

## Supporting Information

### Three Dimensional (3D) Flexible Graphene Foam/Polypyrrole Composite: Towards Highly Efficient Supercapacitors

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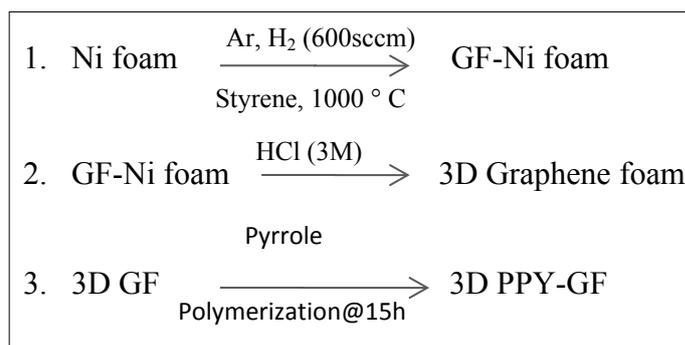


Figure S1: Schematic processes for the preparation of the 3D GF and the 3D PPY-GF composite electrodes.

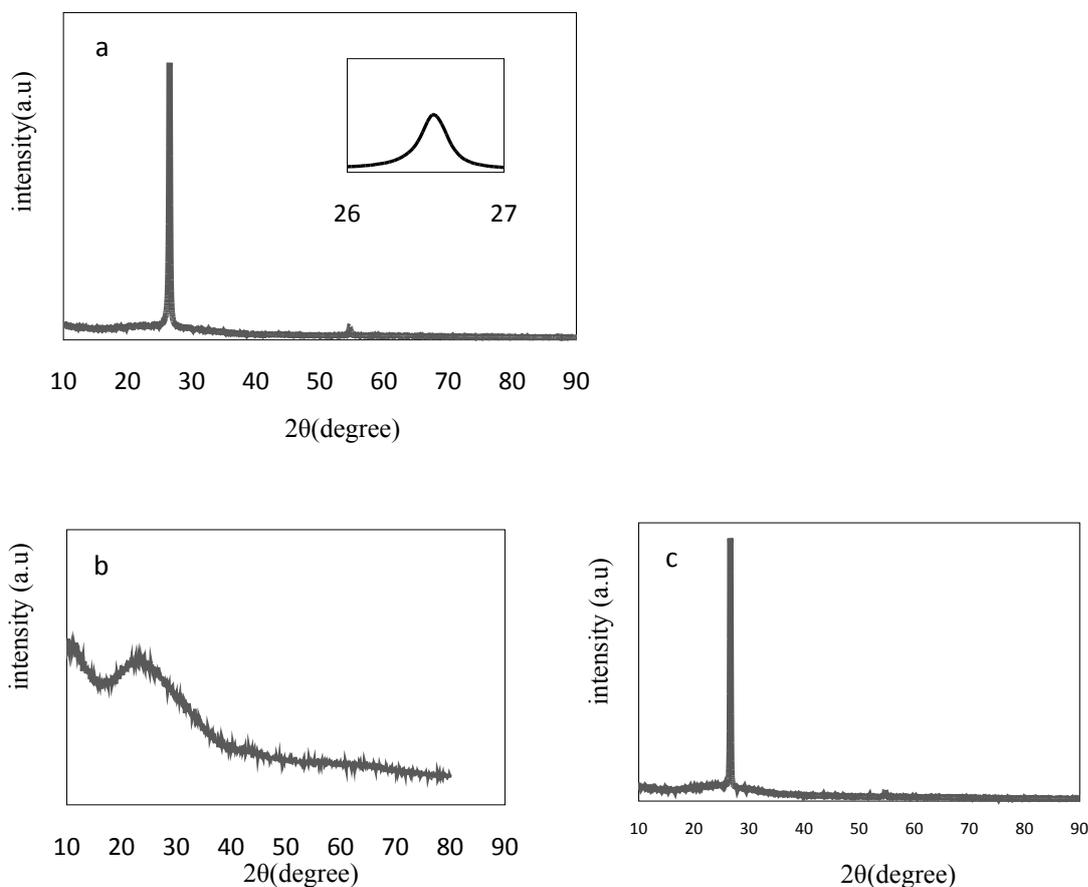


Figure S2: XRD profiles of: (a) GF, (b) PPY, and (c) PPY-GF.

