# Realizing fast plating/stripping of high-performance Zn metal anode with a low Zn loading

## 1. Experimental Section

### 1.1 The preparation of graphene oxide (GO)

Commercial Zn foil (100 um, Zhengying Co., Ltd, China), Copper mesh (500 mesh, Zhengying Co., Ltd, China), $ZnSO_4 \cdot 7H_2O$ (99%, Macklin), $Na_2SO_4$ (99%, Macklin), $KMnO_4$ (99%, Macklin), carbon black (99%, Macklin), HCl (36 wt%), deionized water, and 1000 mesh sandpaper. The preparation of graphene oxide was based on the Hummers method [1].

### 1.2 The preparation of Zn coating copper mesh (CZ) and flat Zn plating copper foil (FCZ)

The electroplating solution of Zn contains 1 M $Na_2SO_4$, 2 M $ZnSO_4$, 0.1 M $H_3BO_3$. The plating current density is 50 mA·cm$^{-2}$. And the deposition duration is extended to 18 min obtaining about 15 mAh·cm$^{-2}$ Zn metal on copper mesh or copper foil.

### 1.3 The preparation of rGO coated CZ (RCZ)

Disperse 25 mg GO in 25 mL deionized water with ultrasonic for 30 min obtaining a transparent and dark yellow solution. Immerse CZ in GO solution for 2 min.

### 1.4 The preparation of AC cathode

The cathode was prepared with 70 wt% active carbon, 20 wt% Supper P, and 10 wt% carboxy methyl cellulose (CMC) on carbon cloth. It should be dried out in a vacuum oven over 12 h. Then cut it into 1 cm × 1 cm rectangular area, the average loading of active materials is about 5 mg·cm$^{-2}$.

### 1.5 Material and electrochemical characterization

The surface morphology of bare Zn coated Zn before and after cycling were detected by SEM with an EDX detector (JEOL JSM-6480, Japan) operated at 15 kV. XRD was carried out on a Rigaku TTR III device operating at 40 kV and 150 mA to ensure the successful preparation of RCZ anode, using Cu target Kα radiation (λ = 0.1514 nm).

CR2032 type coin cells were fabricated with 2 mol·L$^{-1}$ $ZnSO_4$ electrolyte at room temperature. The RCZ and FCZ anodes were cut into 1 cm × 1 cm as well as. Glass fiber that was cut into a circle with a diameter of 16 mm was employed as a separator to segregate anode and cathode. The symmetric cells galvanostatic cycling, coulombic efficiency, and full cells long-term charging/discharging test were all conducted with NEWARE BESTA system battery-testing instrument. Corrosion test and linear sweep voltammetry (LSV) were performed by EC-Lab workstation. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were tested by Ivium electrochemical workstation. The EIS measurements were tested at a frequency from 0.01 Hz to 100 Hz. Symmetric cells with different zinc anodes were assembled to monitor real-time changes during the process of Zn deposition by optical microscopy (RX50M China).

## 2. Simulation

The simulation was employed to simulate the Zn-ion deposition behavior with different morphology on surface. The rectangle of this simplified 2D model represents a tiny area of anode surface and the upper side represents the cathode, the lower side represents the anode. The ion conductivity of the electrolyte was set to 36 S·m$^{-1}$, and the reaction was occurs under the exchange current density of 30 mA·cm$^{-2}$ on anode. The potential of Zn follows the Equation S1.

$$E = -0.763 - \frac{RT}{2F} \cdot log\frac{1}{0.1c} \qquad \text{Equation S1}$$

Where the $E$, $R$, $T$, $F$, and $c$ is potential, ideal gas constant, Kelvin temperature, Faraday constant, and Zn-ion concentration, respectively.

The secondary current density distribution was simulated by finite element simulation, and the results was calculated through the equation from Equation S2 to Equation S5.

$$\nabla \cdot i_l = Q_l \qquad \text{Equation S2}$$

$$\nabla \cdot i_s = Q_s \qquad \text{Equation S3}$$

$$i_l = -\sigma_l \nabla \varphi_l \qquad \text{Equation S4}$$

$$i_s = -\sigma_s \nabla \varphi_s \qquad \text{Equation S5}$$

Where $i$, $Q$, $\sigma$, and $\varphi$ are current density, electric quantity, conductivity, and electric potential, respectively. The simulation constructed mentioned above is an ideal model without the consideration of condition fluctuation.

## Reference


1. D. Li, M. B. Müller, S. Gilje, R. B. Kaner, and G. G. Wallace, "Processable aqueous dispersions of graphene nanosheets," *Nature Nanotechnology*, vol. 3, pp. 101-105, 2008.