Electronic Supplementary Information

Interlayer Engineering-Induced Charge Redistribution in Bi_2Te_3 toward Efficient Zn^{2+} and NH_4^+ Storage

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EXPERIMENTAL SECTION

Preparation of Bi₂**Te**₃, **Bi**₂**Te**₃@**PEDOT-x (x = 1, 2, 3) nanosheets, and PEDOT.** 0.5 mmol Bi₂O₃ (Aladdin, 99%), 1.5 mmol TeO₂ (Bidepharm, 99.99%), and 0.4 g of polyvinylpyrrolidone (Bidepharm, PVP, Mw \approx 40,000) were completely dissolved in 20 mL of ethylene glycol (Kermel, 99.5%) by stirring and sonication, then 2 mL of 2 mol L⁻¹ NaOH (Xilong scientific, 96%) solution was added and stirred continuously for 20 minutes. The mixed solution was transferred to a 50 mL PTFE-lined stainless steel autoclave and kept at 200 °C for 260 min. The products were washed four times with ethanol and dried to obtain Bi₂Te₃ powders, the synthesis yield of Bi₂Te₃ is about 95%.

100 mg Bi₂Te₃ powders and different doses of 3,4-ethylenedioxythiophene (EDOT, Aladdin, 99%) monomer (100, 150, and 200 μ L) were dispersed in 6 mL of acetonitrile (Kermel, 99.5%) and stirred for 30 minutes (Denoted as A). At the same time, different amounts of FeCl₃ (Macklin, 99%) (228, 342, and 456 mg) were dissolved in 4 mL of acetonitrile (Denoted as B). Then B was slowly added to A and stirred for 3 hours. Finally, the sample was obtained by washing with ethanol and drying under vacuum at 80 °C. The products obtained from different doses of EDOT were denoted as Bi₂Te₃@PEDOT-1, Bi₂Te₃@PEDOT-2 (Bi₂Te₃@PEDOT), and Bi₂Te₃@PEDOT-3.

912 mg FeCl₃ was dissolved in 10 mL acetonitrile, then 400 μ L of EDOT was added and stirred continuously for 3 h. The sample obtained was washed with ethanol and then dried at 80 °C to obtain PEDOT powder.

Preparation of free-standing electrodes. To obtain a free-standing electrode, $Bi_2Te_3/Bi_2Te_3@PEDOT-x/PEDOT$ and multi-walled CNTs (purity: >95%, diameter: 8–15 nm, length: \approx 50 µm, XFNANO) with a weight ratio of 7 : 3 were dispersed in ethanol, and the solution was filtered with a microporous filtration membrane. The obtained film was dried in a vacuum oven to form a free-standing and flexible film. It was cut into small disks for electrochemical performance tests. The areal weight of active material was 5 mg cm⁻². Similarly, the high mass loading electrodes with 10 mg cm⁻² were also prepared.

Characterization. The crystal structure, molecular structure, and elemental composition were analyzed through X-ray diffraction (XRD, D-MAX 2200 VPC, Rigaku) pattern with Cu K α radiation ($\lambda = 1.5418$ Å), Raman spectra (inVia, Renishaw) with a laser length of 514 nm, Fourier transform infrared (FT-IR, Nicolet 6700, Thermo Scientific) spectra, and X-ray photoelectron spectroscopy (XPS, ESCALab250, Thermo VG) spectra corrected by C 1s line at 284.6 eV. The scanning electron microscope (SEM, Gemini500, Zeiss) and transmission electron microscope (TEM, Tecnai G2 F30, FEI) were utilized to reveal the morphology evolution of samples. Nitrogen adsorption/desorption isotherms were collected to count the specific surface area and pore volume of samples (ASAP2460, Micromeritics).