Figure S2a shows the XRD profile of the GF powder sample. The sharp peak at  $26.512^\circ$  represents the (002) plane of graphene, and the weak peak at *ca.*  $55^\circ$  belongs to the (004) plane. The XRD pattern of the PPY exhibits one broad peak at about  $21^\circ$ , representing an amorphous structure of the film. The sharp peak at  $26.52^\circ$  for the PPY-GF composite belongs to the (002) plane of the graphene structure.

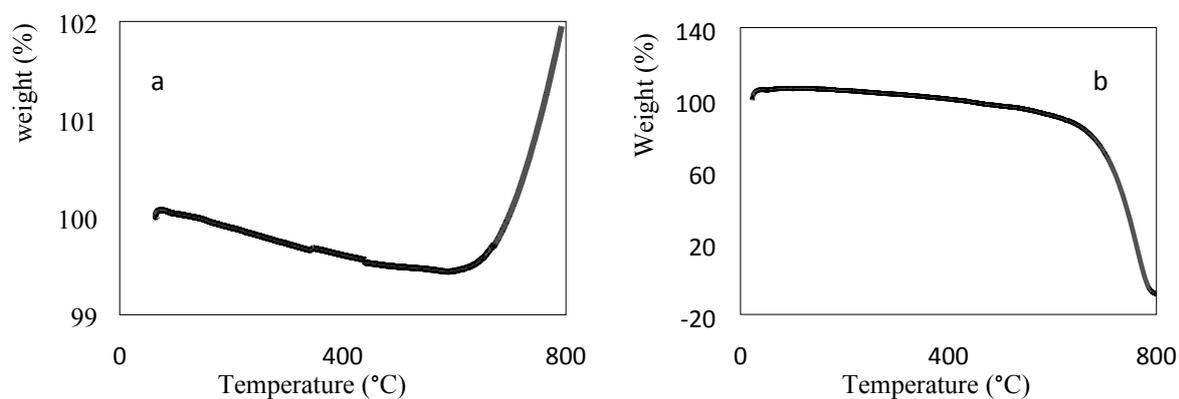


Figure S3: TGA profiles of the GF-Ni (a) and pure GF (b).

Comparing the TGA results of GF-Ni (Figure S3a) with GF (Figure S3b), we have found that the GF accounts for less than 1 wt.% of the GF-Ni composite, and the 100% weight loss for the GF further confirmed the high purity of our GFs. In Figure S3 b, there are two weight loss regions: a minor loss occurred before  $600^\circ\text{C}$ , which was related to the removal of adsorbed water (at about  $100^\circ\text{C}$ ) and oxygen decomposition at higher temperatures; a major loss appeared between  $600\text{-}750^\circ\text{C}$ , owing to the complete oxidation of carbon.

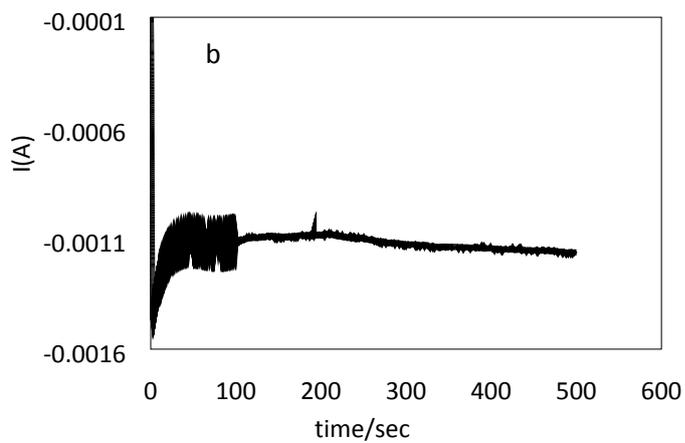
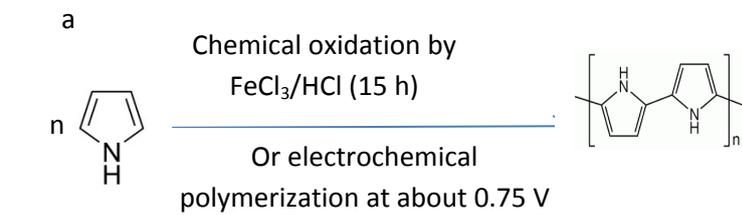


