

Electronic Supplementary Information

In-situ growth of copper rhodizonate complexes on reduced graphene oxide for high performance organic lithium-ion batteries

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1. Experimental section

1.1 Synthesis of CPR and CPR/rGO composites: Disodium rhodizonate (240 mg, > 97%)

without further purification was dissolved into deionization water (15 mL) under stirring. Then excess $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ (268 mg, > 98%) was mixed with GO aqueous suspension (20 mL, 2 mg/mL; 15 mL, 2 mg/mL; 12 mL, 2 mg/mL) under ultrasonic dispersion for 20 min, and GO suspension was prepared through the modified hummer's method.¹ The excess $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ in the water will produce excess copper ions. On the one hand, excess copper ions absorbed on the surfaces of the GO are beneficial for uniform distribution of CPR in the GO sheets; on the other hand, excess copper ions could control the molar ratio of copper ions to rhodizonate ligands in the CPR. Then, the above-mentioned two suspensions were mixed well under stirring for 10 min and added into the hydrothermal synthesis reactor (50 mL). The hydrothermal reaction was carried out at 180 °C for 12 h. Then, the product was obtained by filtration and

washed with water. Finally, the product was dried in a vacuum oven at 80 °C for 24 h. The CPR was prepared similarly without GO suspension in the same way. The pure rGO was prepared similarly without CPR, and different CPR/rGO composites with different rGO contents were prepared in the same way.

1.2 Materials characterization: The Fourier transform infrared spectra of the products were tested by a VERTE 70 (Bruker Company, Germany) in the wavenumber range from 400 to 4000 cm^{-1} at room temperature. The SEM and TEM images of the products were tested by scanning electron microscopy (SEM, FEI, Sirion 200) and transmission electron microscopy (TEM, JEOL 2100F), respectively. The phase of the products was tested by powder X-ray diffraction (XRD, PANalytical B.V., Holland). The thermogravimetric (TG) test of the products was conducted by Pyris1 TGA (PerkinElmer Instruments) in air from room temperature to 800 °C (10 °C min^{-1}). X-ray photoelectron spectroscopy (XPS) test of the products was carried out on a VG Multilab 2000 system using a monochromatic Al $K\alpha$ radiation (Thermo VG Scientific). AAS test was conducted on an atomic absorption spectrometer (Thermo, ICE 3000).

1.3 Electrochemical measurements: The CPR and CPR/rGO electrodes were prepared by mixing 60 wt% active materials, 10 wt% PVDF (dissolved in N-methyl-2-pyrrolidone), and 30 wt% Super P with a mortar. Besides, The pure rGO and Super P electrodes were prepared by mixing 90 wt% active materials and 10 wt% PVDF. Then the Cu foils were covered with the electrode slurry, dried in a vacuum oven at 80 °C for 24 h, and punched into wafers with a diameter of 8 mm. The active material loading are about 0.8-1.2 mg cm^{-2} . The CR2032 half coin cells contain the electrode slices, the Celgard 2300 membrane as separator, 100 μL 1 M LiPF₆ in a mixed solvent of EC (ethylene carbonate) and DEC (diethyl carbonate) (1:1 by

volume) as the electrolyte, and lithium metal foil as the counter electrode. The galvanostatic charge–discharge tests were conducted on a Land Battery Measurement System. Besides, the electrochemical impedance spectroscopy (EIS) measurement and the cyclic voltammetric (CV) measurement were carried out by an electrochemical workstation (CHI 660D, China). For the EIS measurement, the open-circuit potential was used, and the voltage amplitude was 5 mV.

