# Supporting Information

## *Leaf-like porous N-doped carbon structure embedded with $CoS_2$ nanoparticles self-supported on carbon fiber paper as cathode in flexible zinc-air batteries*

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*Materials*

HCP020N carbon fiber paper, 99% thiourea, 98% 2-methylimidazole, 99% $Co(NO_3)_2 \cdot 6H_2O$, 95% KOH, 99% $C_4H_6O_4Zn \cdot 2H_2O$,anhydrous ethanol, deionized water, $RuO_2$, Pt/C, 5% nafion solution, 98% isopropyl alcohol, 99% acrylamide (AM), 98% acrylic acid (AA), average Mw 5 millions-700 millions sodium polyacrylate, 98% N, N '-methylene diacrylamide (ABS), 99% ammonium persulfate (initiator), polyester Film(PET film), zinc plate, copper wire.

*ORR performance measurement*

The ORR has a scanning range of 0.2 to -0.8 V (vs. Ag/AgCl) with a scanning speed of 10 mV $s^{-1}$.

The $CoS_2$-CZ@CFP was cut into 0.25 $cm^2$ squares and placed on a glass carbon rotating disk electrode (diameter RDE, diameter 5 mm). A plexiglass model was tightly fitted to the rotating disk electrode to leak out 0.19625 $cm^2$ of the glass carbon, ensuring a 0.19625 $cm^2$ contact area between the $CoS_2$-CZ@CFP and electrolyte such that the $CoS_2$-CZ@CFP does not slide off. The concentration of the electrolyte is 0.1 M KOH (pH = 13). For the test, the disk electrode was spun at a speed of 1600 rpm. The reversible hydrogen electrode potential $E_{RHE} = E_{Ag/AgCl} + 0.059\ pH + E^{\theta}_{Ag/AgCl}$.

Pt/C catalyst ink was fabricated by distributing uniformly 8 mg of Pt/C in a solvent containing 800 μL of deionized water, 175 μL isopropyl alcohol, and 60 μL Nafion solution (5 wt%). The ORR performance of Pt/C was measured by dropping directly 70 μL ink on rotating disk electrode (Fig. S4 (a)).

250 μL of Pt/C ink was applied to 0.7 $cm^2$ CFP (the contact area between Pt/C and electrolyte is 0.5 $cm^2$) to obtain Pt/C@CFP with a catalyst load of 2 mg $cm^{-2}$. After the above steps were completed, the CFP was dried. The ORR performance of Pt/C@CFP and Co-CZ@CFP were measured with similar method with $CoS_2$-CZ@CFP (Fig. S6.).

*Calculation Methods*

Density functional theory (DFT) calculations was conducted using the Vienna ab-initio simulation package (VASP) in which projector augmented wave (PAW) potentials was adopted to describe ion-electrons interactions. The generalized gradient approximation (GGA) parameterized by the Perdew-Becke-Ernzerhof (PBE) method was used for treating exchange-correlation between electrons. The cut-off energy was 480 eV. The conventional energy ($10^{-4}$ eV) and force (0.02 eV $Å^{-1}$) convergence criteria were employed.

