

## Supporting Information

### **An Aqueous Copper Battery Enabled by $\text{Cu}^{2+}/\text{Cu}^+$ and $\text{Cu}^{3+}/\text{Cu}^{2+}$**

#### **Redoxes Conversion Chemistry**

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## **Experimental**

### **Material preparation**

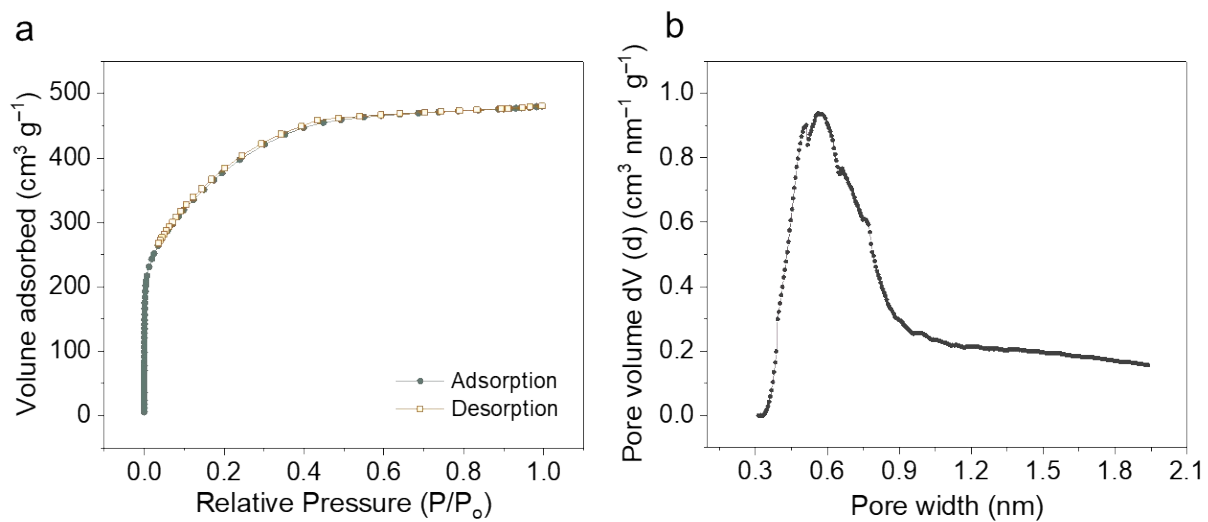
The cathode was prepared with AC, Ketjen Black (KB), and Polytetrafluoroethylene (PTFE) binder in isopropanol with a weight ratio of 7:2:1. The mixed slurry was plated to a self-standing film and dried in an oven at 80°C for 12 hours. The loading mass of AC is controlled to  $\sim 5 \text{ mg cm}^{-2}$  with a round piece in a diameter of 10 mm. The copper anode foil was 0.03 mm in thickness and 10 mm in diameter with a mass of  $\sim 19.5 \text{ mg}$ . Glass fiber of Pall Corporation was used as the separator. All electrolyte samples were prepared by mixing the designated molality of copper chloride ( $\text{CuCl}_2$ , anhydrous,  $\geq 98\%$ , Sigma Aldrich) and choline chloride ( $\text{ChCl}$ , TCI) with de-ionized water.

### **Material Characterization**

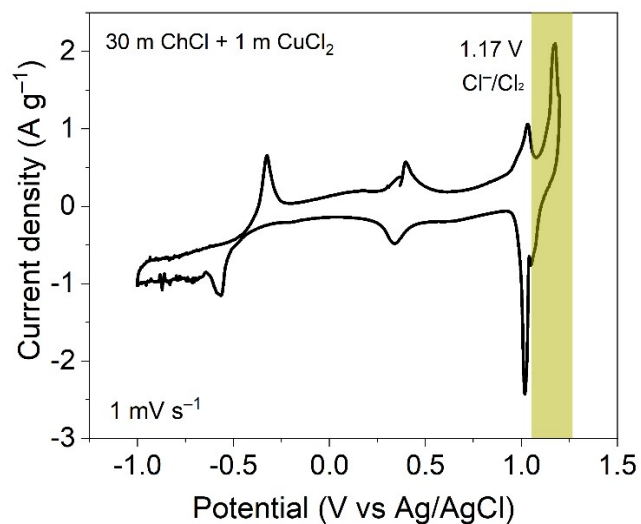
The specific surface area and pore size distribution of the ac were analyzed by Brunauer-Emmett-Teller (BET) on NOVA 4000. Raman spectroscopy measurement was performed by Horiba LabRAM HR Evolution microscope. The morphology and elemental distribution of all samples were examined using the field-emission scanning electron microscopy (SEM) (HITAS-4800). X-ray diffraction (XRD) patterns of all samples were collected on a Rigaku Ultima IV Diffractometer with a  $\text{Cu K}\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ).

