

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

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Potassium dual-ion batteries (K-DIBs) have invoked considerable interest owing to their high safety and power density. However, achieving high-rate and good cyclability anodes for K-DIBs is still a grand challenge. Herein, layered TiS₂ is proposed as an attractive anode for K-DIBs, which achieves a discharge capacity of 91.0 mA h g⁻¹ while being discharged/charged to 2000 cycles in half cells. Interestingly, such a stable capacity is attributed to the mechanism of the K⁺ induced phase transformation. In situ characterizations and first principles calculations reveal that the inserted K⁺ acts as pillar between the Ti-S layers producing the thermodynamically stable K_{0.25}TiS₂ phase eventually. The robust K_{0.25}TiS₂ phase shows enlarged interlayer space, enhanced electronic conductivity, and lower diffusion barrier that enable highly stable and fast storage of K⁺. Moreover, a novel K-DIB based on TiS₂ anode and mesocarbon microbead cathode is reported for the first time. The K-DIB achieves a reversible capacity of 75.6 mA h g⁻¹ at 100 mA g⁻¹ and excellent cyclability with 85.8% capacity retention over 1000 discharge/charge at 5000 mA g⁻¹. Such mechanistic research provides new insights into the reaction process of layered sulfides/selenides and will facilitate their application in safe and high-power K-DIBs.

1. Introduction

The development of the new battery systems is to seek high power density, low-cost, and high safety energy storage devices surpassing the current dominant lithium-ion batteries (LIBs). Among alternative battery chemistries, metal-based dual-ion batteries (M-DIBs) show great potential as electrochemical device. M-DIBs are assembled with battery-type anodes and capacitor-type cathodes, which show the advantages of both batteries and capacitors. Recently, the research of lithium dual-ion batteries (L-DIBs), sodium dual-ion batteries (S-DIBs), potassium dual-ion batteries (K-DIBs), calcium dual-ion batteries (C-DIBs), and zinc dual-ion batteries (Z-DIBs) shows a blowout trend.^[1–5] Among them, K-DIBs show obvious advantages due to abundant resources, low standard electrode potential, and low cost of K.^[4,6–8] However, the current K-DIBs are far from satisfactory due to the imbalance in the capacity and kinetics between anode and cathode materials.

It is still a challenge for K-DIBs to find suitable high-rate and high-capacity anode materials to match the fast-kinetic cathodes.

To date, several types of anode materials (such as carbon, transition metal oxide, alloys, organic material, etc.) have successfully been developed for K-DIBs.^[9–17] However, these materials have serious inherent bottlenecks that limit their application. For examples, voltage polarization and safety issues for K metal plating at low voltages and high rates are the major challenges for carbon materials; the main disadvantage of alloys and transition metal oxide materials is the large volume expansion, resulting in poor cycle performance; the main challenges of organic-based materials are low conductivity and solubility in organic electrolytes. The issues of these materials need to be solved in the practical application of batteries. Recently, 2D layered transition metal dichalcogenides (TMDs) have been particularly desirable anode materials owing to their layered structures similar to graphene and outstanding physicochemical properties.^[18–23] Titanium disulfide (TiS₂) is typical transition metal chalcogenide compound, which is potential electrode for potassium-ion batteries (KIBs) owing to their large interlayer spacing, fast diffusion rate, and high electrical conductivity.^[24,25] Although the

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electrochemical performance of TiS_2 cathodes in KIBs has been reported, the relationship between mechanism and performance are not well established. Also, no research on the application of TiS_2 in K-DIBs is investigated to date.

In this work, we introduce a K-DIB with MCMB cathode and TiS_2 anode. Compared with other K-DIBs, the novel TiS_2 -MCMB-based K-DIB does not involve the design of the nanostructure of the material, and the composite of the carbonaceous material, but it can still show fast K^+ insertion kinetics with superior cycle stability. Such a K-DIB delivers a reversible capacity of 75.6 mA h g^{-1} at 100 mA g^{-1} , as well as excellent cyclability of 1000 discharge/charge at 5000 mA g^{-1} . Intriguingly, an anomaly was discovered, where a phase transition in TiS_2 anode during the first discharge resulted in a more stable structure. Through density functional theory (DFT) calculations and detailed in situ characterizations, we propose that the intercalated K^+ acts as pillars between the layers, stabilizing the layered structure of TiS_2 . $\text{K}_{0.25}\text{TiS}_2$ exhibits a wider ion channel and high electronic conductivity than the pristine TiS_2 . Moreover, $\text{K}_{0.25}\text{TiS}_2$ demonstrates a very high K^+ diffusion coefficient (D_{K^+}) of 10^{-11} – $10^{-10} \text{ cm}^2 \text{ s}^{-1}$ and also displays a very low K^+ diffusion energy barrier of only 0.27 eV .