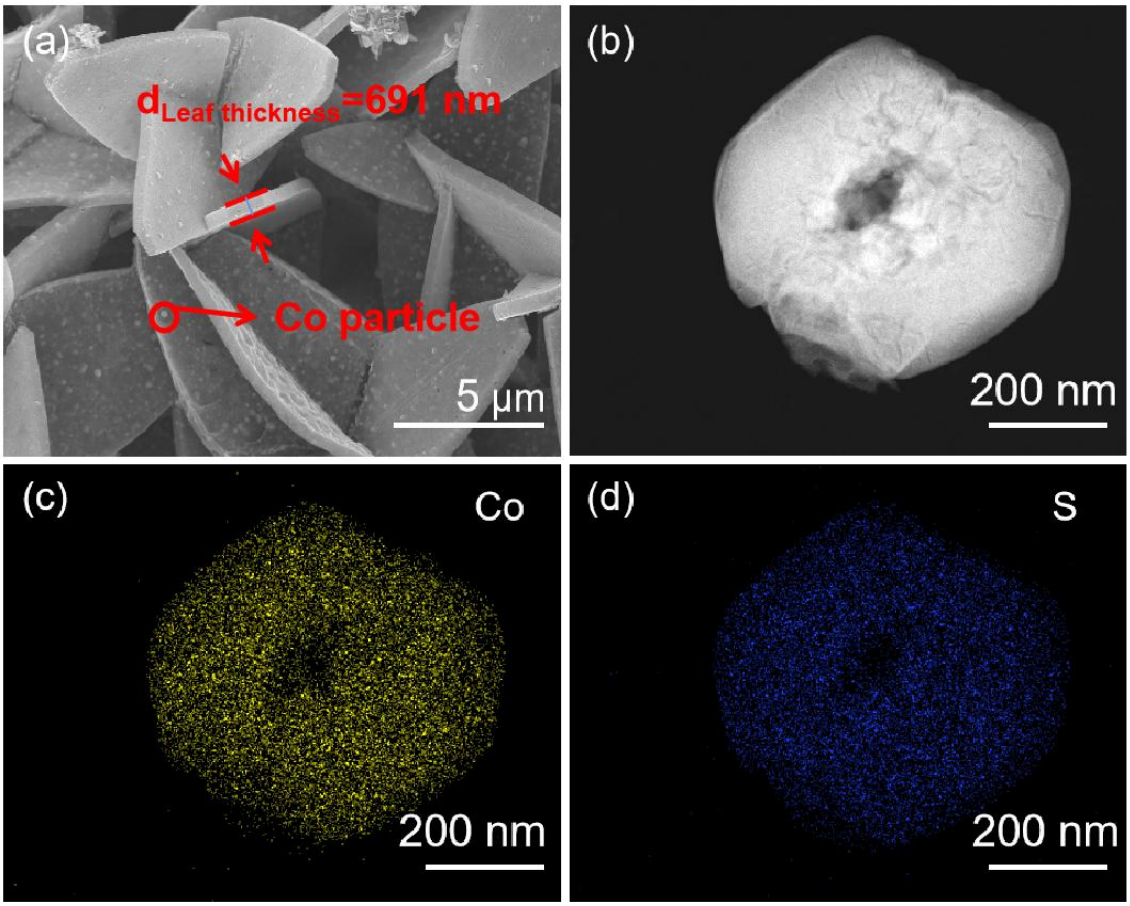


**Fig. S1.** (a) SEM images of Co-CZ@CFP at high magnification. (b) High angle annular dark field scanning transmission (HAADF-STEM) of $CoS_2$ particle. (c) (d) Corresponding elemental mapping diagram of (b).

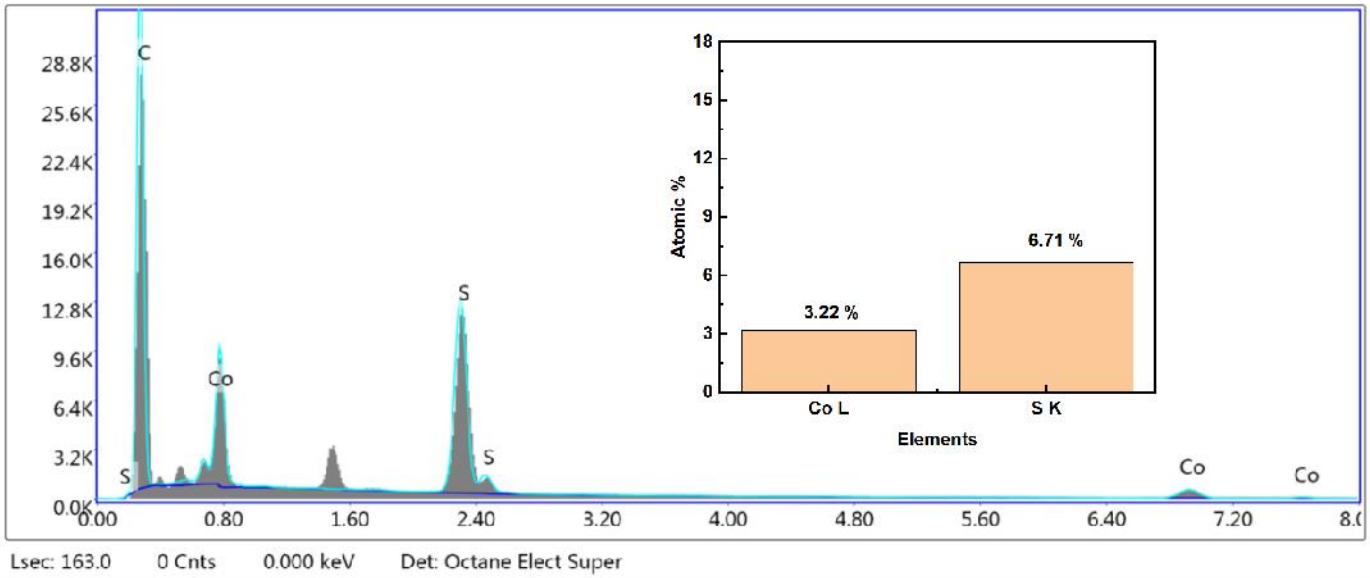


**Fig. S2.** EDS element proportion analysis diagram of Fig. 2 (e)-(f).

**Table S1.** Co and S content measured by plasma emission spectrometer.

| Element | Co | S |
|---|---|---|
| Content (μg $mL^{-1}$) | 0.294 | 0.329 |
| Atomic number (μmol $mL^{-1}$) | 0.00498 | 0.01028 |

