

Supporting Information

A quinone electrode with reversible phase evolution for long-life rechargeable aqueous aluminum metal batteries

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Experimental section

Preparation of electrode. Commercial TCQ was purchased from Shanghai Macklin Biochemical Co., Ltd and directly used for electrode preparation without further purification (purity 98%). The commercial TCQ was used as the active material, and mixed with acetylene black (Sigma-Aldrich) and polyvinylidene fluoride (PVDF, Sigma-Aldrich) in the weight ratio of 6:3:1. The 1-methyl-2-pyrrolidinone (NMP, Sigma-Aldrich) was used as the solvent. The homogeneous mixture was coated onto a carbon paper (Shanghai Hesen Electric Company). The electrode was dried in a vacuum oven at 60 °C overnight. The active mass loading of the electrode is 2 mg cm⁻².

Preparation of Al anode. Al anode was synthesized according to the previous work. The commercial Zn foil (AR 99.9%) was cut into Zn wafers and assembled as a 2032-type symmetric cell by using 1 M aluminum trifluoromethanesulfonate (Al OTf, pH=2.0) as the electrolyte. The galvanostatic charge-discharge test of the symmetric cell was continuously conducted at the current density of 2 mA cm⁻² for 240 hours to form Zn-Al alloy on the surface of Zn wafers. Zn wafers were subsequently disassembled and used as the anodes of the Al//TCQ batteries without further treatment.

Structural characterization: The morphological structure and composition of the electrode materials were characterized by X-ray diffraction (XRD, SmartLab, RIGAKU, with a Cu X-ray source), field-emission scanning electron microscopy (SEM, JSM-6330F). The functional group

distribution was measured through Fourier transform infrared radiation (FT-IR, NICOLET 6700, Thermo) and X-ray photoelectron spectroscopy (XPS, NEXSA, Thermo VG). Cyclic voltammogram (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectra (EIS) measurements were collected by using the electrochemical workstation (CHI760 and PARSTAT MC). The electrochemical performance of the TCQ electrode was characterized by employing a three-electrode system with a graphite rod as the counter electrode and a saturated calomel electrode (SCE) as the reference electrode in 1 M Al OTf aqueous electrolyte. The used area of working electrode was 0.5 cm². The Al//TCQ battery was assembled by using the TCQ electrode as the cathode, the Zn-Al alloy as the anode, and 1M Al OTf as the electrolyte.

Extended data

Supplementary Note 1: Calculation of the relation between the peak current and the scan rate in the CV curves.

The peak current and the scan rate in the CV curves of the chloranil electrode and the Al//TCQ battery follows the formula:

$$i = av^b \quad (1)$$

where a and b are the variable, i is the peak current (mA) and v is the corresponding scan rate (mV s⁻¹).

Upon the further calculation, the (1) can be transformed to:

$$\log i = b \log v + \log a \quad (2)$$

The data of the peak current and the scan rate are assigned to the parameters so the response can be plotted and fitted, the calculated slope of the fitting line is the value of b.

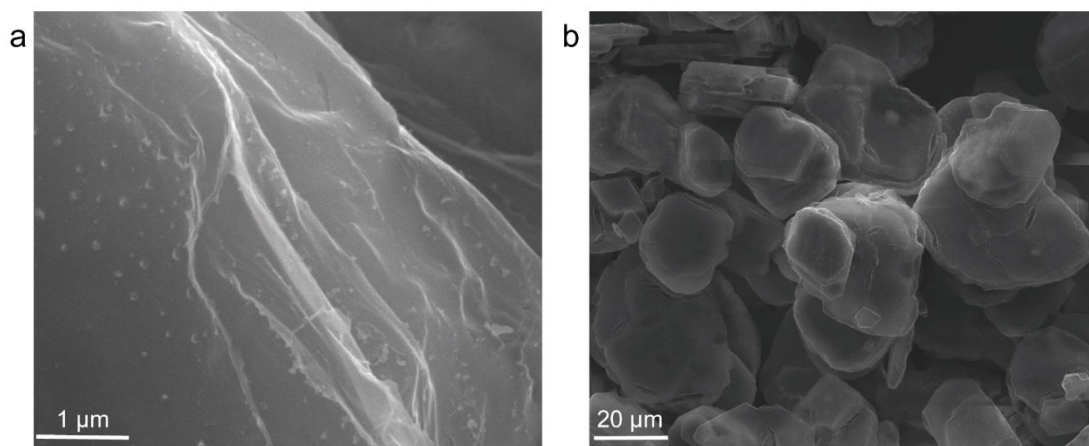


Fig. S1. SEM images of chloranil powder at different magnification.