2. Results and Discussion

The X-ray diffraction (XRD) pattern of the TiS_2 powder indicates its pure crystalline phase (Figure 1a). All diffraction peaks could be indexed with a hexagonal cell with the $P-3m1$ space group ($a = b = 3.4079 \text{ \AA}$, $c = 5.6989 \text{ \AA}$) for TiS_2 . TiS_2 shares the layered framework, which is constructed by the lantern-like units sequenced along the c -axis (Figure 1b,c). The interlayer is composed of two hexagonal closely arranged S atoms and Ti in the center to form an octahedron. The scanning electron microscopy (SEM) image of TiS_2 powders (Figure S1, Supporting Information) shows its irregular morphology with particle size ranging from 2 to $30 \mu\text{m}$. The high-resolution transmission electron microscopy (HRTEM) images depict lattice fringes with 0.26 nm d -spacing, assigning to the (011) planes of TiS_2 (Figure S2a,b, Supporting Information). The presence of diffraction dots in the selected area electron diffraction (SAED) pattern confirms that the existence of TiS_2 nanocrystal (Figure S2c, Supporting Information). Energy dispersive X-ray spectroscopy (EDX) elemental mappings confirm the homogeneous element distribution of Ti and S in TiS_2 (Figure S2d–f, Supporting Information). X-ray photoelectron

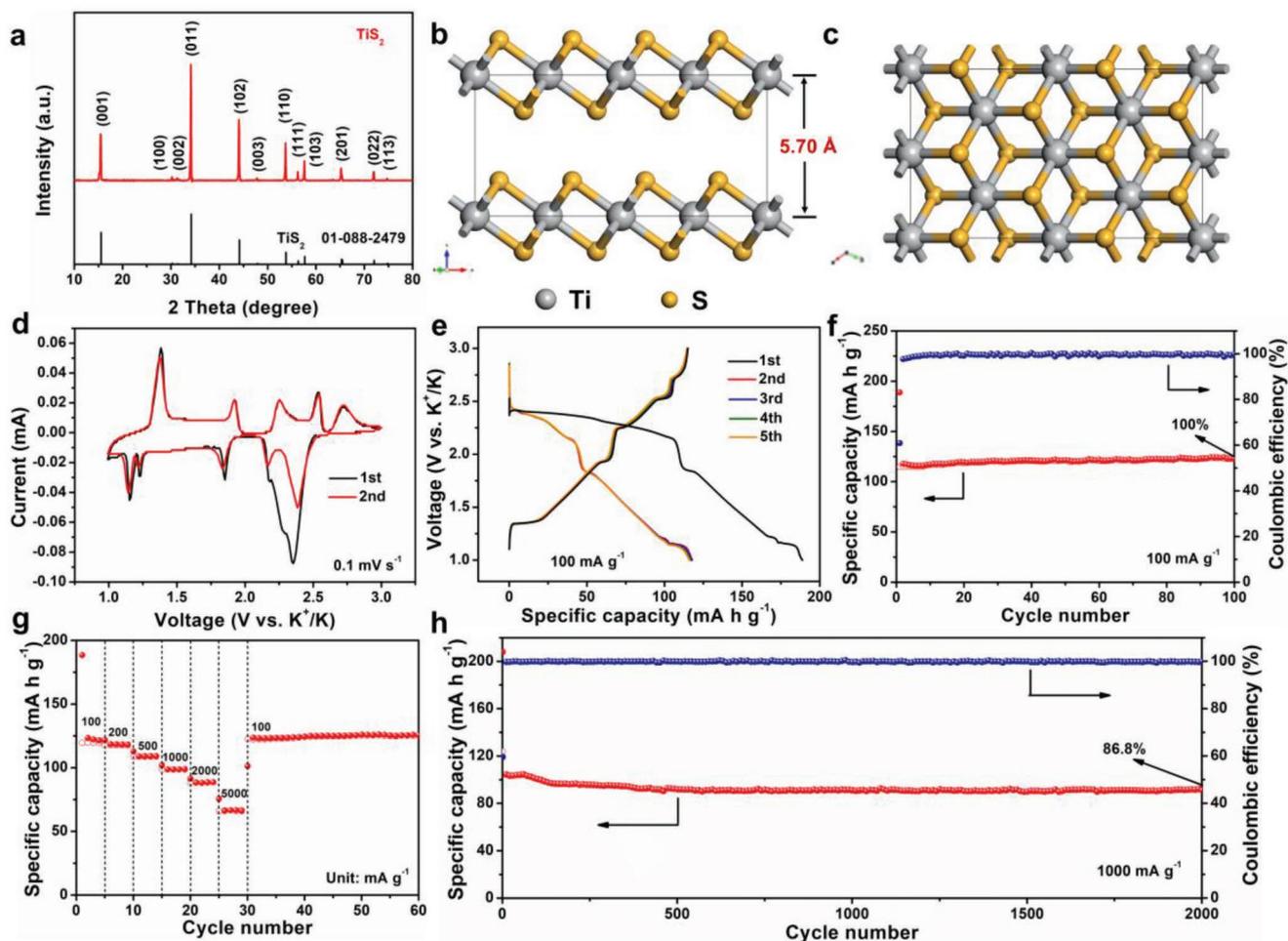


Figure 1. a) XRD pattern and b,c) crystal structure seen along a - b plane and c -axis for TiS_2 . d) CV curves at 0.1 mV s^{-1} , e) GCD profiles at 100 mA g^{-1} , f) cycling stability at 100 mA g^{-1} of TiS_2 anode. g) Rate capabilities and h) long-term cyclability at 1000 mA g^{-1} of TiS_2 anode.

spectroscopy (XPS) was then performed to further confirm the surface chemical structure of TiS_2 (Figure S3, Supporting Information). The Ti XPS spectrum exhibits four peaks of $\text{Ti}^{3+} 2p_{3/2}$ (456.1 eV), $\text{Ti}^{3+} 2p_{1/2}$ (462.1 eV), $\text{Ti}^{4+} 2p_{3/2}$ (458.8 eV), and $\text{Ti}^{4+} 2p_{1/2}$ (464.5 eV), indicates the coexistence of Ti^{3+} and Ti^{4+} in TiS_2 (Figure S3a, Supporting Information).^[26,27] The much lower Ti^{3+} concentration in sample can be ascribed to the less defects generated on the surface.^[28] The peaks at 161.8 (S $2p_{1/2}$) and 160.6 eV (S $2p_{3/2}$) are ascribed to S^{2-} of TiS_2 (Figure S3b, Supporting Information). All the results demonstrate that the obtained samples are TiS_2 particles with high purity and crystallinity.

