

## *Supplementary material*

### **Novel and high-performance asymmetric micro-supercapacitors based on graphene quantum dots and polyaniline nanofibers**

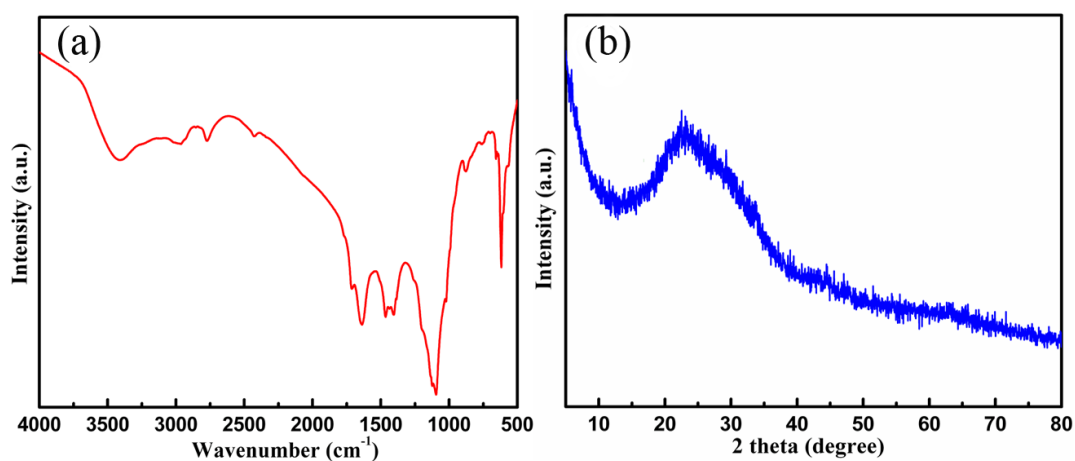
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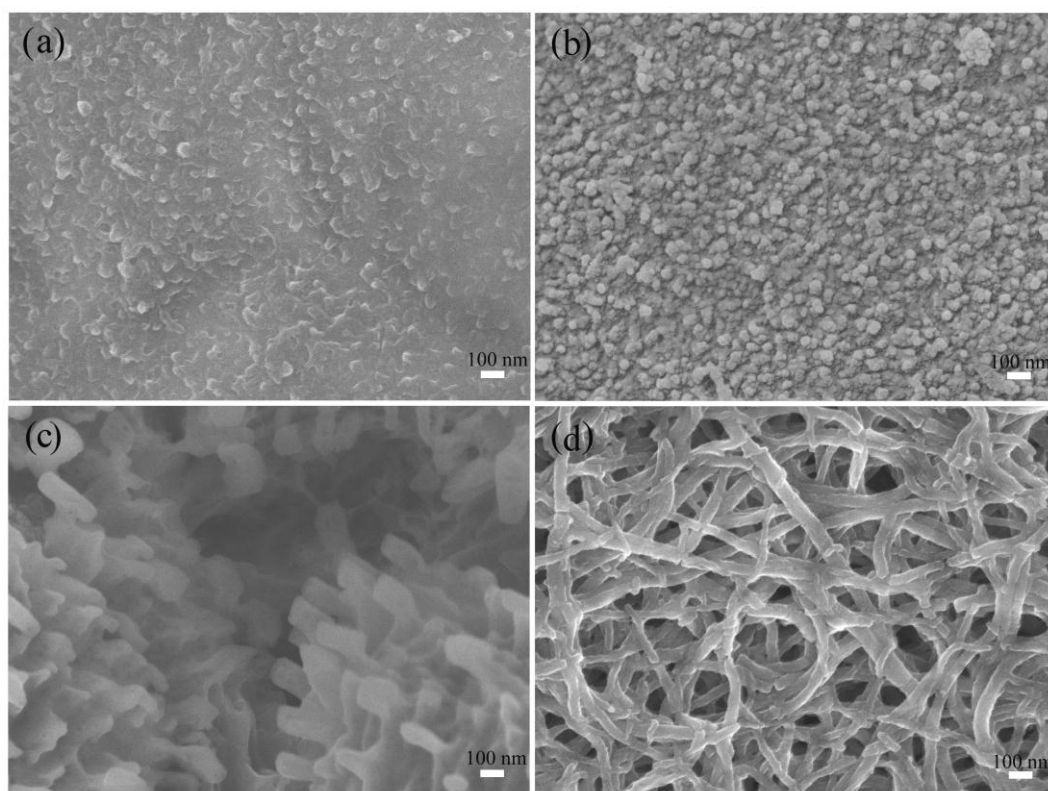
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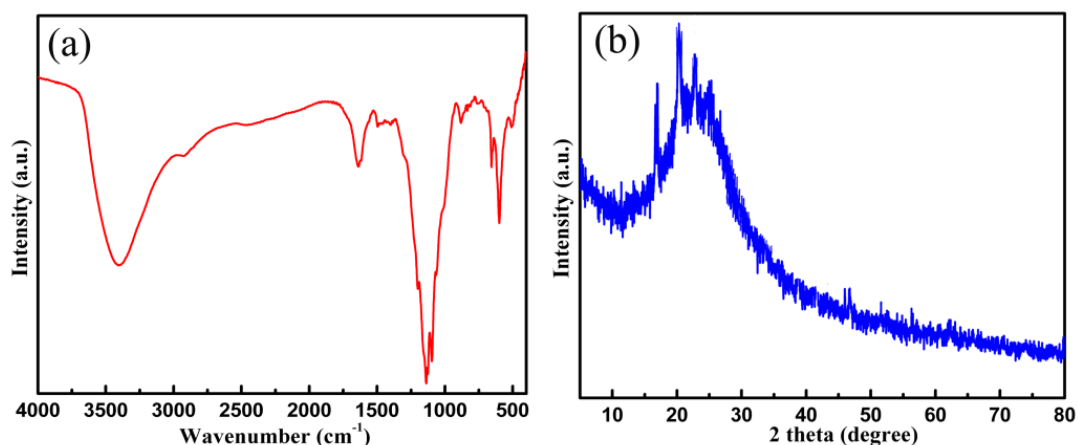


