Electronic supplementary information

Rational Design of Nano-architecture Composite Hydrogel Electrode towards High Performance Zn-ions Hybrid Cell

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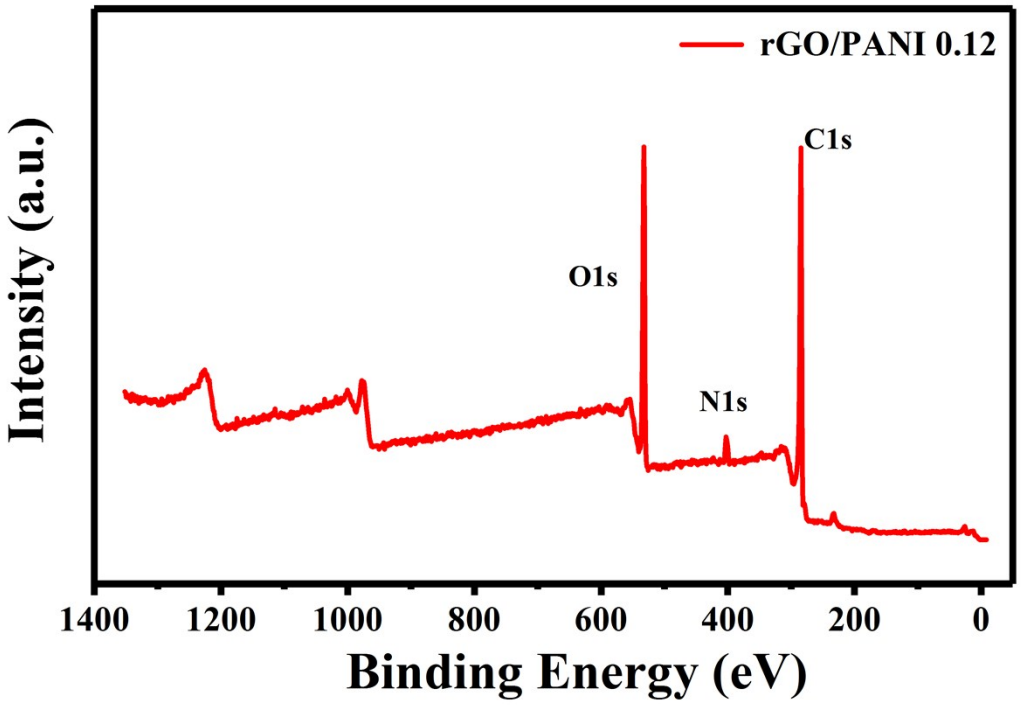

Figure S1. The XPS survey spectrum of Freeze-dried 3D graphene@PANI-0.12M hydrogel.

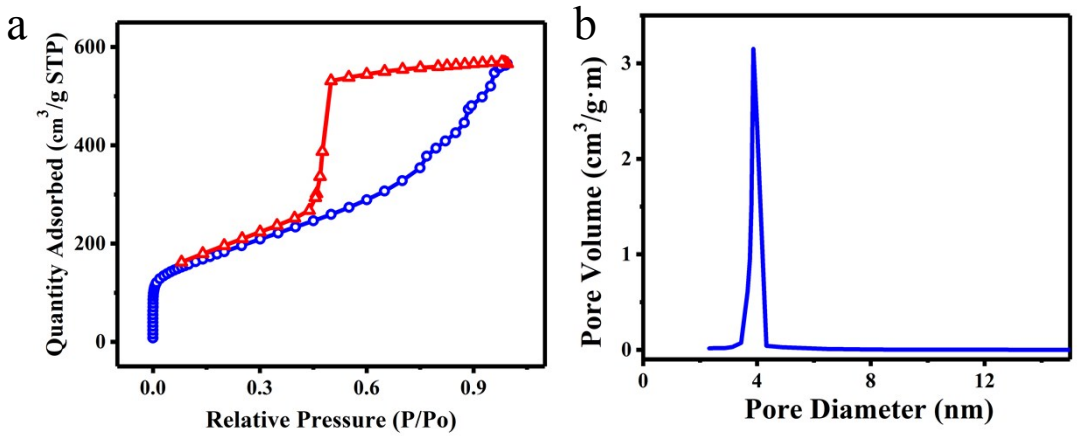

Figure S2. N_2 adsorption/desorpotion measurements: (a) N_2 adsorption-desorption isotherm of the freeze-dried composite hydrogel exhibits a type IV curve with a H4 hysteresis loop. (b) Pores size distribution indicates the composite hydrogel includes large amount of mesopores.

