

## Supplementary Material

### **Synchronously improved graphitization and surface area in 3D porous carbon network as high capacity anode material for lithium/sodium-ion batteries**

Haimi Liu<sup>a,b</sup>, Weihao Zeng<sup>a</sup>, Yue Yang<sup>a</sup>, Junxin Chen<sup>a</sup>, Yuwei Zhao<sup>a</sup>, Shichun Mu<sup>\*a,b</sup>

<sup>a</sup>State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, P. R. China.

<sup>b</sup>Foshan Xianhu Laboratory of the Advanced Energy Science and Technology Guangdong Laboratory, Xianhu hydrogen Valley, Foshan 528200, China.

\* Corresponding author E-mail: [msc@whut.edu.cn](mailto:msc@whut.edu.cn)

### **Synthesis of porous-NPC:**

PANi aerogel was prepared in the presence of phytic acid by oxidative polymerization according to a published procedure. Typically, 5 mL phytic acid (70%, Aladdin) was added into 15 mL deionized water, followed by the addition of 0.6 g of SiO<sub>2</sub> nanoparticles (100 nm) into this solution. After stirring at the room temperature for 20 minute, 5 mL of aniline (99%, Aladdin) was added into this solution. 0.9 g ammonium persulphate (APS, 99%, Aladdin) was dissolved in 10 mL deionized water under stirring at room temperature. The above two solutions were mixed together and kept stewing for 12 h at 4 °C. The resultant hydrogel was washed by plenty of deionized water for two days then freeze-dried for 24 h to produce PANi aerogel for pyrolysis. To prepare porous-NPC foam, the PANi aerogel was annealed at 950 °C for 2 h under Ar atmosphere. The obtained powder was added into moderate HF solution (30% wt, Aladdin) under stirring for 24 h to remove the SiO<sub>2</sub> template. The reaction product was collected by centrifugation, thoroughly washed by deionized water for several times, then freeze-dried for 24 h to obtain the final product. For comparison, SiO<sub>2</sub> nanoparticles was removed from the phytic acid solution and without HF washing, the N, P co-doped carbon (NPC) foam was then prepared by annealing the obtained PANi aerogel at 950 °C for 2 h under Ar atmosphere.

### **Battery assembly and electrochemical measurements:**

A slurry of the active material, Super P, and polyvinylidene fluoride (PVDF) are mixed at the weight ratio of 7:2:1. N-Methyl-2-pyrrolidone (NMP, 99.5%, Aladdin) was

introduction to adjust the viscosity of the slurry. The electrodes were then coated on a copper current collector with a spreader of 120  $\mu\text{m}$ , and transferred to a vacuum oven at 70  $^{\circ}\text{C}$  for 12 h. LIBs used lithium as reference electrodes and 1 M  $\text{LiPF}_6$  in a mixture of ethylene carbonate/diethyl carbonate (EC/DEC, 1:1 by volume) as the electrolyte, and Celgard13501 (Celgard) as the separator. SIBs were also fabricated at the same conditions assembling with sodium as the reference electrodes, 1 M  $\text{NaPF}_6$  in a mixture of ethylene carbonate/diethyl carbonate (EC/DEC, 1:1 by volume) as the electrolyte, and glass microfiber (Whatman) as the separator. The electrochemical performance was measured on the Neware Battery Measurement System with a potential window of 0.01-3 V, the cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were conducted on the CHI660E electrochemical workstation.

#### **Materials characterization:**

The morphology of the sample was characterized by scanning electron microscopy (SEM) on a HITACHI S-4800 and transmission electron microscopy (TEM) on a FEI Tacnai G2 with the accelerating voltage of 200 kV. The crystalline structure of the sample was performed on X-ray diffraction (XRD) with Cu radiation of Dmax 2500 V. X-ray photoelectron spectroscopy (XPS) scans were performed on an ESCALAB 250 photoelectron spectrometer. Raman spectra was collected on Lab RAM HR800. The Brunner-Emmet-Teller (BET) surface area of the sample was measured on Micromeritics ASAP 2020 analyzer. The thermogravimetric curve (TG) of the sample was performed by STA449F3.

