Supporting information

Endowing CuTCNQ a New Role: a High Capacity Cathode for K-ion Batteries

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Experimental details

Synthesis of CuTCNQ

7,7,8,8-tetracyanoquinodimethane (TCNQ) and acetonitrile were purchased from Aladdin Incorporation and were used as received. CuTCNQ films were prepared according to the reported procedures and the powders were obtained by scraping the CuTCNQ film surface.[1]

Characterization of CuTCNQ

The X-ray diffraction measurement (XRD, X'Pert Pro MPD) was carried out by using Cu Kα radiation (λ=1.54056Å) with the scanning rate of 0.03^{o} s^{-1} in the 2θ range of 5-80^{o}. The Fourier transform infrared spectrometer (FT-IR, Shimadzu, IR Prestige-21) test was operated with the wave number range of 400-4000 cm^{-1}. The morphology was observed by field-emission scanning electron microscopy (FE-SEM, Hitachi, S3400N). The X-ray photoelectron spectroscopy (XPS, Kratos XSAM800) tests were carried out by using Al-Kα (1486.6 eV, 150 W) radiation as the primary excitation source.

Electrochemical characterization

The cathode (~2 mg cm^{-2}) was prepared by casting the slurry onto a clean Al foil, where the slurry electrode was composed of CuTCNQ (60 wt%), Super P (30 wt%) and carboxymethyl cellulose (CMC) as the binder (10 wt%). The coin cells (CR2032) were assembled in a glove box filled with Ar (99.999%). The reference/counter electrode was metallic K foil and the separator was glass fiber. A carbon interlayer was inserted between cathode and glass fiber, which was prepared by casting the slurry (Super P and poly(vinylidenefluoride) with the mass ratio of 3:1) onto the Celgard poly(propylene) (PP) separator.[2] A modified interlayer was composed of the

mixture of carboxylic multi-walled carbon nanotube and graphene oxide (CNT/GO, the weight ratio is 4:1) and poly(vinylidenefluoride) (with the mass ratio of 3:1). The mass and thickness of the interlayers were same, ~ 1.5 mg cm^{-2} and ~ 35 μm, respectively. The electrolyte was 1 M KPF_6 in ethylene carbonate (EC) /propylene carbonate (PC) (1:1 v/v). The electrochemical tests were carried on a CT2001A instrument with the voltage range of 2-4.1 V (*vs.* K^+/K). Cyclic voltammograms (CVs) were tested on a CHI 660C electrochemical workstation with a scan rate of 0.1 mV s^{-1}. The mass of CuTCNQ electrode used for *ex-situ* measurement was ~ 5 mg cm^{-2}.

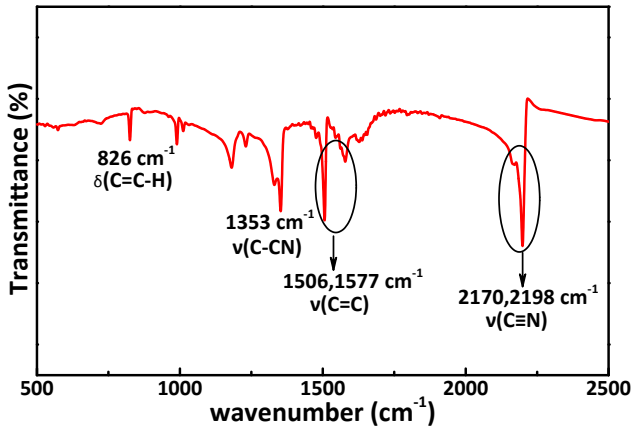

Fig. S1 The IR spectrum for the obtained CuTCNQ sample.

