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Advanced MXene materials: Paving the way for next-generation lithium-sulfur batteries in energy storage

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Abstract. In recent years, lithium-sulfur (Li-S) batteries, with high theoretical energy density and low cost, have become one of the most promising energy storage systems. However, the insulation of sulfur, the "shuttle effect" of polysulfides and volume expansion hinder their practical applications. MXenes have excellent electrical conductivity, high specific surface area, and a strong adsorption capacity for polysulfides, which helps to overcome the shortcomings of traditional Li-S batteries. In this paper, the structure, properties and preparation methods of MXenes are reviewed, and the applications of MXenes in the cathode, separator and anode of Li-S batteries are introduced. Finally, the future directions of MXenes in the field of Li-S batteries are prospected.

Keywords: MXene, lithium-sulfer battery, shuttle effect, polysulfides

1. Introduction

Industrial development, environmental concerns, and economic considerations have put forward higher demand for battery systems. Lithium-ion batteries (LIBs) have become one of the best choices due to their advantages such as high energy efficiency, no memory effect and long cycle life.[1] Nevertheless, they face challenges such as limited energy density and safety concerns, resulting in the pursuit of novel energy storage devices that go beyond LIBs.[2] As a kind of rechargeable battery, the charge and discharge process of lithium-sulfur (Li-S) battery is carried out through S-S bond breaking and bonding in S8 molecule. At the same time, the dissolution/deposition of lithium happens on the anode surface.[3] Li-S batteries have been considered highly promising because of their high energy density, environmental friendliness, and the abundance of sulfur.[4] When elemental sulfur is completely reduced to lithium sulfide (Li2S), the specific capacity of sulfur can theoretically reach 1675 Ah·Kg−1, and the energy density can theoretically reach 2600 Wh·Kg−1, which is three to five times higher than that of LIBs (Figure 1).[5]

However, the practical application of this type of battery has been hindered by several critical issues. The intrinsically insulating sulfur reduces the electron transfer rate, resulting in slower reaction kinetics, which lowers the charge and discharge rate and reduces the efficiency.[6] Besides, the dissolution of lithium polysulfides leads to the shuttle effect, which affects the cycle life and efficiency of the battery, and causes lithium corrosion.[7,8] Moreover, The density of the product (Li2S) is larger than that of sulfur, so sulfur cathodes undergo considerable structural and volume changes during cycling.[9]

Volume expansion exposes the battery to mechanical stress, which can result in electrode deterioration and shorten the cycle life of the batteries.

As a new type of transition metal carbon/nitrogen two-dimensional material, MXenes have been more and more widely used in the cathode, separator and anode of Li-S batteries in recent years, and some good results have been achieved. This paper introduces the structures, properties and synthesis methods of MXenes and their applications in (Li-S) batteries.

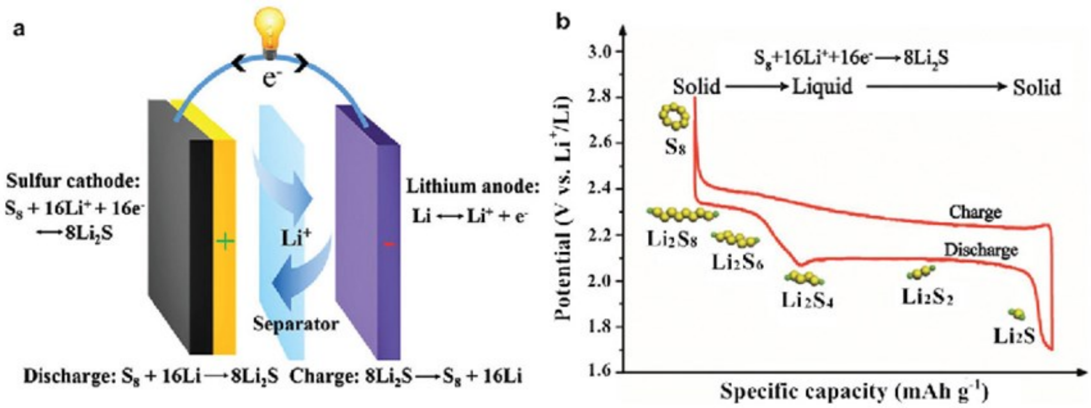

Figure 1. (a) Schematic of the electrochemistry for Li–S batteries. (b) A typical charge–discharge voltage profile of Li–S batteries in ether-based electrolyte.[10] Copyright 2021, John Wiley and Sons.