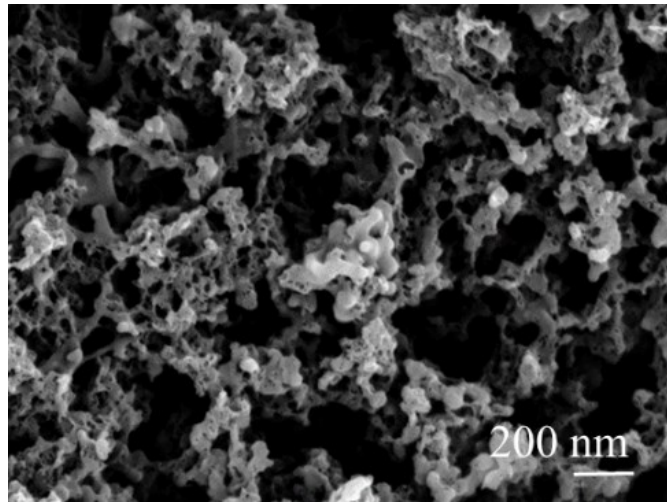


Figure S1. Low-magnification SEM image for porous-NPC.

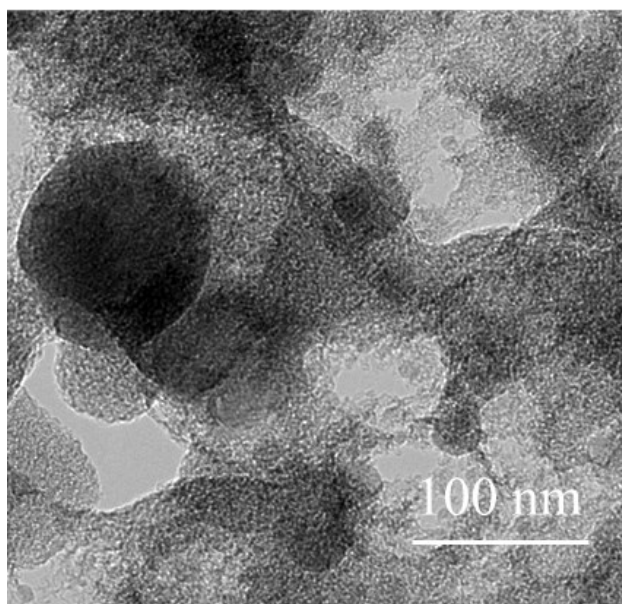


Figure S2. High-magnification TEM image for NPC

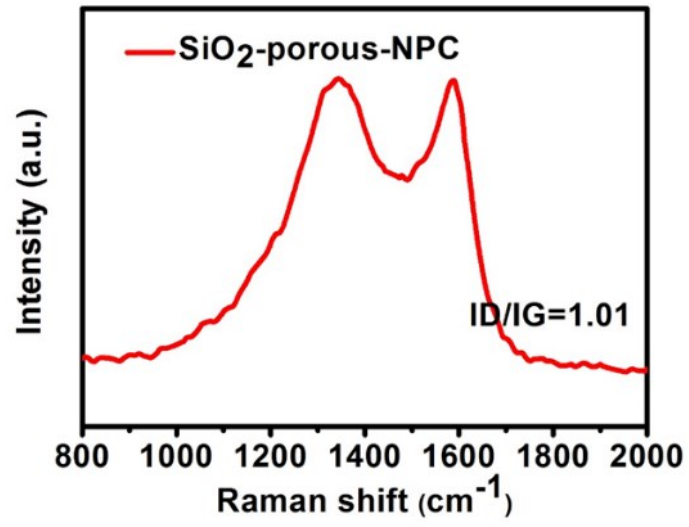


Figure S3. Raman spectra of the SiO<sub>2</sub>-porous-NPC.

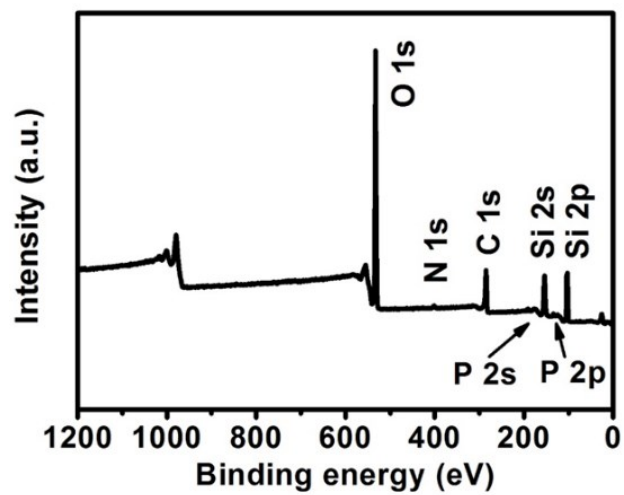


Figure S4. XPS survey spectrum of the SiO<sub>2</sub>-porous-NPC.