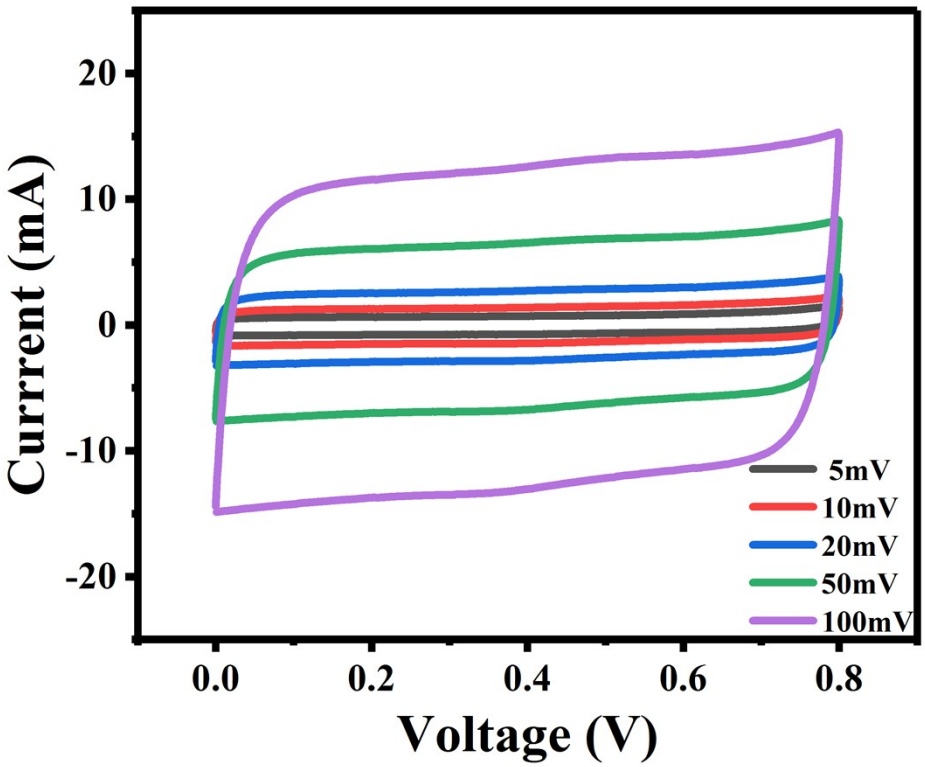

Figure S3. The CV curves of 3D graphene hydrogel film electrodes based supercapacitor

