Research article

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Facile synthesis of sulfur@titanium carbide Mxene as high performance cathode for lithium-sulfur batteries

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Abstract: The design of sulfur hosts with polar, sulfurphilic, and conductive network is critical to lithium-sulfur (Li-S) batteries whose potential applications are greatly limited by the lithium polysulfide shuttle effect. Mxenes, possessing layered-stacked structures and high electrical conductivities, have a great potential in sulfur hosts. Herein, sulfur nanoparticles uniformly decorated on titanium carbide Mxene ($S@Ti_3C_2T_x$ Mxene) are synthesized via a hydrothermal method and then utilized as a cathode for lithium-sulfur batteries. This unique architecture could accommodate sulfur nanoparticles expansion during cycling, suppress the shuttling of lithium polysulfide, and enhance electronical conductivity. Consequently, the $S@Mxene$ with a high areal sulfur loading ($\sim 4.0 \text{ mg cm}^{-2}$) exhibits a high capacity ($1477.2 \text{ mAh g}^{-1}$) and a low capacity loss per cycle of 0.18% after 100 cycles at 0.2 C. This work may shed lights on the development of high performance sulfur-based cathode materials for Li-S batteries.

Keywords: cathode; electrochemical performance; Li-S battery; Mxene; sulfur.

1 Introduction

The rapid developments of electric vehicles industry and electric grid require secondary batteries that have high energy density, high power density, and long cycling life. Because of limited energy density, the conventional lithium-ion batteries (LIBs) with $LiFePO_4$ as cathodes and graphite as anodes cannot meet the requirement [1–4]. Hence, there is a great need for exploiting new battery systems with high energy density [5]. Among some new rechargeable batteries, lithium-sulfur batteries (LSBs) are regarded as one of the most promising next generation energy storage systems due to their high theoretical specific capacity ($\sim 1675 \text{ mAh g}^{-1}$) and theoretical energy density ($\sim 2567 \text{ Wh kg}^{-1}$) which are 3–5 times higher than those of state-of-art LIBs [6–9]. Moreover, sulfur also has the advantages of low cost, nontoxic, and abundant reserves. Therefore, LSBs have great application potential in the transportation, electronic devices, and electric grid fields.

Despite considerable advantages, LSBs were plagued by several serious issues. Firstly, the conversion reaction from elemental sulfur to lithium sulfide is accompanied by a large volume change of about 80% which may destroy the cathode structure during cycling and cause low cycling performance. Secondly, poor ionic and electronic conductivities of sulfur and the discharged product polysulfides (Li_2S_x) increase the internal resistance of the battery, leading to poor high-rate performance. In addition, the adverse "shuttle effect" caused by the dissolution and diffusion of reaction intermediate polysulfides in organic electrolytes will cause capacity fading and low coulombic efficiency [10–12]. In such a context, new sulfur cathode host materials should be taken to overcome these problems. An ideal sulfur cathode host should not only have an interconnected architecture to encapsulate sulfur, keeping the polysulfides from dissolving into the electrolyte as well as preventing its' volume expansion, but also have the high electrical conductivity to compensate for the poor conductivity of elemental sulfur.

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Various carbon materials such as amorphous carbon, carbon nanofibers, carbon nanotubes, and graphene have been used as sulfur cathode host materials. The LSBs based on S/C cathodes exhibit high capacities owing to excellent electronical conductivities of carbon materials. However, their capacities always quickly decay because the interaction between carbon materials and polysulfides intermediate is weak, which cannot buffer the large volume changes of sulfur compound cathodes during the cycling processes. Therefore, efforts to explore ideal sulfur cathode hosts for suppressing the polysulfide shuttle effect are still requested. In recent years, a novel type of two-dimensional transition metal carbides or carbonitrides has emerged, which is known as Mxenes [13, 14]. Because of high electrical conductivities and 2D structures, Mxenes have been widely exploited for electrochemical sensors [15], LIBs [16], and supercapacitors [17]. Moreover, the Mxene family has a great potential to suppress the polysulfide shuttle effect and buffer the volume expansion of sulfur cathode during lithiation because of its layered-stacked structure and high pore volume. In a recent paper, N-doped Mxene/S composites were prepared by a thermal annealing and found to demonstrate improved performance [18].

In this paper, we have developed a simple method to prepare S@Ti₃C₂T_x Mxene composite. The S@ Mxene composite synthesized by a hydrothermal method displays a sandwich-like structure in which S nanoparticles were uniformly coated and decorated onto Ti₃C₂T_x Mxene's surface. Thanks to the coating structure design, a three-dimensional electronic transport pathway formed, which promoted electron transfer during cycling processes. Importantly, the as-fabricated S@Mxene composite cathode has demonstrated outstanding electrochemical performances with high reversible capacity and extended cycling stability. Since the process is simple and easy to scale up, our work may foster large scale production of high performance cathodes for LSBs.