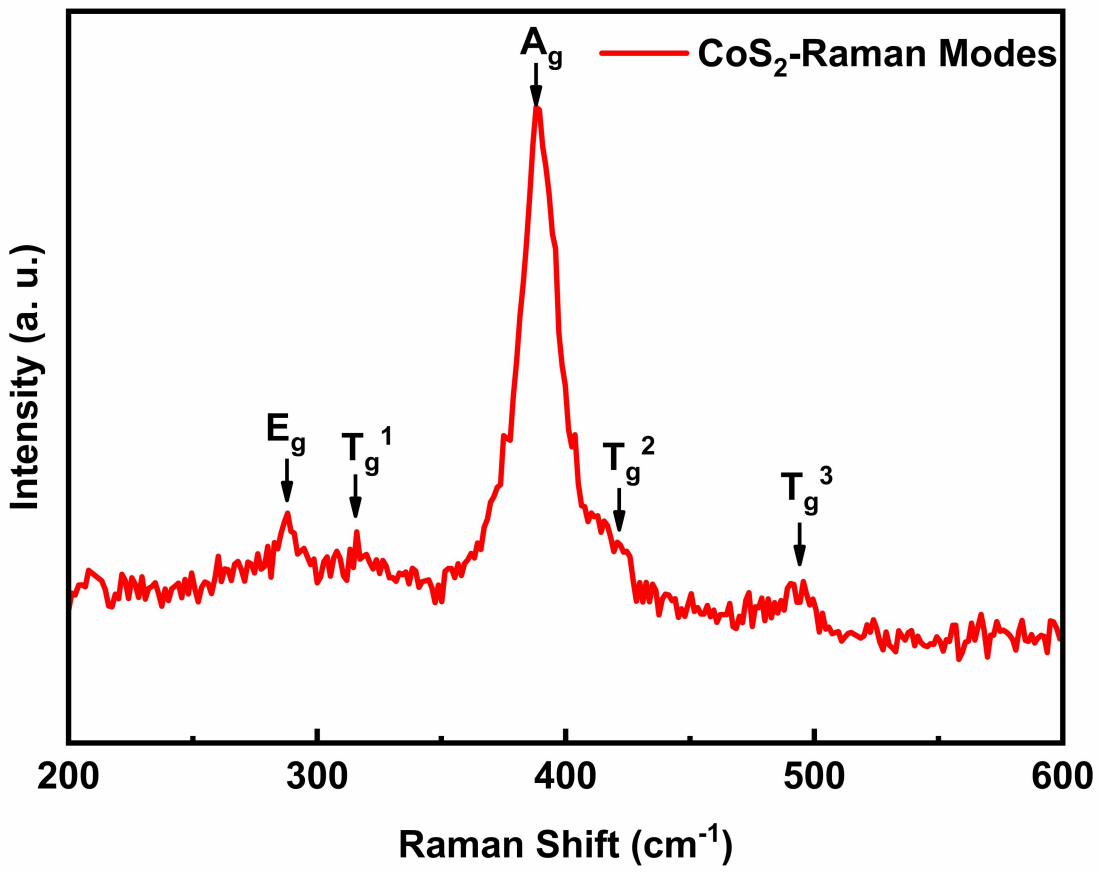


**Fig. S3.** Raman spectra of $CoS_2$.

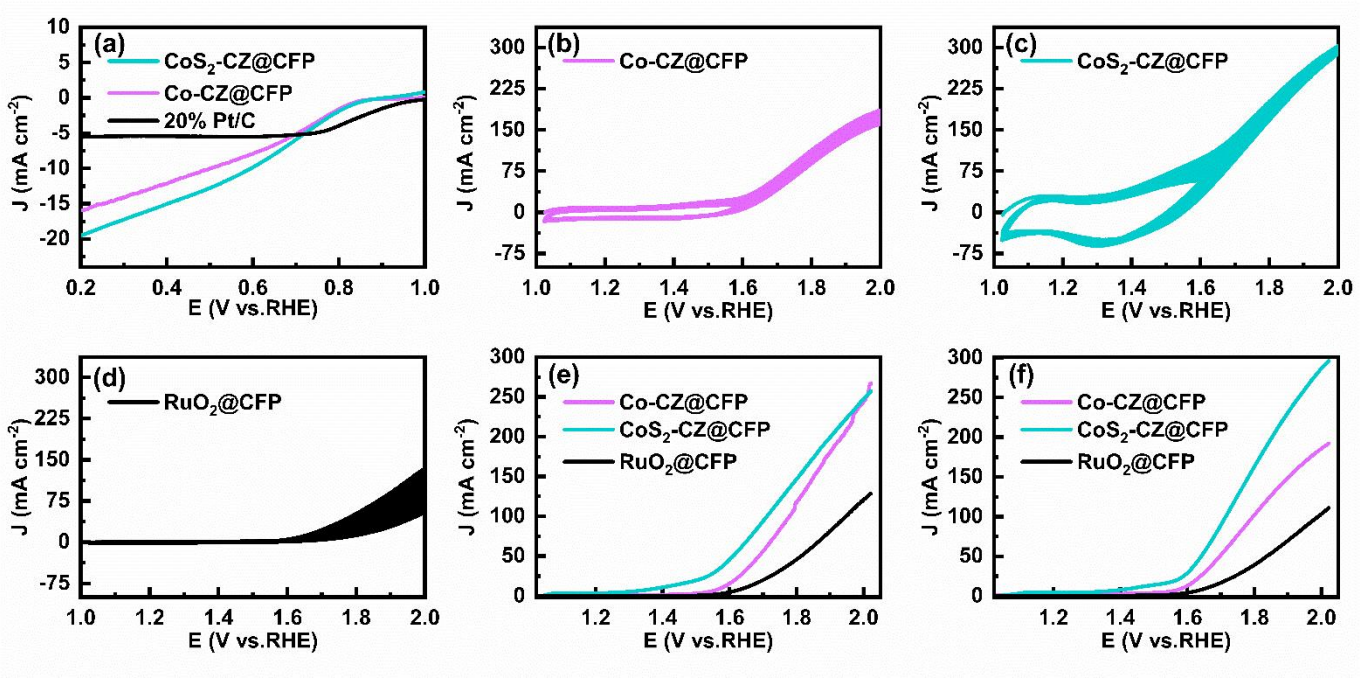


**Fig. S4.** (a) ORR tests of $CoS_2$-CZ@CFP, Co-CZ@CFP and 20% Pt/C by rotating disk electrode. (b)-(d) 1000 cycle CV of the samples. (e) Samples were tested for OER before 1000 cycles of CV. (f) Samples were tested for OER after 1000 cycles of CV.