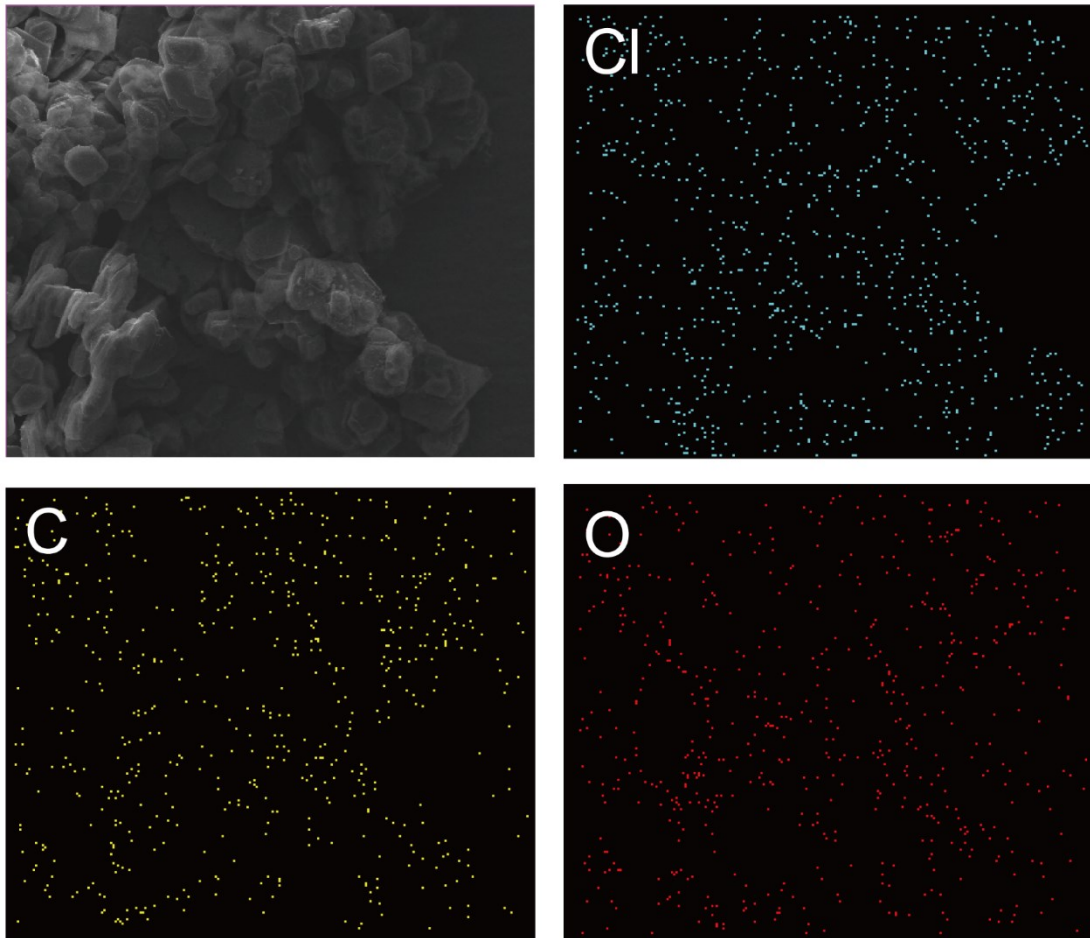


Fig. S2. The elemental mapping images of the TCQ powder by SEM.