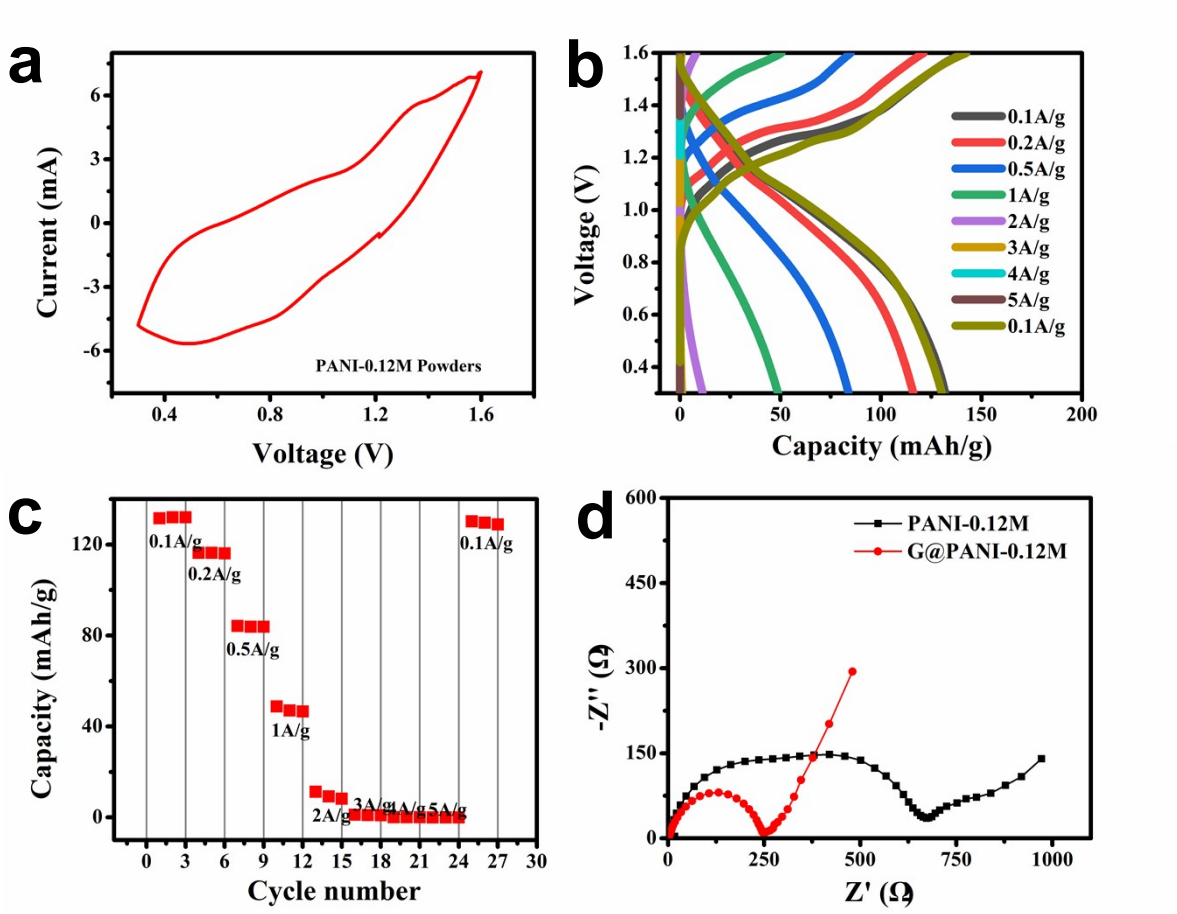

Figure S4. For Zinc-ion battery with conventional PANI cathode: (a) CV curves of PANI at the scan rate of 1 mV/s. (b) Voltage profiles curves at the different current densities. (c) Discharge capacity of different current. (d) EIS plots of pure PANI cathode and 3D graphene hydrogel film cathode based Zn-ions hybrid cells.

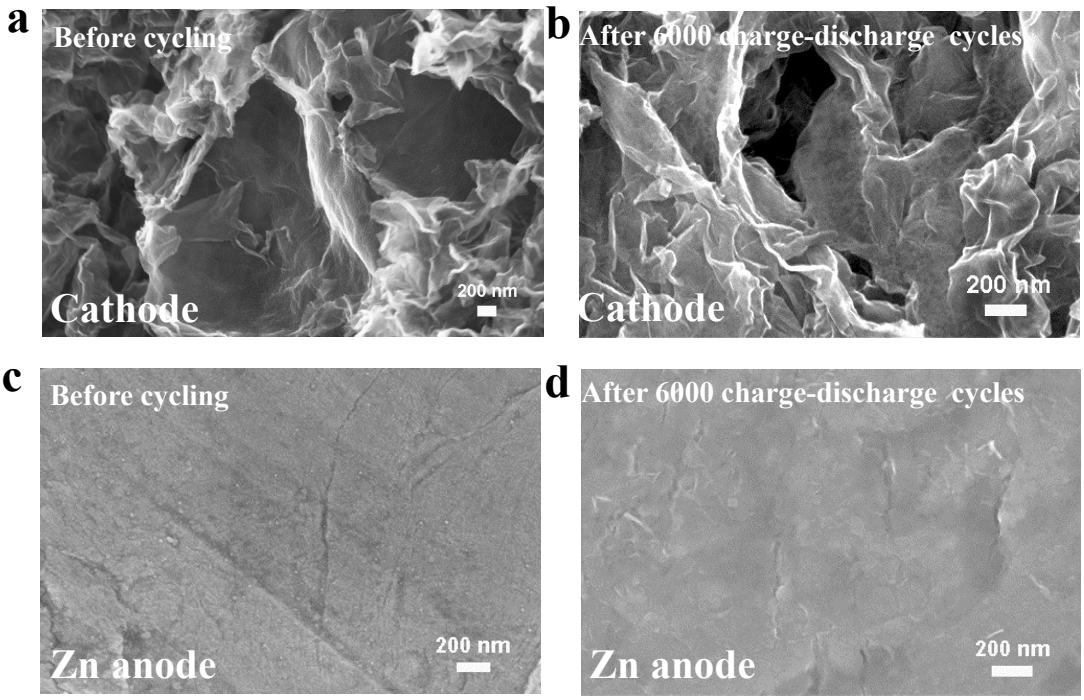

Figure S5. The morphology evolution of the (a) cathode and (b)anode. There is no obvious change for composite hydrogel cathode material before and after 6000 charge-discharge cycles. For Zn anode, the surface of Zn foil becomes smooth and a few Zn flakes present after cycling, which may be resulted from the repeated Zn stripping/plating cycles. However, no large amount of Zn dendritic were observed on the Zn foil surface based on our experiments, representing that Zn dendritic is not a serious issue in this Zn hybrid cell with mild electrolyte.