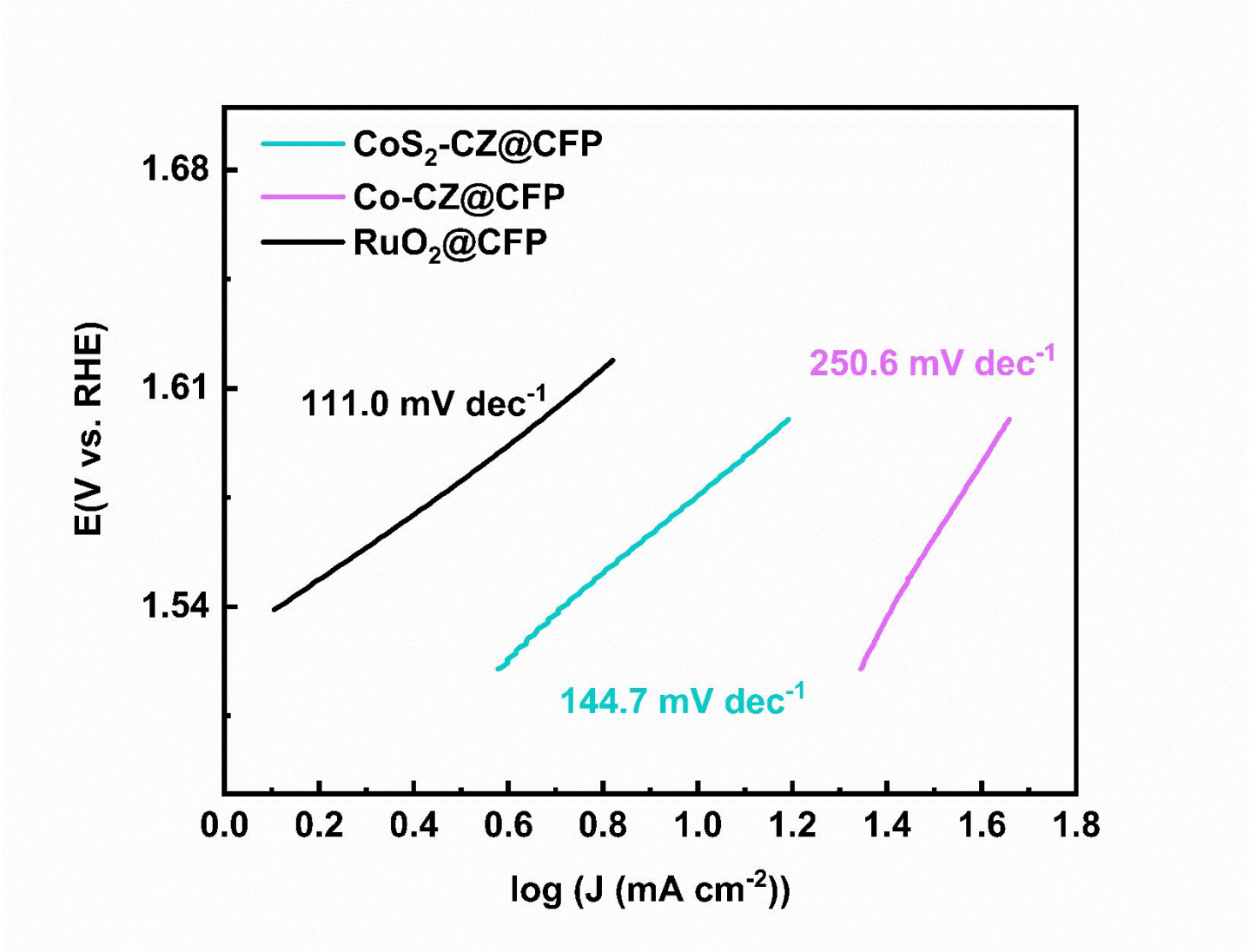


**Fig. S5.** The Tafel slopes of $CoS_2$-CZ@CFP, Co-CZ@CFP and $RuO_2$@CFP before 1000 laps of CV.

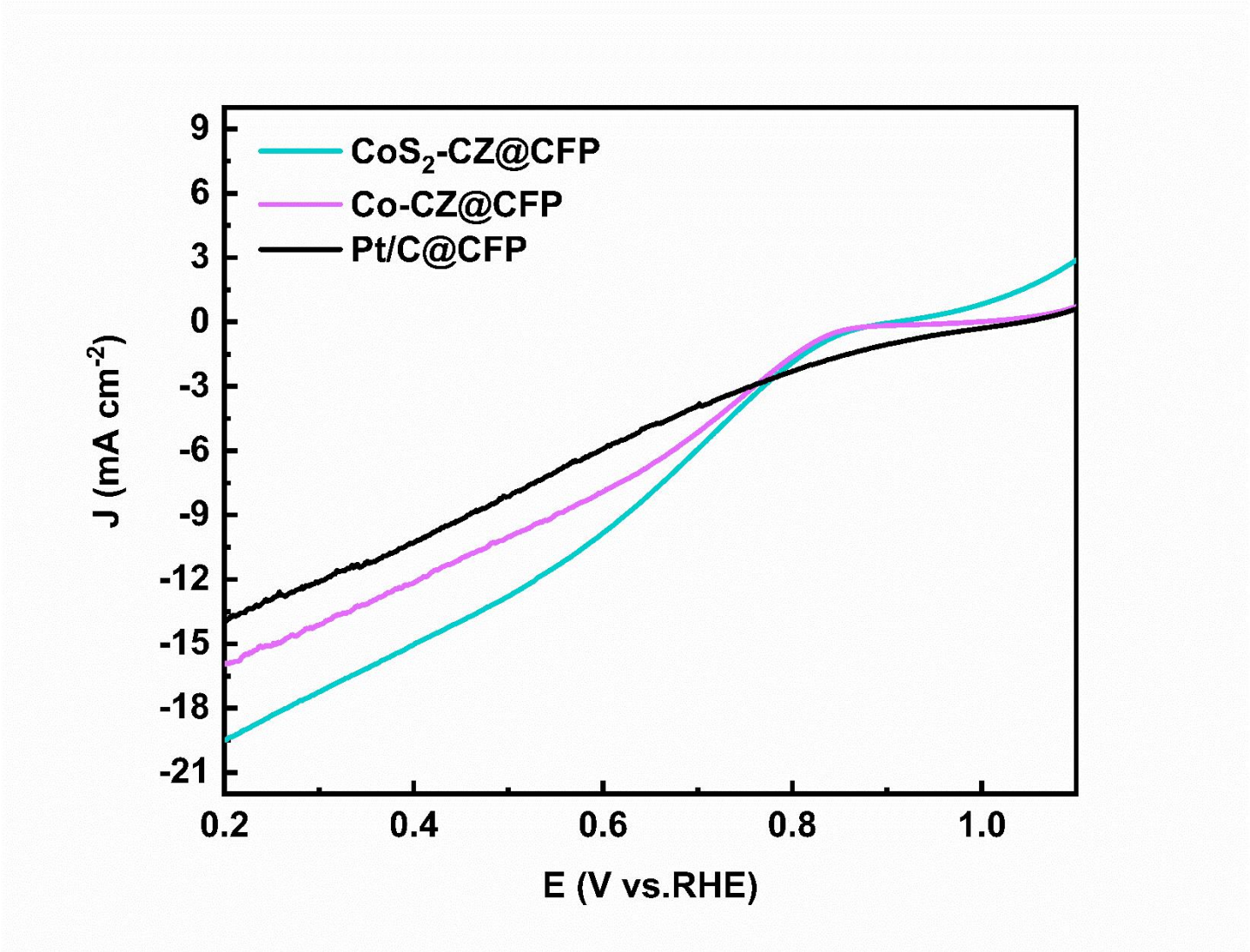


**Fig. S6.