Supporting Information for

Built-In Electric Field-Driven Ultrahigh-Rate K-Ion Storage via Heterostructure Engineering of Dual Tellurides Integrated with Ti₃C₂T_x MXene

Long Pan^{1, #}, Rongxiang Hu^{1, #}, Yuan Zhang¹, Dawei Sha¹, Xin Cao¹, Zhuoran Li¹, Yonggui Zhao², Jiangxiang Ding³, Yaping Wang^{1, *}, ZhengMing Sun^{1, *}

¹Key Laboratory of Advanced Metallic Materials of Jiangsu Province, School of Materials Science and Engineering, Southeast University, Nanjing 211189, P. R. China

²Department of Chemistry, University of Zurich, Winterthurerstrasse 190, CH-8057 Zurich, Switzerland

³School of Materials Science and Engineering, Anhui University of Technology, Ma'anshan, Anhui 243002, P. R. China

[#] Long Pan and Rongxiang Hu contributed equally to this work.

* Corresponding authors. E-mail: <u>zmsun@seu.edu.cn</u> (ZhengMing Sun); <u>ypwang2011@seu.edu.cn</u> (Yaping Wang)

Supplementary Figures and Tables



Fig. S1 Charge differences of K⁺ adsorption in CoTe₂, ZnTe, and CoTe₂/ZnTe



Fig. S2 (a) K^+ adsorption energy and (b) K^+ adsorption charge differences of CoTe₂/Ti₃C₂T_x (CT) and ZnTe/Ti₃C₂T_x (ZT)



Fig. S3 Side views of the K⁺ diffusion routes in CoTe₂, ZnTe and CoTe₂/ZnTe



Fig. S4 SEM images of (**a**) Ti₃AlC₂ MAX phase, (**b**) multi-layered Ti₃C₂T_{*x*}, and (**c**) exfoliated Ti₃C₂T_{*x*} nanosheets. (**d**) XRD patterns of Ti₃AlC₂ MAX phase, multi-layered Ti₃C₂T_{*x*}, and exfoliated Ti₃C₂T_{*x*} nanosheets. The inset of (**c**) shows TEM image of exfoliated Ti₃C₂T_{*x*} nanosheets



Fig. S5 Elemental mappings of CoZn-MOF/Ti₃C₂T_x



Fig. S6 Size distribution of CoTe₂/ZnTe particles. 100 particles were counted



Fig. S7 SEM images of (a) CT and (b) ZT

Table S1 Atomic ratios of Co and Zn in CoZn-MOF/Ti₃C₂T_x and CZT

Sample	Co (at%)	Zn (at%)	Co/Zn Atomic Ratio
CoZn-MOF/Ti ₃ C ₂ T _x	13.9	10.0	1.39
CZT	8.89	6.09	1.45



Fig. S8 XRD patterns of (a) CT and (b) ZT



Fig. S9 (a) Nitrogen adsorption-desorption isotherms, (b) BET surface area, and (c) pore size distribution of CoZn-MOF/Ti₃C₂T_x. The CoZn-MOF/Ti₃C₂T_x exhibits a pore volume of 0.76 cm³ g⁻¹ with an average pore size of 34.2 nm



Fig. S10 (a) Pore size distribution and (b) pore volume ratios of CZT

XPS peak		CT (eV)	ZT (eV)	CZT (eV)
Co 2p	Satellite	802.32	-	801.95
	2p _{1/2}	796.40	-	795.90
	Satellite	785.57	-	785.23
	2p _{3/2}	780.45	-	780.18
Zn 2p	2p _{1/2}	-	1044.60	1044.39
	2p _{3/2}	-	1021.49	1021.28
Te 3d	Oxide	586.24	586.32	586.05
	3d _{3/2}	582.97	583.45	582.94
	Oxide	575.84	575.95	575.68
	3d _{5/2}	572.58	572.58	572.57

Table S2 Comparation of binding energy in CT, ZT, and CZT



Fig. S11 The first three CV curves of CT and ZT at a scanning rate of 0.1 mV s^{-1}



Fig. S12 TEM images of CZT discharged to 0.01 V and corresponding lattice spacing information

Figure S12 shows the TEM characterization and lattice fringe test results of CZT after discharging to 0.01 V. It can be observed that there are many lattice fringes characterized in the electrode material, and the lattice spacing is approximately 0.225 nm after local magnification and measurement. Corresponding to the (311) crystal plane of K₂Te, the CZT composite material exhibits conversion-type negative electrode characteristics. Combining with the previous studies on K⁺ storage of ZnTe and CoTe₂ separately, it is concluded that the K⁺ storage reaction of CZT is:

$$ZnTe + 2K^+ + 2e^- \rightarrow Zn + K_2Te$$

$$CoTe_2 + 4K^+ + 4e^- \rightarrow Co + 2K_2Te$$



Fig. S13 Cycle performance of CZT at a current density of 0.5 and 2.0 A g^{-1}



Fig. S14 Postmortem SEM images of CZT tested at 0.5 A g^{-1} after various cycles



Fig. S15 The voltage-time curves of CZT, CT, and ZT in the GITT test

The K⁺ diffusion coefficients were calculated based on the diffusion formula:

$$\mathsf{D}_{\mathsf{K}^+} = \frac{4}{\pi\tau} (\frac{m_B V_M}{M_B S})^2 (\frac{\Delta E_s}{\Delta E_t})^2$$

Where τ represents the relaxation time, m_B and S refer to the mass and area of the electrode. V_M and M_B denote the molar volume and molar mass of the electrode. ΔEs represents the steady-state voltage change, and ΔE_t represents the transient voltage change.



Fig. S16 (**a-c**) CV curves of (**a**) CZT, (**b**)CT and (**c**) ZT at various sweeping rates; (**d-f**) Relationship between peak current and sweeping rate of (**d**) CZT, (**e**) CT and (**f**) ZT; (**g,h,i**) Capacitive contribution of (**g**) CZT, (**h**) CT and (**i**) ZT at a sweeping rate of 1.0 mV s⁻¹



Fig. S17 Cycling performance of KPB half cells at a current density of 100 mA g^{-1} (note that the cell was cycled at 50 mA g^{-1} for the first 5 cycles). Half cells were assembled using K metal as the counter electrode





Fig. S18 Cycle performances of KPB CZT full cells at a current density of 500 mA g⁻¹



Fig. S19 Cycling performance of KPB^ICZT full cells at a current density of 200 mA g⁻¹ without pre-potassiation of anodes