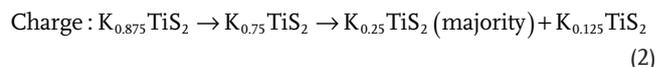
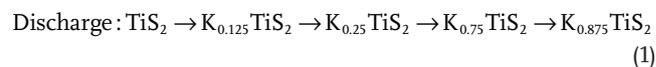
The K^+ storage performance of TiS_2 anode was first evaluated in the half-cell configuration. Figure 1d shows the initial two cyclic voltammetry (CV) profiles of TiS_2 anode at 0.1 mV s^{-1} from 1.0 to 3.0 V versus K^+/K . The first CV curve of TiS_2 anode displays six reduction peaks at 2.35, 2.28, 2.18, 1.85, 1.23, and 1.15 V and five oxidization peaks at 1.39, 1.94, 2.27, 2.56, and 2.74 V, respectively. Obviously, in the second cathodic scan, a peak located at 2.28 V disappears, which is ascribed to the irreversible reaction. The CV curves in the second anodic scan almost overlap and no obvious irreversible process can be found. Figure 1e shows the galvanostatic discharge/charge (GCD) profiles of TiS_2 anode at 100 mA g^{-1} . The plateaus in the corresponding GCD profiles well coincide with the characteristic peaks of CV curves. The low initial Coulombic efficiency (CE) of 60.88% for TiS_2 anode is also due to the formation of the irreversible reaction.^[12]

The cyclability and corresponding CEs at 100 mA g^{-1} of TiS_2 anode are presented in Figure 1f. TiS_2 anode demonstrates a discharge capacity of 123.0 mA h g^{-1} and nearly 100% capacity retention (compared with the 10th discharge/charge) over 100 cycles. The rate capabilities of TiS_2 anode were assessed under various current densities (Figure 1g; Figure S4, Supporting Information). TiS_2 anode exhibits reversible capacities of 122.4, 118.4, 109.0, 98.7, and 88.4 mA h g^{-1} at 100, 200, 500, 1000, and 2000 mA g^{-1} , respectively. The reversible capacity of TiS_2 anode achieves 66.4 mA h g^{-1} even at 5000 mA g^{-1} (54.2% capacity retention). Moreover, when the current density is switched abruptly to 100 mA g^{-1} , TiS_2 anode can still exhibit 123.2 mA h g^{-1} , demonstrating $\approx 100\%$ capacity recovery rate. In contrast, the similar layered titanium diselenide (TiSe_2) as an anode exhibits poor rate performance, only a discharge capacity of 20.8 mA h g^{-1} at 5000 mA g^{-1} (Figure S5, Supporting Information). Intrigued by the excellent rate capability, the high-rate cycling stability of TiS_2 anode was also assessed at 1000 mA g^{-1} . As provided in Figure 1h and Figure S6 (Supporting Information), TiS_2 anode exhibits a high discharge capacity of 91.0 mA h g^{-1} over 2000 discharge/charge and a capacity retention of 86.8% (compared with the 10th discharge/charge, only 0.0066% capacity loss per discharge/charge). Such cycle life is longer than the most reported TiS_2 -based electrodes in the literature (Figure S7, Supporting Information). Likewise, TiSe_2 anode exhibits poor long-cycle stability, which only achieves a low discharge capacity of 21.9 mA h g^{-1} with 43.6% capacity retention over 2000 discharge/charge (Figure S8, Supporting Information).

To explore the reaction mechanism during cycle, in situ XRD for TiS_2 and TiSe_2 anodes with different cycles were performed.

Figure 2a shows the first GCD profiles on the left and the in situ XRD patterns of TiS_2 anode on the right. During the first discharging, TiS_2 anode first experiences a phase transition reaction at the beginning. The (001) peak of TiS_2 (phase 1) disappears at 15.5° and some new peaks of a buffer phase begin to appear. Upon further K^+ insertion, the buffer phase gradually converts to $\text{K}_{0.125}\text{TiS}_2$ (phase 2) through the phase transition. With the further potassiation, a solid-solution reaction among $\text{K}_{0.125}\text{TiS}_2$, $\text{K}_{0.25}\text{TiS}_2$ (phase 3), and $\text{K}_{0.75}\text{TiS}_2$ (phase 4) occurs. At the end of the potassiation process, the diffraction peaks of $\text{K}_{0.875}\text{TiS}_2$ (phase 5) appear, accompanied by the second phase transition. Ex situ TEM characterization of the potassiated TiS_2 electrode reveals the good crystalline nature of the intermediate phase (Figure S9, Supporting Information). During the whole charging process, the $\text{K}_{0.875}\text{TiS}_2$ phase begins to turn back into $\text{K}_{0.75}\text{TiS}_2$ phase, and the $\text{K}_{0.25}\text{TiS}_2$ phase and a little $\text{K}_{0.125}\text{TiS}_2$ phase finally appears again, i.e., the electrode will not turn back into the original TiS_2 phase. The crystal structures of TiS_2 , $\text{K}_{0.125}\text{TiS}_2$, $\text{K}_{0.25}\text{TiS}_2$, $\text{K}_{0.75}\text{TiS}_2$, and $\text{K}_{0.875}\text{TiS}_2$ are intuitively illustrated to demonstrate the structural evolution during the potassiation/de-potassiation process (Figure 2b; Figure S10, Supporting Information).