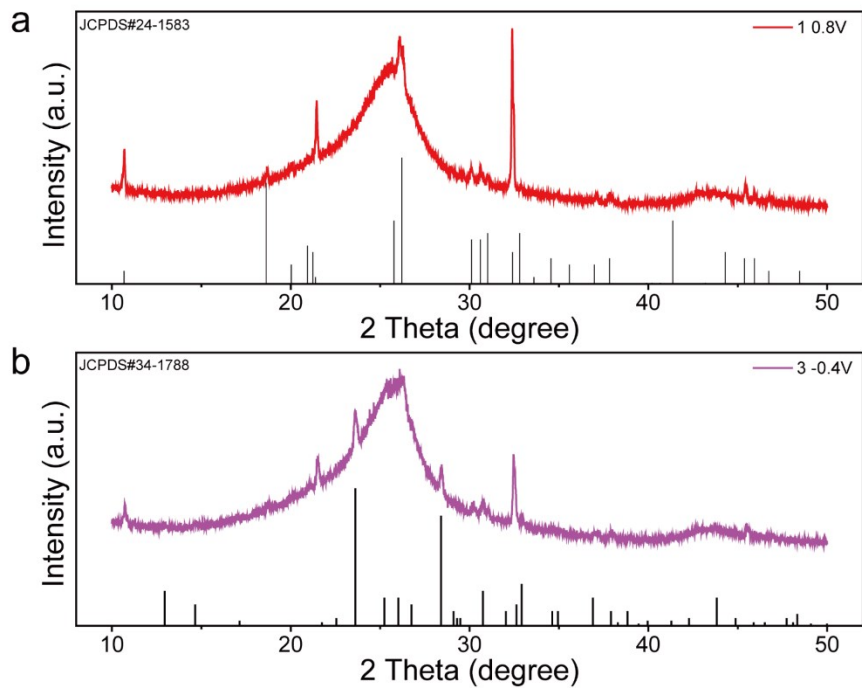


Fig. S3. The XRD profile of the chloranil samples at the voltage state of a) 0.8 V and b) -0.4 V.

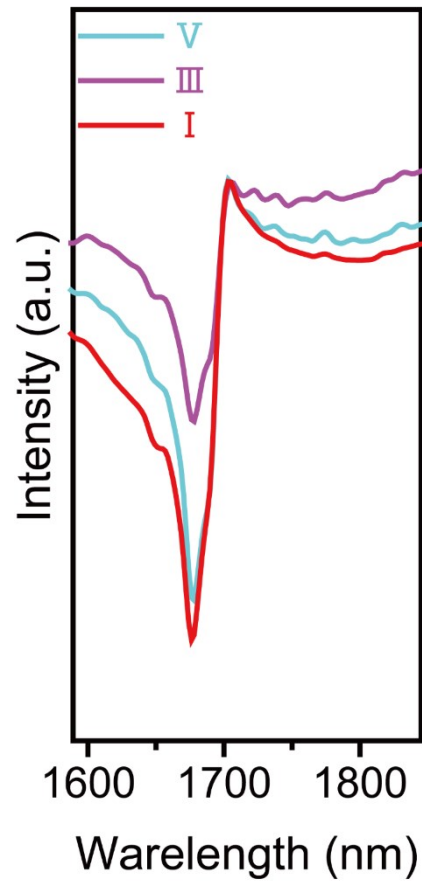


Fig. S4 Ex-situ FT-IR at different charging and discharging states of the TCQ electrode.

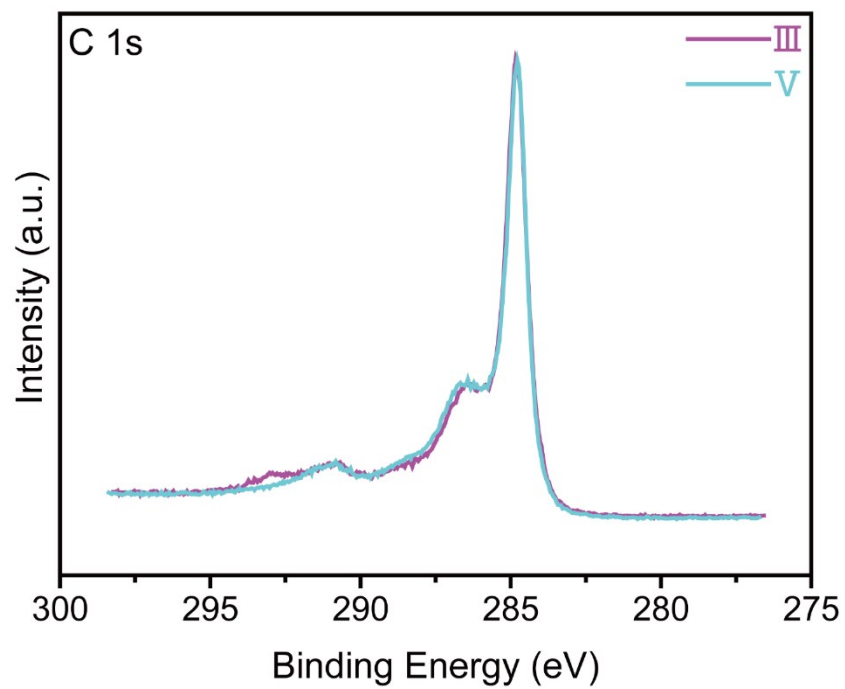


Fig. S5. The XPS spectra of C 1s at discharge and charge states.

