

## **Supporting Information**

### **Artificial Solid-Electrolyte Interface Facilitating Dendrite-Free Zinc Metal Anodes via Nano-Wetting Effect**

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## Experimental Section

*Preparation of UiO-66:* The synthesis of UiO-66 MOF is based on the procedure in the previously reported literature.<sup>1</sup> The average particle size of MOF was measured to be approximately 100 nm.

*Preparation of MOF coated Zn foil:* A slurry was prepared by mixing UiO-66 and PVDF in a weight ratio of 9:1 in N-methyl pyrrolidone (NMP) solvent, followed by at least 6 hours of magnetic stirring. Then, the prepared slurry was coated on Zn foil and dried in vacuum at 80 °C for 8 hours. The thickness of the coating layer was approximately 8-10  $\mu\text{m}$ .

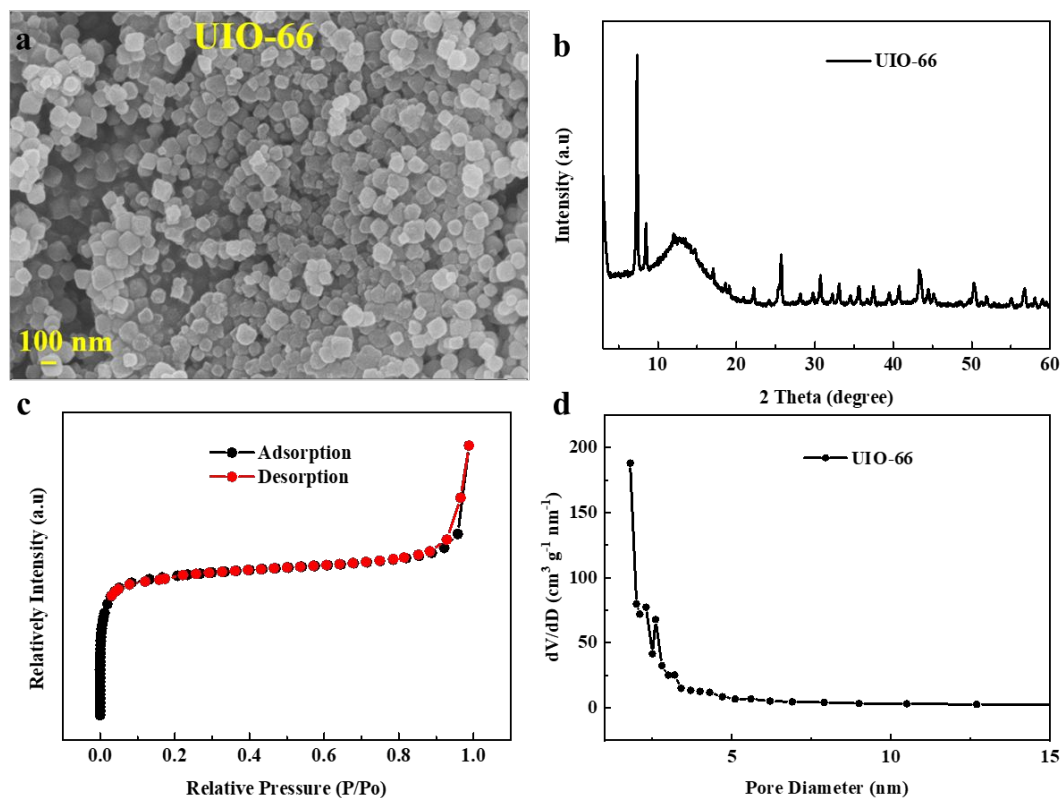
*Preparation of MnO<sub>2</sub> electrode:* MnO<sub>2</sub> was synthesized according to the previous literature.<sup>2</sup> The cathode was prepared by mixing 70 wt% of the as-prepared MnO<sub>2</sub>, 20 wt% acetylene black, and 10 wt% PVDF, stirred in NMP for 8 hours to form a homogenous slurry. Then, the slurry was coated on Ti foil and dried in vacuum at 80 °C for 8 hours. The areal loading of MnO<sub>2</sub> was approximately 1.6 mg cm<sup>-2</sup>.