Overall, the structural evolution of TiS_2 anode for K^+ storage during the first cycle can be expressed by the following sequences.



The layer spacing can more intuitively reflect the structural changes during K^+ insertion process (Figure S11, Supporting Information). In the early potassiation stage of TiS_2 anode, the layer spacing increases from 5.70 to 8.13 Å, indicating new K^+ insertion phases. The layer spacing decreases from 8.09 to 7.65 Å upon further K^+ insertion and then finally to 7.57 Å, which is attributed to the interlayer coulombic force, making the intercalation of K^+ increasingly difficult in the meantime. In order to reveal the reversibility of the entire intercalation–deintercalation reaction, the intercalation energy was calculated (Figure 2c). As the discharge process, the intercalation of K^+ becomes more and more difficult, indicating that the intercalation of K^+ is limited. In addition, as the reversible process of the intercalation process, the deintercalation process of K^+ also becomes difficult in the final stage, and the energy of K^+ deintercalation reaches >2.6 eV in $\text{K}_{0.25}\text{TiS}_2$. The large energy barrier hinders the further desorption of K^+ from $\text{K}_{0.25}\text{TiS}_2$ phase, which avoids the large structural variation in the following cycling processes, though it causes non-negligible irreversible capacity decay in the initial discharge/charge.

After the first cycle, the structural evolution of TiS_2 anode changes significantly (Figure 3a,b). The initial state of the reaction is the $\text{K}_{0.25}\text{TiS}_2$ and $\text{K}_{0.125}\text{TiS}_2$ phase, indicating that the buffer phase reaction is almost bypassed. During potassiation process, the diffraction peaks shift to higher angles till the phase of $\text{K}_{0.75}\text{TiS}_2$ forms, manifesting a solid-solution reaction between $\text{K}_{0.25}\text{TiS}_2$ and $\text{K}_{0.75}\text{TiS}_2$ occurs. Upon further K^+

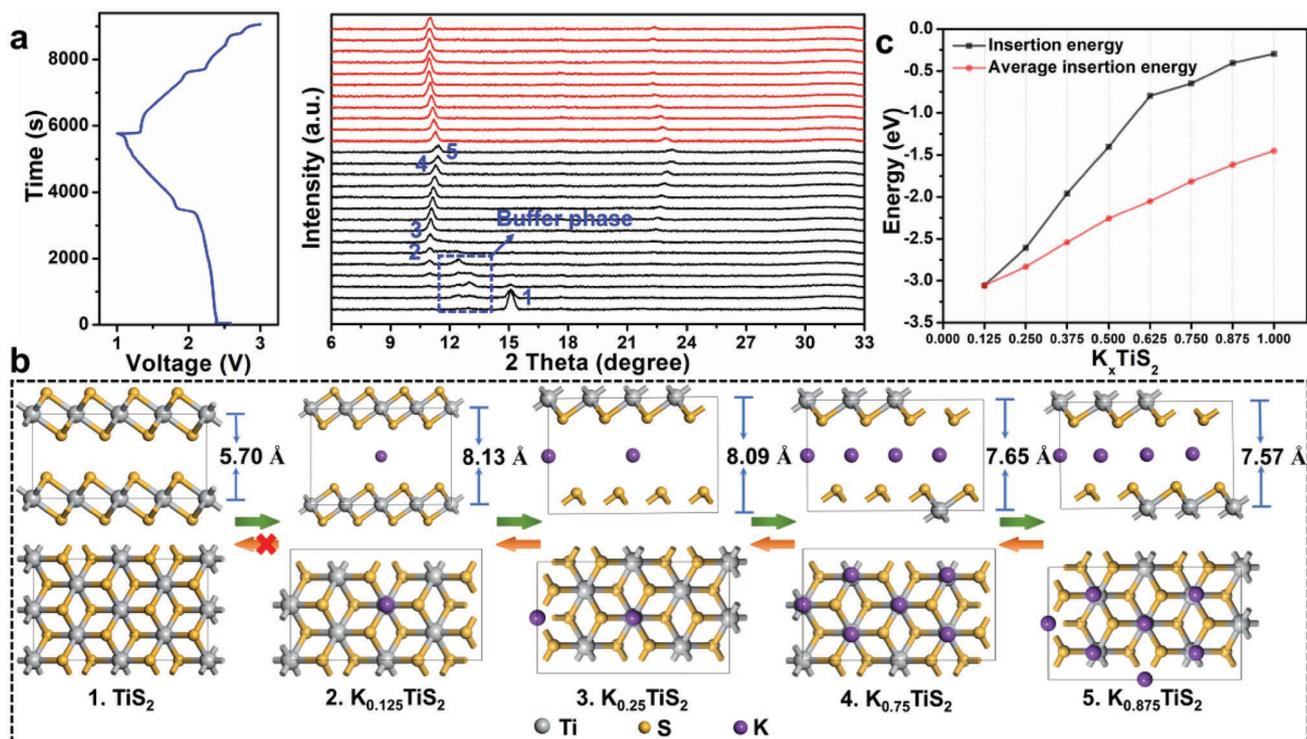


Figure 2. a) The first discharge/charge profiles and the corresponding in situ contour maps for TiS_2 anode. b) Schematic diagram of the structural evolution for TiS_2 anode (in top views, the upper TiS_2 layers of K^+ inserted structures are hidden). c) The calculated insertion energy and average insertion energy of K^+ during the first potassiation process.

