Supplementary Information (SI) for ChemComm.

Achieving Dendrite-Free Growth of Zn Anode by Electrodepositing on Zincophilic Gold-Furnished Mesh for Aqueous Zinc-Ion Batteries

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Experimental

Materials

Bare GFM was received from XGD Innovation Technology Service Studio. The Au coating layer has a thickness of approximately 1 um, contributing to 14% of the total mass. Bare Zn foil (80 um), Ti foil (20 um), Cu foil (20 um), Cu foam (0.5 mm), Ni Foam (0.5 mm), Zinc sulfate heptahydrate ($ZnSO_4 \cdot 7H_2O$, AR), Vanadium pentoxide (V_2O_5, AR), Oxalic acid ($H_2C_2O_4$, 99%), Magnesium acetate [$Mg(CH_3COO)_2$, 98%], and Poly(vinylidene fluoride) (PVDF) were purchased from Aladdin Biochemical Technology Co., Ltd. And 1-Methyl-2-pyrrolidinone (NMP, AR) was purchased from Chron Chemicals Co., Ltd. No further treatment was conducted on these chemicals before use.

Fabrication of Zn@GFM

The Zn@GFM anode was synthesized by an electrochemical reduction process at room temperature. The GFM was sonicated in ethanol to clean the surface and employed as the working electrode in a three-electrode system with the Zn foil as the counter electrode, Ag/AgCl as the reference electrode, and $ZnSO_4$ aqueous solution (2 M) as the electrolyte. The Zn was deposited on the cleaned GFM by chronopotentiometry at the current density of 40 mA cm^{-2} for 1800 s to obtain the electrode with an electrodeposition capacity of 20 mAh cm^{-2}. Then the obtained electrode was washed with deionized water to remove the residual electrolyte. Finally, the prepared anode was dried at room temperature (Zn@GFM).

Fabrication of VO_2 cathode

The VO_2 was prepared by the hydrothermal method[1]. Firstly, 3 mmol V_2O_5 and 9 mmol $H_2C_2O_4$ were placed in a beaker with 70 mL deionized water and stirred for 4 h. Then, 3 mmol $Mg(CH_3COO)_2$ was added to the above solution. After dissolution, the mixture was transferred into a 100 mL Teflon-lined autoclave and sealed. The sealed autoclave was heated to 160 °C and maintained for 72 h. After cooling, the precipitates were collected by centrifugation and washed several times with deionized water and then with ethanol. Finally, the products were dried in a vacuum at 70 °C for 24 h. To prepare the cathode, VO_2 powder, acetylene black and PVDF were firstly mixed in NMP solvent at a weight ratio of 7: 2: 1. Then the slurry was coated on the Ti foil and dried at 80 °C. The average mass loading of VO_2 per electrode was about 1.0 mg.

Fabrication of batteries and electrochemical measurements

In symmetrical cells, two identical Zn@GFM or bare Zn were used as the anode and cathode of the cell. The Zn deposition process was explored in asymmetrical cells using bare Zn as the anode. In Zn/Cu asymmetrical cells, Cu foil was used as the working electrode and bare Zn or Zn@GFM as the counter electrode. In the Zn/VO_2 full cells, Zn@GFM or bare Zn was used as the anode and VO_2 as the cathode. All electrodes were cut into round disks of 12 mm diameter and assembled into CR2032-type coin cells in the air, using 2 M $ZnSO_4$ aqueous electrolyte (90 uL) and glass fiber (0.68 mm, Whatman, GF/D) separator. The galvanostatic charging-discharging (GCD) of the cells was tested by a Land CT3002A test system (Wuhan LAND Electronic Co. Ltd., Wuhan, China). Linear sweep voltammetry (LSV), cyclic voltammetry (CV), and electrochemical impedance spectroscopy (EIS) measurements were conducted on the CHI660E electrochemical workstation. The LSV test was conducted at a scanning rate of 2 mV s^{-1} in Na_2SO_4 (1 M) electrolyte. The scan rate for CV testing of Zn/VO_2 full cells was 1 mV s^{-1}. The frequency of EIS tests for symmetric cells ranges from 100 kHz to 0.02 Hz and for full cells from 100 kHz to 0.01 Hz, both with an amplitude of 5 mV. The limiting current tests were performed in a three-electrode system, with a glassy carbon electrode as the working electrode, Zn foil as the counter electrode, and an Ag/AgCl electrode as the reference electrode. The scan rate was 5 mV s^{-1}. The Sand's time tests under overlimiting current conditions were performed using GMF, Cu foam, and Ni foam as working electrodes, with Zn foil as the counter electrode and an Ag/AgCl electrode as the reference electrode.

