ADVANCED FUNCTIONAL MATERIALS

Supporting Information

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K^+ Induced Phase Transformation of Layered Titanium Disulfide Boosts Ultrafast Potassium-Ion Storage

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Experimental Section

Materials characterization

TiS_2 powder was purchased from Nanjing MKNANO Tech. Co., Ltd.. MCMB powder was purchased from SZKEJING. The crystal structure of TiS_2 was characterized via XRD (D-MAX/2000-PC, Rigaku), and HRTEM (HR-TEM, JEM-ARM200F/JEOL, operating at 200 kV) combined with EDX (system attached to the HR-TEM instrument). Morphology was analyzed by SEM (JSM-6500, JEOL, acceleration voltage: 20 kV).

Electrochemical characterization

The working electrode was prepared by casting slurry containing TiS_2 (70 wt%), acetylene black (20 wt%), and PVDF binder (10 wt%) of onto aluminum (Al) foil. The loading content of the anode materials on Al foil was about 1.0 mg cm^{-2}. K metal was used as the counter electrode for half cells. The glass fiber (GF/D) was elected as separator. 1.0 M solution consist of KPF_6 in a 4/3/2 (v/v/v) mixture of ethylene carbonate (EC), dimethyl carbonate (DMC), and ethyl methyl carbonate (EMC) was used as the electrolyte. The full-cells were assembled with MCMB cathode and TiS_2 anode. To prepare the cathodes, MCMB, acetylene black, and PVDF binder were mixed with a weight ratio of 8:1:1. For the full cells, the mass ratio of MCMB cathode to TiS_2 anode was fixed at 4:1. The CV test was carried out using a ZIVE-MP2A. The galvanostatic discharge/charge test was conducted on LAND CT2001A. The GITT data were collected by applying a current of 20 mA g^{-1} for 3 minutes, followed by 1 hour of rest.

Calculation method

Materials Studio 2020 CASTAP module was used for density functional theory calculations.[1] The intercalation energy of K^+ at different concentration (K_nTiS_2, n = 0.125, 0.25, 0.375, 0.5, 0.625, 0.75, 0.875) and corresponding variation of layer spacing were evaluated. In addition, some electronic structures and the migration path of K between the interlayers of TiS_2 were also simulated.[2] The generalized gradient approximation (GGA) and the Perdew−Burke−Ernzerhof (PBE) exchange−correlation function were used.[3] The calculations were performed with fine quality, and the dispersion force was corrected by

Grimme method.[4] In detail, the electronic self-consistent field (SCF) tolerance was 1×10^{-6} eV/atom, while the force tolerance in geometry optimization was 0.03 eV/Å, and the plane wave basis energy cutoff was set as 435.4 eV. The insertion energies were calculated with a supercell, $K_nTi_8S_{16}$ (n=1, 2, 3, 4, 5, 6, 7), and the insertion energy and average insertion energy were calculated by following equations, respectively.

$$E = E(K_nTi_8S_{16}) - E(K_{n-1}Ti_8S_{16}) - E(K)$$

$$E_{avg} = \frac{E(K_nTi_8S_{16}) - E(Ti_8S_{16})}{n}$$

In above equations, $E(x)$ represents the energy of the structure, x, and $E(K)$ is the normalized energy of bulk K metal.

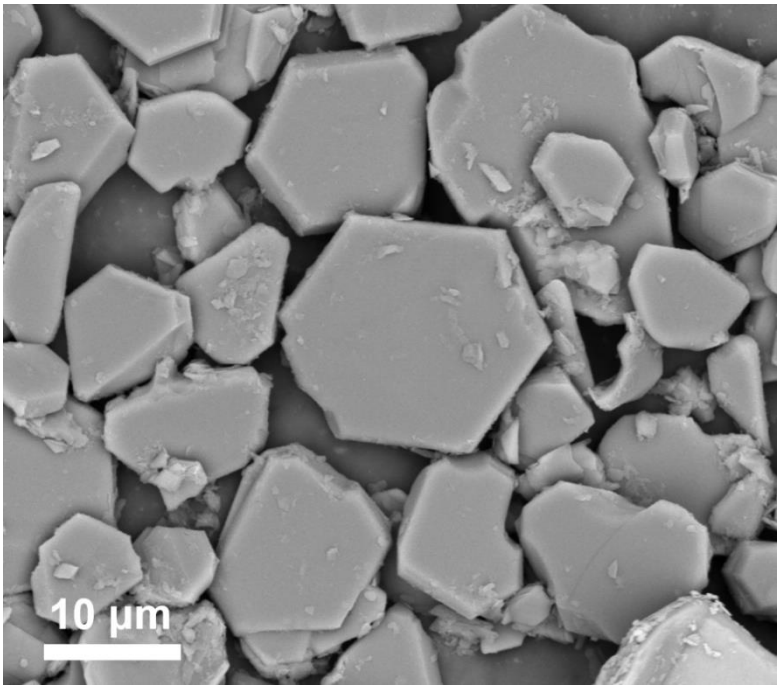

Figure S1. SEM image of TiS_2.