Electrochemical measurements. Battery tests were carried out with a 2025-type coin cell. For half-cell, $Bi_2Te_3/Bi_2Te_3@PEDOT$ -x flexible film was used as working electrode, Zn foil and active carbon acted as counter/reference electrode for Zn^{2+} and NH_4^+ storage respectively. The electrolytes were aqueous solution with 2 mol L^{-1} ZnSO₄ (Aladdin, 99%) and hybrid solution with 2 mol L^{-1} CH₃COONH₄ (Aladdin, 99%) (the volume ratio of deionized water to 1,3-Dimethyl-2-imidazolidinone (Aladdin, 99%) is 1 : 1). Glass fiber membrane (Whatman, GF/D) worked as separator. MnO₂ (Aladdin, 99%) and ZnMn₂O₄ cathodes were pasted on stainless steel foils via preparing slurry with 70% active material, 20% acetylene black, and 10% poly (vinylidene fluoride). The "rocking-chair" ZIBs/AIBs were assembled using MnO₂/ZnMn₂O₄, $Bi_2Te_3@PEDOT$, and 2 mol L^{-1} ZnSO₄ and 2 mol L^{-1} CH₃COONH₄ (V (H_2O) : V (DMI) = 1 : 1) as cathode, anode, and electrolyte. The N/P values of full cells were about 1.00. The battery tests were performed in the voltage windows of 0.2-1.5/–1-0.6 V (half-cell) and 0.1-1.6/0-1.2 V (full cell) by a NEWARE battery test system (CT-4008-5V20mA-164, Shenzhen, China). Cyclic voltammetry (CV) curves and electrochemical impedance spectra (EIS, Frequency: 0.1-100000 Hz) were tested through utilizing electrochemical workstation (DH7000, Jiangsu Donghua Analytical Instrument Co., Ltd.). Galvanostatic intermittent titration technique (GITT) tests were collected via a NEWARE battery test system with a current pulse of 30 mA g⁻¹ and a potential relaxation step for 2 h at open-circuit voltage.

Details of calculation. The dominant capacitive behavior or diffusion process of the samples could be evaluated according to the following equation: $i = av^b$, where *i* and *v* represented peak current and scan rate. The capacitive behavior or diffusion process dominated when *b* was about 1 or 0.5. The Zn²⁺ diffusion coefficients of electrodes are counted by GITT curves

$$D = \frac{4}{\pi \tau} \left(\frac{m_B V_M}{M_B S} \right)^2 \left(\frac{\Delta E_S}{\Delta E_z} \right)^2$$

and Fick's second law: $\pi\tau (M_B S)^{-\Delta E_{\tau}}$, where m_B , S, V_M and M_B represent weight, surface area, the molar volume and the molar mass of active material, τ corresponds to the time of current pulse, ΔE_S represents the steady-state voltage change by the current pulse, ΔE_{τ} is the potential difference of constant current pulse charging or discharging.

DFT calculations. All the first-principles calculations in this work were performed by using the Vienna ab initio simulation package (VASP),¹ in which the interactions of core electrons were described by the projector augment wave (PAW) potentials and the exchange and correlation functional were modeled by the Perdew-Burke-Ernzerhof (PBE) functionals derived from the generalized gradient approximation (GGA).²⁻⁴ A cut-off energy of 500 eV

was used for the plane-wave basis set. The Brillouin zone was sampled by using a $13 \times 13 \times 2$ Monkhorst-Pack k-point mesh for structural optimizations of unit cell BiTe, a $2 \times 1 \times 1$ k-point mesh for structural optimizations of $2 \times \sqrt{3} \times 1$ supercells, and a $7 \times 4 \times 2$ k-point mesh for electronic structure calculations of supercells. The convergence criteria for energy is set to 10^{-5} eV and The convergence criteria for force is set to 0.01 eV/A for unit cell and 0.04 eV/A for supercells, respectively. The van der Waals (vdW) interaction is considered by adopting the DFT-D3 method (an empirical dispersion correction scheme of Grimme).⁵



Figure S1. (A) XRD pattern, (B) Raman spectrum, and (C) FT-IR spectrum of PEDOT.

The pure PEDOT is characterized by means of multiple methods. The XRD peak at about 26° is attributed to the interchain planar ring stacking (Fig. S1a). The characteristic peaks of PEDOT are located at 1263, 1365, 1435, 1510, and 1566 cm⁻¹ in the Raman spectrum (Fig. S1b). These are assigned to the C-O-C deformation, symmetric C-S-C deformation, oxyethylene ring deformation, C α -C α inter-ring stretching, symmetric C α = C β (-O) stretching, respectively. In the FT-IR spectrum, the peaks at about 1515 and 1315 cm⁻¹ match with asymmetric stretching mode of C=C and inter-ring stretching mode of C-C, respectively. The peaks at around 1187, 1138, 1083, and 1048 cm⁻¹ correspond to the C-O-C bending vibration in ethylenedioxy group. Moreover, the peaks at 972, 915, 832, and 674 cm⁻¹ are attributed to the characteristic bands of stretching vibrations of the C-S-C bond in thiophene ring (Fig. S1c).



Figure S2. (A) Bi 4f XPS spectra of Bi₂Te₃ and Bi₂Te₃@PEDOT. (B) C 1s and (C) S 2p XPS spectra of Bi₂Te₃@PEDOT. (D) FT-IR spectra of Bi₂Te₃ and Bi₂Te₃@PEDOT.