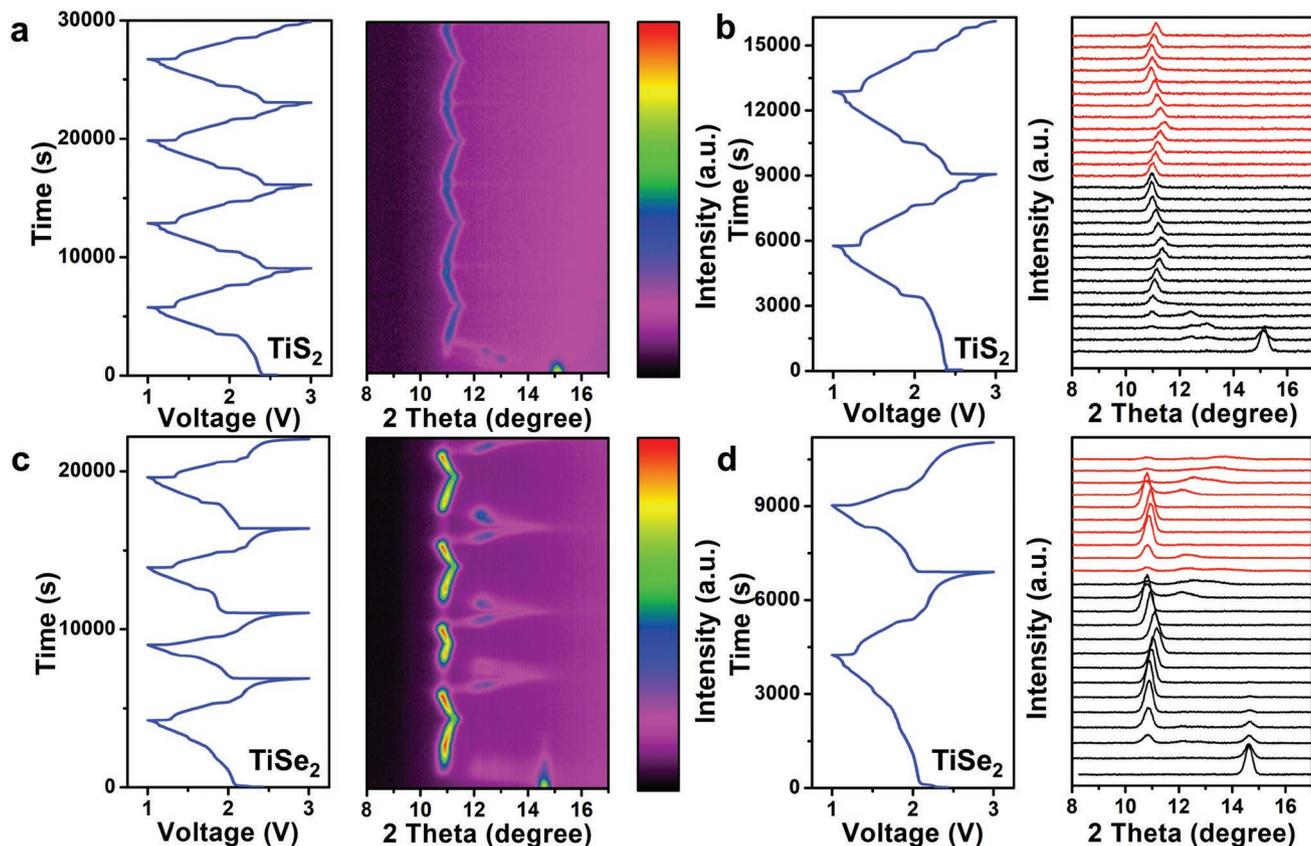
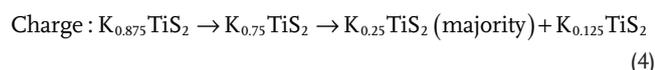
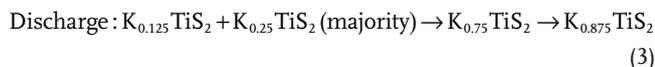


Figure 3. The discharge/charge profiles and the corresponding in situ contour maps for a,b) TiS_2 and c,d) TiSe_2 anodes.

insertion, a phase transition between $K_{0.75}TiS_2$ and $K_{0.875}TiS_2$ can be observed. During the charging, the position of peaks varies reversely. In the subsequent discharge/charge, the evolution of diffraction peaks is completely reversible, and it is expected to achieve more stable intercalation and deintercalation of K^+ .

Hence, the overall potassiation/de-potassiation mechanism of TiS_2 anode after the first cycle can be summarized as follows:



However, the partially irreversible electrochemical behavior does not occur in $TiSe_2$ anode. The insertion/extraction of K^+ in $TiSe_2$ anode is completely reversible (Figure 3c,d). $TiSe_2$ anode experiences two phase transformations and two solid-solution reactions with a completely reversible formation of $K_{0.25}TiSe_2$, $K_{0.58}TiSe_2$, and K_xTiSe_2 ($x > 0.58$) during K^+ insertion/extrac-

tion. This indicates that there is no K^+ as a “pillar” to stabilize the layered structure after the first cycle for $TiSe_2$ anode, which may cause relatively poor cycle and rate performance in the above electrochemical tests.