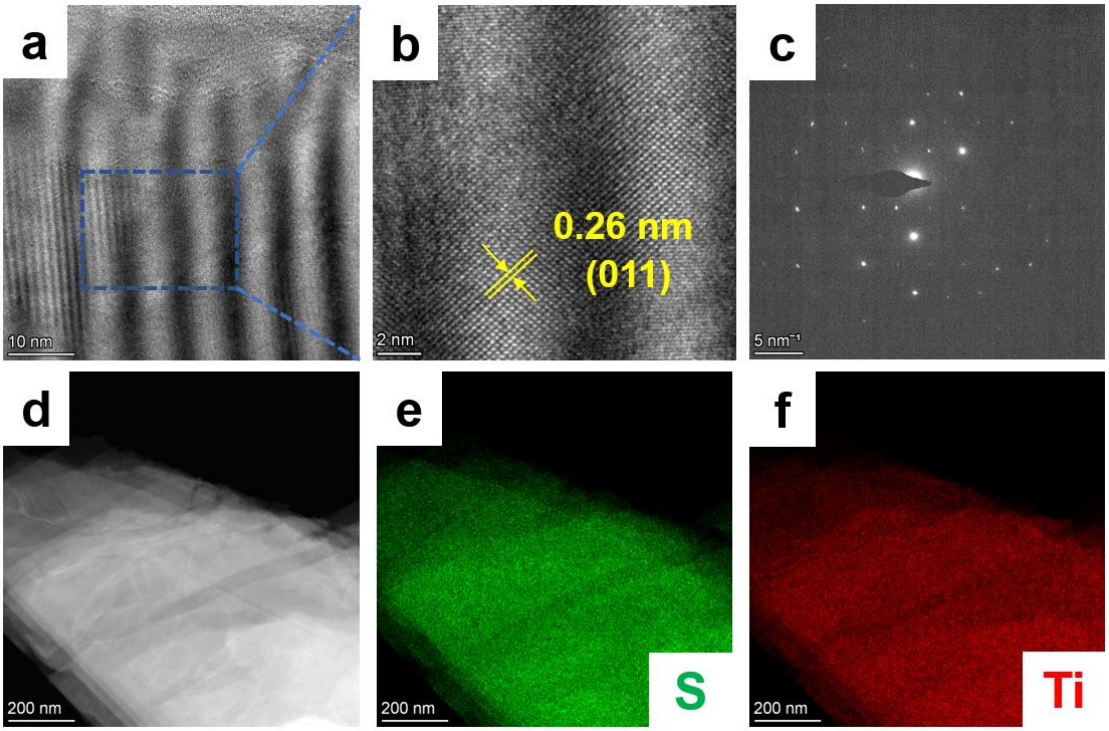

Figure S2. (a-b) HRTEM image and (c) the corresponding SAED pattern of TiS_2. (d-f) The corresponding elemental mapping images of S and Ti.

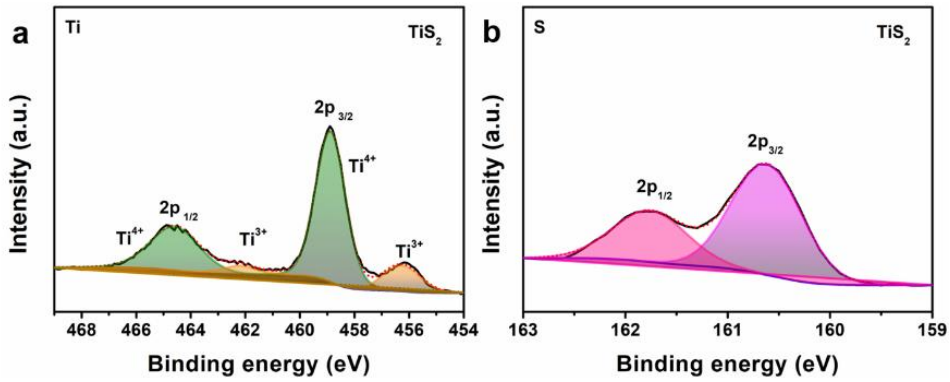

Figure S3. XPS spectra (a) Ti 2p and (b) S 2p of TiS_2.