2 Experimental

2.1 Preparation of S@Ti₃C₂T_x Mxene

Ti₃C₂T_x Mxene was prepared by HF etching of Al from Ti₃AlC₂ powder. 1.0 g Ti₃AlC₂ powder was added to a mixture solution of 20 mL hydrochloric acid (6 M) and 1.32 g LiF at 40 °C for 72 h. The resulting suspension was centrifuged and washed with DI water until the PH of the supernatant reached to 6. The Ti₃C₂T_x Mxene sample was dried at 100 °C for 12 h in oven.

S@Ti₃C₂T_x Mxene composite was prepared by a hydrothermal method. 150 mg Ti₃C₂T_x Mxene powder was dispersed in a mixture solution of 22.5 mL ethanol and 150 mL DI water using magnetic stirring and ultrasound. Then 4 mL CS₂ (Sigma-Aldrich, ≥99.0%) was used to dissolve 600 mg S (Sigma-Aldrich, ≥99.5%). Subsequently, the S solution was added into the former mixture. After mixing all the materials together, the mixture was then transferred to a 300 mL Teflon lined autoclave and kept in an oven at 180 °C for 12 h. After the reaction, the product was washed with distilled water for 3 times. Finally, the product was dried at 60 °C oven for 12 h. A schematic of the preparation of S@Mxene composite is shown in Figure 1. For a comparison experiment, a S/Mxene composite was prepared by a ball-milling method, in which Ti₃C₂T_x Mxene and S powder were mixed at the same mass ratio of Ti₃C₂T_x Mxene to S in the S@Mxene composite and ball-milled at 600 rpm for 1.0 h.

2.2 Characterizations

The structures of Ti₃AlC₂ MAX, Ti₃C₂T_x Mxene, and S@Mxene composite were characterized by X-ray diffraction (XRD, Panalytical XPert PRO Alpha-1) with Cu Kα radiation and transmission electron microscopy (TEM,

Figure 1: The schematic of preparation of S@ Mxene composite.

Hitachi HT7700). The morphologies of Ti_3AlC_2 MAX, $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene, and S@Mxene composite were determined using scanning electron microscopy (SEM, Hitachi SU 8230). The weight percentage of sulfur in S@Mxene composite was analyzed from the weight loss curves measured under air atmosphere on a thermogravimetric analysis instrument (TGA, TA instrument, Q500) with a heating rate of $5^\circ\text{C}/\text{min}$ to 600°C . The elemental mapping was obtained on HRTEM (JEM-2100F).

2.3 Electrochemical tests

A sulfur cathode was prepared by mixing 60 wt% S@Mxene composite, 30 wt% conductive carbon black and 10 wt% polytetrafluoroethylene (PTFE) into a paste, followed by an operation of pressing the paste onto a nickel foam. The sulfur mass loading is 4.0 mg cm^{-2} . The electrolyte used in this study was 1 M lithium bis(trifluoromethane sulfonyl) imide (LiTFSI, Sigma-Aldrich, 99.95%) dissolved in dimethoxyethane (DME, (Sigma-Aldrich, $\geq 99\%$) and 1,3-dioxolane (DOL, Sigma-Aldrich, $\geq 99\%$) (1:1, v:v), with the addition of 1 wt% lithium nitrate (Sigma-Aldrich, $\geq 99\%$). Celgard 2400 membrane was used as the separator. Lithium foil was used as counter electrode. The coin cells were assembled in an Ar-filled glove box. The galvanostatic discharge-charge experiments were performed on a Land CT2001 battery tester in the potential window of 1.6–2.8 V (vs Li/Li⁺). Cyclic voltammetry (CV) was measured at a scan rate of 0.1 mV s^{-1} within 1.6–2.8 V (vs Li/Li⁺) on VMP3 electrochemical workstation (Bio-logic).