The electrochemical reaction kinetics of TiS_2 anode after K^+ insertion was investigated by CV measurements. The CV curves at various scan rates from 0.1 to 1.0 $mV s^{-1}$ with 1.0–3.0 V were plotted in Figure 4a. According to $i = av^b$, the b values of peaks 1–9 are 0.65, 0.82, 0.83, 0.75, 0.67, 0.64, 0.77, 0.92, and 0.69, respectively, revealing that the K^+ storage process over TiS_2 during the phase transition process was mainly controlled by capacitive behavior (Figure 4b,c).^[29] The diffusion coefficients (D) of K^+ in TiS_2 and $TiSe_2$ anodes were further estimated by the galvanostatic intermittent titration (GITT) measurement. On the basis of the GITT curves, the K^+ diffusion coefficient in TiS_2 anode was calculated to be around 10^{-10} – $10^{-11} cm^2 s^{-1}$ according to Fick's second law, much higher than $TiSe_2$ anode (10^{-11} – $10^{-12} cm^2 s^{-1}$), implying the faster diffusion kinetics of K^+ in TiS_2 anode (Figure 4d; Figure S12, Supporting Information).

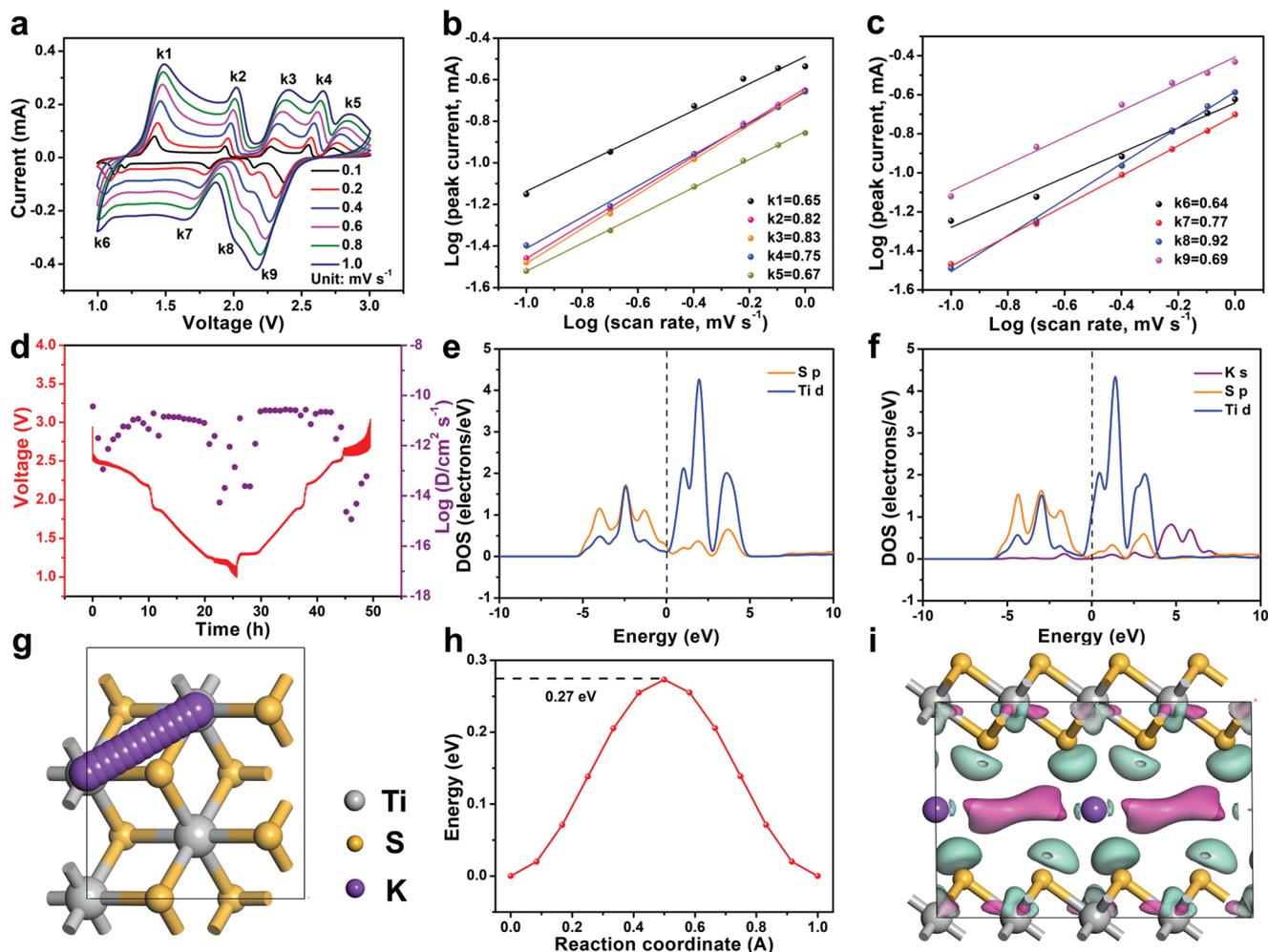


Figure 4. a) CV curves for TiS_2 anode at different scan rates and b,c) the relationship between scan rate and peak current. d) GITT voltage profiles and the corresponding diffusion coefficients for TiS_2 anode. e,f) DOS diagram for TiS_2 and $K_{0.25}TiS_2$ calculated by VASP. The proposed g) K^+ diffusion path (the upper TiS_2 layer is hidden for a better vision) and h) energy barriers in $K_{0.25}TiS_2$. i) The charge density difference contour of $K_{0.25}TiS_2$ with respect to TiS_2 and K^+ (the isosurface value is $0.008 e bohr^{-3}$).