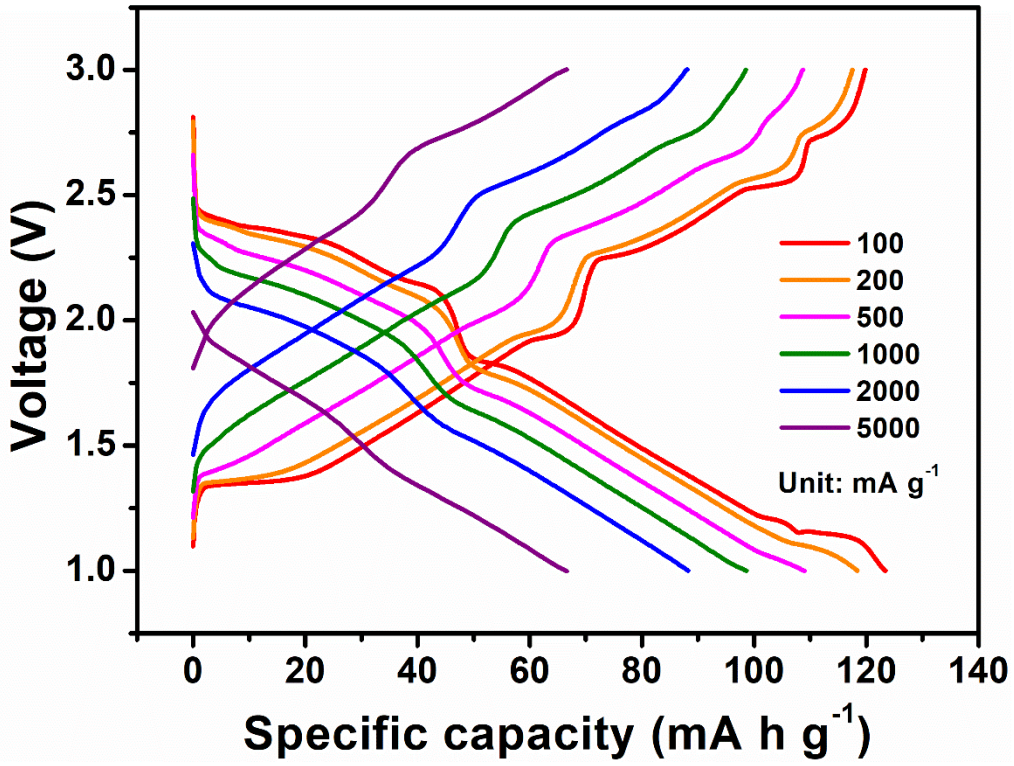

Figure S4. GCD curves of TiS_2 anode in half cells at different current densities.

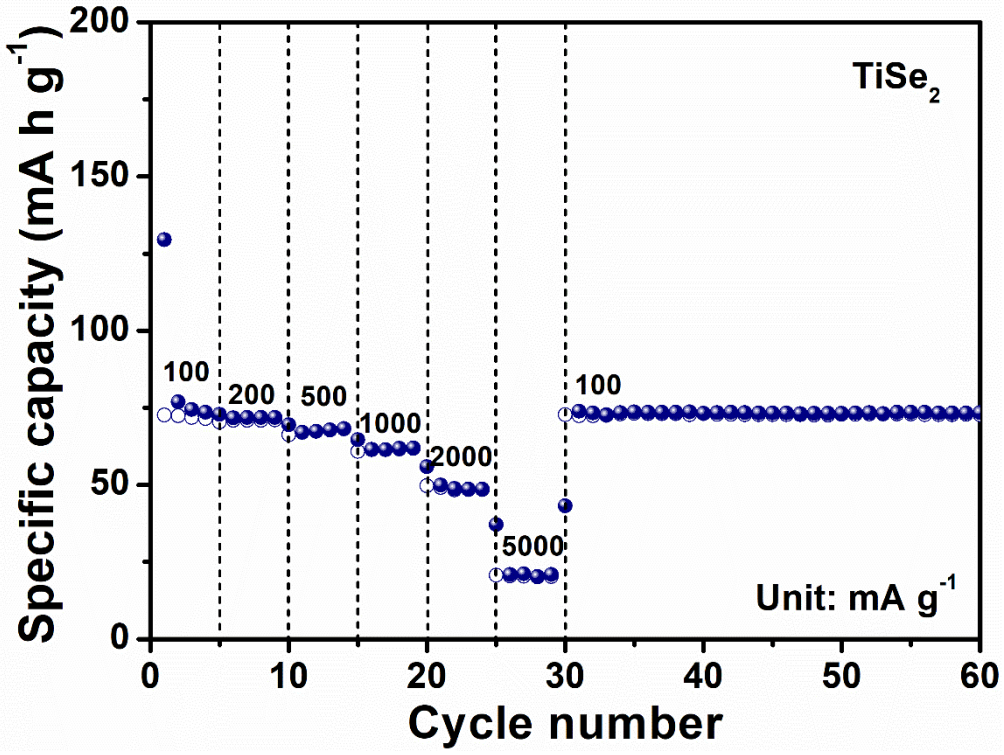

Figure S5. Rate capability of $TiSe_2$ anode.