** ORR tests of $CoS_2$-CZ@CFP, Co-CZ@CFP and Pt/C@CFP.

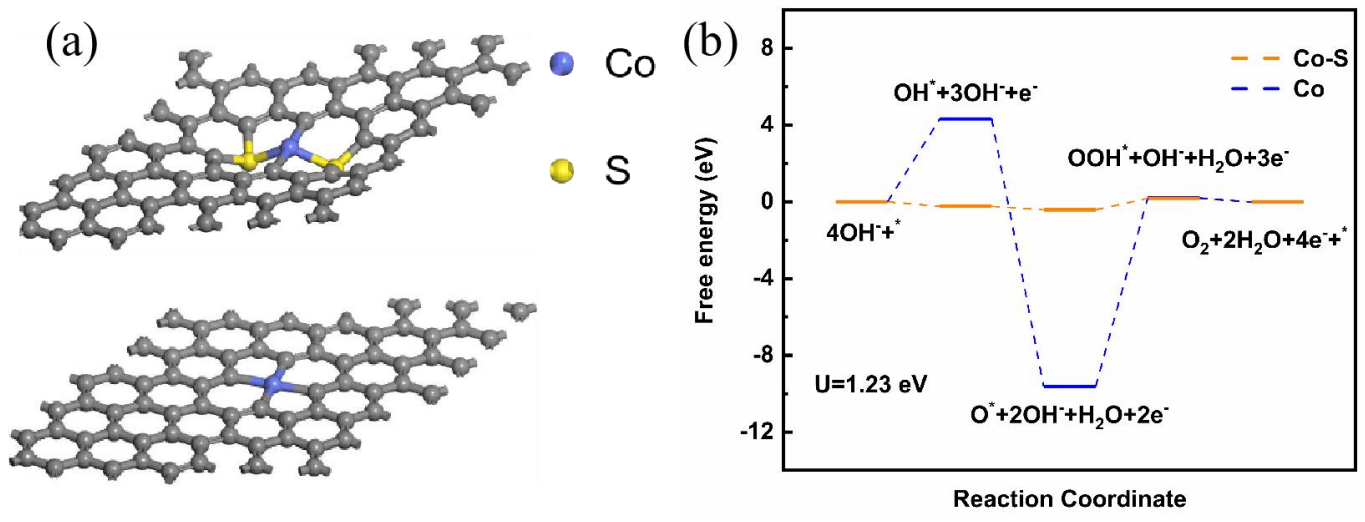


**Fig. S7.** (a) The model of Co-graphene and Co-S-graphene. (b) Density functional theory (DFT) of bonding between Co and S atoms (Co-S) and Co atoms.