Material Characterization

The components and crystalline structures of samples were characterized by X-ray diffraction (XRD, Empyrean, PANalytical B.V., Almelo, The Netherlands) with Cu Kα radiation at a scanning rate of 5° min^{-1}. The morphologies of the electrodes were characterized by scanning electron microscopy (SEM, Helios 5 CX, Thermo Scientific, Waltham, MA, USA).

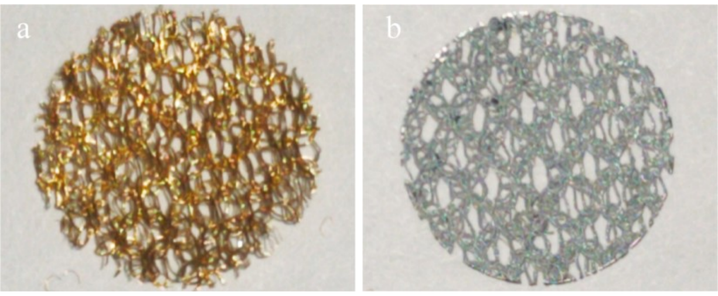

Fig. S1 Photographs of a) GFM and b) Zn after electrodeposition on the GFM surface.

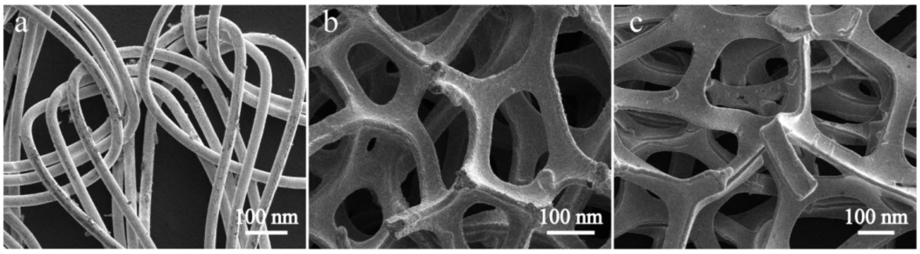

Fig. S2 SEM images of a) GFM, b) Cu foam, and c) Ni foam.

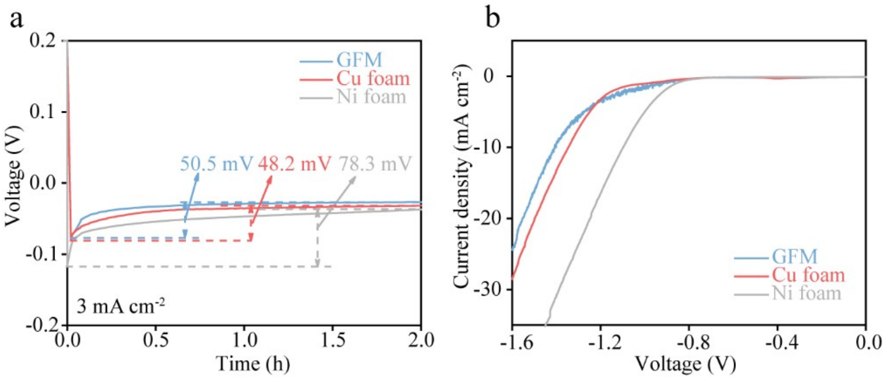

Fig. S3 a) Voltage profiles of Zn nucleation on different substrates at 3 mA cm^{-2}. b) LSV curves of different substrates in 1 M Na_2SO_4 electrolyte.