2. MXenes

2.1. Structure

Since the discovery by Gogotsi et al., MXenes, a unique type of two-dimensional materials with accordion-like morphology and multi-layer structure, have been extended to various energy applications.[11-14] They have high electrical conductivity, appreciable Young's modulus, and inherent two-dimensional flexibility. The layered structure of MXenes can also help to achieve a large specific surface area, which is critical for achieving abundant active sites and high energy density storage. In addition, they enable a wide range of energy storage mechanisms under a variety of conditions, especially under acidic conditions where functional transformations occur on the surface of MXene.[15]

By introducing metal vacancies and doping nitrogen atoms, the structure of MXenes can be further customized to improve their pseudocapacitive performance. These modifications improve the electrochemical performance of MXenes by increasing its ability to store and transfer charges, enabling it to be applied more efficiently in energy storage systems.[16] To overcome the self-stacking problem, researchers have developed several methods to design MXene electrodes, including introducing "spacers" such as carbon nanotubes or graphene. These spacer layers maintain separation between the MXene layers, which enhances ion transport and maximizes surface utilization, improving their electrochemical performance.[17] Researchers have also successfully combined MXenes with other materials, such as polymers and metal oxides, to create composites suitable for specific functions. These composites exhibit improved performance, further extending the benefits of MXene for energy storage applications.[18,19]

2.2. Synthesis

The synthesis of MXenes involves a variety of techniques to produce materials with specific properties suitable for different applications, which reflects their adaptability. These synthesis methods can be divided into top-down and bottom-up types.[20] In the top-down approach, the MAX phase is etched the MAX phase with various chemicals to customize the end groups and inherent properties of the resulting MXenes (Figure 2). This approach is generic and allows the properties of MXenes to be

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modified for specific applications.[21-24] The selection of precursor synthesis, etching and delamination methods determines the wafer size, defect level, synthesis yield and surface function of MXenes, and the selection of appropriate methods can optimize their application-specific performance.[25] The bottom-up synthesis of MXenes involves methods such as chemical vapor deposition (CVD). In CVD, gases such as methane react at high temperatures on a substrate to form MXene layers, providing precise control over thickness and morphology, ideal for producing high-quality films with minimal defects.[26]

The field of MXene synthesis is developing rapidly with the introduction of innovative methods using non-aqueous etchants, molten salts, fluoride salts and acid halogens. These new methods play a key role in optimizing the surface chemistry of MXenes, thus expanding their functional applications.[27] Synthetic processes have also been customized to manufacture specific types of MXenes, such as vanadium carbide MXenes.[28] At the same time, there is a growing focus on developing more environmentally friendly and sustainable methods of MXene synthesis. These efforts aim to reduce the environmental impact while maintaining or enhancing the desired performance of MXenes through green synthetic technologies.[29]

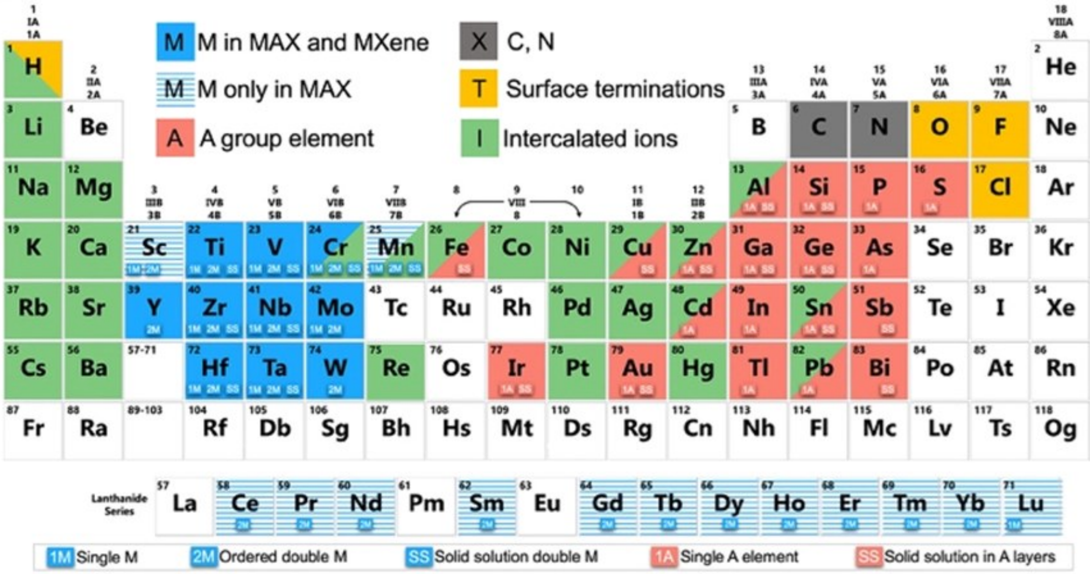

Figure 2. Elements used to build MAX phases, MXenes, and their intercalated ions.[30] Copyright 2019, American Chemical Society.