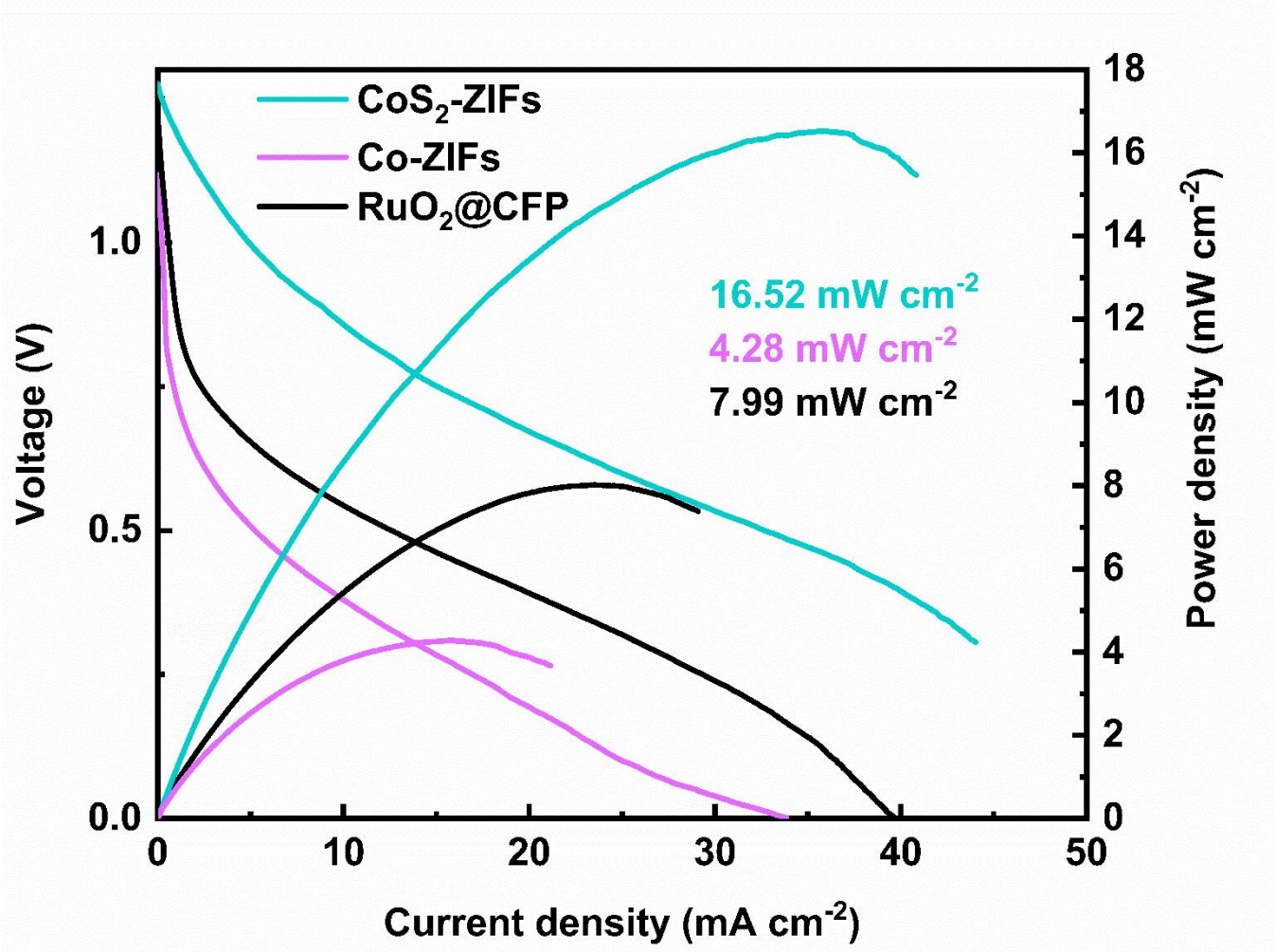


**Fig. S8.** Discharge polarization spectra and corresponding power density maps of $CoS_2$-CZ@CFP, Co-CZ@CFP and $RuO_2$@CFP batteries.