**Fig. S1** (a) FTIR and (b) XRD spectra of as-made GQDs.

In the FTIR spectrum, the broad band centered at 3392 cm<sup>-1</sup> represents free O–H bonding. The bands at 1360 and 2927 cm<sup>-1</sup> reveal the presence of C–H bonding. Furthermore, the bands at around 1641 and 1417 cm<sup>-1</sup> are correlated to C=C and C=O bonds, respectively. In addition, the bands centered at 1027 and 1076 cm<sup>-1</sup> represent the C–O group. The XRD pattern of the GQDs exhibits a broad (002) peak corresponding to the interlayer spacing of 0.374 nm, which is larger than the interlayer of graphite (0.334 nm). It is due to the disordered stacking of some GQDs.

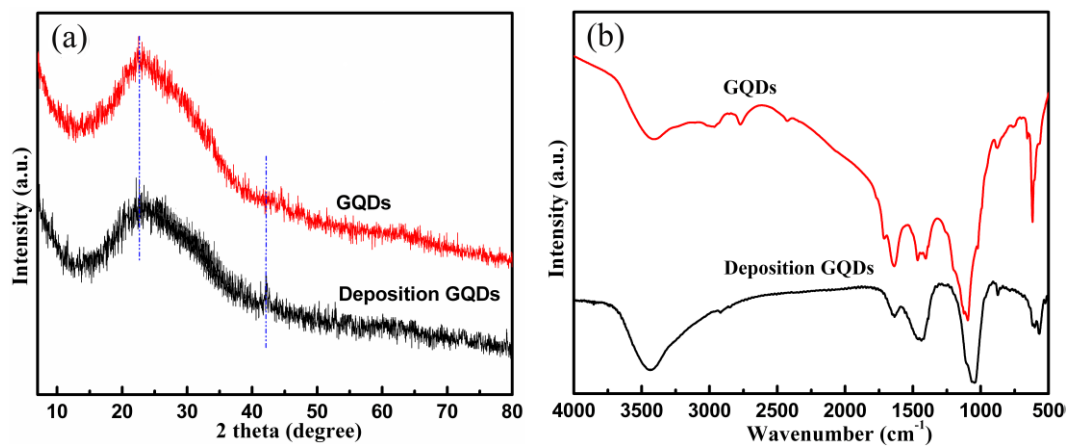


**Fig. S2** SEM images of the surface topography of PANI coatings electro-deposited in solution of 0.1 M aniline (pH=5) with the current density of  $1 \text{ mA}\cdot\text{cm}^{-2}$  for different times: (d) 1 min , (e) 5 min, (f) 10 min, and (g) 15 min.



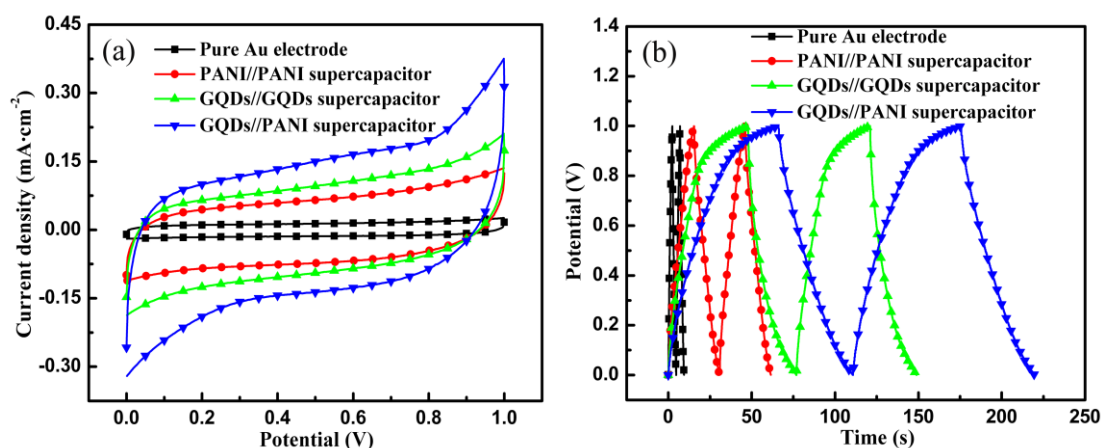
**Fig. S3** (a) FTIR spectrum and (b) XRD pattern of electrochemical deposited PANI nanofibers.