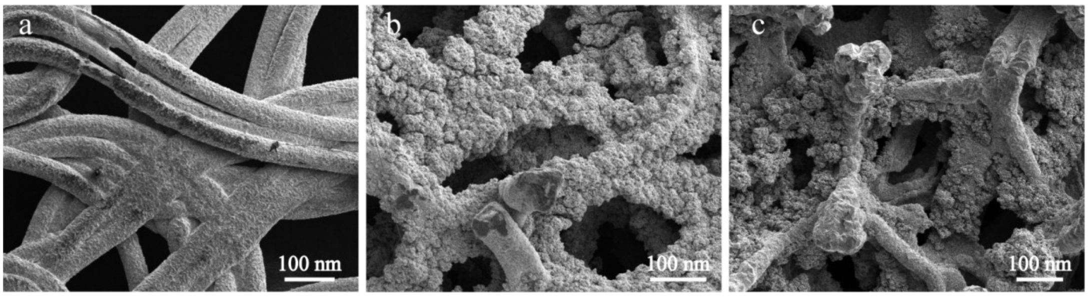

Fig. S4 SEM images of Zn electrodeposited on the a) GFM, b) Cu foam, and c) Ni foam at 40 mA cm^{-2}/10 mAh cm^{-2}.

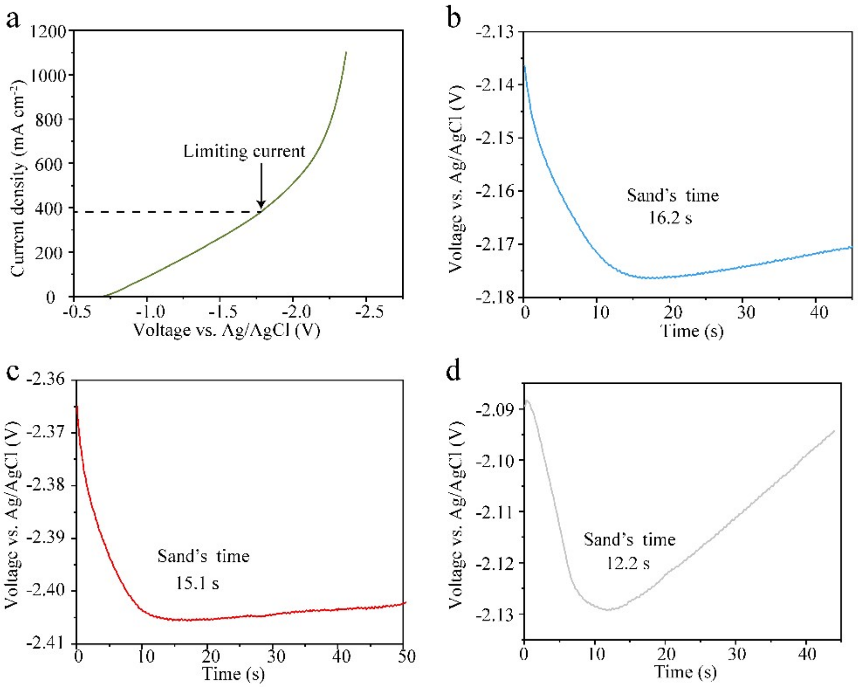

Fig. S5 Electrochemical measurements of a) the current density-voltage curve tested in a three-electrode system to determine the limiting current density. Comparison of the V-t profiles of the b) GFM, c) Cu foam, and d) Ni foam.

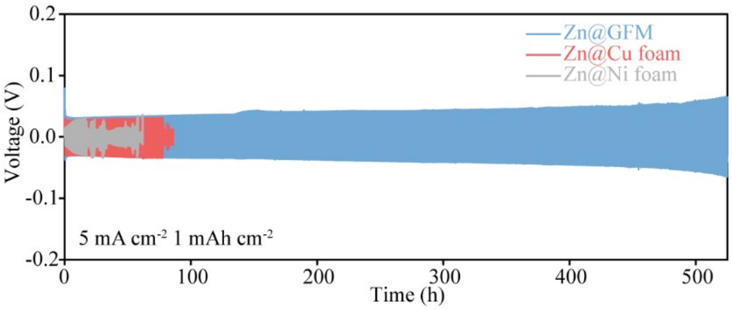

Fig. S6 The cycling performance of symmetrical cells assembled with electrodes made of different current collectors at 5 mA cm^{-2}/1 mAh cm^{-2}.