*Material Characterization:* Scanning electron microscope (SEM, SUPRA 55 SAPHIRE, Carl Zeiss AG, Germany) and energy dispersive spectrometer (EDS, 51-XXM1004, Oxford Instruments Inc., America) were used to observe the surface morphology and elements distribution of the samples. Powder X-ray diffraction (XRD, D8 ADVANCE, Bruker Co., Germany) was used to record the data of MOF by using Cu K $\alpha$ ,  $\lambda=1.541 \text{ \AA}$ , with  $2\theta$  from 3° to 60°. Brunner–Emmet–Teller (BET) measurements of N<sub>2</sub> absorption/desorption isothermal and the pore size distributions were measured by a Micromeritics ASAP 2020 HD88 instrument.

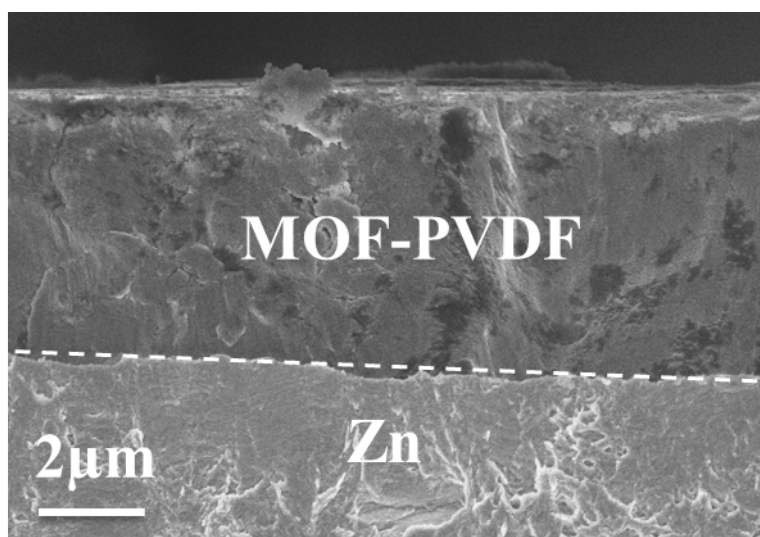
*Electrochemical Tests:* Coin cells (CR2032) were assembled in ambient atmosphere. Zn pellets ( $\Phi = 15 \text{ mm}$ ) and fibre paper (55% cellulose + 45% polyester) ( $\Phi = 17 \text{ mm}$ ) were used as anode and separator, respectively. The electrolyte was prepared by dissolving 3 M zinc sulfate heptahydrate (ZnSO<sub>4</sub>·7H<sub>2</sub>O) and 0.1 M manganese sulfate (MnSO<sub>4</sub>) in DI water. Electrochemical impedance spectroscopy (EIS, 1–10<sup>6</sup> Hz) and cyclic voltammetry (CV) data were collected on a CHI600E electrochemical

workstation. Galvanostatic cycling was performed in a NEWARE battery testing instrument. The onset potential of Zn deposition/stripping was measured in a three-electrode cell, where Pt and Ag/AgCl electrode were used as counter electrode and reference electrode, respectively.