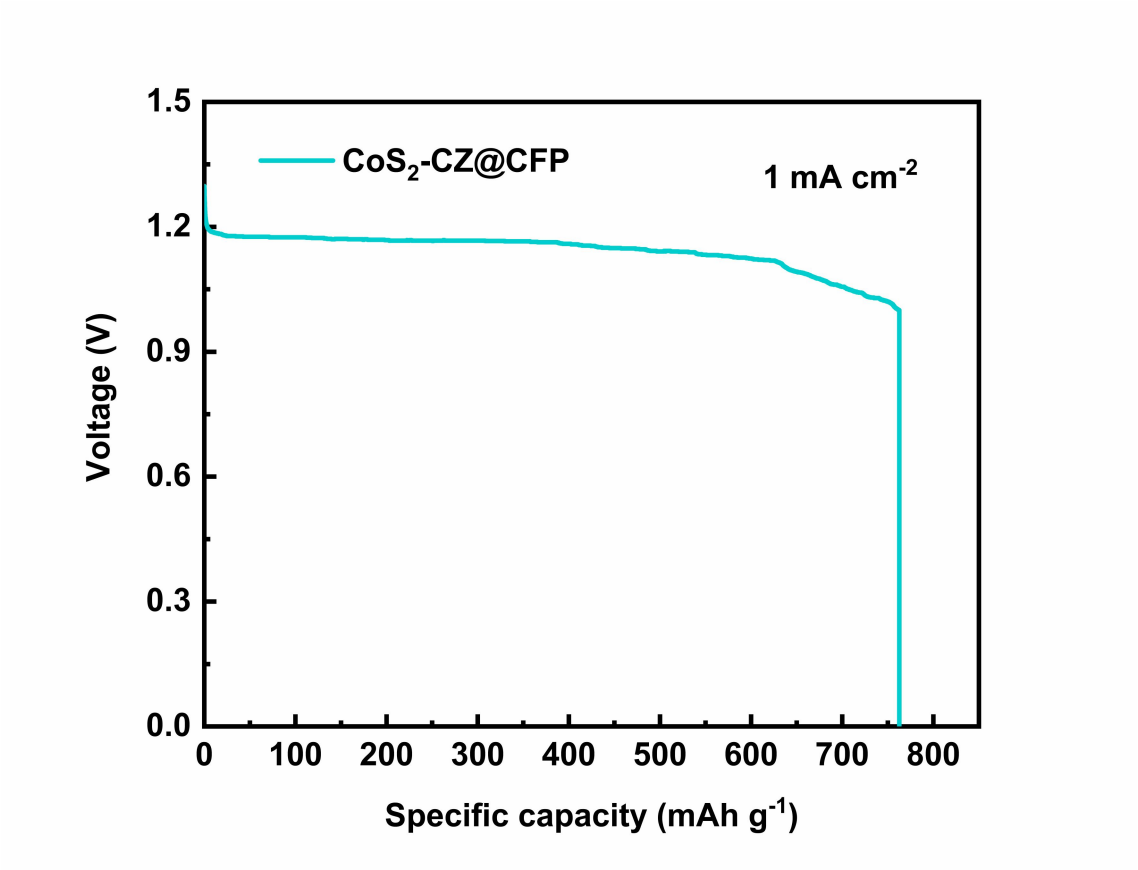


**Fig. S9.** The discharge profiles under 1 mA $cm^{-2}$ constant current of $CoS_2$-CZ@CFP.