2.3. Properties

Due to their outstanding characteristics, MXenes have received a lot of attention in the field of Li-S batteries. One of the distinctive features of MXenes that makes them unique, and therefore best suited for Li-S batteries is their unique terminal metal sites. This feature is the reason behind MXenes' effective binding of different sulfur species such as lithium-polysulfides via Lewis acid strength and polarity effects, contributing to a new way to improve battery chemistry and functions.[31] This is basically a core feature why MXenes can prevent the dissolution of polysulfides into the electrolyte-a big and complicated issue in the life of a Li-S battery. Furthermore, the high conductivity and abundant active sites of MXenes also offer great improvements in battery performance through the efficient transport of electrons, the rapid transformation of polysulfides and the formation of uniform and regular lithium metal, which is a target factor that also notably improves the cyclic stability and rate capability.[32] MXenes have excellent mechanical performance as well,[15] ensuring the structural stability of the electrodes.[33]

The surface functional groups of MXene, such as oxygen and fluorine atoms on the end groups, exhibit significant electronegativity, resulting in a high negative charge on the surface of MXene. This property enhances their ability to polarize the material on contact, which is conducive to promoting the interaction of sulfur species and other components in Li-S batteries, thereby improving electrochemical performance.[34] The combination of these properties makes MXenes ideal for Li-S battery electrodes, promoting efficient reaction kinetics and providing the necessary chemical activity and stability during cycling.

In terms of its structural properties, MXenes are able to inhibit the growth of lithium dendrites, thereby improving the performance of lithium metal anodes in Li-S batteries. This suppression is essential to maintain the integrity of the anode and improve battery safety.[35] The MXene/ carbon nanotube (CNT)/MXene sandwich layers form multiple physical barriers, as well as the chemical trapping and catalytic activity of MXenes, effectively preventing the shuttle of polysulfides under high sulfur loading conditions. This greatly enhances the confinement of 3D hosts, addressing one of the key challenges associated with polysulfide shuttling and improving the stability of high-load Li-S batteries.[36]

MXenes have a number of remarkable properties that make them ideal for use in Li-S batteries. Their polar surface properties, high electrical conductivity, mechanical robustness, and ability to effectively inhibit polysulfide shuttling make them promising candidates for improving the performance, stability, and energy density of Li-S batteries. The combination of these features highlights the transformative potential of MXenes in the next generation of battery technology.

3. Applications of MXenes in Li-S batteries

3.1. Applications in cathodes

As mentioned above, MXenes has high conductivity, adsorption and catalytic conversion of polysulfides, and the application of MXenes to the main material of sulfur cathode helps to improve the rate performance and cycle stability of the battery.[37-39] Mxene-based materials also have good compatibility with sulfur nanoparticles, which can promote the efficient utilization of sulfur during the operation of Li-S batteries.[35]

Ti3C2Tx has been effectively used to prepare nano-carbon hybrid cathodes. In this structure, Ti3C2Tx, as an excellent sulfur host, strongly inhibits the diffusion of polysulfide and promotes rapid electron transfer.[40] This structural and electrochemical performance enhancement has led to the production of high-performance Li-S batteries capable of providing higher power output and longer battery life. This approach, which focuses on optimizing the structural arrangement and lamination of nanostructured carbon hybrid cathodes, represents a significant advance in battery technology, ensuring greater efficiency and performance.[41] The Ti3C2Tx three-dimensional hybrid structure integrated with carbon nanotubes (CNTs) greatly enhanced the Li+/e transport pathway and provided a wealth of electroactive sites (Figure 3). These properties are essential for the efficient binding and retention of sulfur in the battery structure, thereby optimizing its capacity and lifetime.[42]