### **Electrochemical measurements**

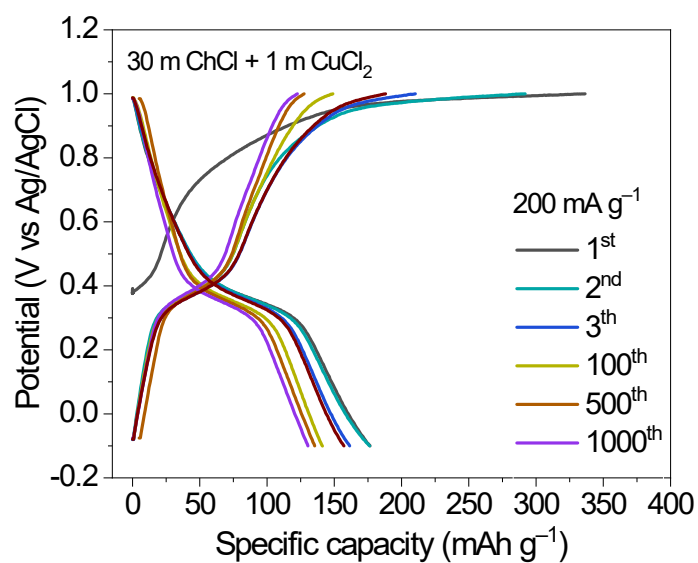
The electrochemical performance was evaluated using a Swagelok® cell (a T-cell) configuration. Galvanostatic charge/discharge tests were carried out on a LAND-CT2001A system at room temperature. Cyclic voltammetry (CV) was carried out on a Bio-logic electrochemical workstation (SP-150). The galvanostatic of Cu plating/stripping was measured in  $\text{Cu}||\text{Cu}$  symmetric cells, and the coulombic efficiency (CE) of Cu plating/stripping was studied in  $\text{Ti}||\text{Cu}$  cells.



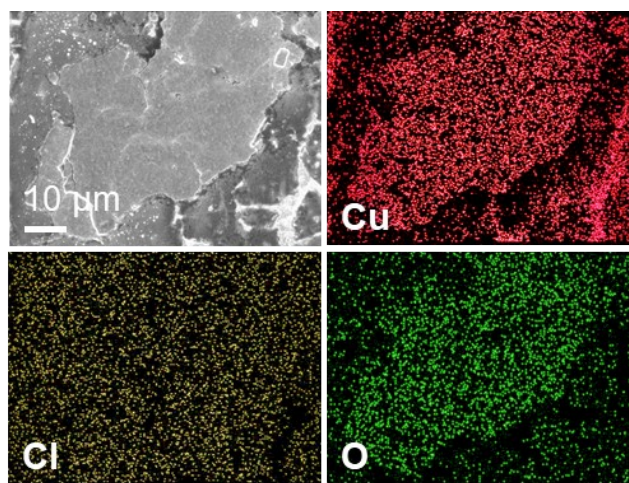
**Figure S1** (a) Volume of gas adsorbed vs relative pressure and (b) BET pore size distribution of activated carbon.



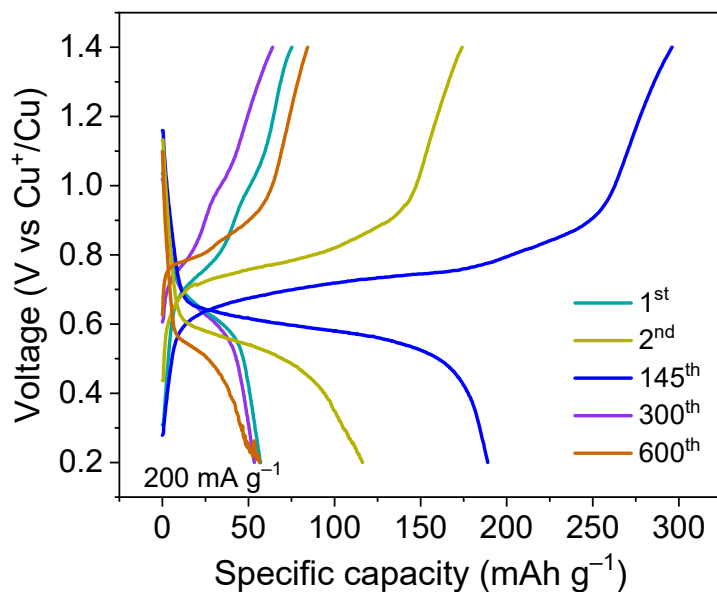
**Figure S2** CV curves of the AC cathode in a three-electrode cell with AC cathode as the counter electrode at the 1<sup>st</sup> cycle in the 30 m ChCl + 1 m CuCl<sub>2</sub> electrolyte.