Table S1 the metal content in CPR

Sample	Metal content	Molar ratio of copper ions to rhodizonate ligands
CPR	25.88%	1 : 1.09

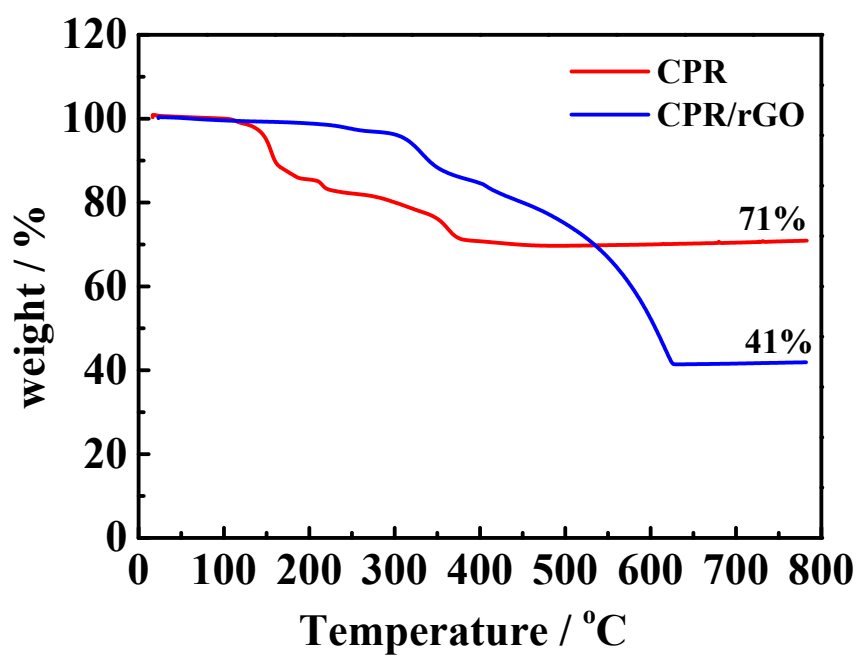


Fig. S1 TG curves of CPR and CPR/rGO in the air ($10\text{ }^{\circ}\text{C min}^{-1}$).

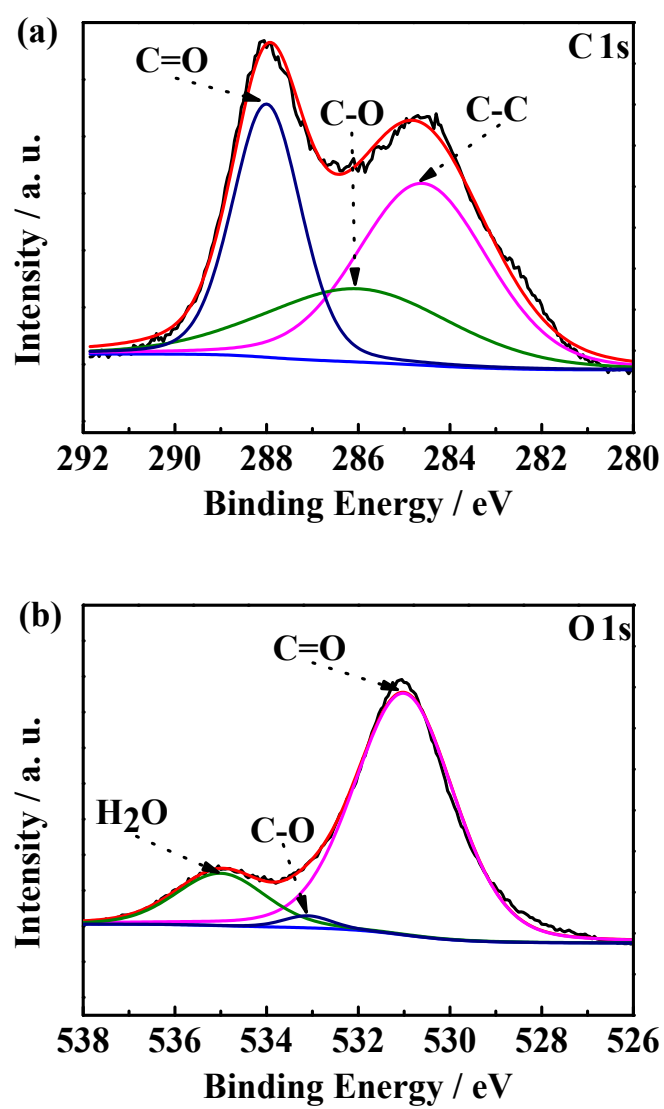


Fig. S2 (a) Fitted XPS spectra for C1s of disodium rhodizate; (b) Fitted XPS spectra for O1s of disodium rhodizate.