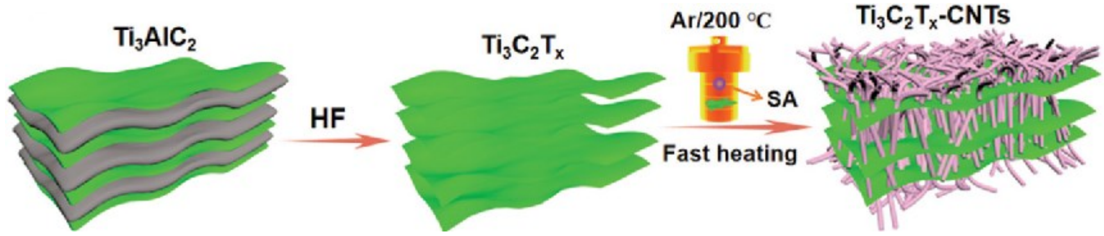

Figure 3. Schematic illustration of the Ti3C2Tx-CNTs hybrids.[42] Copyright 2021, John Wiley and Sons.

Bao et al. demonstrated a one-step method to synthesize crumpled N-doped MXene nanosheets with a well-defined porous structure. These structural features not only increase the surface area, but also improve the physical and chemical co-adsorption properties for polysulfides, which helps prevent the dissolution of these compounds in the battery electrolyte. This configuration results in a high sulfur load of 5.1 mg·cm-2, a high reversible capacity of 1144 mAh·g⁻¹ at a rate of 0.2 C (Figure 4).[43]

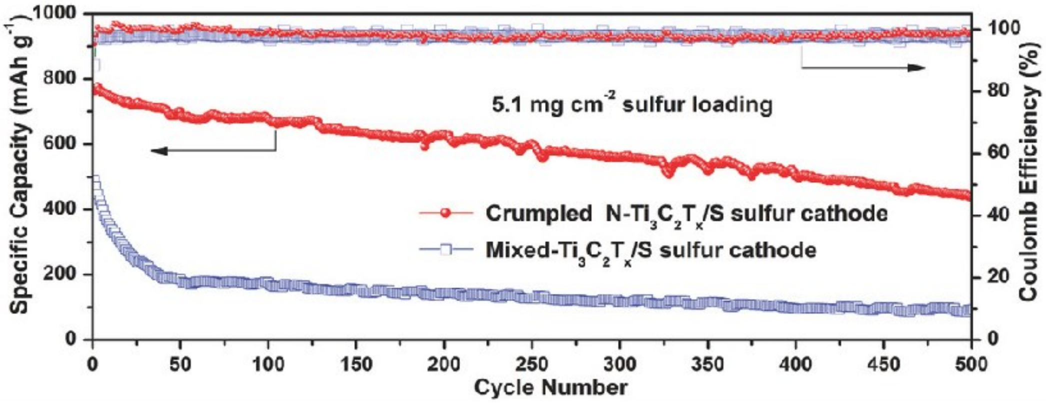

Figure 4. Cycling performances of crumpled N-Ti3C2Tx/S electrodes and mixed-Ti3C2Tx/S electrodes with 5.1 mg·cm−2 sulfur loadings at 0.2 C for 500 cycles (1 C = 1673 mA·g–1).[43] Copyright 2018, John Wiley and Sons.

Zhong et al. developed a sandwich MXene-based cathode for flexible Li-S batteries (Figure 5). Using a scalable drop-casting method, they fabricated a Ti3C2Tx film that enhances adhesion to the sulfur layer and maintains flexibility. The TiO2 anchored on the porous Ti3C2Tx improves lithium ion transport and prevents polysulfide shuttling. This configuration achieved a high capacity of 740 mAh·g⁻¹ g⁻¹ at 2 C, with 81% capacity retention after 500 cycles, and maintained 95% capacity after bending 500 times.[44]