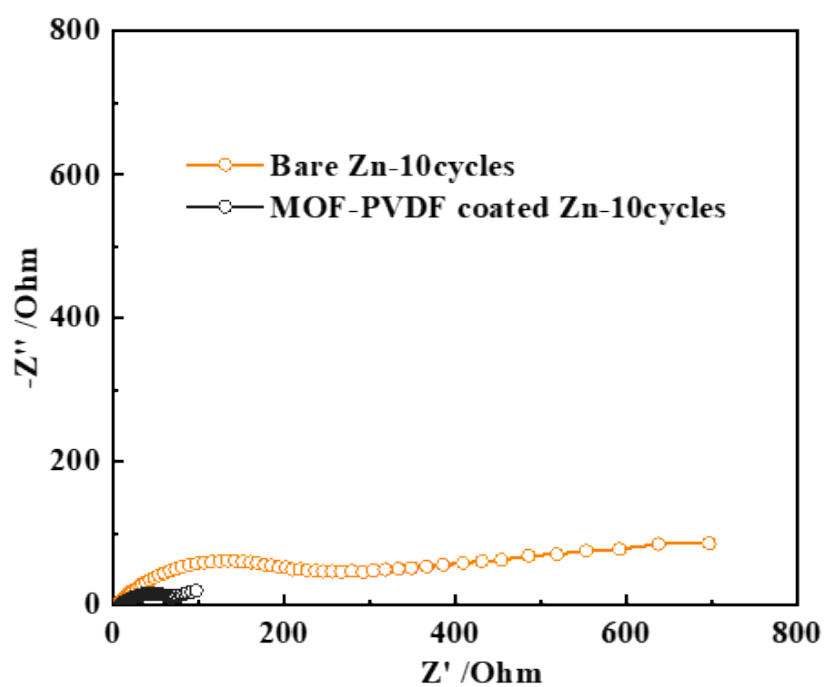
## Additional Figures



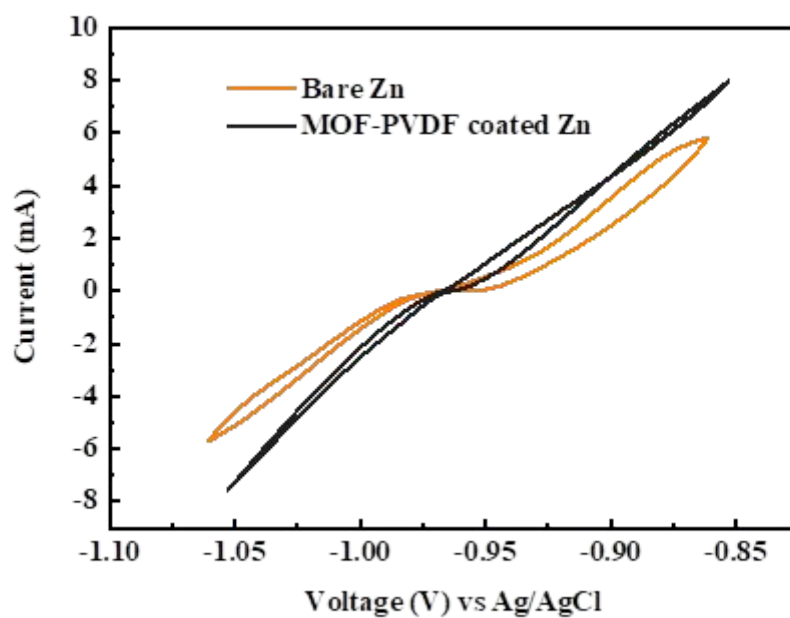
**Figure S1.** (a) SEM image, (b) XRD patterns, (c)  $N_2$  adsorption-desorption curves and (d) pore size distribution of UIO-66 MOFs.



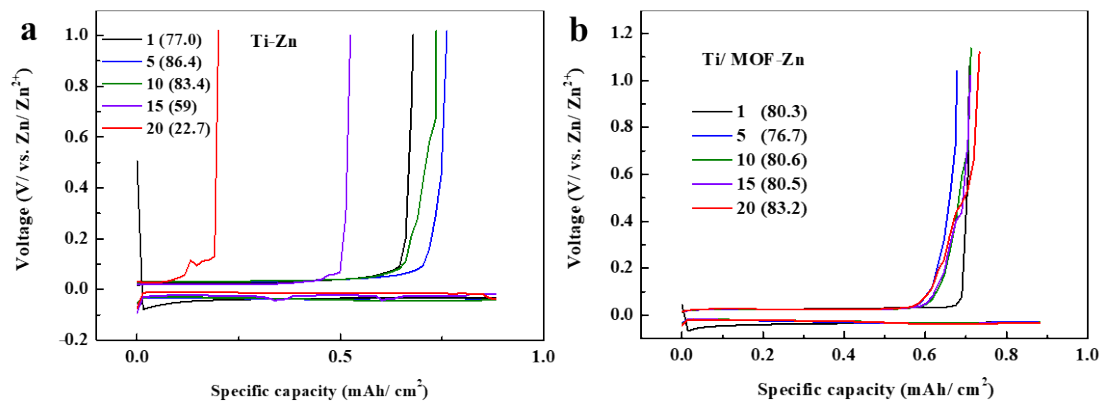
**Figure S2.** Cross section image of MOF-PVDF coated Zn electrode.