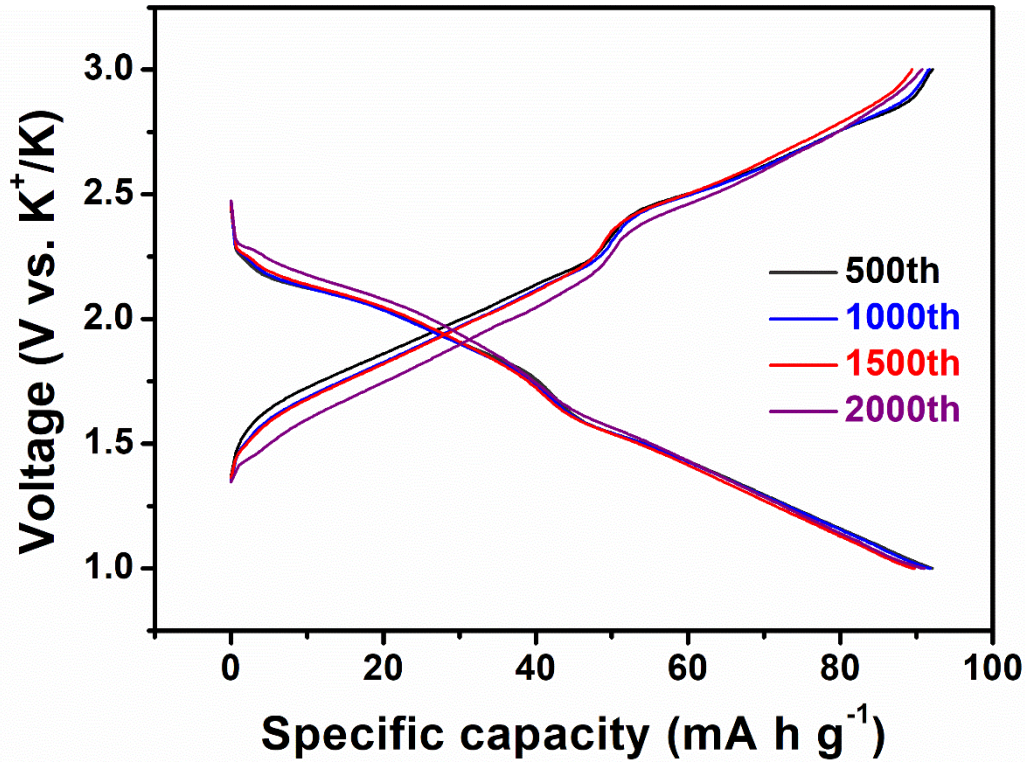

Figure S6. GCD Profiles of the TiS_2-K battery at different cycles at 1000 mA g^{-1}.

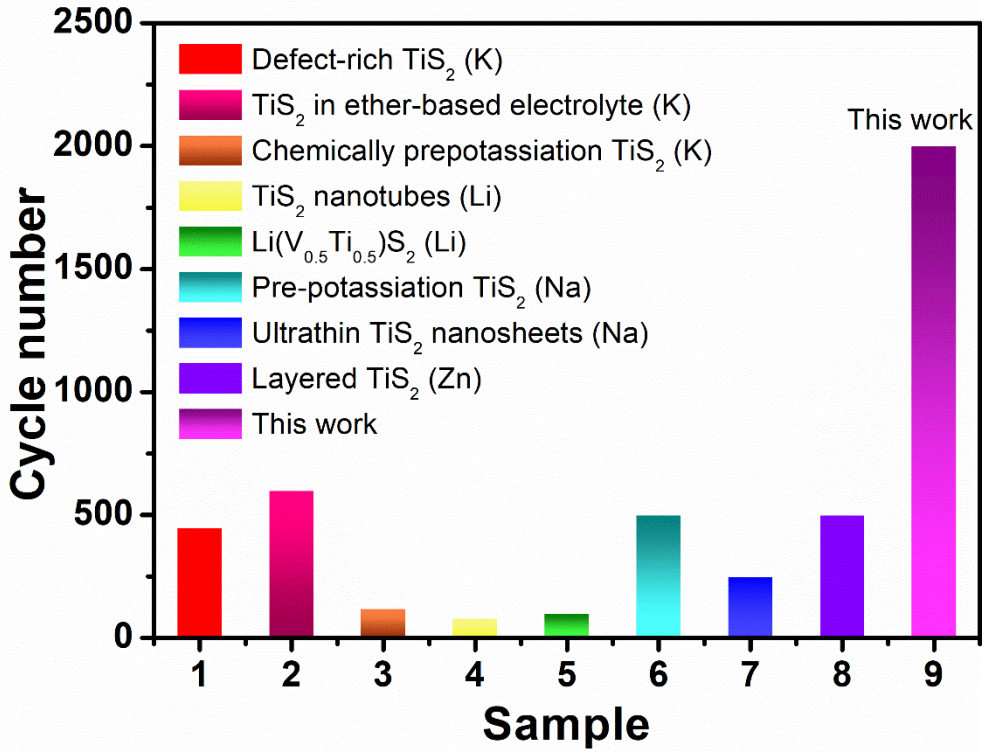

Figure S7. Cycle performance comparison with previous literature.

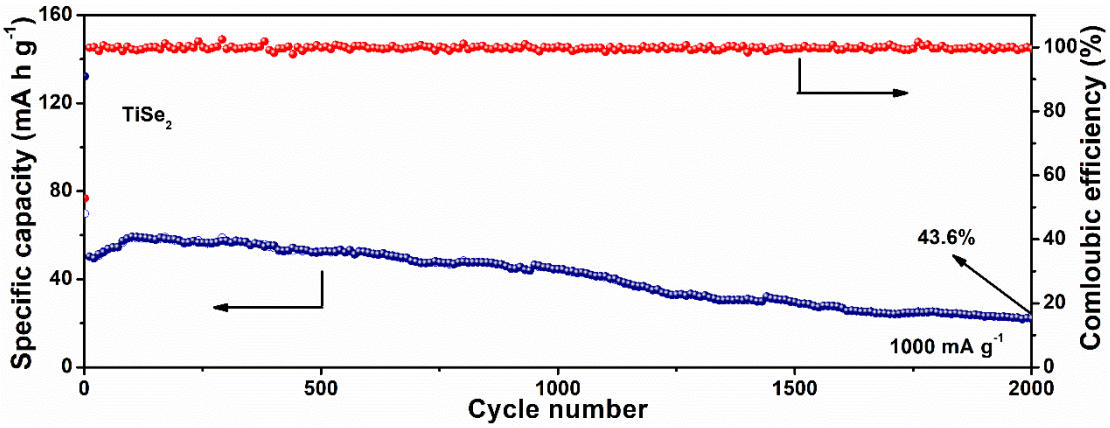

Figure S8. Long-term cycle performance at 1000 mA g^{-1} of $TiSe_2$ anode.