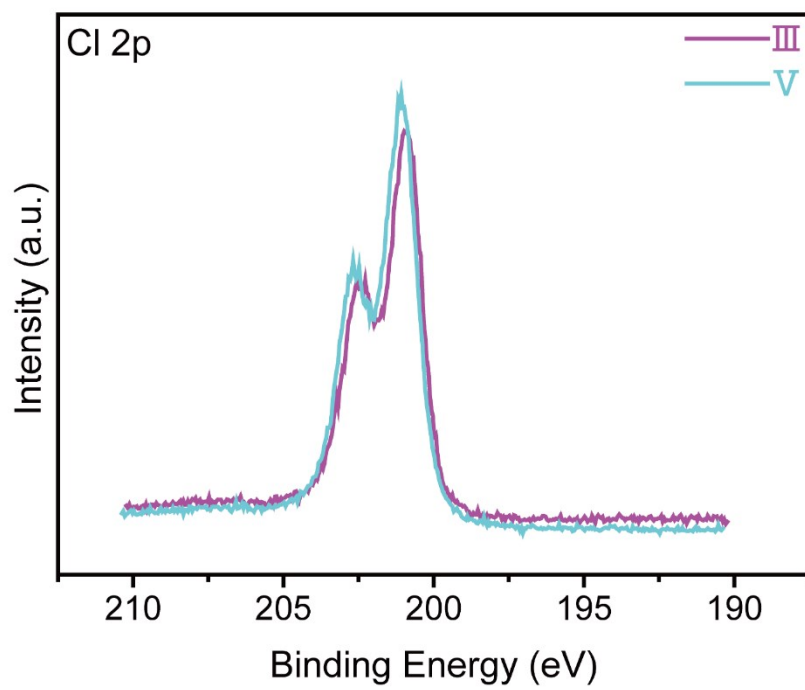


Fig. S6. The XPS spectra of Cl 2p at discharge and charge states.

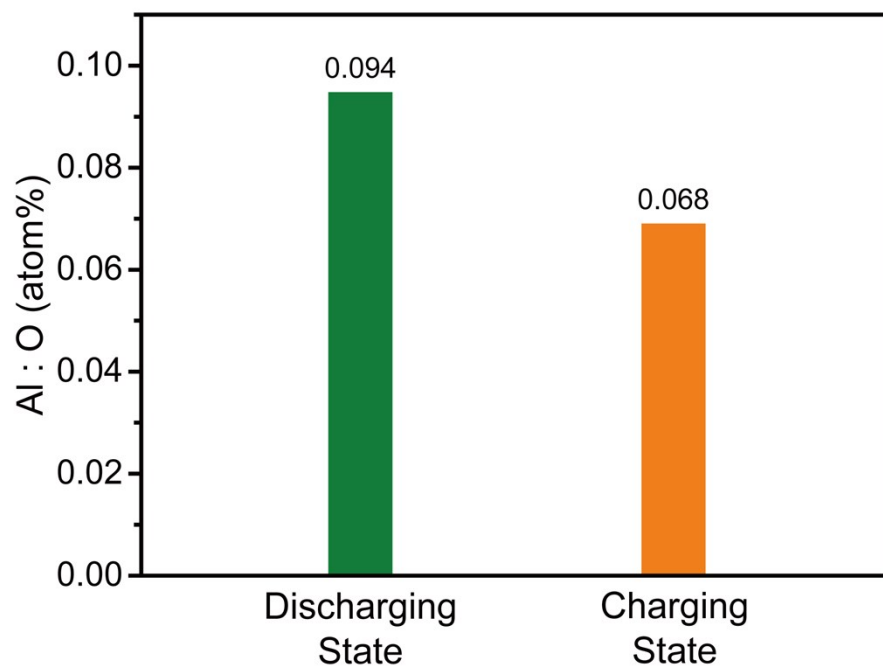


Figure S7. Al:O element ratio for TCQ in charge and discharge state.

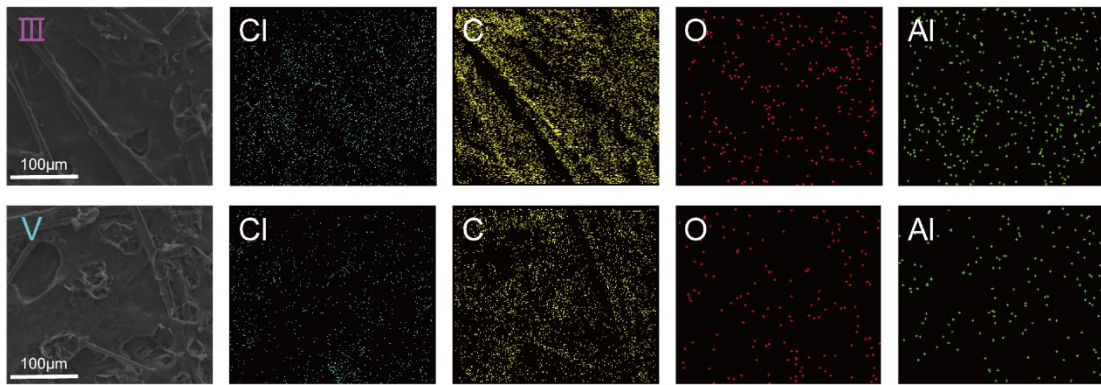


Fig. S8. The elemental mapping images of the TCQ electrode at discharge and charge states by SEM.