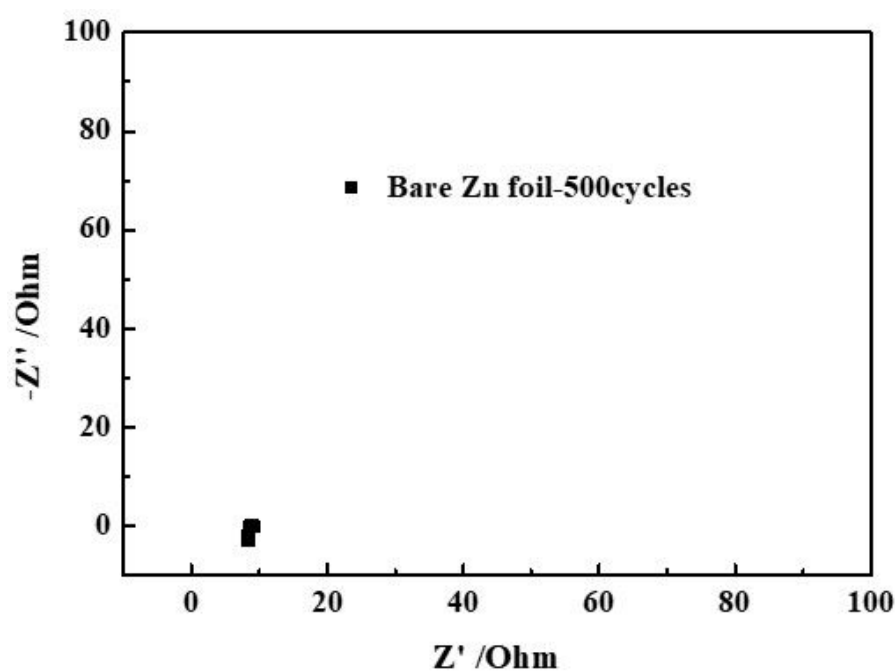
**Figure S3.** Electrochemical impedance spectra of Zn-symmetric cells with different anodes.



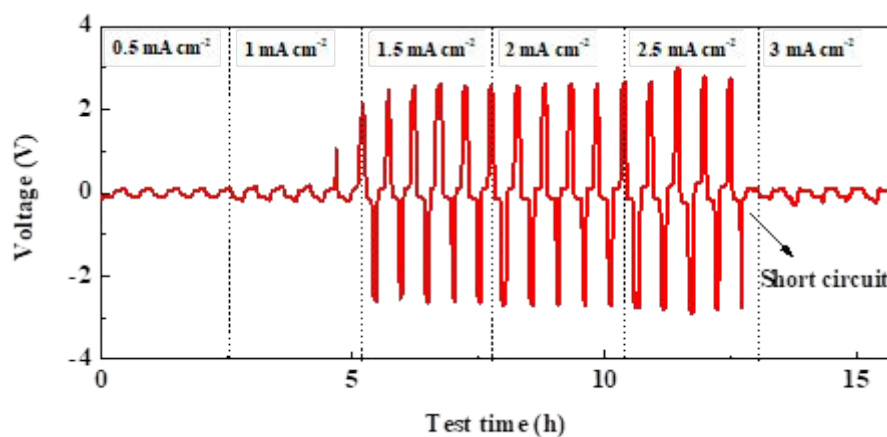
**Figure S4.** Cyclic votammogrammes of Zn plating/stripping behaviors for different electrodes in a three-electrode system.