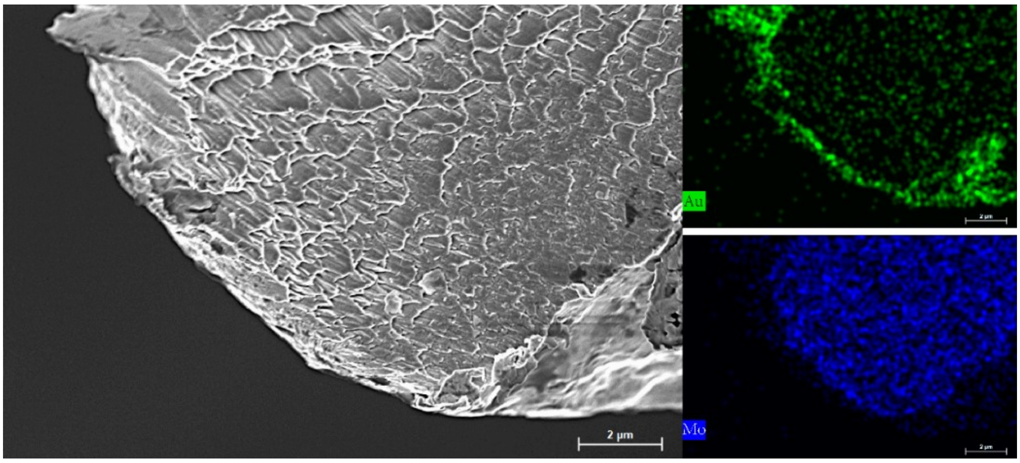

Fig. S7 SEM image and EDS elemental mapping of GFM

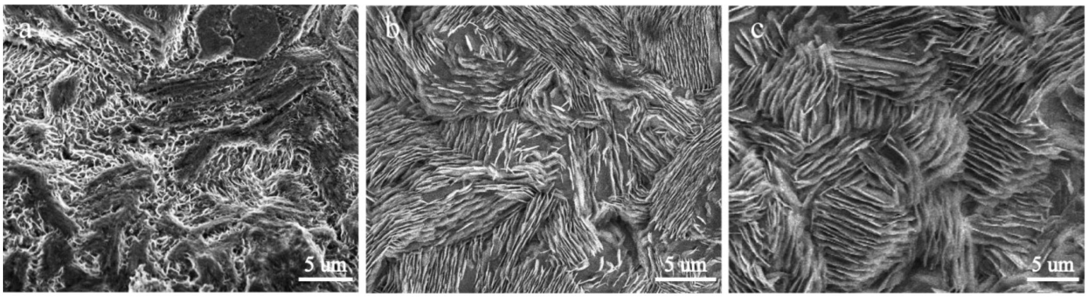

Fig. S8 SEM images of electrodes prepared at different current densities of a) 20 mA cm^{-2}, b) 60 mA cm^{-2}, and c) 80 mA cm^{-2} with an electrodeposition capacity of 10 mAh cm^{-2}.

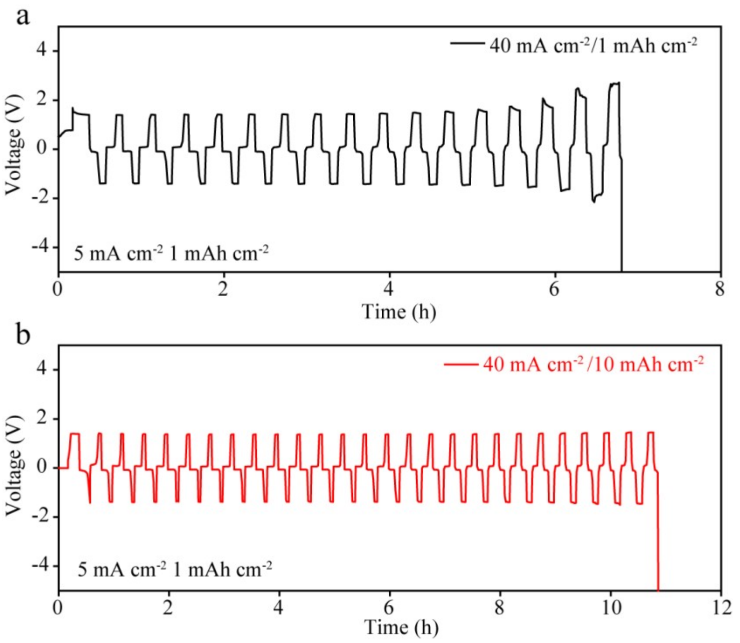

Fig. S9 The cycling performance of symmetrical cells assembled with electrodes prepared at different electrodeposition capacities of a) 1 mAh cm^{-2} and b) 10 mAh cm^{-2} with the current density of 40 mA cm^{-2}.