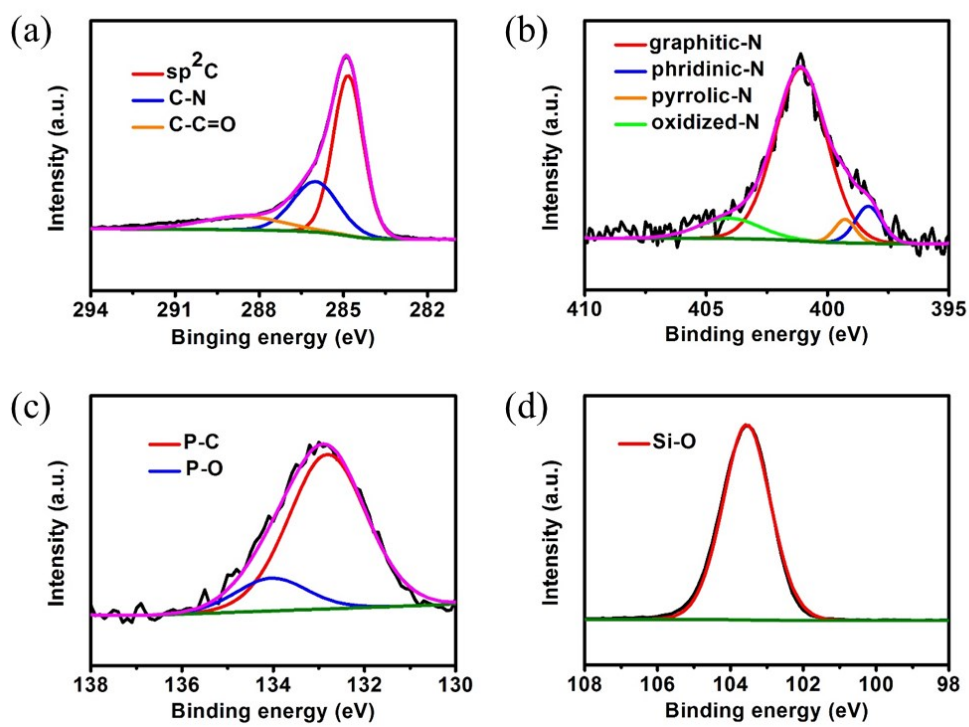


Figure S5. XPS high-resolution spectra of (a) C 1s, (b) N 1s, (c) P 2p, and (d) Si 2p of SiO<sub>2</sub>-porous-NPC.