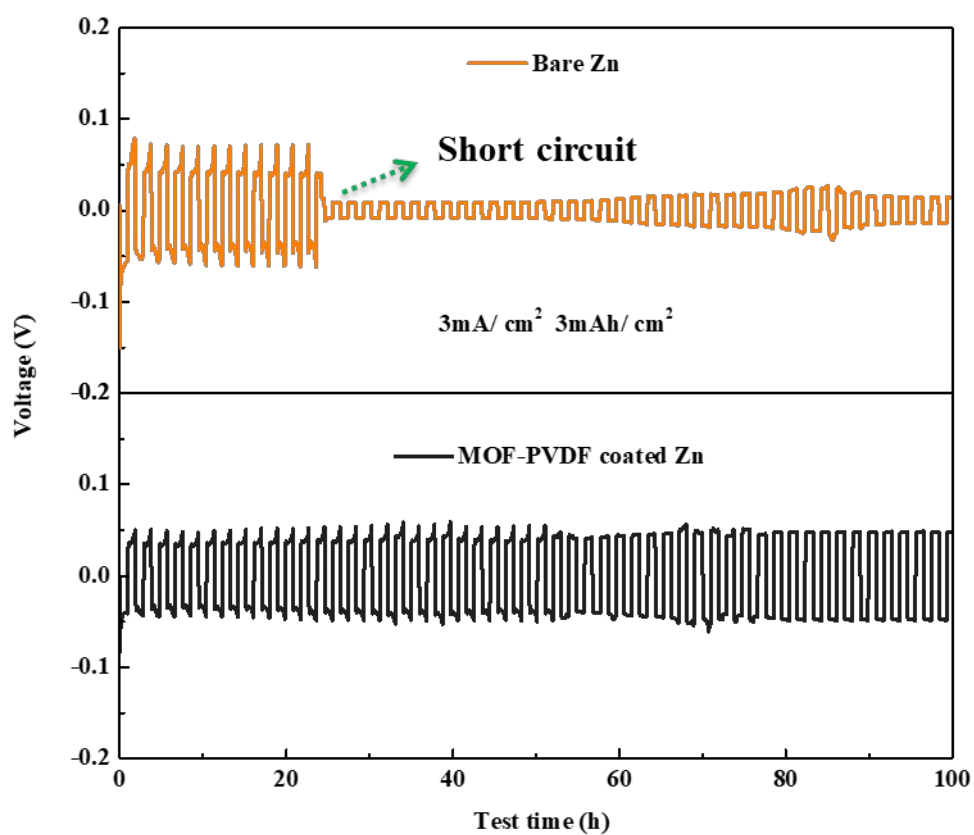
**Figure S5.** Coulombic efficiencies and voltage curves of Zn plating/stripping on bare (a) Ti and (b) coated Ti at 0.5 mA cm<sup>-2</sup> for different cycle numbers.



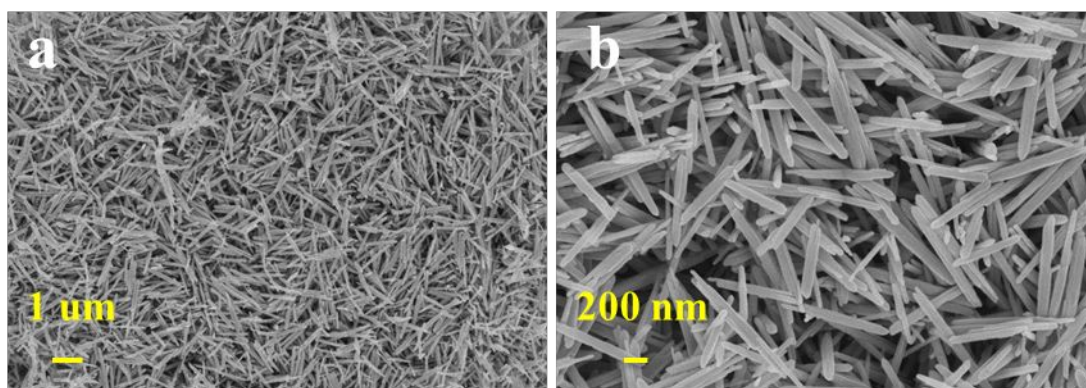
**Figure S6.** Electrochemical impedance spectra of Zn-symmetric cell after sudden voltage drop.