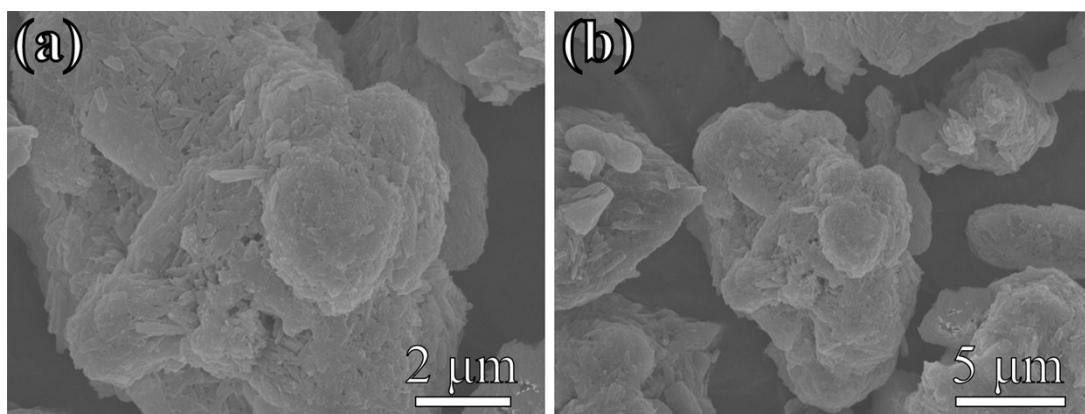


Fig. S3 (a-b) SEM images of disodium rhodizonate.

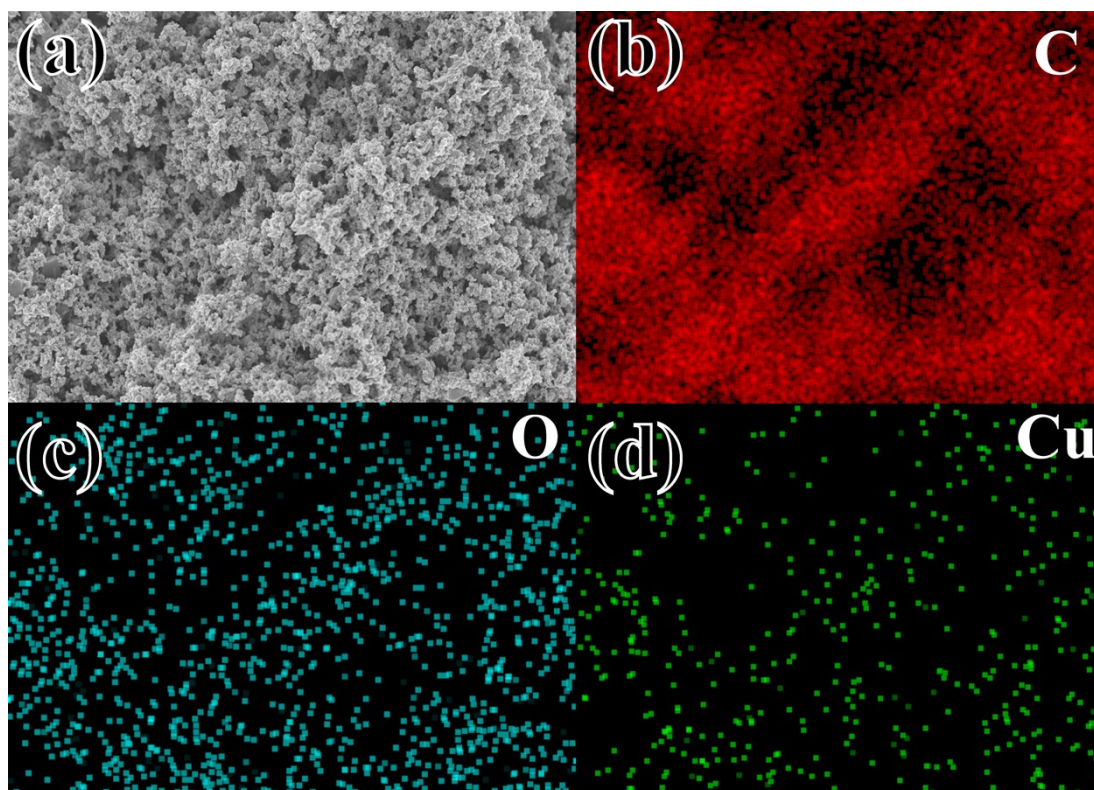


Fig. S4 (a) SEM images of CPR and (b-d) Corresponding EDX elemental mapping of C, O, and Cu.