3 Results and discussion

Scanning electron microscopy (SEM) was used to image the morphology of Ti_3AlC_2 MAX and $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene. Figure 2a,d and Figure 2b,e show SEM images of Ti_3AlC_2 MAX and the synthesized $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene, respectively. Figure 2a,d reveals the solid structure and smooth surface of Ti_3AlC_2 MAX. After HF etching, Ti_3AlC_2 MAX was exfoliated and converted to a multilayered structure (Figure 2b) which consists of many $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene nanosheets and the average distance of two neighbouring nanosheets is about 100 nm. Figure 2e shows the $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene has a rough surface, which facilitates the uniform distribution and anchoring of nanoparticles between layers. The structures of Ti_3AlC_2 MAX and the $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene were characterized by X-ray powder diffraction (XRD). Figure 2c shows the XRD patterns of Ti_3AlC_2 MAX before and after HF etching. Strong characteristic peaks at $2\theta = 9.6^\circ, 19.1^\circ, 34.1^\circ, 36.9^\circ,$

$38.9^\circ, 42.0^\circ, 45.0^\circ, 48.5^\circ, 52.5^\circ, 56.6^\circ,$ and 60.2° in blue correspond to (002), (004), (101), (103), (104), (105), (106), (107), (108), (109), and (110) planes of Ti_3AlC_2 (JCPDS No. 52-0875) [19], respectively. After HF etching, the XRD pattern shows a significant decrease in peak intensity, indicating a lower degree of crystallinity and a less ordered structure. The diffraction peaks at $2\theta = 36.9^\circ, 38.9^\circ, 42.0^\circ$ cannot be observed after etching, which suggests the removal of Al atoms from Ti_3AlC_2 MAX. The (002) diffraction peak at $2\theta = 9.6^\circ$ shifted to lower area ($2\theta = 8.6^\circ$), indicating the c-lattice parameter increased in the etching process. And the (002) diffraction peak became broadened, which is mainly because of the decrease in the degree of crystallinity. Moreover, the intensity of (110) plane increased, which resulted from the structural expansion in etching processes and substitution of Al with F and $-\text{OH}/=\text{O}$ terminating groups. The changes in XRD patterns of Ti_3AlC_2 MAX before and after HF etching indicate successful preparation of $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene.

The nitrogen adsorption-desorption measurements are carried out to investigate the porosity of the obtained $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene and the result is shown in Figure 2f. The nitrogen adsorption-desorption isotherms show that the pore volume and specific surface area of the $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene were $0.09\text{ cm}^3\text{ g}^{-1}$ and $39.23\text{ m}^2\text{ g}^{-1}$ according to the Brunauer-Emmett-Teller (BET) method. In contrast, the Ti_3AlC_2 MAX has a low pore volume of $0.008\text{ cm}^3\text{ g}^{-1}$ and a low specific surface area of $0.42\text{ m}^2\text{ g}^{-1}$. These results suggest that HF etching has resulted in lots of pore and active surface. Figure 2g–i shows SEM energy-dispersive X-ray spectroscopy and element mappings of the $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene. Element mappings reveal that Ti, C, O, and F atoms are uniformly distributed throughout the entire structure (Figure 2h), where F and O atoms are from the introduced $-\text{F}$, $-\text{OH}$, $=\text{O}$ group. The "T" in $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene is the introduced surface groups ($-\text{F}$, $-\text{OH}$ and $=\text{O}$ groups), and the "x" in $\text{Ti}_3\text{C}_2\text{T}_x$ is number of surface group per formula unit. According to EDS analysis, the $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene contains $13.1 \pm 0.4\text{ wt\% C}$, $61.9 \pm 1.7\text{ wt\% Ti}$, $13.9 \pm 0.3\text{ wt\% O}$, $10.8 \pm 0.3\text{ wt\% F}$, and $0.3 \pm 0.1\text{ wt\% Al}$. The much lower content of Al in $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene reveals that Al removal was almost completed. Besides, the mole ratio of Ti/C is calculated as ~ 1.3 , which is close to the stoichiometry of Ti_3C_2 .

To determine the content of sulfur in S@Mxene composite, the as-synthesized composite was examined by thermal gravimetric analysis (TGA) in N_2 atmosphere from room temperature to 600°C , and the result is presented in Figure 3a. The TGA curve shows a two-stage weight loss. During the first stage at $25\text{--}110^\circ\text{C}$, the weight loss of 0.62% is due to the evaporation of water and HF residue attached

Figure 2: (a, d) SEM images of Ti₃AlC₂ MAX. (b, e, g) SEM images of Ti₃C₂T_x Mxene. (c) XRD patterns of Ti₃AlC₂ MAX and Ti₃C₂T_x Mxene. (f) Nitrogen adsorption-desorption isotherms of Ti₃AlC₂ MAX and Ti₃C₂T_x Mxene. (h) Element mappings of Ti₃C₂T_x: Ti, C, O, F. (i) EDX spectrum of Ti₃C₂T_x Mxene.