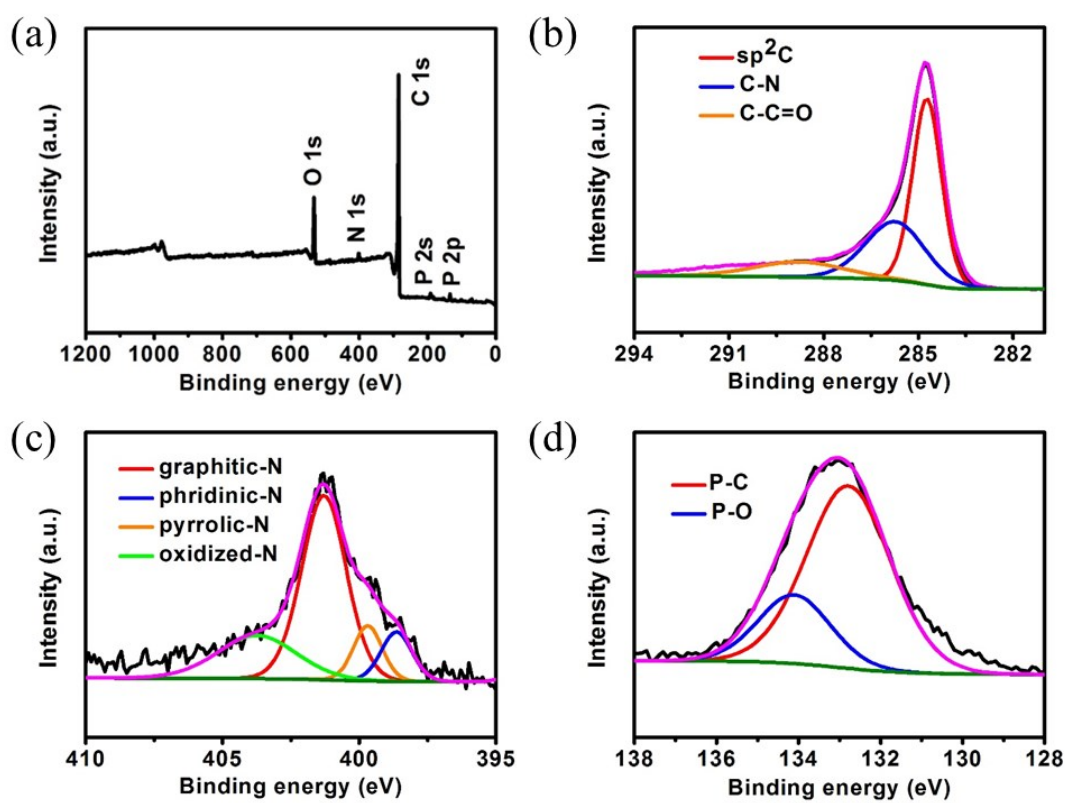


Figure S6. XPS high-resolution spectra of (a) C 1s, (b) N 1s, and (c) P 2p of NPC.

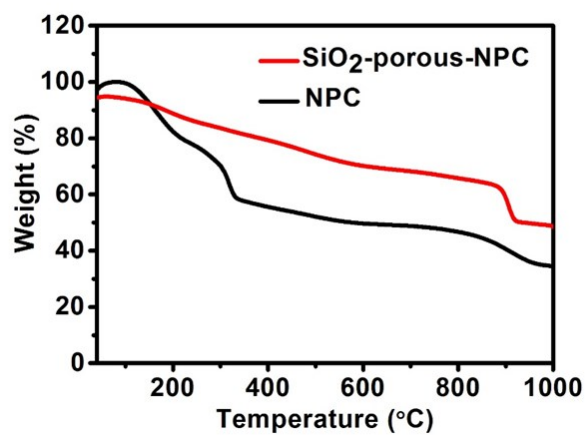


Figure S7. TG curves of the SiO<sub>2</sub>-porous-NPC and NPC precursor.

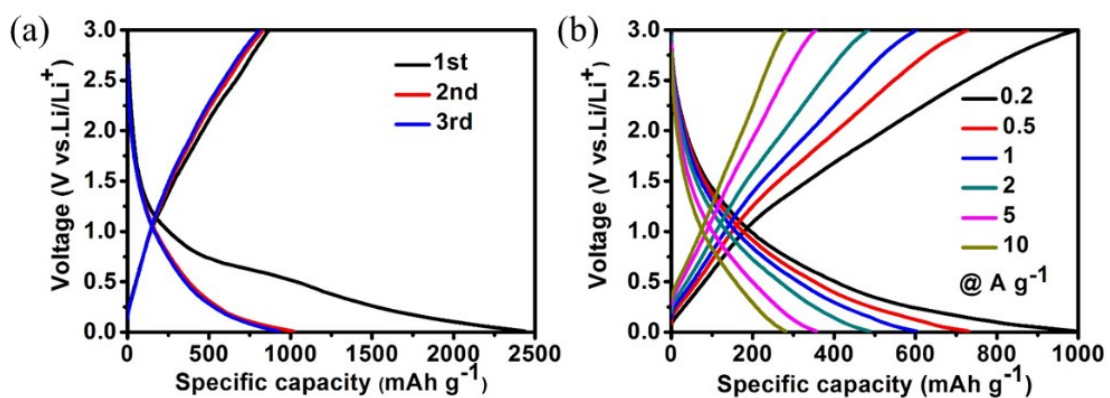


Figure S8. Electrochemical performance of the porous-NPC electrode for LIBs: (a) Charge-discharge voltage profiles at 1 A g<sup>-1</sup>, and (b) Charge-discharge voltage profiles at various current densities.