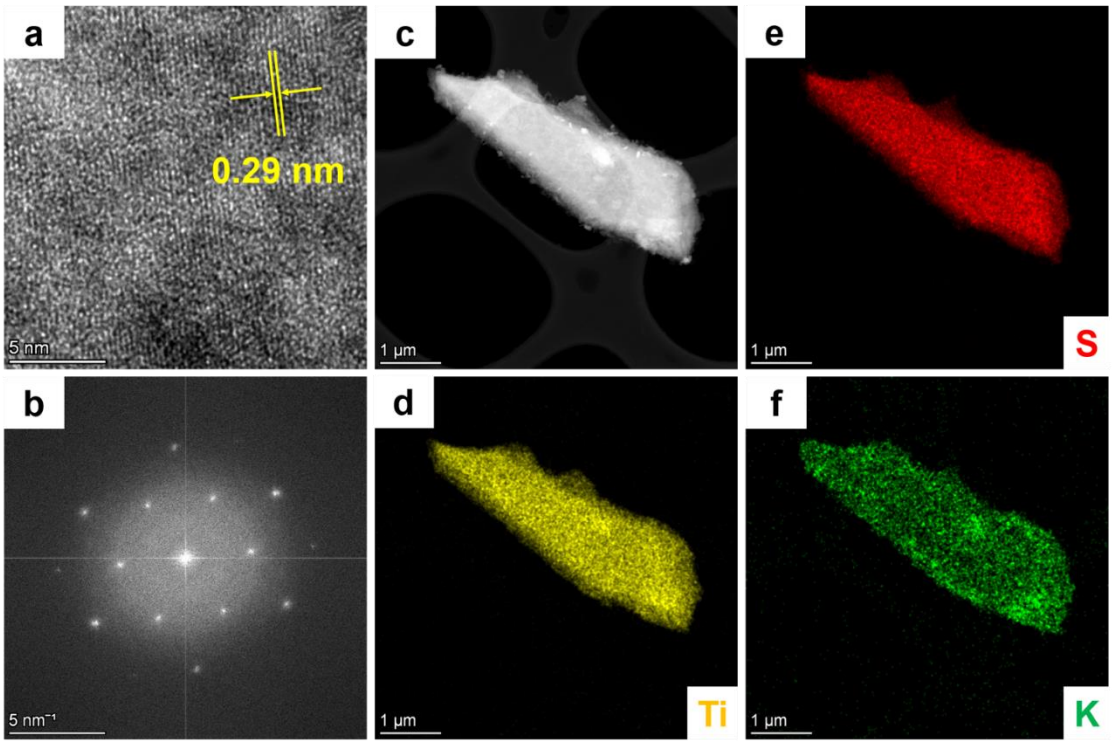

Figure S9. (a) HRTEM image and (b) the corresponding SAED pattern of TiS_2 at potassiation state. (c-f) The corresponding elemental mapping images of Ti, S, and K.

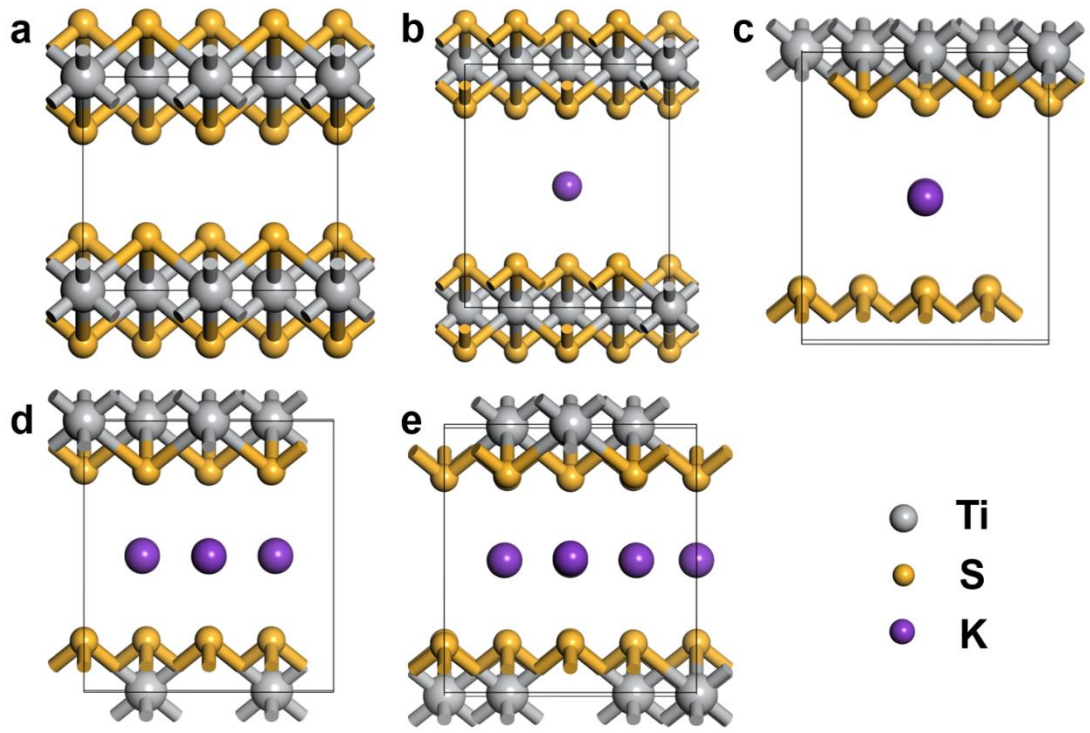

Figure S10. The crystal structure of (a) TiS_2, (b) $K_{0.125}TiS_2$, (c) $K_{0.25}TiS_2$, (d) $K_{0.75}TiS_2$, and (e) $K_{0.875}TiS_2$.

