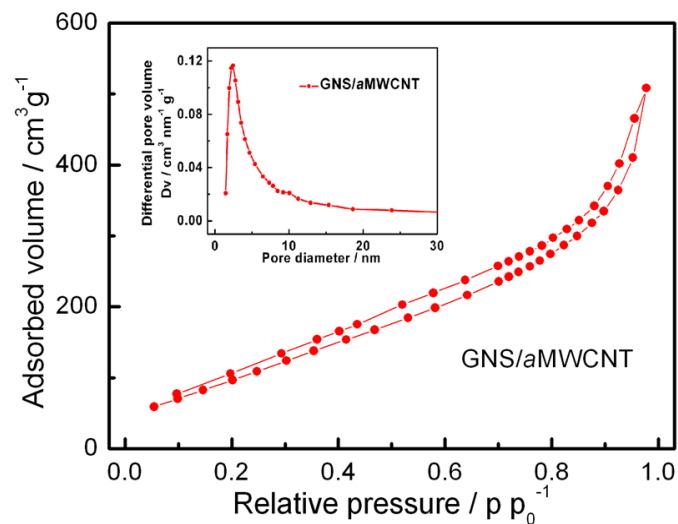**Fig. S1** Raman spectra of GONS/$a$MWCNT and GNS/$a$MWCNT.



**Fig. S2** N$_2$ adsorption/desorption isotherm of GNS/$a$MWCNT. The inset is the corresponding pore-size distribution.
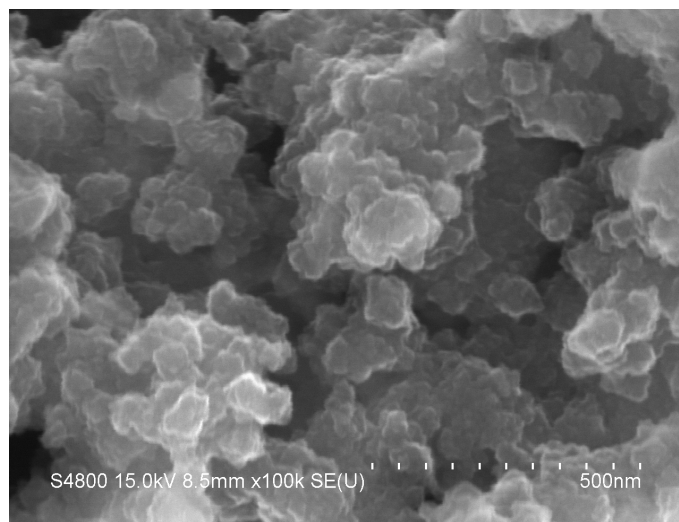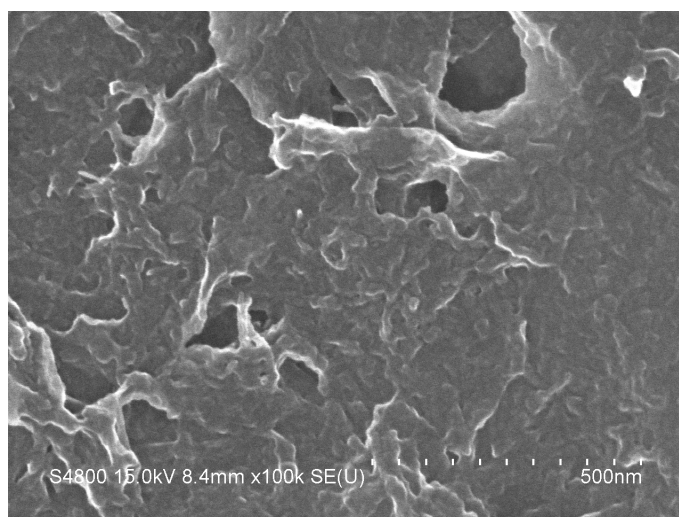
**Fig. S3** FE-SEM image of pure PDAA.