Figure S4: Schematic polymerization of pyrrole (a), and the chronoamperometry result of PPY in the potential windows of 0-0.8 V (b).

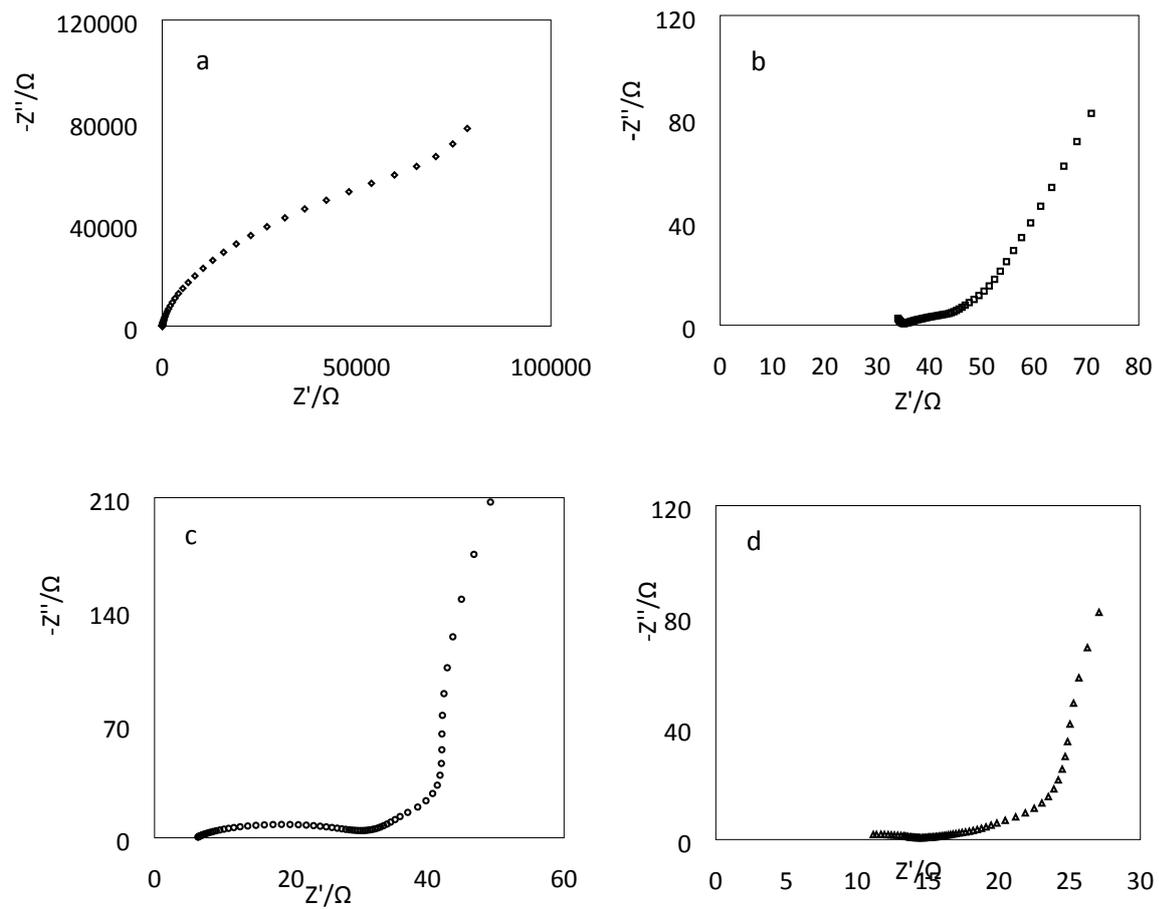


Figure S5: Nyquist plots of different electrodes: (a) GF, (b) PPY, (c) PPY-GF in 0.5 M KCl, and (d) PPY-GF in 0.5 M H<sub>2</sub>SO<sub>4</sub>.

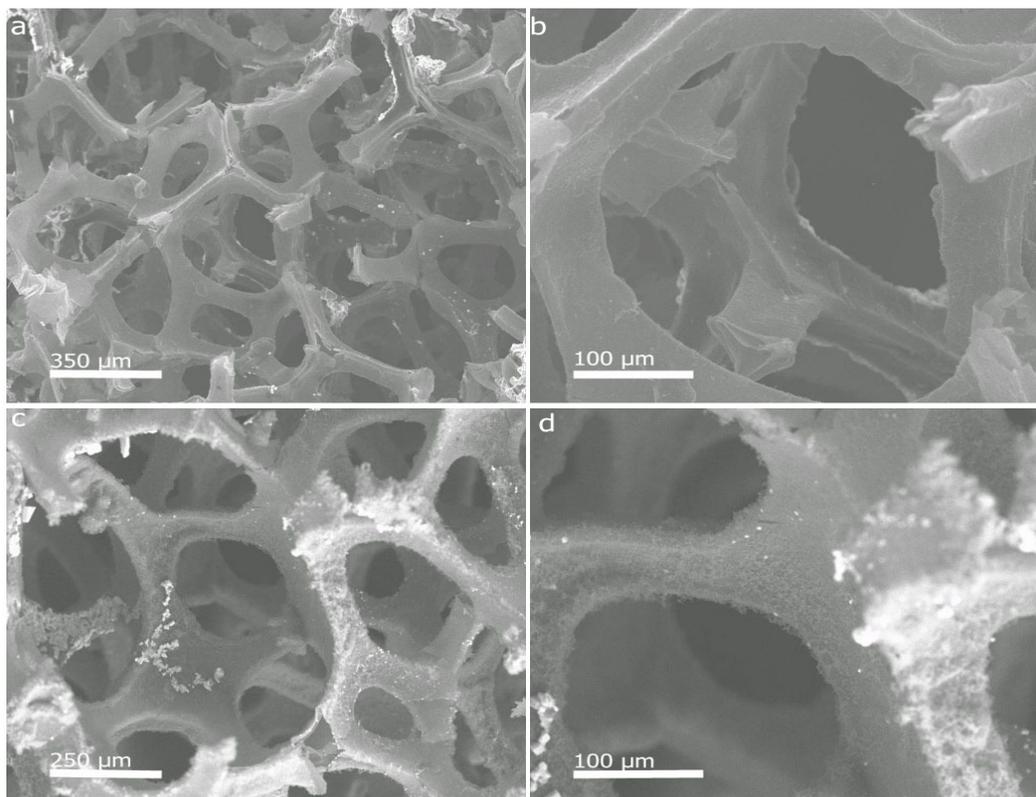


Figure S6: (a, b) SEM images of the GF after 10,000 cycles of CV test, and (c, d ) SEM images of PPY-GF after 6,000 cycles of CV test.

Figure S6 shows the SEM images of GF after 10,000 and PPY-GF after 6,000 cycles of CV test. Both electrodes almost kept the same network structures as prior to the tests. White spots on the image are electrolyte ions. As seen from the image, there are some broken struts on the surface of GF, which we believe were mainly formed during the handling, such as the separation from electrochemistry cell after the CV test, and during the SEM sample preparation.