In the C 1s spectrum, peaks at 285.8 and 287.4 eV correspond to the C-O and C-S bonds of PEDOT, respectively (Figure S2B).⁶ The S 2p spectrum shows a peak at approximately 163.5 eV, indicating the presence of the C-S-C bond, which confirms the existence of PEDOT in $Bi_2Te_3@PEDOT$ (Figure S2C).⁷ Additionally, the FT-IR spectrum (Figure S2D) displays characteristic peaks of PEDOT, further supporting the coating of PEDOT on $Bi_2Te_3@PEDOT.^8$ Therefore, these tests demonstrate the successful synthesis of $Bi_2Te_3@PEDOT$.



Figure S3. (A) Nitrogen adsorption/desorption isotherms and (B) pore distributions of Bi_2Te_3 and $Bi_2Te_3@PEDOT$.



Figure S4. (A, B) TEM images and (C) SAED pattern of Bi₂Te₃@PEDOT.



Figure S5. Crystal structures of (A) Bi₂Te₃ and (B) PEDOT-Bi₂Te₃.



Figure S6. Calculated DOSs of (A) Bi₂Te₃ and (B) PEDOT-Bi₂Te₃.



Figure S7. CV curves of (A) Bi₂Te₃ and (B) Bi₂Te₃@PEDOT.



Figure S8. GCD curves of (A) Bi₂Te₃ and (B) Bi₂Te₃@PEDOT.



Figure S9. Rate performance of PEDOT (5 mg cm^{-2}).



Figure S10. (A) Rate performances and (B) cyclic life of $Bi_2Te_3@PEDOT-x$ (5 mg cm⁻²) at 10 A g⁻¹.



Figure S11. CV curves from 0.4 to 1.0 mV s⁻¹ of (A) Bi₂Te₃ and (B) Bi₂Te₃@PEDOT. (C) Relationship between peak currents (inset of A,B) and scan rates in logarithmic format.



Figure S12. GITT curves of (A) discharge and (B) charge processes and corresponding diffusion coefficients of Bi_2Te_3 and Bi_2Te_3 @PEDOT.



Figure S13. (A) Digital photos and (B) XRD patterns of Bi_2Te_3 and Bi_2Te_3 @PEDOT after immersion in 2 mol L⁻¹ ZnSO₄ solution for 2 h.



Figure S14. Ex-situ Zn 2p XPS spectra of (A) Bi₂Te₃, (B) Bi₂Te₃@PEDOT.



Figure S15. Ex-situ O 1s XPS spectra of (A) Bi₂Te₃ and (B) Bi₂Te₃@PEDOT.



Figure S16. Discharge curves of Bi_2Te_3 @PEDOT in different electrolytes.



Figure S17. GCD curves of (A) Bi₂Te₃ and (B) Bi₂Te₃@PEDOT at different current densities.



Figure S18. (A) Rate performance and (B) cyclic life of PEDOT (5 mg cm⁻²) at 1 A g⁻¹.



Figure S19. CV curves of (A) Bi₂Te₃ and (B) Bi₂Te₃@PEDOT.



Figure S20. CV curves from 0.4 to 1.0 mV s⁻¹ of (A) Bi₂Te₃ and (B) Bi₂Te₃@PEDOT. (C) Relationship between peak currents (inset of A,B) and scan rates in logarithmic format.



Figure S21. GITT curves of (A) discharge and (B) charge processes and corresponding diffusion coefficients of Bi_2Te_3 and Bi_2Te_3 @PEDOT.



Figure S22. (A) Bi 4f and (B) O 1s XPS spectra of Bi₂Te₃@PEDOT at different voltages.



Figure S23. Top views of (A) Zn^{2+} and (B) NH_4^+ diffusion paths in the PEDOT-Bi₂Te₃ bulk.



Figure S24. GCD curves (A) $Bi_2Te_3@PEDOT//MnO_2$ and (B) $Bi_2Te_3@PEDOT//ZnMn_2O_4$ full cells.