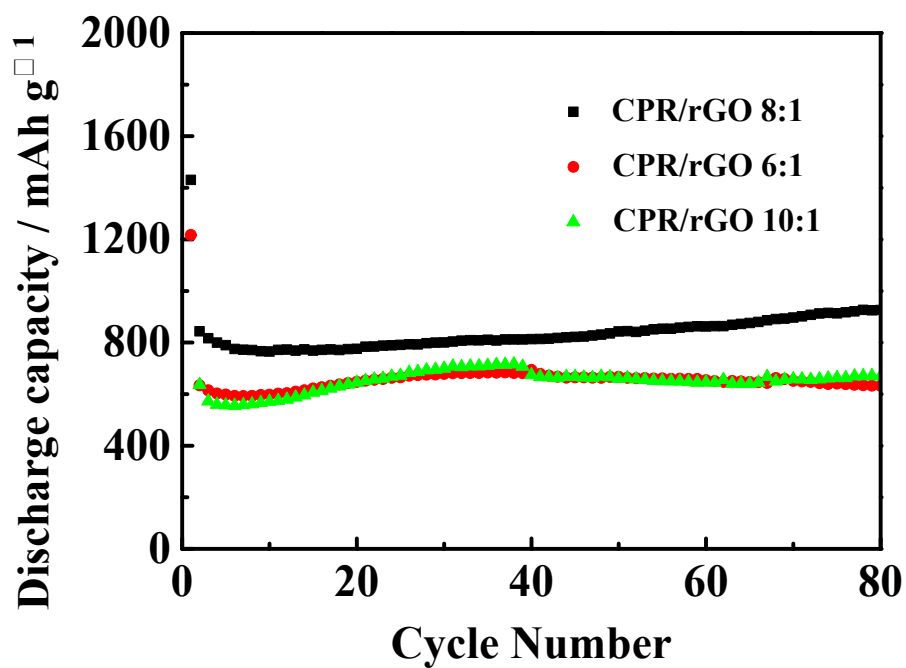


Fig. S5 Cycling performances of CPR/rGO with different weight ratio of reactants.

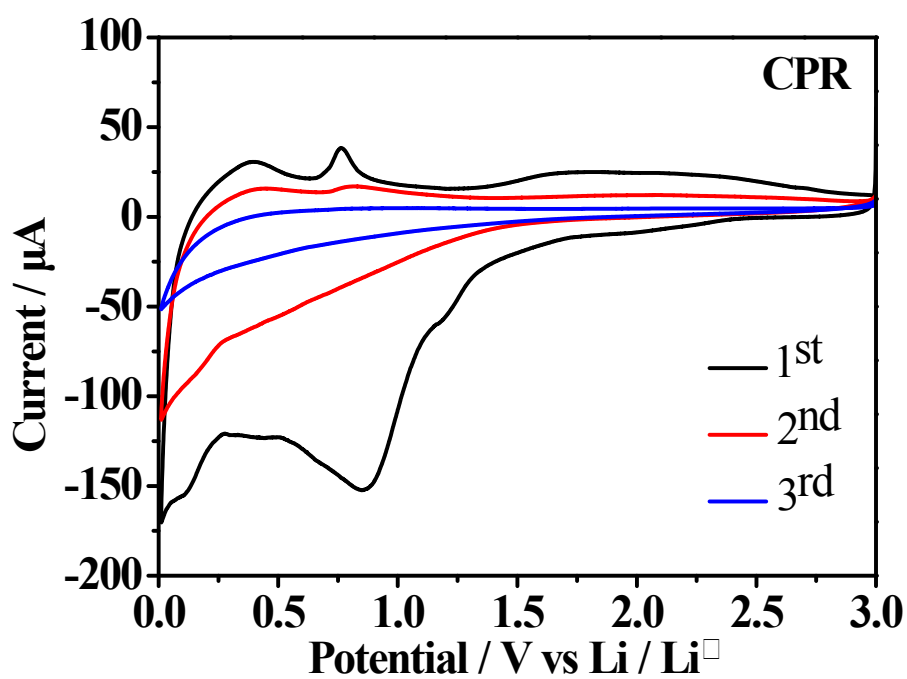


Fig. S6 Initial CV curves of CPR.