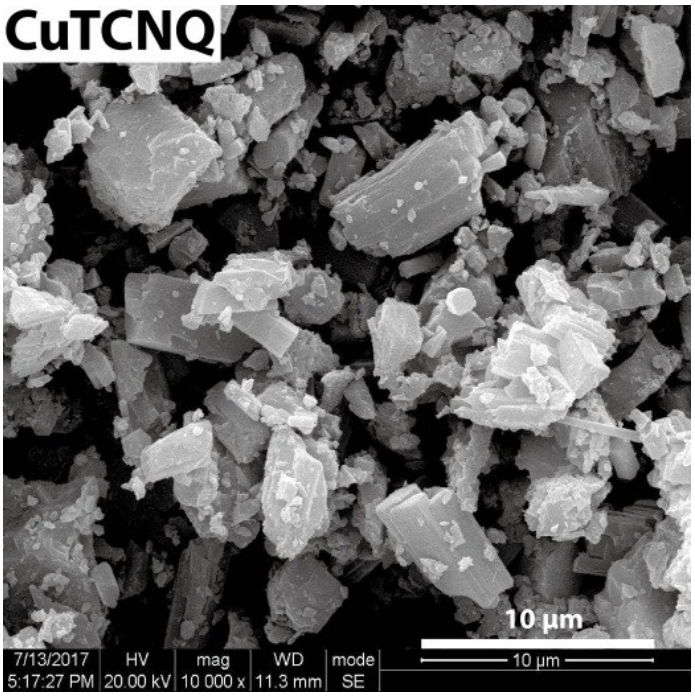

Fig. S2 The SEM image for the obtained CuTCNQ sample.

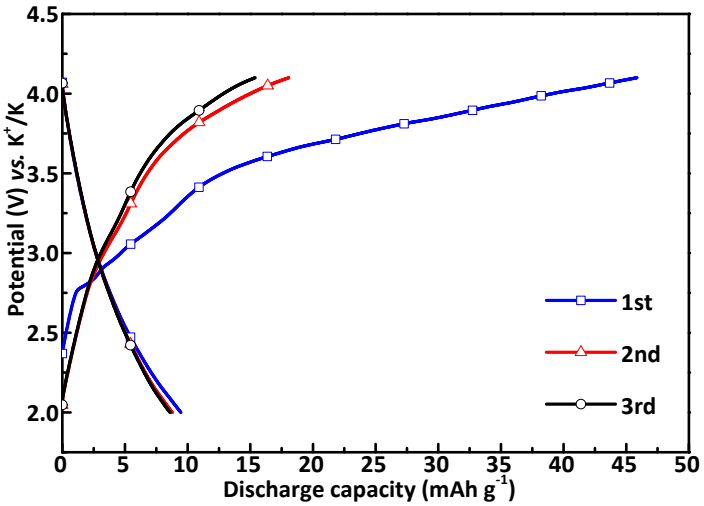

Fig. S3 The initial three charge-discharge profiles for neat Super P. The carbon interlayer is composed of Super P. The current density is 20 mA g^{-1} and the voltage window is 2-4.1 V.

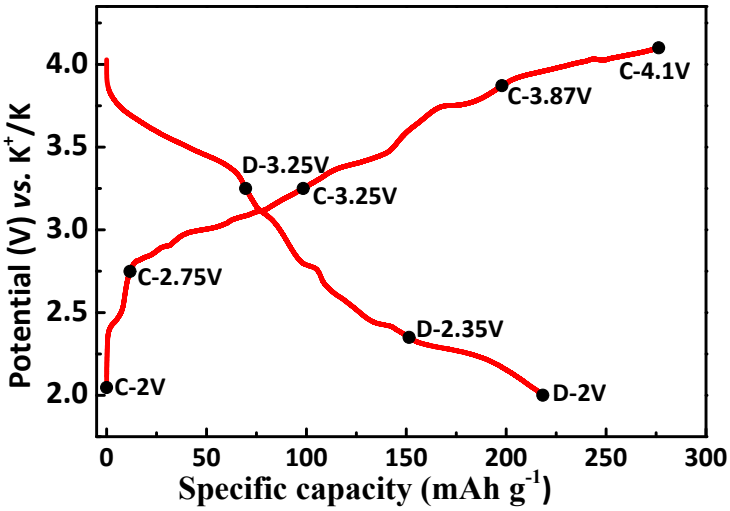

Fig. S4 The selected points during the 2nd charge-discharge cycle for IR measurement.

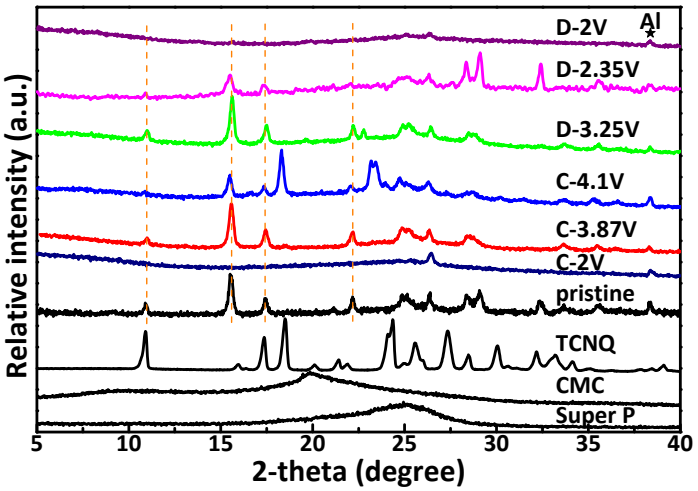

Fig. S5 The *ex-situ* XRD curves of CuTCNQ at different states during the 2nd cycle.