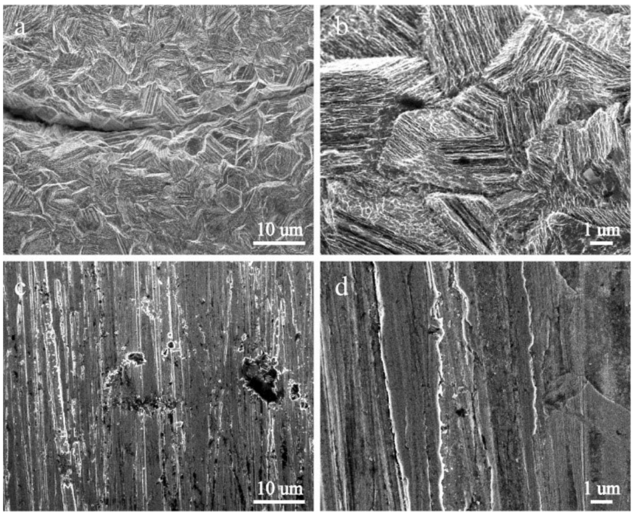

Fig. S10 SEM images of the surface of a, b) Zn@GFM and d, e) bare Zn at different magnifications.

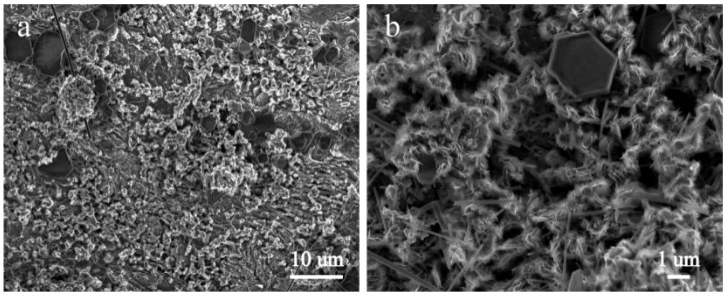

Fig. S11 SEM images of bare Zn after 100 cycles at 5 mA cm^{-2}/1 mAh cm^{-2}.

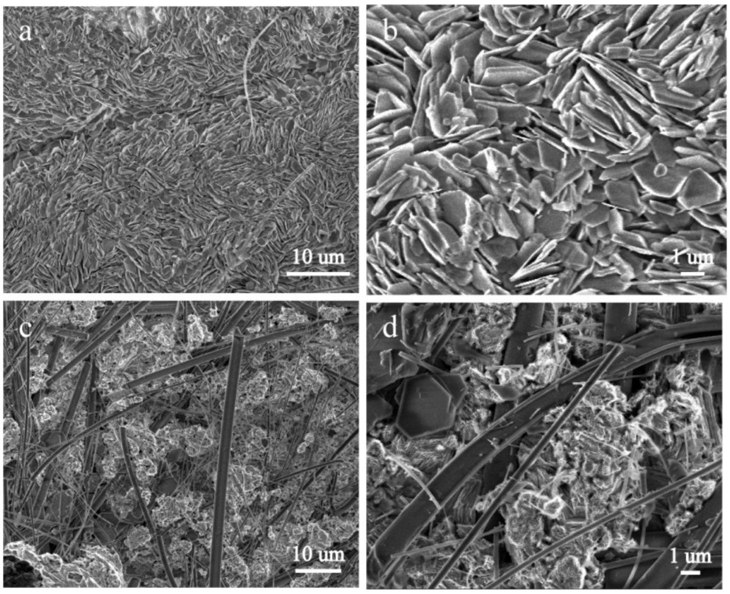

Fig. S12 SEM images of a, b) Zn@GFM and c, d) bare Zn after 400 cycles at 5 mA cm^{-2}/1 mAh cm^{-2}.

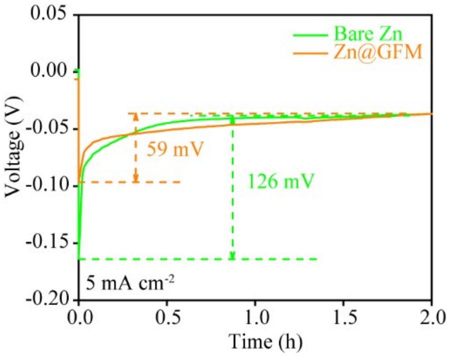

Fig. S13 Voltage profiles of Zn nucleation on Zn@GFM and bare Zn electrodes at 5 mA cm^{-2}.

