

Supporting Information

Micropore Confinement for a Highly Rechargeable Aqueous Zn-Hydroquinone
Battery with Exceptional Capacity

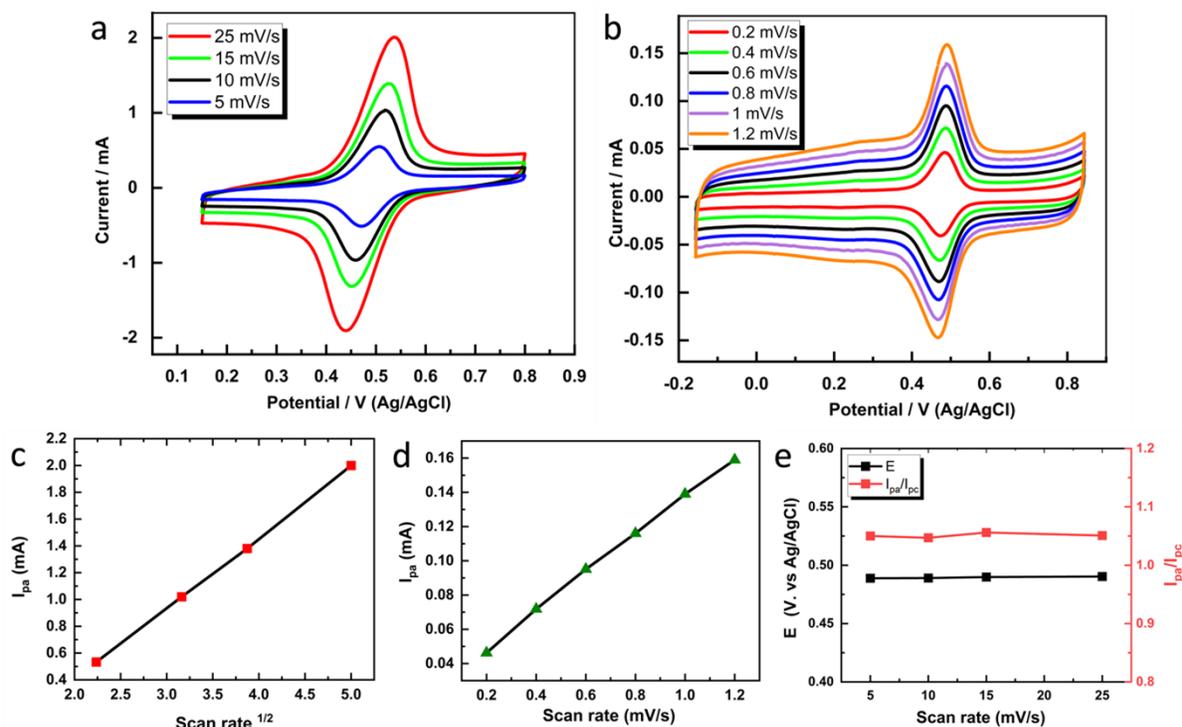


Figure S1. CV scans of QH2@AC at (a) high scan rates and (b) slow scan rates, (c,d) relationship between peak current (I_{pa}) vs. scan rate demonstrating linearity with $v^{1/2}$ for the high scan rates and linear dependence on v for slow scan rates, (e) plot of QH2/Q redox potential and I_{pa}/I_{pc} current ratio at different scan rates, in 1M H_2SO_4 .

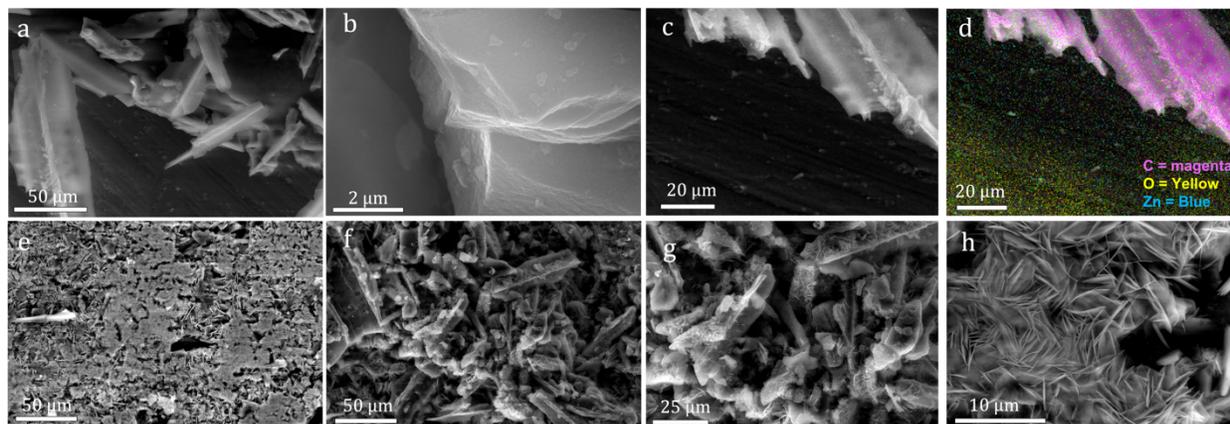


Figure S2. SEM images after battery cycling showing (a) AC particles from the cathode side, (b) higher magnification of an AC particle showing graphene platelets at the surface, (c, d) SEM image and EDX elemental mapping of the cathode (e) morphology of the AC layer coating the Zn foil anode after cycling showing smooth surface with no significant dendrite formation, (f, g) AC particles deposited on anode showing homogeneous deposition of Zn platelets tightly anchored to the AC surface (h) magnified morphology of the Zn platelets deposited uniformly on the AC.