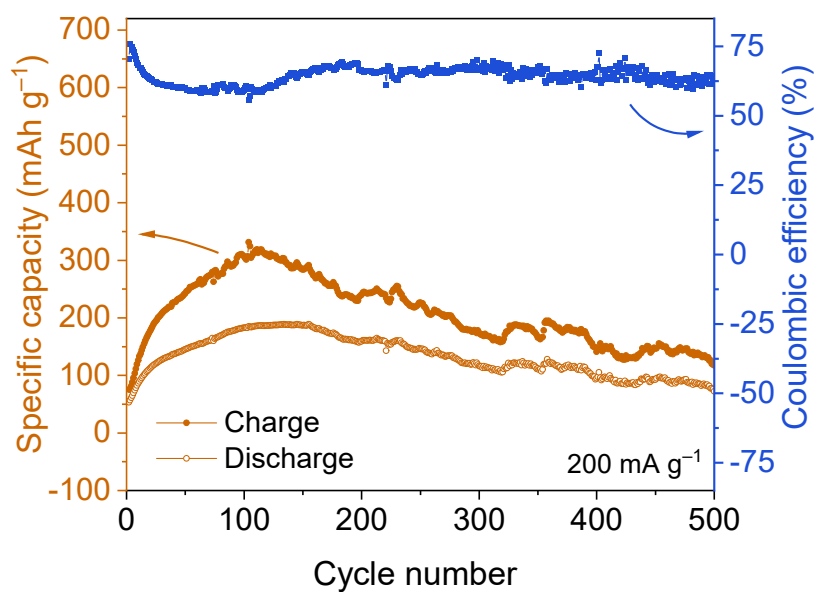
**Figure S3** GCD potential profiles of AC in the 30 m ChCl + 1 m CuCl<sub>2</sub> electrolyte in a three-electrode cell with Ag/AgCl as a reference electrode with a charge capacity of 200 mAh g<sup>-1</sup> within (a) -0.1–1.0 V.



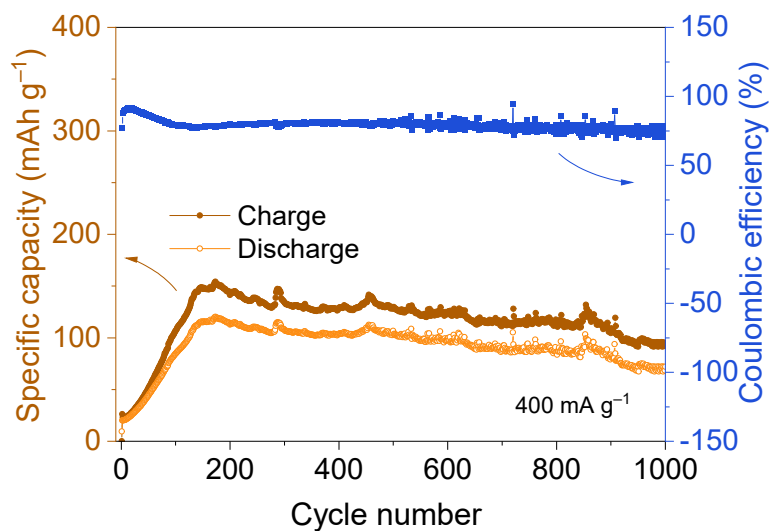
**Figure S4** SEM image of the plated Cu metal.