To further understand the effect of K^+ induced phase transition on the physical properties of TiS_2 , the DFT calculations were conducted to simulate the density of states (DOS), mass transport channels, and the migration barrier of K^+ . Compared with TiS_2 , the Fermi level of $K_{0.25}TiS_2$ is shifted to the conduction band, resulting in enhanced electronic conductivity of $K_{0.25}TiS_2$ (Figure 4e,f). The electrochemical impedance spectroscopy (EIS) results of TiS_2 anode before and after cycling are shown in Figure S13 (Supporting Information). The charge transfer resistance (R_{ct}) after cycling of TiS_2 anode is lower than that before cycling, indicating that TiS_2 anode has a faster charge transfer process after K^+ insertion. A model of bilayer framework for the K^+ diffusion process was constructed. The reasonable K^+ transport path in $K_{0.25}TiS_2$ structural model is presented in Figure 4g. The diffusion barrier in $K_{0.25}TiS_2$ was supported to investigate the K^+ diffusion property (Figure 4h). The value of $K_{0.25}TiS_2$ (0.27 eV) was lower than that of VS_2 (0.51 eV), CoV_2O_6 (0.5 eV), Sn (0.6 eV), K_2TP (0.46 eV), and other reported electrodes, supporting the fast kinetics of $K_{0.25}TiS_2$ (Table S1, Sup-

porting Information).^[16,17,30–32] Even with higher concentration of inserted K^+ ($K_{0.5}TiS_2$), the transport energy barrier of K^+ is still as low as 0.41 eV, indicating TiS_2 is a good K^+ conductor (Figure S14, Supporting Information). Furthermore, the calculation of the charge density difference contour of $K_{0.25}TiS_2$ suggests that K^+ intercalation increases the electron density in the middle of the layer, resulting in enhanced electron mobility (Figure 4i; Figure S15, Supporting Information).

The above electrochemical results show that TiS_2 is a promising anode for K-DIB due to the generation of robust $K_{0.25}TiS_2$ phase induced by the first insertion of K^+ . Benefited from the enlarged interlayer space, enhanced electronic conductivity, and lower diffusion barrier, the resulting $K_{0.25}TiS_2$ shows high capability and good stability for K^+ storage. Inspired by these results, we proposed a novel K-DIB based on this TiS_2 anode and MCMB cathode. Figure 5a shows a schematic diagram of a full cell using a traditional KPF_6 -based electrolyte. The crystal structure and microscopic morphology of the used MCMB cathode are shown in Figure S16 (Supporting Information).

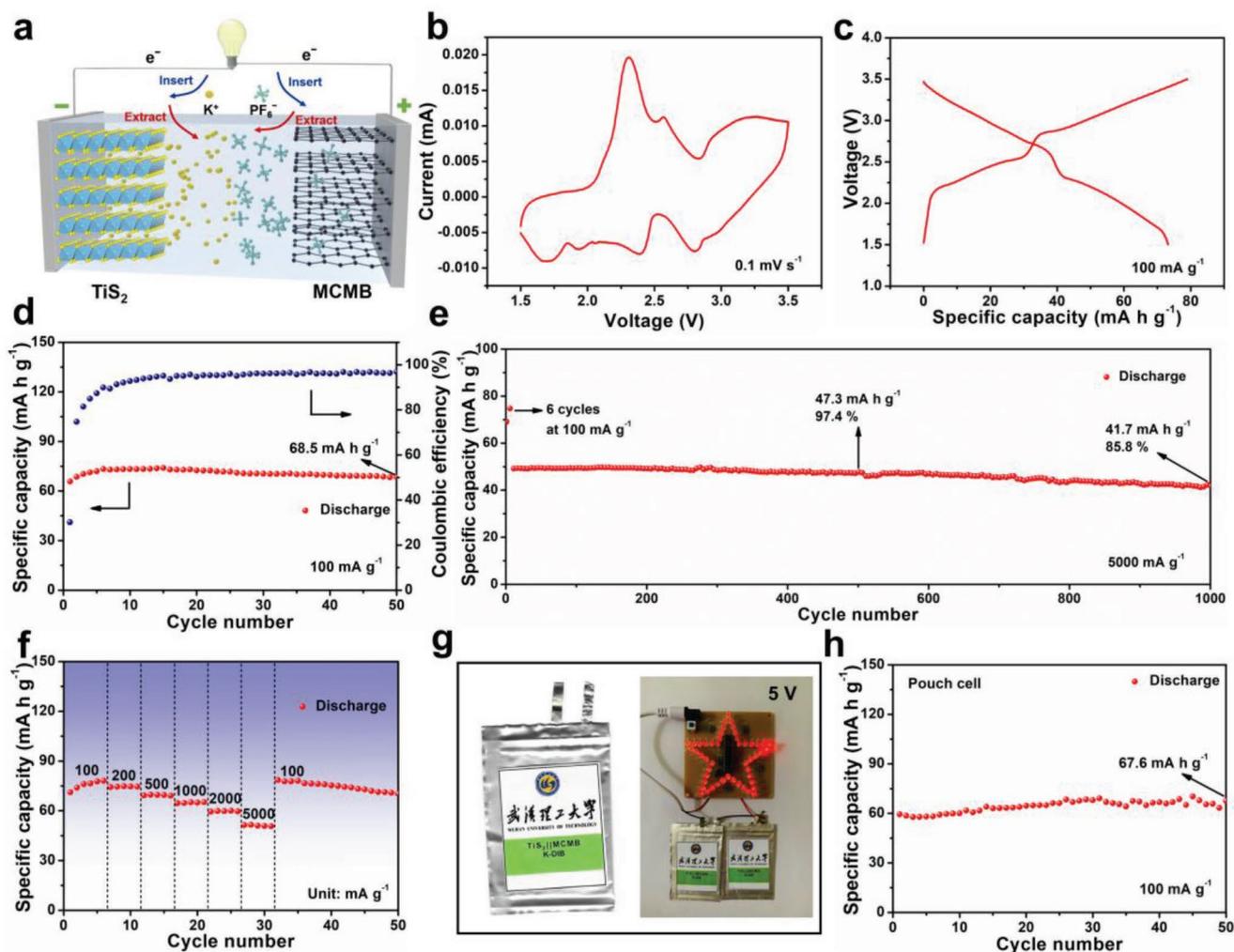
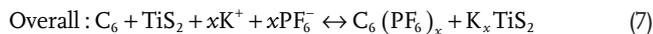
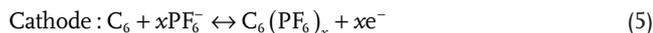