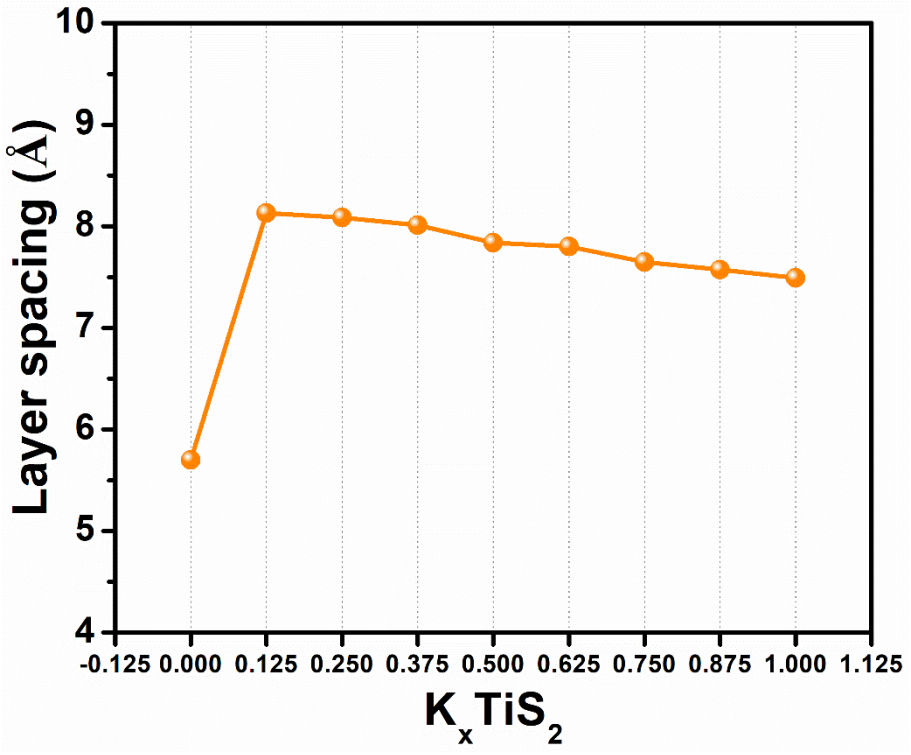

Figure S11. The calculated layer spacing variation function of K content.

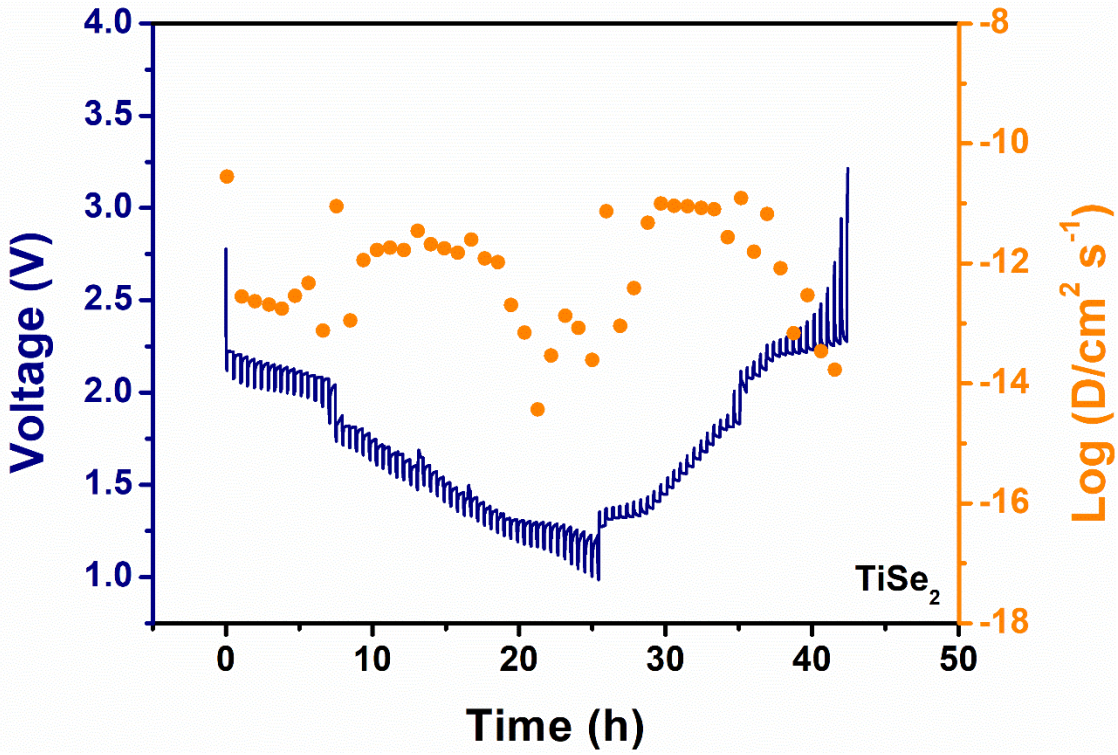

Figure S12. GITT voltage profiles and diffusion coefficients versus state of charge and discharge of $TiSe_2$ anode.