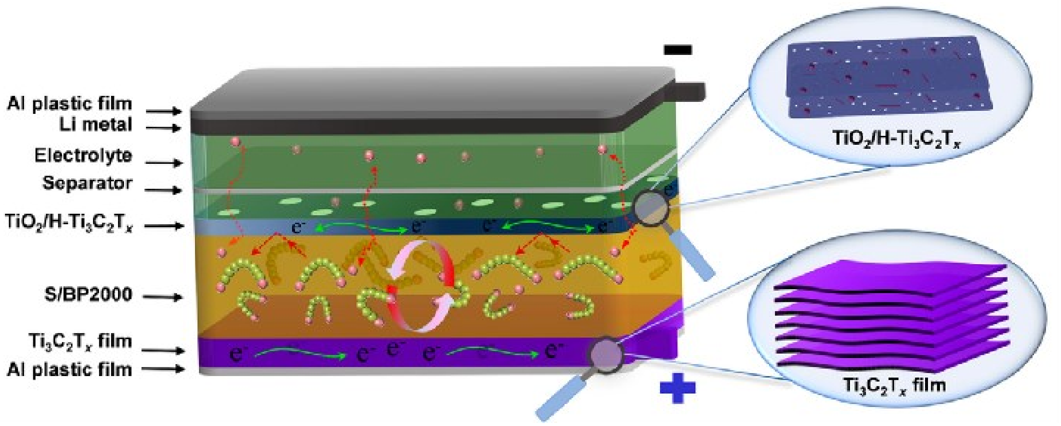

Figure 5. Schematic of flexible Li–S batteries using the Ti3C2Tx film as the current collector and TiO2/H–Ti3C2Tx as the coating layer.[44] Copyright 2022, American Chemical Society.

Qiu et al. developed a hierarchical MXene@TiO2 nanoarray for Li-S batteries through an in situ solvothermal strategy followed by heat treatment. This advanced structure significantly enhances charge transfer and sulfur encapsulation due to its robust dual chemisorption capability to trap polysulfides. This is achieved through the intense polarity of TiO2 and the abundant acid metal sites of MXene. As a result, the MXene@TiO2 nanoarray as a sulfur host demonstrated a highly stable discharge capacity of 612.7 mAh·g⁻¹ after 500 cycles at a rate of 2 C, with a very low fading rate of 0.058 % per cycle.[45]

Chen et al. prepared CeO2/MXene as a cathode material for Li-S batteries. CeO2 chemically adsorbs lithium polysulfides, while MXene, anchors sulfur, through its layered structure, effectively suppressing the shuttle effect. This synergistic action allows the battery to achieve an initial discharge specific capacity of 505.5 $mAh \cdot g^{-1}$ at 0.2 C. After 50 cycles, the capacity retention rate is 99.41%, with a discharge specific capacity of 502 $mAh \cdot g^{-1}$ and an average coulombic efficiency of 98.04%.[46]

3.2. Applications in separators

MXenes show great promise in optimizing Li-S battery separatorsby helping to mitigate the shuttle effect of polysulfides between the anode and cathode. Several studies have investigated the functionality of MXenes in separators and focused on various aspects of battery performance, including charge and discharge rates, cycle durability, and the overall stability.

Lee et al. demonstrated the use of CO2-oxidized Ti3C2Tx-MXenes components to inhibit shuttling through physical and chemical adsorption of lithium polysulfides (Figure 6). This modification method involves using CO2 oxidation delamination transition metal carbides (MXenes) to generate Oxi-d-MXenes, which are then used as modified separators coated on glass fibers without the need for conductive materials and adhesives. This method effectively inhibits the diffusion of lithium polysulfides and solves a key problem of Li-S batteries.[47]

Oschinski et al. highlighted the catalytic effect of well-dispersed Zn atoms on MXene (Ti3C2). This catalytic activity helps to increase the redox rate of polysulfide, thereby improving redox kinetics and overall cell efficiency.[48]

Huang et al. discuss the effective synergies of chemisorption and Wackenroder reactions for Li-S batteries by heterostructure La2O3-Ti3C2Tx-embedded carbon nanofibers (Figure 7). The results show that the La2O3 domain has strong adsorption capacity, which can adsorb lithium polysulfides for subsequent catalytic conversion, and the hydroxyl terminal group on the surface of MXene promotes the rapid conversion of lithium polysulfides to Li2S via the Wackenroder reaction. This approach results in excellent cycle stability and high capacity retention in Li-S batteries.[49]