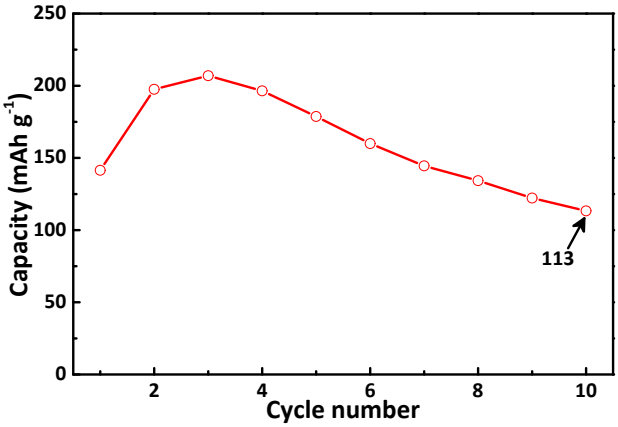

Fig. S6 The cycling performance of CuTCNQ in PIBs with Super P as the carbon interlayer. The current density is 50 mA g^{-1} and the voltage window is 2-4.1 V.

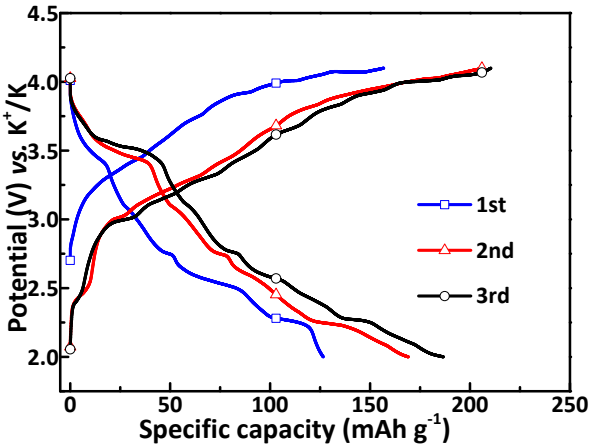

Fig. S7 The initial three galvanostatic charge-discharge curves of CuTCNQ. The carbon interlayer is the mixture of carboxylic multi-walled carbon nanotube and graphene oxide (CNT/GO). The current density is 50 mA g^{-1} and the voltage window is 2-4.1 V.

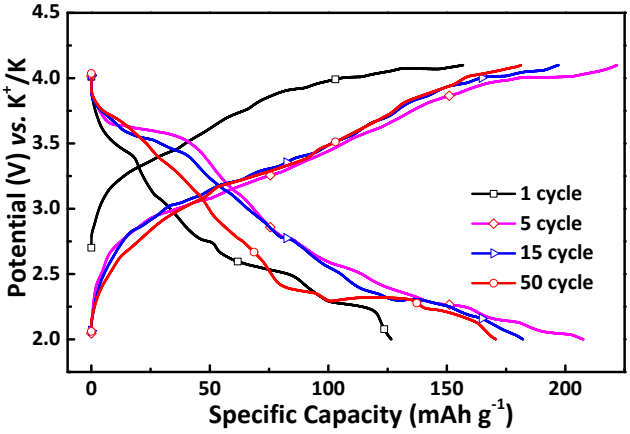

Fig. S8 The charge-discharge curves of CuTCNQ in different cycles. The carbon interlayer is CNT/GO. The current density is 50 mA g^{-1} and the voltage window is 2-4.1 V.

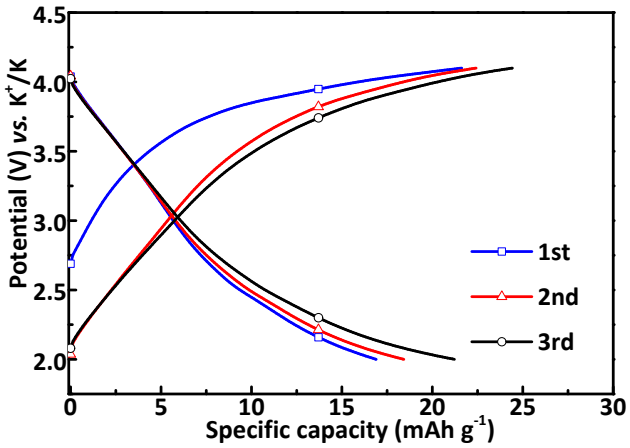

Fig. S9 The initial three charge-discharge profiles for neat Super P. The carbon interlayer is CNT/GO. The current density is 50 mA g^{-1} and the voltage window is 2-4.1 V.