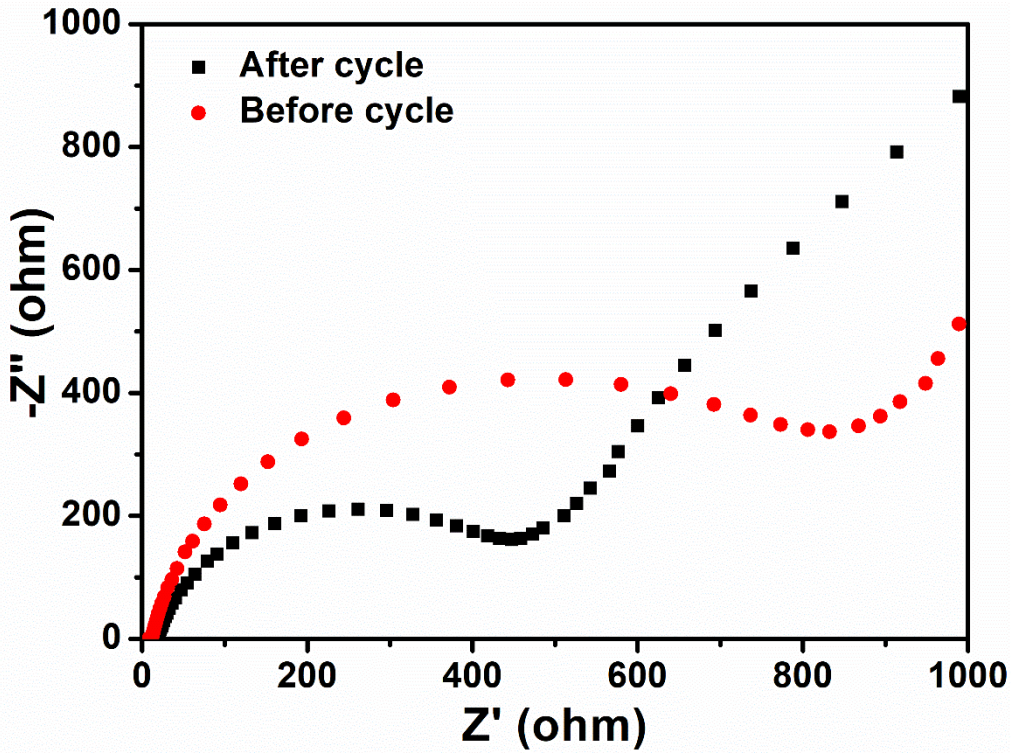

Figure S13. Nyquist plots of TiS_2 before and after cycling.

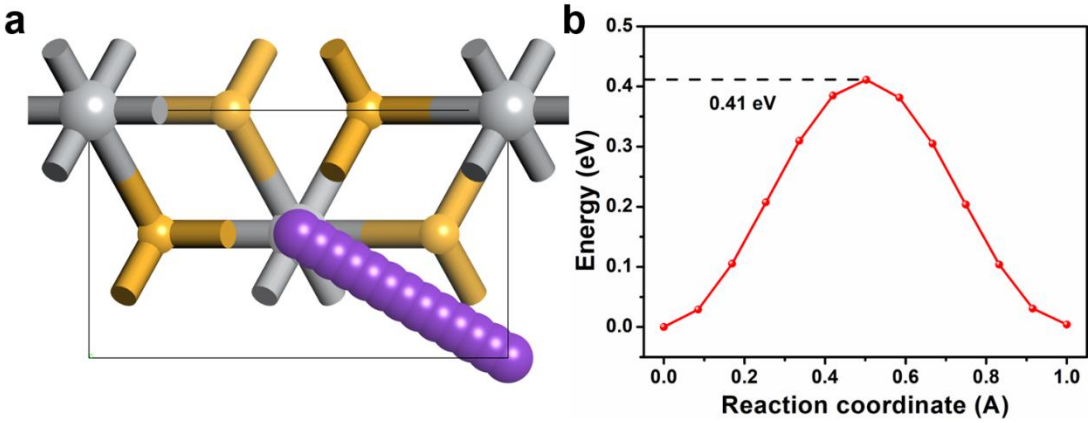

Figure S14. The proposed K^+ (a) diffusion path and (b) energy barriers in $K_{0.5}TiS_2$.

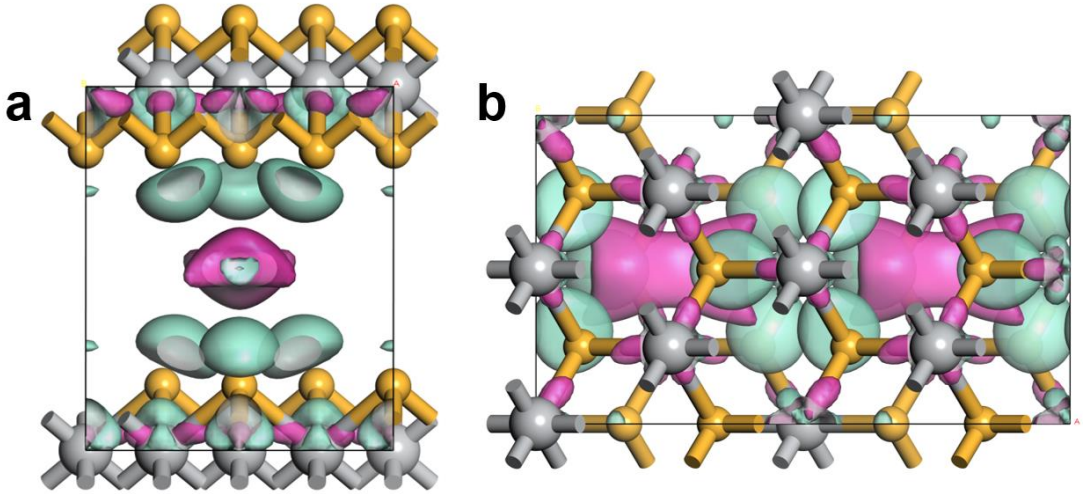

Figure S15. The local charge density difference isosurface.