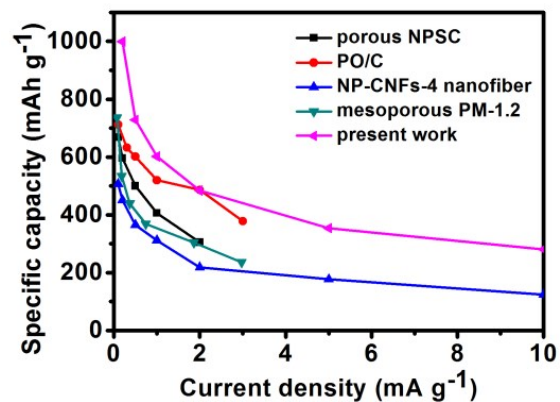


Figure S9. The comparison of electrochemical performances for the porous-NPC electrode with other reported results for LIBs.

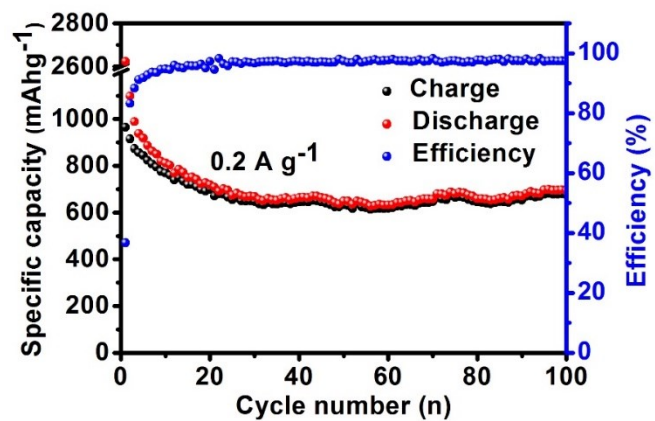


Figure S10. Long-term cycling performance of the porous-NPC electrode at a current of  $0.2 \text{ A g}^{-1}$  for LIBs.

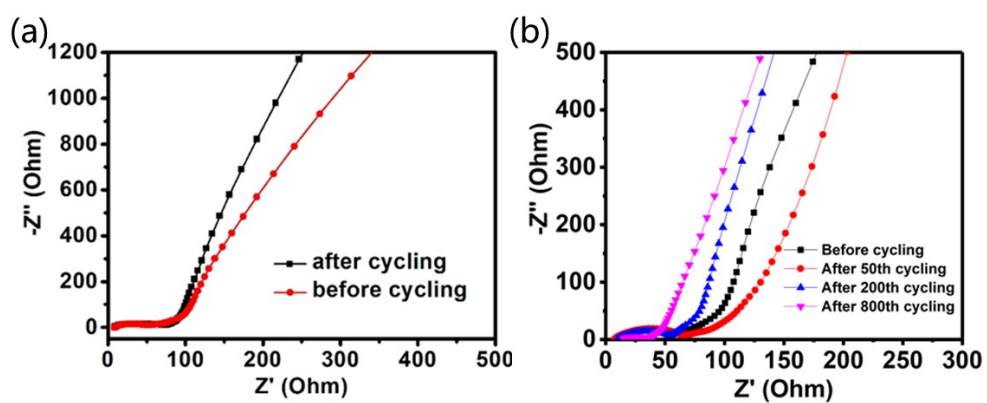


Figure S11. Nyquist plots of the porous-NPC electrode (a) before and after the cycle for LIBs, (b) after circulating different cycles for LIBs.