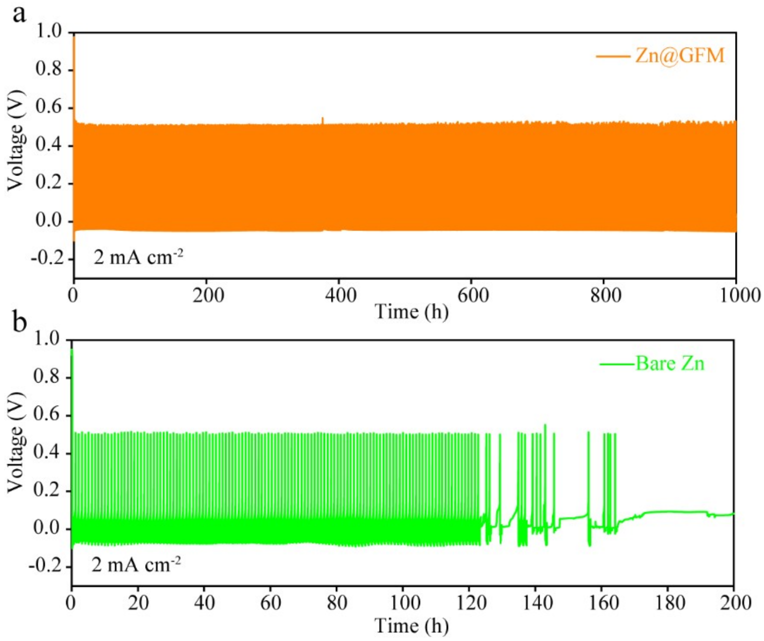

Fig. S14 Cycling performance of Zn plating/stripping process of a) Zn@GFM/Cu and b) bare Zn/Cu half cells, respectively.

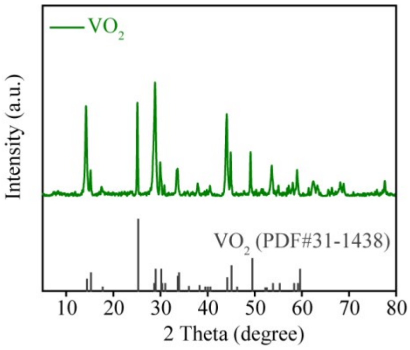

Fig. S15 XRD pattern of VO_2.

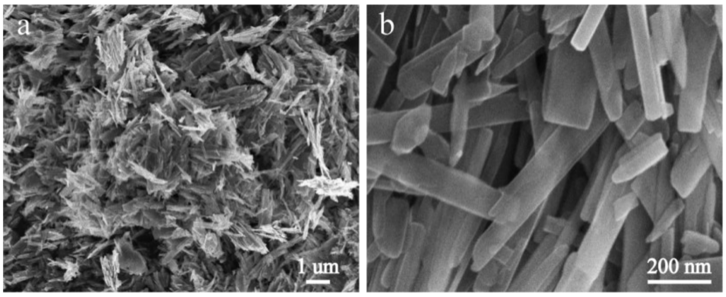

Fig. S16 SEM images of VO_2.

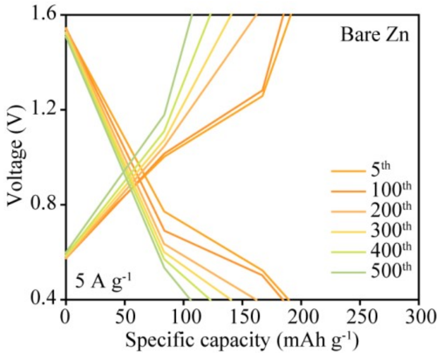

Fig. S17 The charging/discharging profiles of the bare Zn/VO_2 full cell at different cycles.

Notes and references

1 K. Guan, L. Tao, R. Yang, H. Zhang, N. Wang, H. Wan, J. Cui, J. Zhang, H. Wang and H. Wang, *Adv. Energy. Mater.*, 2022, **12**, 2103557.