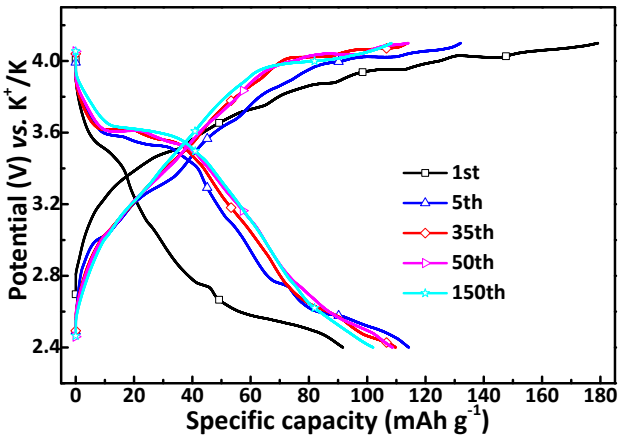

Fig. S10 The charge-discharge curves of CuTCNQ in different cycles. The carbon interlayer is CNT/GO. The current density is 50 mA g^{-1} and the voltage window is 2.4-4.1 V.

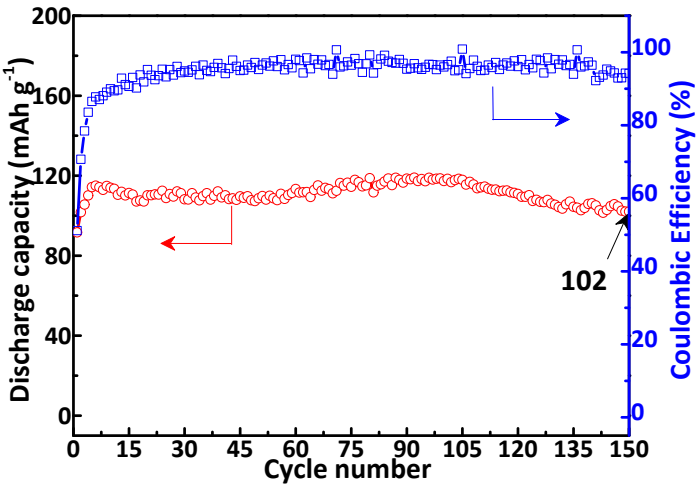

Fig. S11 The long cycling performance of CuTCNQ in PIBs. The carbon interlayer is carboxylic CNT/GO. The current density is 50 mA g^{-1} and the voltage window is 2.4-4.1 V.

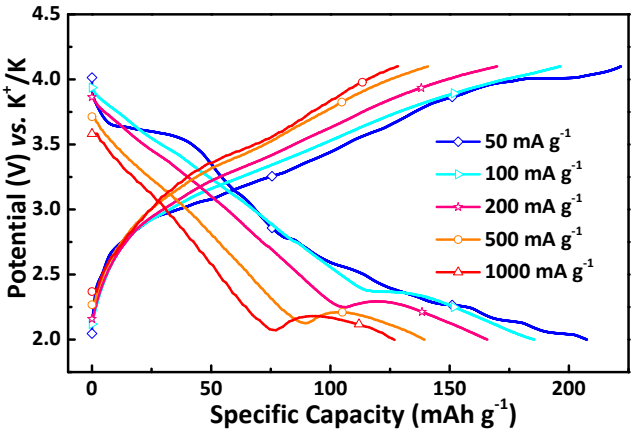

Fig. S12 The charge-discharge profiles of CuTCNQ at different rates. The carbon interlayer is CNT/GO and the voltage window is 2-4.1 V.

References

1. R. A. Heintz, H. H. Zhao, O. Y. Xiang, G. Grandinetti, J. Cowen and K. R. Dunbar, *Inorg. Chem.*, 1999, **38**, 144-156.
2. Y.-S. Su and A. Manthiram, *Nat. Commun.*, 2012, **3**, 1-6.