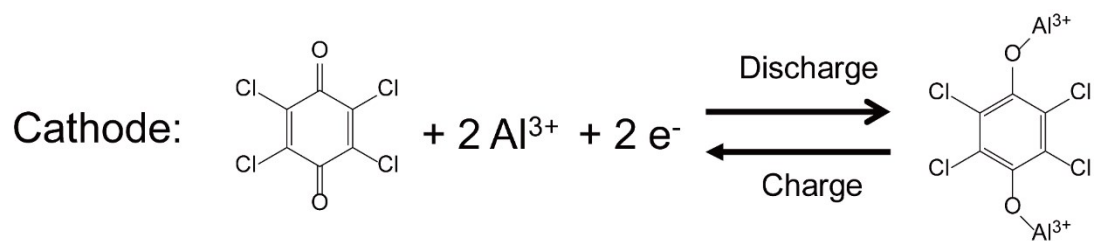


Fig. S9. The electrochemical reaction formula of the Al//TCQ battery.

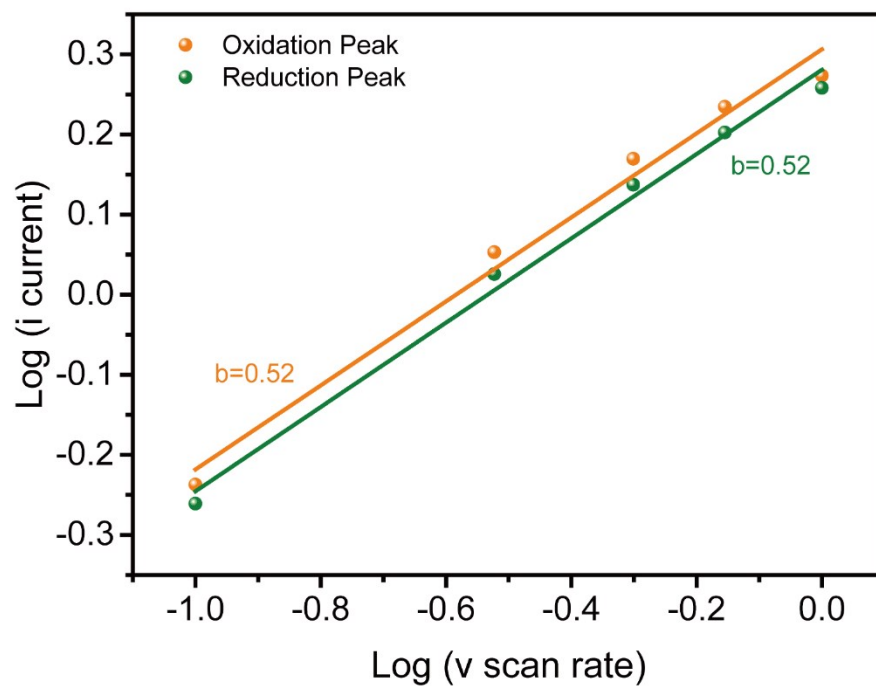


Fig. S10 Log i – log v plots derived by the peaks current and scan rates of the Al//TCQ battery.

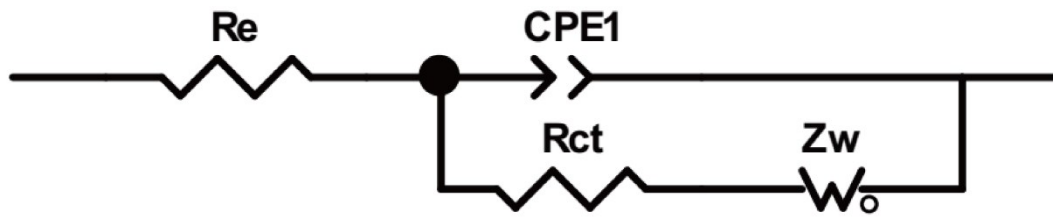


Fig. S11. The equivalent circuit of EIS.

Table S1. The impedance of the Al//TCQ battery.

R_s (Ω)	1.18
R_{ct} (Ω)	47.4
Z_w (Ω)	33.1