In the FTIR spectrum, the broad band centered about 3440 cm<sup>-1</sup> corresponds to the free N–H stretching vibration of primary (–NH<sub>2</sub>) and secondary amine (–NH–) groups. The peak at 1584 cm<sup>-1</sup> is characteristic of the quinone diimine ring-stretching deformation, while the 1500 cm<sup>-1</sup> band indicates benzenoid diamine ring-stretching. The 1300 cm<sup>-1</sup> band is assigned to the C–N stretching of the secondary aromatic amine. The peaks at 1246 cm<sup>-1</sup> and 1144 cm<sup>-1</sup> correspond to C–N stretching (–N–benzenoid–N–) and C=N stretching (–N=quinoid=N–), respectively. The bands at 772 cm<sup>-1</sup> and 1024 cm<sup>-1</sup> are assigned to aromatic C–H out of plane bending vibration and aromatic C–H in plane bending vibration, respectively. In the XRD pattern, four peaks centered at 16.9 °, 20.5 °, 25.3 ° and 27.2 °, are characteristic for nanocrystalline PANI. The peak at 16.9 ° is attributed to parallel repeat units of PANI. The peaks at 20.5 ° and 25.3 ° are usually ascribed to the periodicity parallel and perpendicular to the polymer chains of PANI, as well as the peak at 27.2 ° is assigned to a periodicity caused by H-bonding between PANI chains.



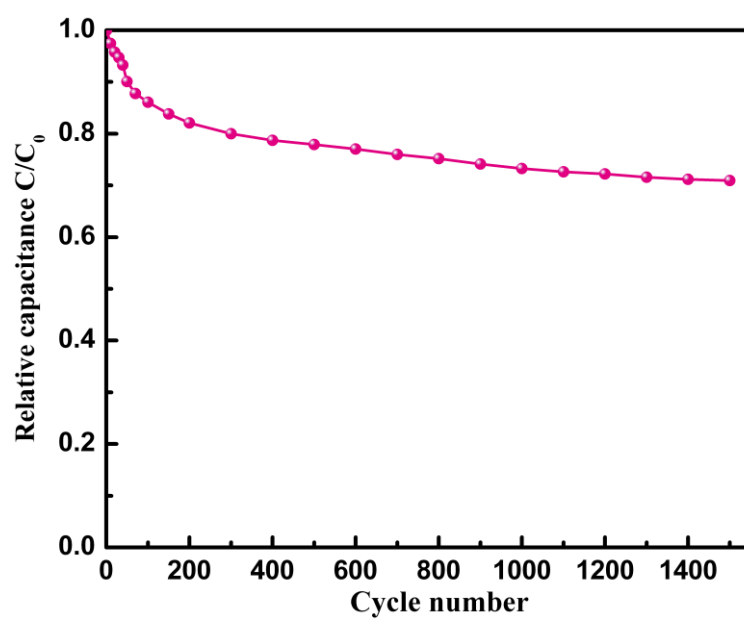
**Fig. S4** The structure change of GQDs after the electro-deposition: (a) XRD patterns and (b) FT-IR spectra.

It can be found that the electro-deposition has negligible influence on the intrinsic crystallinity and carbon-carbon conjugated backbones of GQDs.



**Fig. S5** (a) CV curves and (b) the charge/discharge curves of bare interdigital finagle Au electrodes, symmetric PANI//PANI, symmetric GQDs//GQDs and asymmetric GQDs//PANI micro-supercapacitors in 0.5 M Na<sub>2</sub>SO<sub>4</sub> electrolyte at the scan rate of 1 V·s<sup>-1</sup> and at the current densities of 15 μA·cm<sup>-2</sup>, respectively.

It is clear seen that the specific capacitance of pure Au interdigital finger electrode is negligibly small and does not contribute much to the total capacitance. Also, it indicates that GQDs//PANI asymmetric micro-supercapacitor possesses much better supercapacitive properties (higher specific capacitance and higher energy density) compared with symmetric GQDs//GQDs micro-supercapacitor and symmetric PANI//PANI micro-supercapacitor.



**Figure S6** Cycle stability of PANI//PANI symmetric micro-supercapacitor in 0.5 M  $\text{Na}_2\text{SO}_4$  electrolyte at the scan rate of  $1 \text{ V}\cdot\text{s}^{-1}$ .