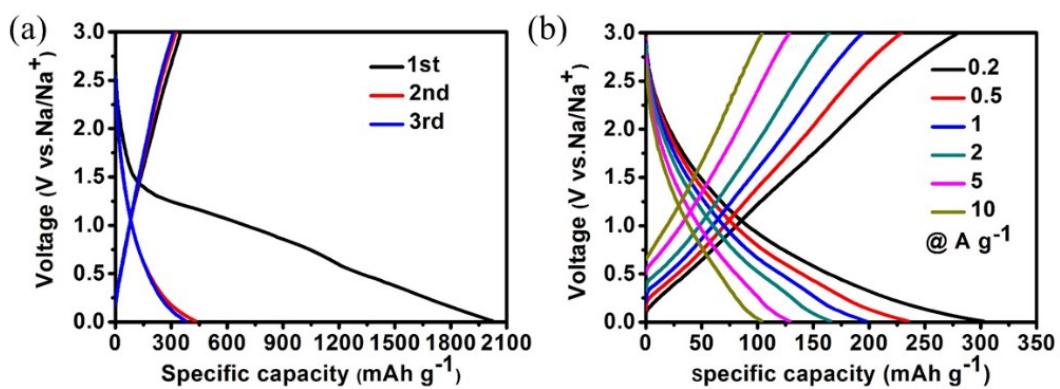


Figure S12. Electrochemical performance of the porous-NPC electrode for SIBs: (a) Charge-discharge voltage profiles at 1 A g<sup>-1</sup>, and (b) Charge-discharge voltage profiles at various current densities.

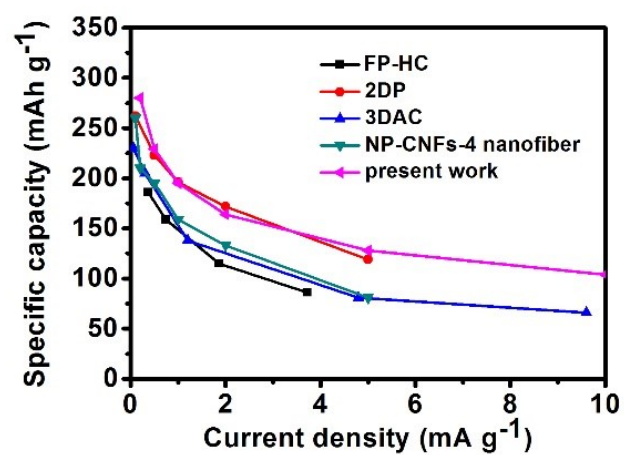


Figure S13. The comparison of electrochemical performances for the porous-NPC electrode with other reported results for SIBs.



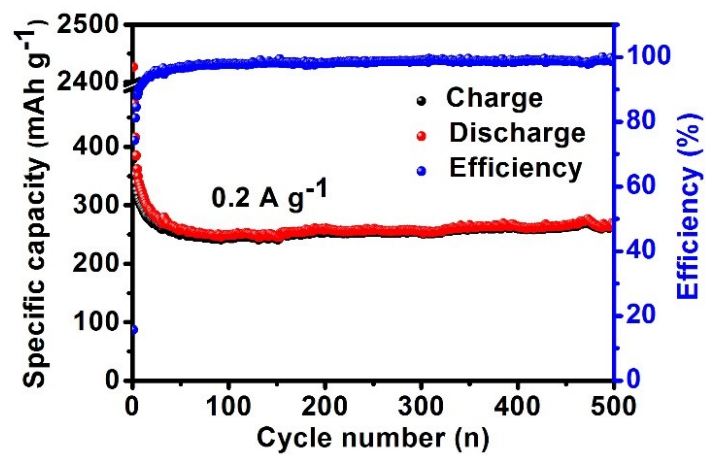


Figure S14. Long-term cycling performance of the porous-NPC electrode at a current of 0.2 A g<sup>-1</sup> for SIBs.

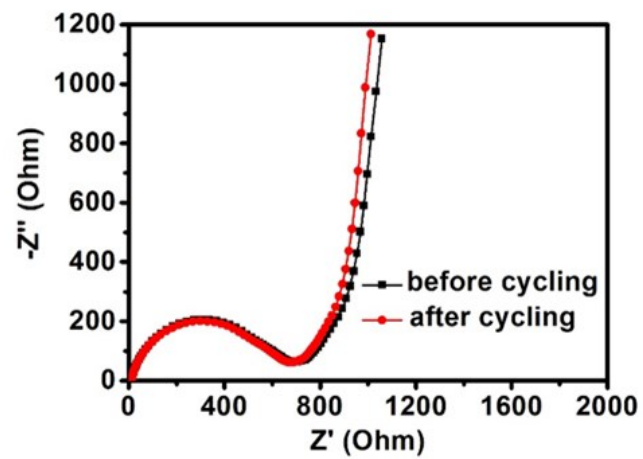


Figure S15. Nyquist plots of the porous-NPC electrode before and after the cycle for SIBs.