on the composite surface. Subsequently during the second stage at 110–300 °C, a significant weight loss of about 67.7% originates from the evaporation of sulfur stored in the composite, as the melting point of bare sulfur is 112.8 °C. Thus the sulfur content is determined to be 67.1 wt %. Figure 3b shows the XRD patterns of bare S and the S@Mxene composite. A high similarity between the two patterns is observed, which is attributed to the high content of sulfur in the S@Mxene composite. In addition, sulfur nanoparticles cover the whole surface of Mxene and hence the XRD pattern of S@Mxene composite mainly shows the characteristic diffraction peak of sulfur. Figure 3d,e presents SEM images of the S@Mxene composite. It can be seen that all Ti₃C₂T_x Mxene were uniformly covered with sulfur nanoparticles. From Figure 3e, we can observe that the majority of sulfur nanoparticles are uniformly distributed between the layers of Mxene whereas the remaining nanoparticles cover the outside surface. In order to analyze

the dispersion of sulfur nanoparticles in Ti₃C₂T_x Mxene, EDS and element mapping of S@Mxene composite are performed (Figure 3c,f–i). These figures exhibit element distributions of S, Ti, and C, confirming the uniform distribution of S nanoparticles throughout the entire Ti₃C₂T_x Mxene (Figure 3g–i). EDS analysis reveal the S@Mxene composite contains 67.1 ± 1.2 wt% S, 20.4 ± 0.7 wt% Ti, and 12.5 ± 0.5 wt% C, which is consistent with the TGA result. The Ti₃C₂T_x Mxene consists of many Ti₃C₂T_x Mxene nanosheets with an average sheet distance of about 100 nm. The particle size of sulfur nanoparticles is lower than the sheet distance of the Mxene, that's why sulfur nanoparticles can be inserted into the space of Mxene and coated on the Mxene's nanosheets. Furthermore, the hydrothermal reaction is an effective route towards sandwich-like nanostructures. A hydrothermal method is very helpful to control the reaction between Mxene and sulfur. Reactants could be uniformly mixed at molecular level and thus

Figure 3: (a) TGA of S@Mxene. (b) XRD patterns of bare sulfur and S@Mxene. (c) EDX pattern of S@Mxene. (d–f) SEM images of S@Mxene. Element mappings of S@Mxene: S (g), Ti (h), C (i).

much small sulfur nanoparticles were synthesized, then a uniform distribution both between the layers and on the outside surface of Mxene was achieved.

TEM images of the $\text{Ti}_3\text{C}_2\text{T}_x$ in Figure 4a,b show that the $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene possesses a typical 2D multilayered structure and an inter-lamellar spacing of ~ 0.98 nm, the latter of which is attributed to the (002) plane of $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene [20]. HRTEM image (Figure 4c) indicates high crystallinity of $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene and the lattice fringe distance of 0.31 nm for the (110) plane of $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene [20]. Thus these results are in line with the XRD result above. Figure 4d,e presents the schematic diagram of three-dimensional network structure between Sulfur and $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene, and Figure 4f displays a schematic of the molecular model of $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene to appear the two main atoms of $\text{Ti}_3\text{C}_2\text{T}_x$. In order to clearly characterize the dispersion of sulfur nanoparticles in the Mxene-based composite, the microstructure of the

composite was also studied by TEM. As shown in Figure 4g,h, a large number of sulfur particles uniformly filled in the space between the $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene nanosheets and coated on $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene's surface. The whole $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene were uniformly covered and decorated with sulfur nanoparticles, showing a non-layered structure. Figure 4i shows the HRTEM image of the S@Mxene composite, where the 0.20 nm lattice spacing for S_8 -{025} [21] is observed, indicating the successful homogeneous loading of sulfur particles in $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene by the hydrothermal reaction.

Figure 5 illustrates the electrochemical performances of S@Mxene composite. Figure 5a presents the cyclic voltammetry (CV) curves of the S@Mxene composite at the scan rate of 0.1 mV/s with the voltage of 1.6–2.8 V (vs. Li/Li^+). The S@Mxene composite displays the typical CV curves of LSBs with two distinct reduction peaks and two oxidation peaks.

Figure 4: (a, b) TEM images of $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene. (c) HR-TEM image of $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene. (d, e) The schematic diagram of three-dimensional network structure between sulfur and $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene. (f) Schematic of the molecular model of $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene. (g, h) TEM images of S@Mxene. (i) HR-TEM image of S@Mxene.