Anode	Specific capacity	Cycle stability	Mass loading
Bi ₂ Te ₃ @PEDOT (This work)	370/325 mA h g ⁻¹ @0.2 A g ⁻¹	≈82%@10 A g ⁻¹ @1000 cycles/84%@10 A g ⁻¹ @500 cycles	5/10 mg cm ⁻²
Cu ₇ Te ₄ 9	157 mA h g ⁻¹ @0.2 A g ⁻¹	≈95%@0.5 A g ⁻¹ @4000 cycles	1.6-2 mg cm ⁻²
VSe ₂ ¹⁰	114.6 mA h g ⁻¹ @0.2 A g ⁻¹	≈75.3%@0.5 A g ⁻¹ @500 cycles	2 mg cm ⁻²
Polyimides/CNT ¹¹	115 mA h g ⁻¹ @0.2 A g ⁻¹	≈80%@5A g ⁻¹ @50000 cycles	1.2 mg cm ⁻²
NHVO@Ti ₃ C ₂ T $_x^{12}$	428 mA h g ⁻¹ @0.2 A g ⁻¹	≈84.2%%@5 A g ⁻¹ @6000 cycles	1-2 mg cm ⁻²
MoO ₂ @NC ¹³	138 mA h g ⁻¹ @0.2 A g ⁻¹	≈63%@5 A g ⁻¹ @3000 cycles	1-1.5 mg cm ⁻²
CuS-CTAB ¹⁴	210 mA h g ⁻¹ @0.2 A g ⁻¹	87.6%@10 A g ⁻¹ @3400 cycles	1 mg cm ⁻²
TiS ₂ ¹⁵	118/96 mA h g ⁻¹ @0.4 A g ⁻¹	≈85%@2 A g ⁻¹ @5000 cycles	3/6 mg cm ⁻²
$H_2Ti_3O_7$ ·x H_2O^{16}	86 mA h g ⁻¹ @0.2 A g ⁻¹	87%@0.2 A g ⁻¹ @1400 cycles	4 mg cm ⁻²
PTCDI/rGO ¹⁷	135 mA h g ⁻¹ @0.2 A g ⁻¹	98%@1 A g ⁻¹ @1000 cycles	/
h-WO ₃ /3DG ¹⁸	108.9 mA h g ⁻¹ @0.2 A g ⁻¹	≈89%@2 A g ⁻¹ @10000 cycles	0.8-1 mg cm ⁻²

Table S1. A comparison of electrochemical performances of reported anodes in ZIBs.

Anode	Specific capacity	Cycle stability	Mass loading
Bi ₂ Te ₃ @PEDOT (This work)	235/219 mA h g ⁻ ¹ @0.2 A g ⁻¹	72%@1 A g ⁻¹ @700 cycles/≈68%@2 A g ⁻ ¹ @300 cycles	5/10 mg cm ⁻²
PNNI ¹⁹	140 mA h g ⁻¹ @0.2 A g ⁻¹	80.2%@5 A g ⁻¹ @10000 cycles	3.5 mg cm ⁻²
VO _x @PPy ²⁰	195.4 mA h g ⁻¹ @0.2 A g ⁻¹	84.9%@1 A g ⁻¹ @2000 cycles	3.9 mg cm ⁻²
$PTCDA/Ti_{3}C_{2}T_{x}$ $MXene^{21}$	202.8 mA h g ⁻¹ @0.5A g ⁻¹	74.3%@15 A g ⁻¹ @10000cycles	0.7 mg cm ⁻²
Alloxazine ²²	138.6 mA h g ⁻¹ @1 A g ⁻¹	≈76%@10 A g ⁻¹ @1500 cycles	1-2 mg cm ⁻²
<i>h</i> -WO ₃ ²³	82 mA h g ⁻¹ @1 A g ⁻¹	86%@20 A g ⁻¹ @20000 cycles	3.9 mg cm ⁻²
K-V-Fe PBAs ²⁴	92.8 mA h g ⁻¹ @2 A g ⁻¹	91.4%@2 A g ⁻¹ @2000 cycles	2 mg cm ⁻²
VS_2/VO_x^{25}	170 mA h g ⁻¹ @0.2 A g ⁻¹	43%@1 A g ⁻¹ @1000 cycles	2 mg cm ⁻²
PNNI ²⁶	147.7 mA h g ⁻¹ @0.1 A g ⁻¹	80.2%@5 A g ⁻¹ @10000 cycles	3.5 mg cm ⁻²
a-MoO ₃ / Ti ₃ C ₂ T _z ²⁷	117 mA h g ⁻¹ @0.2 A g ⁻¹	74.8%@10 A g ⁻¹ @10000 cycles	1.5-1.9 mg cm ⁻²
MoS ₂ /MXene ²⁸	118.7 mA h g ⁻¹ @0.2 A g ⁻¹	91.5%@0.1 A g ⁻¹ @200 cycles	/

 Table S2. A comparison of electrochemical performances of reported anodes in AIBs.

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