SEM characterization

The scanning electron microscopy (SEM) images of the cathode and anode after testing the battery for 1,000 cycles are shown in **Fig. S2**. SEM images in **Fig. S2a-b** demonstrate the morphology of the cathode at different magnification, where the AC particles demonstrate a smooth surface with no notable residues, further confirming the impregnation of QH2 within the pores of the AC. At higher magnification, **Fig. S2b**, the graphene platelets added to the cathode to enhance its electronic conductivity are visible at the surface of the AC particles. The energy-dispersive X-ray elemental mapping of the cathode is shown in **Fig. S2c-d**, demonstrating high C content and low Zn content, in agreement with the XPS results. Interestingly, the Zn foil surface after 1,000 cycles of charge-discharge does not demonstrate formation of Zn dendrites, **Fig. S2e**. In the assembled battery, a slurry of AC wetted by the electrolyte was coated on the Zn foil to enhance the Zn utilization and to provide nucleation sites for Zn deposition to avoid dendrite formation. The higher magnifications of the SEM images of the AC coating layer on the Zn foil are shown in **Fig. S2f-h**, demonstrating notable formation of Zn flakes tightly attached to the AC particles. This observation is in good agreement with the postulated role of the AC anode-coating layer, to facilitate nucleation and uniform deposition of Zn during cycling of the battery. Overall, the SEM images of the cathode and anode demonstrated the successful impregnation of AC with QH2 molecules and prevention of dendrite formation due to the nature of the electrolyte used and the role of AC coating layer deposited on the anode surface, facilitating more uniform deposition of Zn and better utilization of the anode.

Methods

All reagents were of commercial grade used without further purification. Scanning electron microscopy images were acquired on Thermofisher Quattro FE-ESEM instrument without coating. Electrochemical measurements were conducted in three electrode system using Ag/AgCl as reference electrode (Hanna Instruments, HI 5314, 3.5M KCl), glassy-carbon electrode as the working electrode, and graphite rod as counter electrode. Electrochemical measurements were recorded on Bio-Logic potentiostat/galvanostat SP-50 and data were recorded with EC-lab software. The XPS measurements were carried out under an ultra-high vacuum with a base pressure of 1×10^{-9} mbar. Core-level spectra were recorded under normal emission with a Scienta R4000 analyzer using Al-K α radiation. For every sample the survey XP spectrum was firstly measured, and no unexpected contaminations were observed in these spectra. Activated carbon was purchased from Carlo Erba (BET surface area ~ 750 m²/g), Hydroquinone (QH2) was purchased from Adwic Pharmaceuticals (99%), H₂SO₄ (ACS grade) was purchased from Fisher Scientific, 2-Propanol (iPrOH) HPLC grade were purchased from Fisher Scientific, Zn(SO₄)•7H₂O was purchased from ADVENT.

Electrochemical testing procedure: The electrochemical characterization in three electrode setup was conducted in 1M H₂SO₄ aqueous solution using graphite rod, Ag/AgCl (3.5 M KCl) (Hanna HI 5314), and a glassy carbon electrode (GCE) as counter, reference and working electrodes, respectively. The ink was prepared from a solid suspension of QH2@AC (8 mg suspended in 1 mL isopropanol, added to it 7 μ L Nafion, 3wt% dispersion as a binder). 30 μ L of the solid suspension was casted on the GCE and was let to dry at room temperature. The tests were performed on Bio-Logic SP-50 potentiostat/galvanostat at different sweep rates as indicated. EIS data were collected on a PalmSens4 galvanostat/potentiostat at 10 mV perturbation potential at frequency range of 0.1 Hz-100 kHz.

Battery assembly:

The cathode was prepared by mixing QH2@AC (30 mg of the QH2@AC, equivalent to 5 mg QH2) with 2M Zn(SO₄) or H₂SO₄ (100 μ L) followed by addition of graphene powder (10 mg), and the mixing was continued until a homogeneous slurry was formed. The slurry was casted on the current collector (circular, diameter 1.7 cm). The anode was a Zn foil coated with a slurry of AC (22 mg) wetted with 2M Zn(SO₄) or 2M H₂SO₄ (70 μ L). The separator was a Nylon filtration membrane wetted with the same electrolyte used to prepare the electrodes. The battery cell used was a reusable PAT-Cell purchased from EL-Cell® fitted to a battery testing docking station.