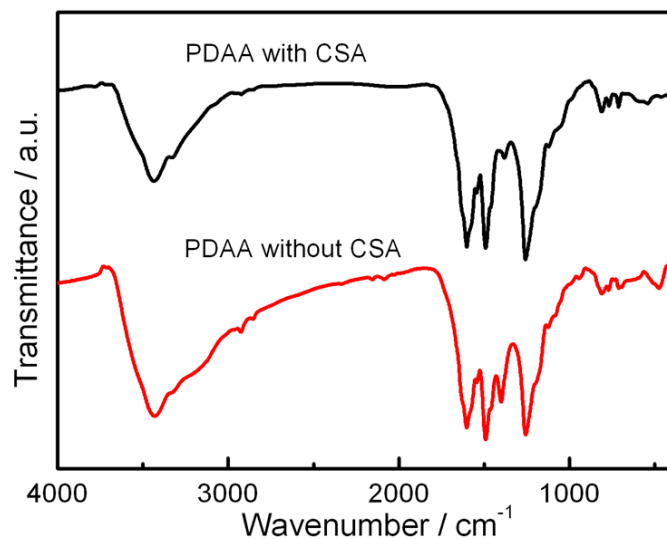**Fig. S4** FE-SEM image of GNS/*a*MWCNT@PDAA without CSA.

**Fig. S5** FTIR spectra of PDAA doped with CSA and PDAA without CSA.

**Tab. S1** XPS results of PDAA and GNS/*a*MWCNT@PDAA for element content, bond assignment and so on.

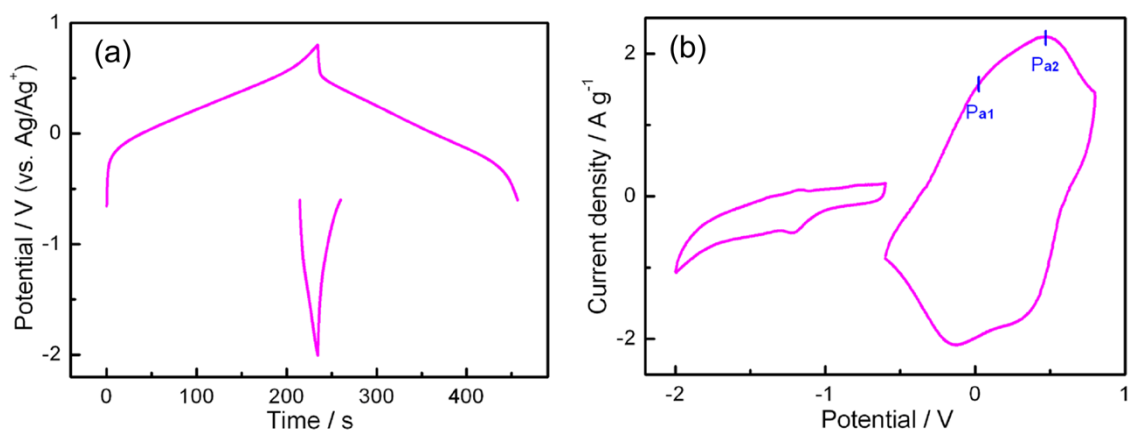| Samples | Element content (%) | | | Bond assignment (%) | | | | $(=N^+ + -N^+)$ / $N_{total}$ (%) | $=N-$ / $-NH-$ (%) |
|---|---|---|---|---|---|---|---|---|---|
| | C | N | O | =N– 398.9eV | –NH– 399.5eV | =N$^+$– 400.2eV | –N$^+$– 401.2eV | | |
| PDAA | 70.0 | 11.3 | 18.7 | 31.0 | 43.3 | 18.6 | 7.1 | 0.26 | 0.72 |
| GNS/*a*MWCNT@PDAA | 72.1 | 8.3 | 19.6 | 31.2 | 34.0 | 26.1 | 8.7 | 0.35 | 0.92 |