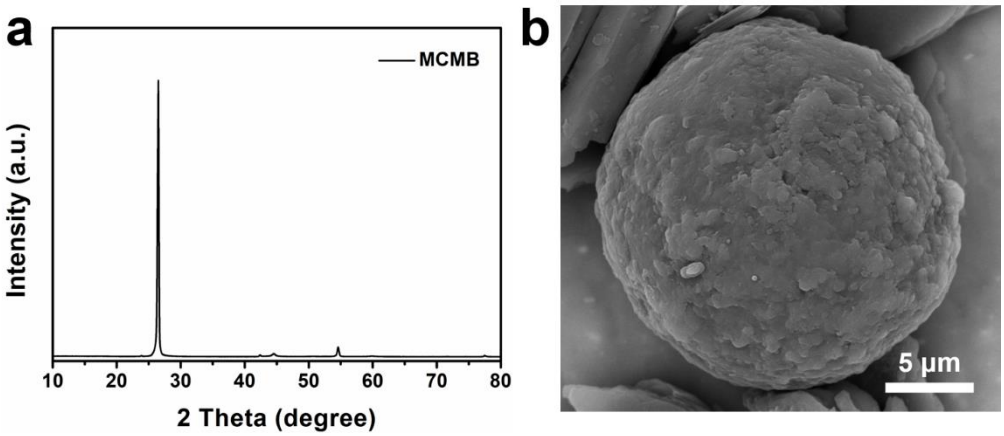

Figure S16. (a) The XRD pattern and (b) SEM image of MCMB powder.

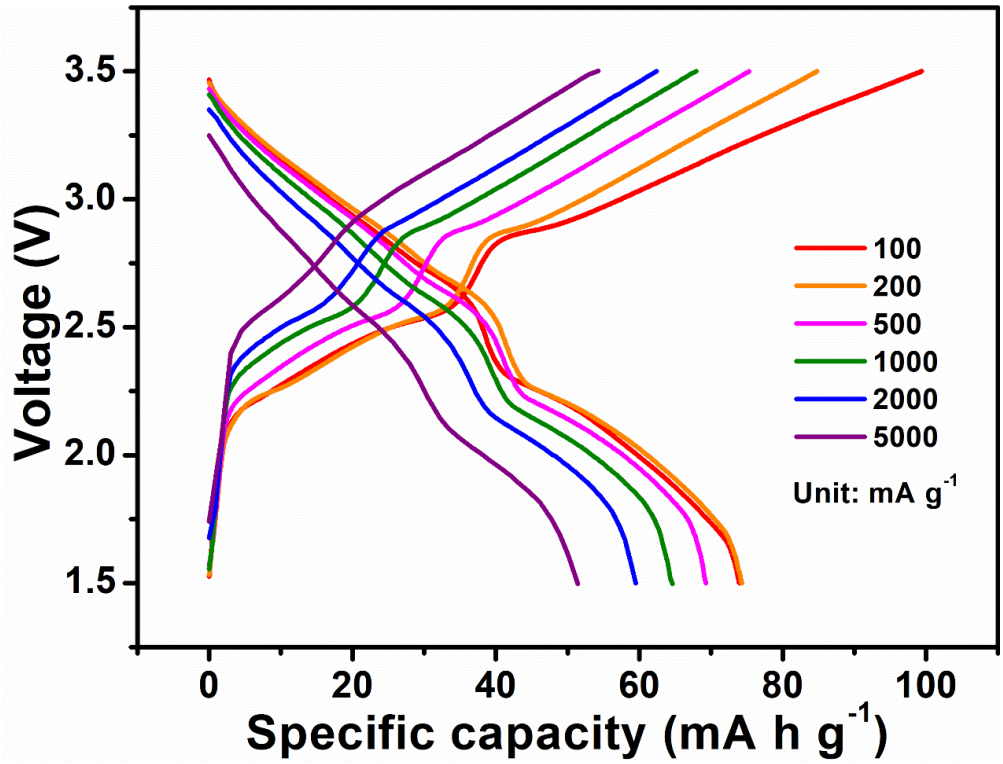

Figure S17. The GCD curves of TiS_2 anode in full cells at different current densities.

Table S1. Diffusion barriers comparison with previous literature.

Materials	**Diffusion barriers (eV)**	**Ref.**
Defect-rich TiS_2	2.446	[5]
VS_2 nanosheets	0.51	[6]
CoV_2O_6 nanosphere@graphene oxide (GO)	0.5	[7]
Sn	0.6	[8]
Dipotassium terephthalate (K_2TP)	0.46	[9]
$K_{0.25}TiS_2$	**0.27**	**This work**

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