Figure 5. a) Schematic illustration of K-DIB based on TiS_2 anode and MCMB cathode. b) CV curves, c) GCD profiles, d) cyclic stability at 100 mA g^{-1} , e) long-term cyclic stability at 5000 mA g^{-1} , and f) rate ability of K-DIB full-cell. g) Digital photos of TiS_2 -MCMB K-DIB pouch cell and LED light band light up by pouch cell. h) Cycling performance of TiS_2 -MCMB K-DIB pouch cell at 100 mA g^{-1} .

During charge of this K-DIB, the K^+ is intercalated into TiS_2 anode, while PF_6^- is inserted into MCMB cathode at the same time. The discharging process is a reversed process. The electrochemical reaction process in the full cell is proposed as follows:



Multiple redox peaks are presented in the CV curves of K-DIB full-cell, which are consistent with the electrochemical behaviors of TiS_2 anode and MCMB cathode (Figure 5b). Figure 5c shows typical GCD profiles of the K-DIB at 100 $mA\ g^{-1}$ from 1.5 to 3.5 V, which displays several voltage plateaus during the charging/discharging process. TiS_2 -MCMB K-DIB holds a discharge capacity of 68.5 $mA\ h\ g^{-1}$ over 50 cycles when utilized at 100 $mA\ g^{-1}$ (based on the anode material) (Figure 5d). Moreover, the K-DIB exhibits excellent long-term cyclability at 5000 $mA\ g^{-1}$ (Figure 5e). After 1000 discharge/charge, the K-DIB possesses a discharge capacity of 41.7 $mA\ h\ g^{-1}$ with a capacity retention of 85.8% and only 0.0143% capacity loss per discharge/charge. Figure 5f and Figure S17 (Supporting Information) illustrate the discharge capacities of K-DIB at different current densities, which are 75.6, 74.6, 69.4, 64.9, 59.8, and 51.1 $mA\ h\ g^{-1}$ at 100, 200, 500, 1000, 2000, and 5000 $mA\ g^{-1}$, respectively, demonstrating excellent rate capability with a capacity retention of 67.6% at 5000 $mA\ g^{-1}$. TiS_2 -MCMB K-DIB reported in this work not only exhibits excellent rate capability, but also outstanding cycle stability. Furthermore, the TiS_2 -MCMB K-DIB pouch cell was successfully assembled (Figure 5g). This pouch cell can light up a LED light band in parallel, indicating its practical application. When tested at 100 $mA\ g^{-1}$, the pouch cell achieves an initial specific discharge capacity of 59.5 $mA\ h\ g^{-1}$ based on the anode mass (Figure 5h). After 50 cycles, a high capacity of 67.6 $mA\ h\ g^{-1}$ is remained, demonstrating good electrochemical stability.

3. Conclusion

In summary, the in situ phase transition from TiS_2 to $K_{0.25}TiS_2$ was found through the insertion of a small amount of K^+ in the early stage, and the obtained material was very stable for further storage of K^+ . The phase transition induced by K^+ enlarges the lattice spacing of TiS_2 , allowing intercalation and deintercalation of K^+ . The new phase $K_{0.25}TiS_2$ exhibits high electronic conductivity and low diffusion barrier, resulting in fast reaction kinetics. K^+ acts as a “pillar” to stabilize the layered structure, thereby ensuring long cyclability. As a result, the capacity retention of TiS_2 anode reaches 86.8% after 2000 discharge/charge at 1000 $mA\ g^{-1}$. Moreover, the as-assembled TiS_2 -MCMB K-DIB exhibits superior rate capability (75.6 and 51.1 $mA\ h\ g^{-1}$ at 100 and 5000 $mA\ g^{-1}$, respectively) as well as excellent cyclability (only 0.0143% capacity loss per discharge/charge at 5000 $mA\ g^{-1}$). Furthermore, the TiS_2 -MCMB K-DIB pouch cell also exhibits outstanding electrochemical stability,

suggesting it to be promising for large scale electronics applications in future. These findings will provide guidance for in-depth understanding of anode chemistry in K-based batteries and for the design of future K-DIB materials.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

K^+ induced phase transformations, potassium dual-ion batteries, reaction mechanisms, TiS_2 , ultrafast potassium-ion storage

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