The reduction peaks at 1.72 and 2.24 V correspond to the lithiation process whereas the oxidation peaks at 2.38 and 2.51 V correspond to the delithiation process. There are almost no peak shifts during the initial three cycles, indicating good electrochemical stability. Figure 5b shows the discharge-charge performance of S@Mxene composite electrode in a voltage range of 1.6–2.8 V versus Li/Li^+ at 0.2 C, including the first cycle, second cycle, 10th cycle, 50th cycle and 100th cycle. In all cycles, S@Mxene composite displayed three discharge potential plateaus at ~ 2.38 , ~ 2.12 , and ~ 1.80 V. The potential plateau at 2.38 V is due to the transformation from sulfur to higher order lithium polysulfides (Li_2S_x , $4 \leq x \leq 8$); while the low potential plateau at 2.12 V is ascribed to the reduction of higher order lithium polysulfides to lower order lithium polysulfides (Li_2S_2 or Li_2S) [22]. Another potential plateau at 1.80 V is attributed to the intercalation of Li^+ to $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene [23, 24]. The charge curves display two charge potential plateaus at ~ 1.86 and ~ 2.23 V. The potential plateau at ~ 2.23 V is due to the oxidation of Li_2S_2 and Li_2S to Li_2S_x ($4 \leq x \leq 8$) and eventually to S_8 [22]. The plateau at ~ 1.86 V is ascribed to the extraction of Li^+ from $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene [23, 24]. During the first cycle, the S@Mxene composite cathode delivered a discharge capacity of 1556.4 mAh g^{-1} and a charge capacity of 1544.4 mAh g^{-1} . And the capacity decreased slowly as the cycle number increased.

Figure 5c compares the specific capacity of S@Mxene cathode with that of S/Mxene cathode at 0.2 C. For the S/Mxene, the first reversible specific capacity was 840 mAh g^{-1} and rapidly dropped to 399.3 mAh g^{-1} after 100 cycles. For the S@Mxene composite, the first specific reversible capacity was 1477.2 mAh g^{-1} and retained 82.1% of the capacity after 100 cycles, which is higher than that of some similar S/C cathodes in previous works [25–27]. Compared with mechanical mixture of sulfur and Mxene, the advantage of S@Mxene composite by the hydrothermal reaction is that a more uniform distribution of sulfur nanoparticles between layers of Mxene can be achieved. With a multilayered 2D structure and high electrical conductivity (1.1×10^5 S/m), Mxene as a host material can greatly enhance the electron transport of sulfur and thus increase the first specific reversible capacity. The cyclability is improved due to the following reasons. The layers of Mxene have good ductility, which can buffer the volume expansion and shrinkage of sulfur nanoparticles, and thus to maintain the structural stability of the composite during cycling. Meanwhile, sulfur nanoparticles distributed between the layers help to keep the layers spaced out, which prevent Mxene from stacking up. In conclusion, the synergistic effect between Mxene host and sulfur particles greatly enhances the cyclability performance of S@Mxene.

Figure 5: (a) Cyclic voltammetry (CV) of S@Mxene composite at the scan rate of 1.6–2.8 V at 0.1 mV/s. (b) Galvanostatic charge/discharge curves of S@Mxene composite at 0.2 C at 1.6–2.8 V, (c) Cycling performance of S@Mxene composite and mixed-S/Mxene composite at 0.2 C. (d) Rate performance of S@Mxene composite.

composite. The rate capability of S@Mxene composite is also superior (Figure 5d), when the current density increases to 2 C, the S@Mxene composite delivers a capacity of 860.2 mAh g^{-1} . After the current density returns to 0.2 C, the capacity is recovered to be $1180.3 \text{ mAh g}^{-1}$, revealing outstanding lithium ion storage reversibility.

4 Conclusions

In summary, a composite of S@Mxene is facilely synthesized by the hydrothermal method. SEM and TEM images show that the $\text{Ti}_3\text{C}_2\text{T}_x$ Mxene possesses layered-stacked structure which can accommodate the large volume changes of sulfur during cycling. And sulfur nanoparticles are homogeneously embedded in the Mxene sheets and coated on the outside surface of Mxene. Because of Mxene's high electrical conductivity and multilayered 2D structure, S@Mxene composite shows improved reversible specific capacity and cycling performance. The S@Mxene composite with a high areal sulfur loading ($\sim 4.0 \text{ mg cm}^{-2}$) shows a first specific reversible capacity of $1477.2 \text{ mAh g}^{-1}$ and a specific reversible capacity of $1212.2 \text{ mAh g}^{-1}$ after 100 cycles at 0.2 C (0.18% capacity loss per cycle). These results suggest a scalable preparation of high electrochemical performance cathode for LiS battery with low cost and simplicity.

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