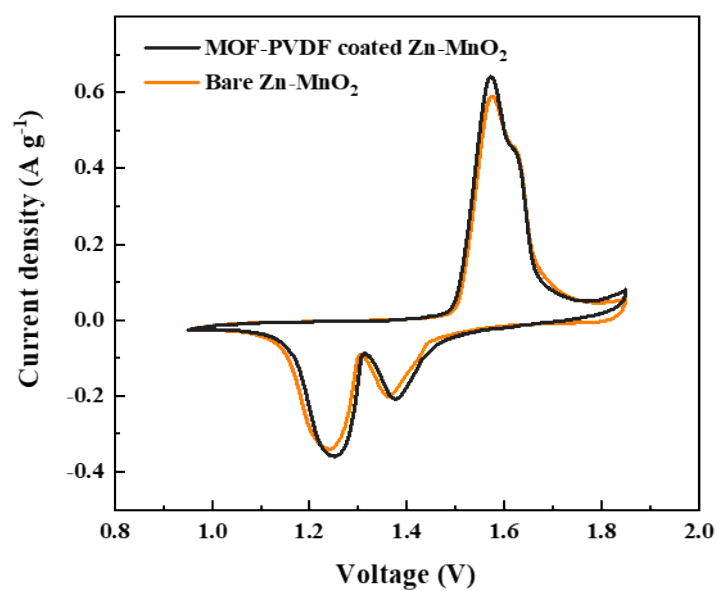
**Figure S7.** Voltage profiles of Zn-symmetric cell with PVDF-coated Zn electrode using step-up Zn plating/stripping current densities.



**Figure S8.** Voltage profiles of Zn-symmetric cell with bare Zn and PVDF-coated Zn at a current density of 3 mA/ cm<sup>2</sup> for 3 mAh/ cm<sup>2</sup>.

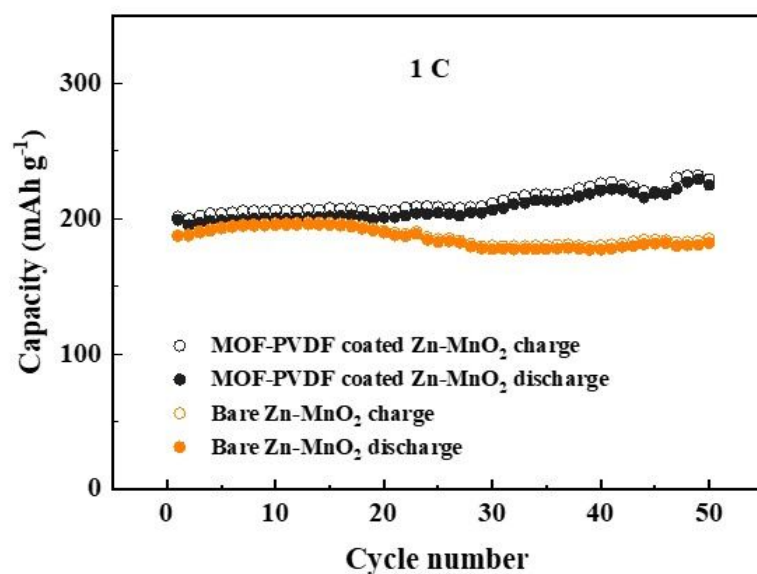


**Figure S9.** SEM images of as-prepared  $\beta$ - $\text{MnO}_2$  cathode material.



**Figure S10.** Cyclic voltammograms of Zn/MnO<sub>2</sub> cells with different anodes.





**Figure S11.** Cycling performance of Zn/MnO<sub>2</sub> cell with MOF-PVDF coated anode.

## Reference

- (1) Wang, S.; Morris, W.; Liu, Y.; McGuirk, C. M.; Zhou, Y.; Hupp, J. T.; Farha, O. K.; Mirkin, C. A. Surface-Specific Functionalization of Nanoscale Metal-Organic Frameworks. *Angew. Chemie - Int. Ed.* **2015**, *54*, 14738–14742.
- (2) Zhang, N.; Cheng, F.; Liu, J.; Wang, L.; Long, X.; Liu, X.; Li, F.; Chen, J. Rechargeable Aqueous Zinc-Manganese Dioxide Batteries with High Energy and Power Densities. *Nat. Commun.* **2000**, *8*, 405.