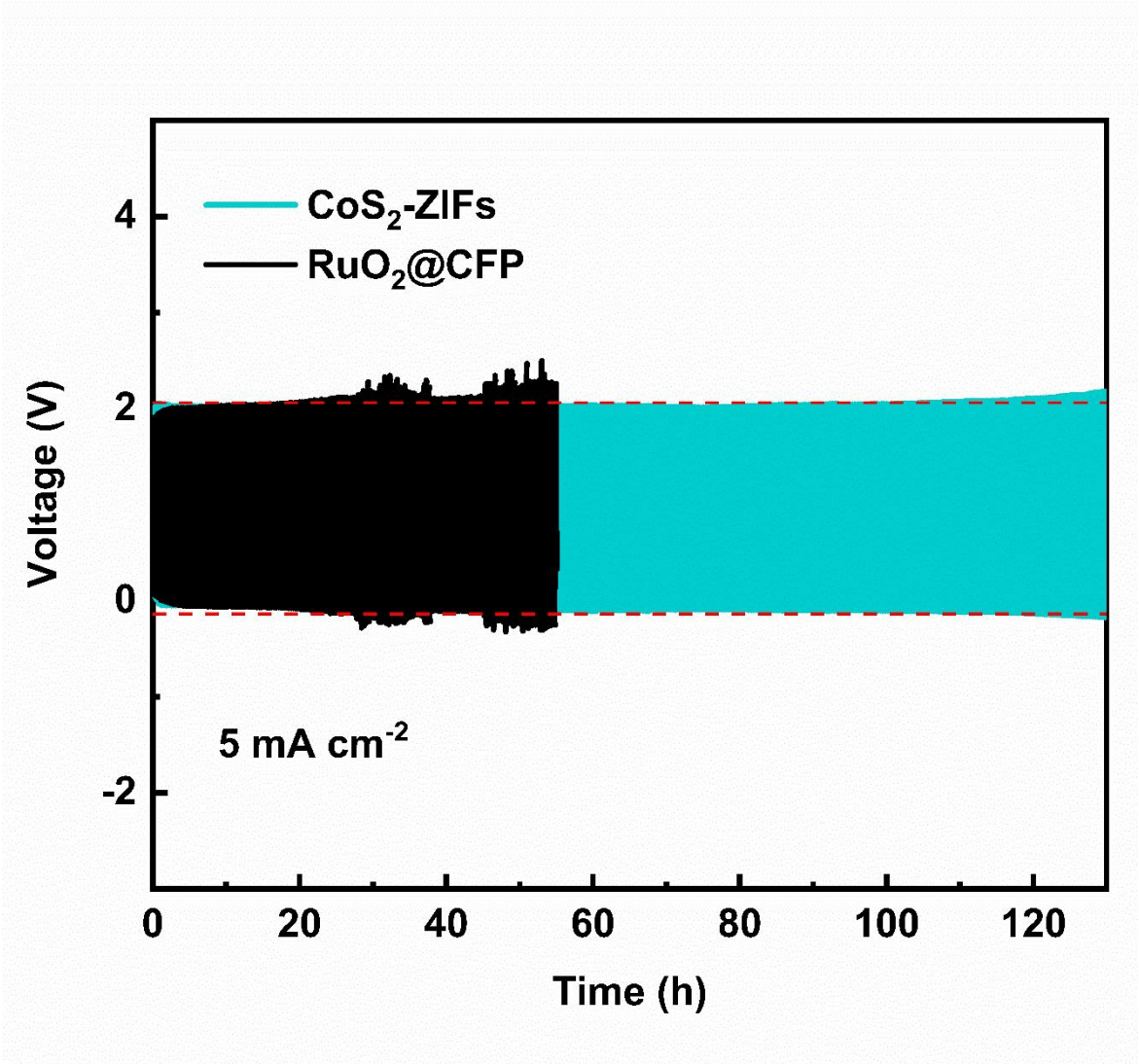


**Fig. S10.** Galvanostatic charge and discharge cycling curve of the two-dimensional solid-state ZABs at 5 mA $cm^{-2}$.