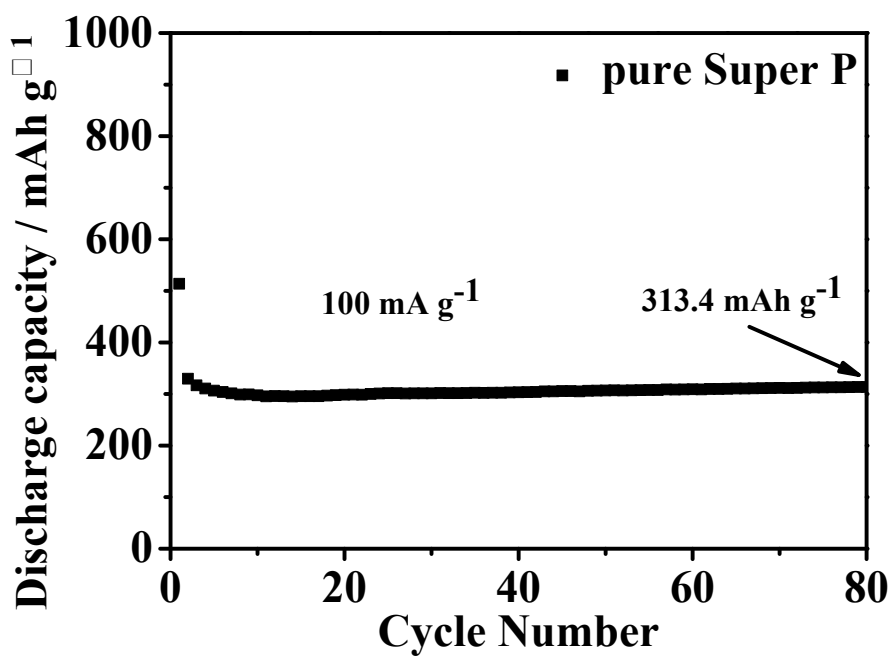


Fig. S7 Cycling performances of pure Super P.

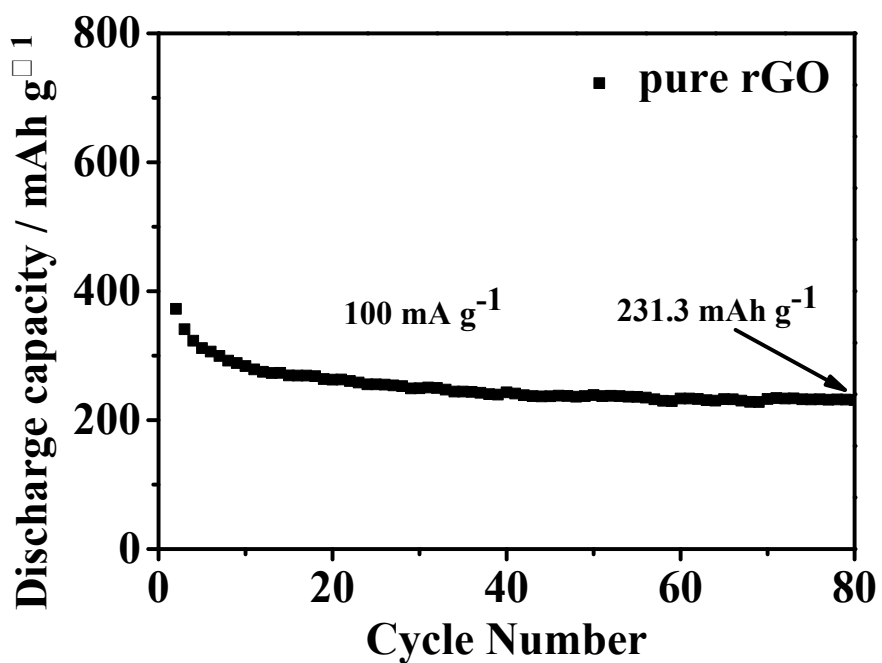


Fig. S8 Cycling performances of the pure rGO.