**Fig. S6 (**a) Comparative charge/discharge curves at a current density of 1 A g$^{-1}$ and (b) comparative CV curves at a scan rate of 10 mV s$^{-1}$ for GNS/*a*MWCNT@PANI recorded by a three-electrode system in half potential window for the negative and positive part in 1 M Et$_4$NBF$_4$-AN electrolyte.
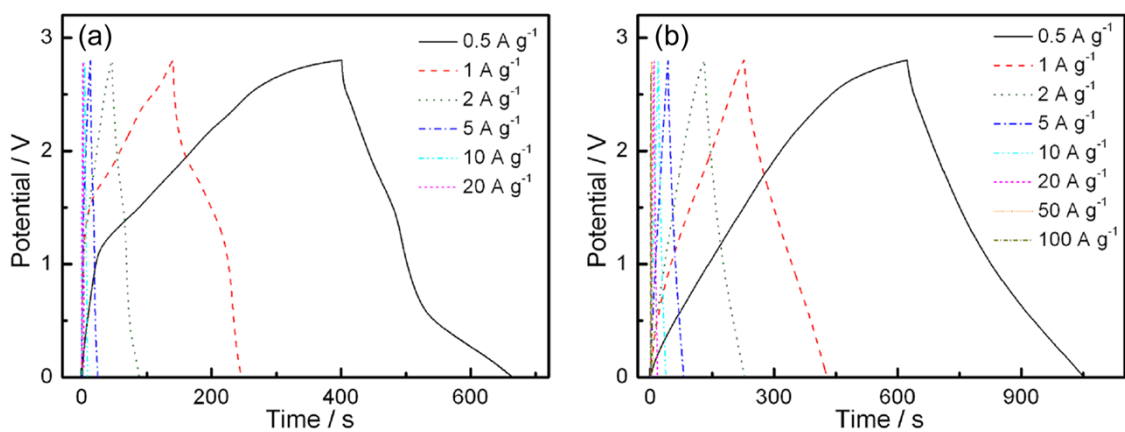


**Fig. S7** Galvanostatic charge/discharge curves of (a) PDAA and (b) GNS/*a*MWCNT@PDAA at different current densities ranging from 0.5 A g$^{-1}$ to 100 A g$^{-1}$.
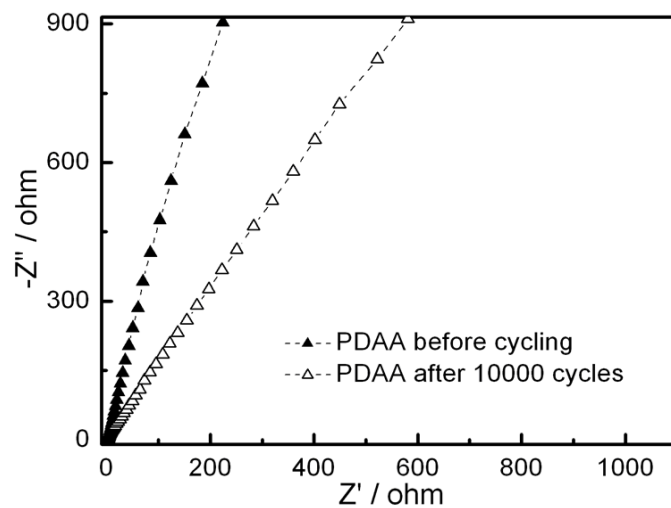
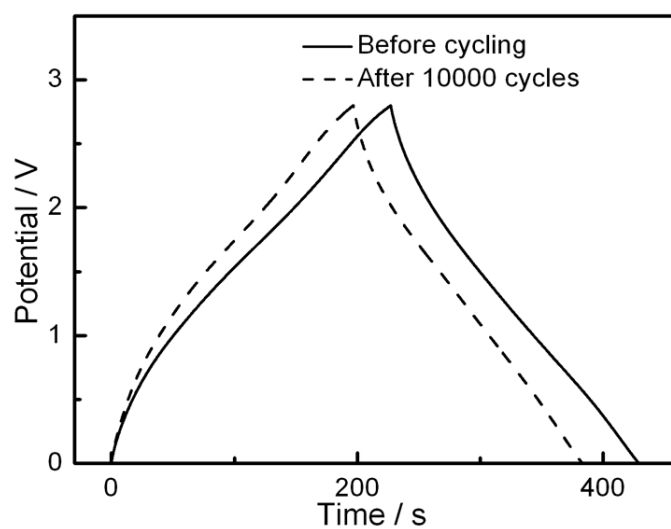**Fig. S8** Nyquist plots of PDAA before cycling and after 10000 cycles.



**Fig. S9** Galvanostatic charge/discharge curves of GNS/*a*MWCNT@PDAA before cycling and after 10000 cycles at a current density of 1 A g$^{-1}$.