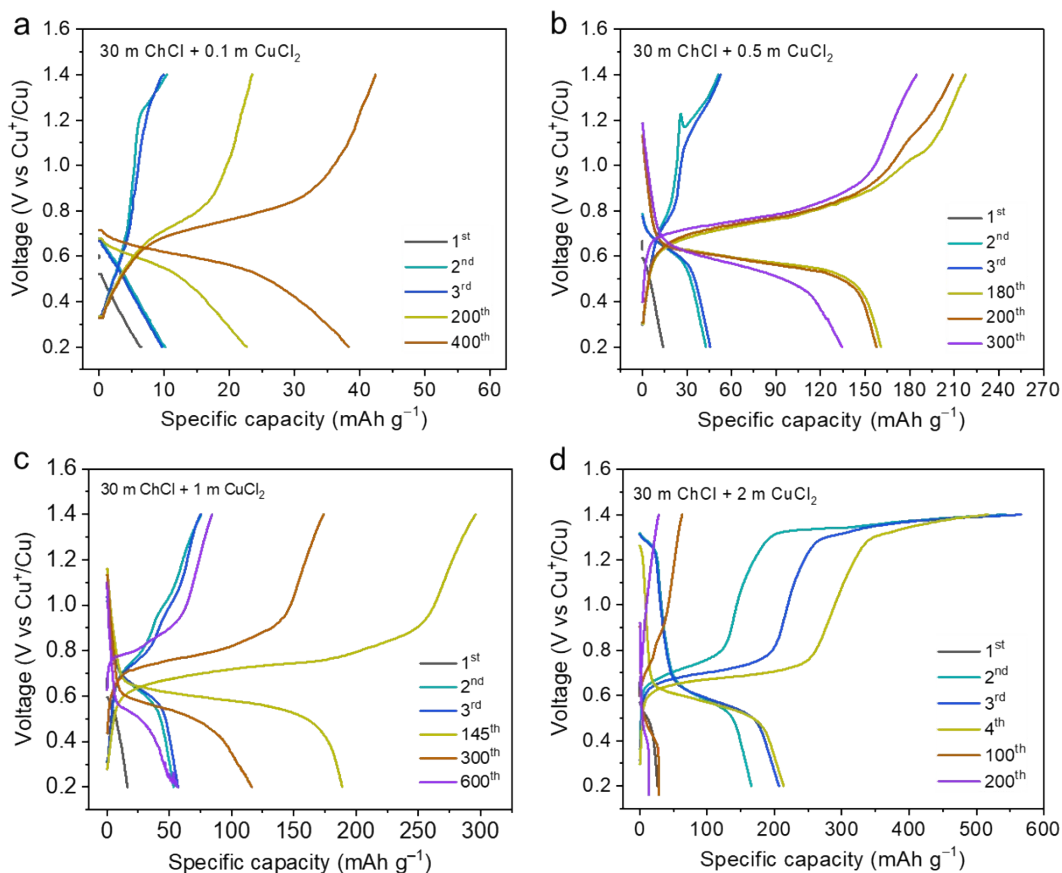
**Figure S5** GCD potential curves of the AC||Cu cell in the 30 m ChCl + 1 m CuCl<sub>2</sub> electrolyte within 0.2–1.4 V, where Cu foil is used as a counter electrode.



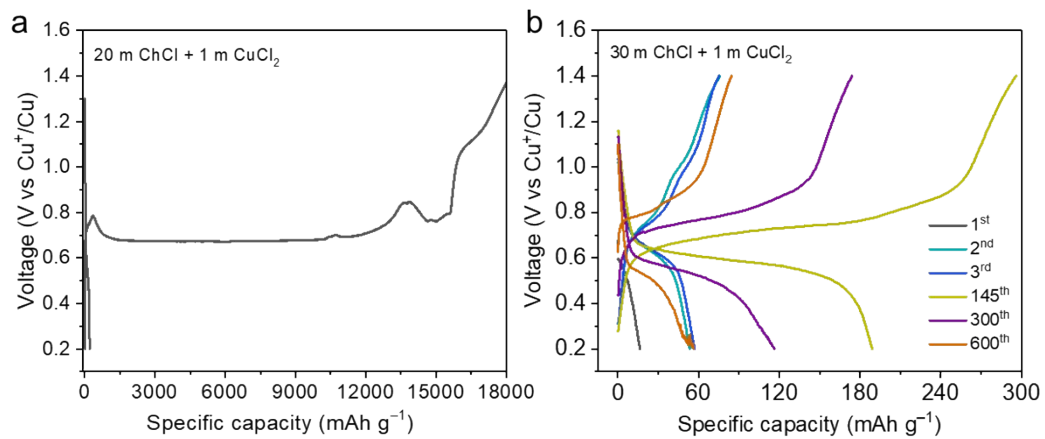
**Figure S6** Cycling performance of the AC||Cu cell in 30 m ChCl + 1 m CuCl<sub>2</sub> electrolyte within 0.2–1.4 V at 200 mA g<sup>-1</sup>.



**Figure S7** Cycling performance of the AC||Cu cell in 30 m ChCl + 1 m CuCl<sub>2</sub> electrolyte within 0.2–1.4 V at 400 mA g<sup>-1</sup>.



**Figure S8** GCD profiles of the AC||Cu cells in the electrolytes of (a) 30 m ChCl + 0.1 m CuCl<sub>2</sub>, (b) 30 m ChCl + 0.5 m CuCl<sub>2</sub>, (c) 30 m ChCl + 1 m CuCl<sub>2</sub> and (d) 30 m ChCl + 2 m CuCl<sub>2</sub> at a current density of 200 mA g<sup>-1</sup>.



**Figure S9** GCD profiles of the AC||Cu in the electrolytes of (a) 20 m ChCl + 1 m CuCl<sub>2</sub> and (b) 40 m ChCl + 1 m CuCl<sub>2</sub> at a current density of 200 mA g<sup>-1</sup>.