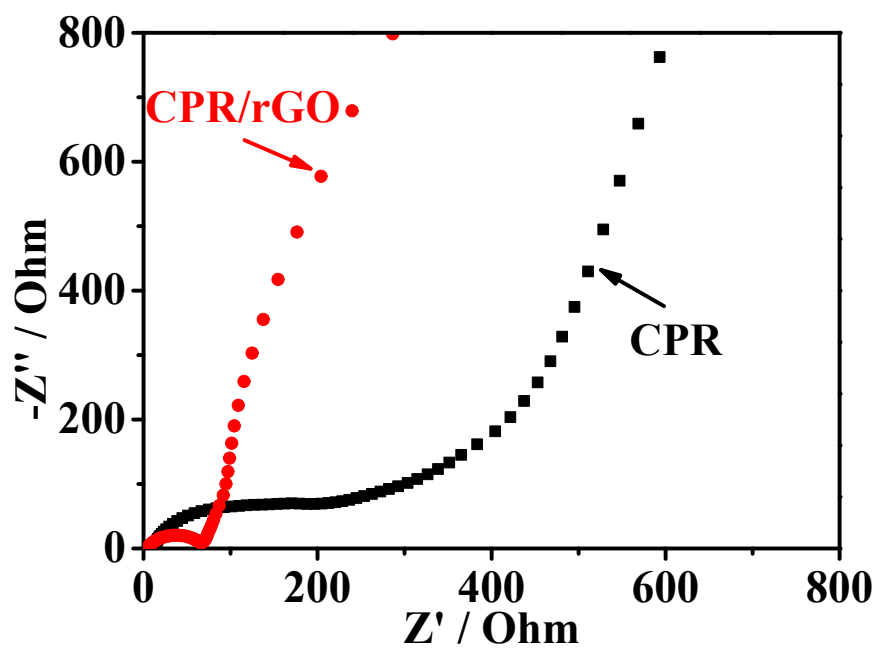


Fig. S9 EIS spectra of the CPR and CPR/rGO electrodes.

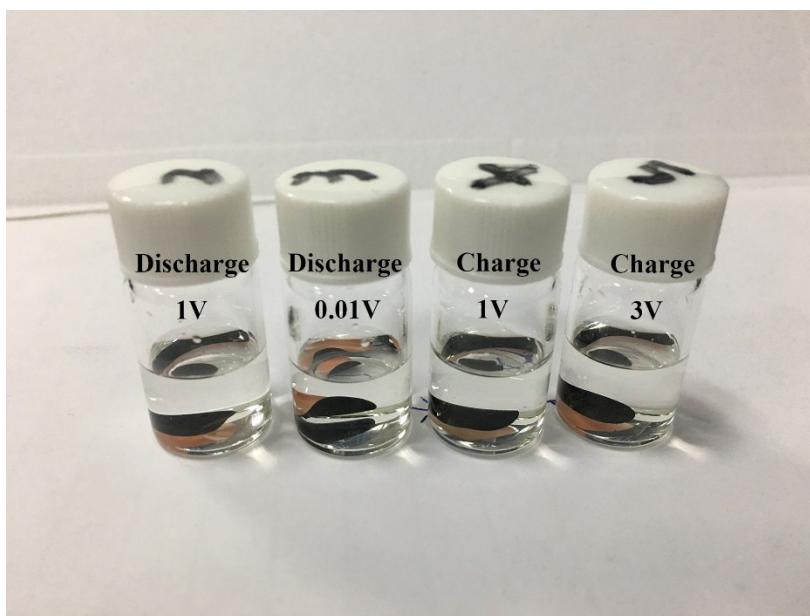


Fig. S10 Digital photographs for the CPR/rGO electrode slices in the electrolyte recorded at different potentials.