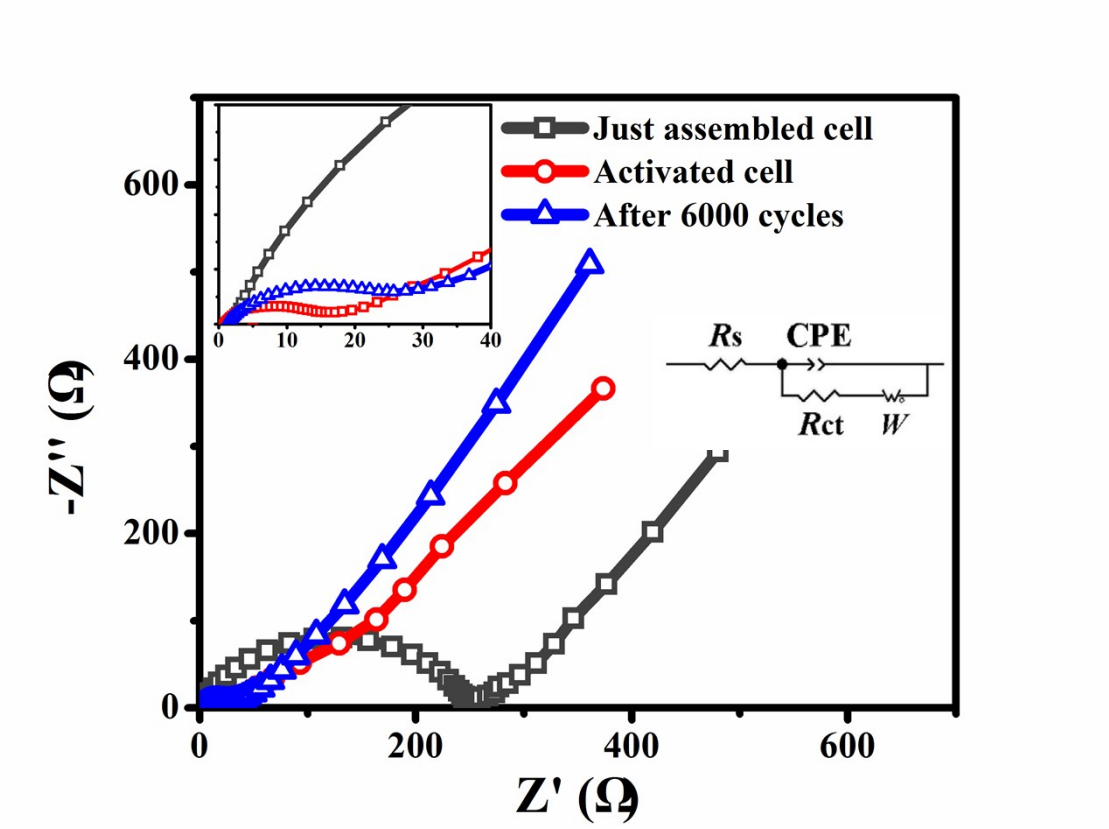

Figure S6. EIS plots of the Zn-ion hybrid cell before and after 6000 charge-discharge cycles. The semicircles represent the charge transfer resistance (Rct) and the diagonal lines represent the solid state diffusion resistance. The x-intersection at high-frequency region of Nyquist plots is the solution resistance (Rs) of the cell, which is around 1Ω and shows no clearly change before and after cycling. This indicates that the ionic conductivity of the solution is constant during long term cycling, and no seriously irreversible electrolyte depleting involved in the charge-discharge process. For the freshly assembled cell (before electrochemical charge-discharge), the charge transfer resistance (Rct) is 219Ω, which largely decreases (18Ω) after electrochemical activation by several

charge-discharge cycles resulted from the formation of stable solid-liquid interface. The Rct becomes around 26Ω after 6000 charge-discharge cycles, which is relative to the electrode reaction process. The increase of Rct indicates the physical and chemical properties of the cathode and anode has changed a little bit after long-term cycles, which is also consistence with that the cell capacity shows a slightly decrease after 6000 cycles.