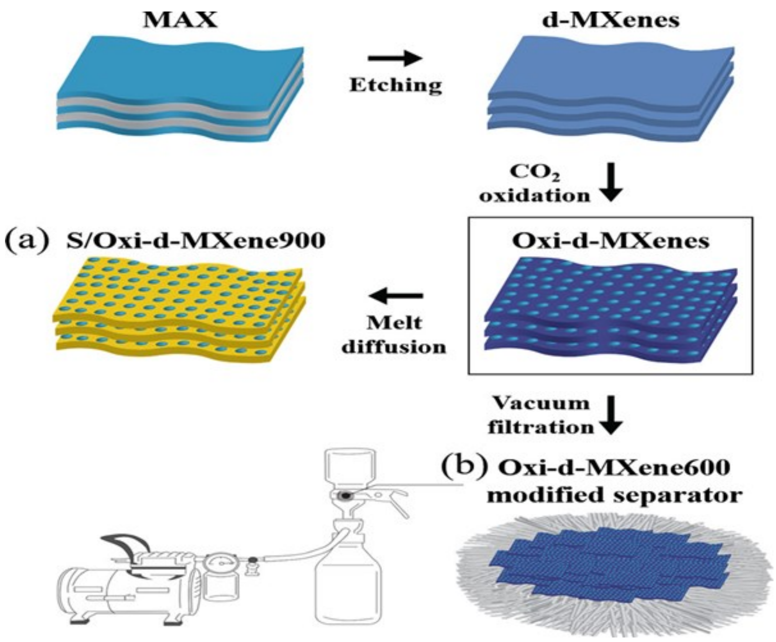

Figure 6. Schematic showing the synthesis procedure for the Oxi-d-MXenes: (a) S/Oxi-d-MXene900 composites prepared by melt diffusion of S; (b) Oxi-d-MXene600-modified separator prepared by facile vacuum filtration on a glass fiber (GF) separator.[47] Copyright 2020, American Chemical Society.

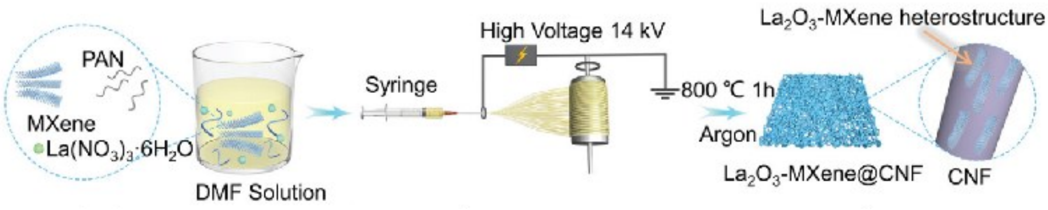

Figure 7. Schematic illustration for the preparation process.[49] Copyright 2023, John Wiley and Sons.

Song et al. improved the performance of Li-S batteries by coating commercial separators with Ti3C2Tx MXene nanosheets (Figure 8). This method is designed to fix the polysulfides, thereby stabilizing the chemical environment of the battery and prompting the redox reaction that is critical to the efficiency of the battery.[50]

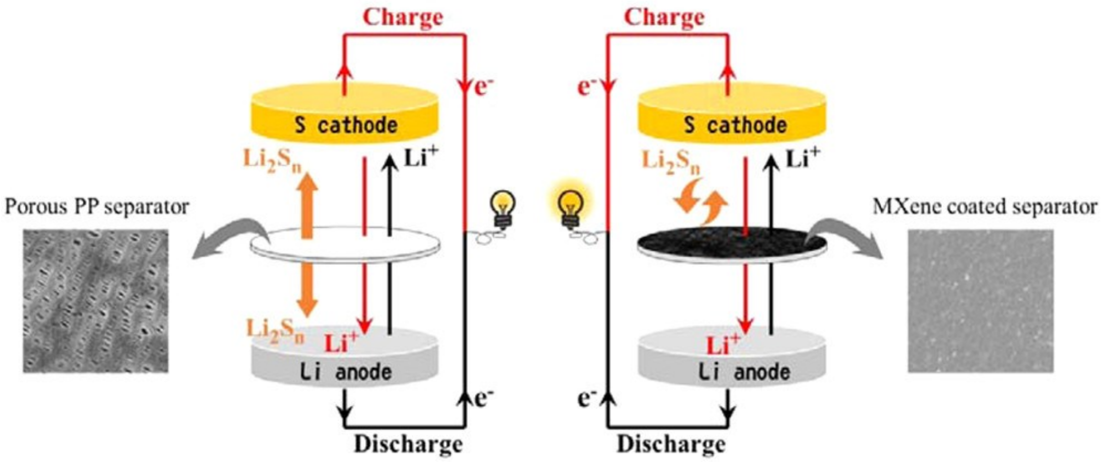

Figure 8. Schematic configuration of the Li–S cells using PP and MPP separators.[50] Copyright 2016, American Chemical Society.

Wang et al. developed a biomimetic composite separator for Li-S batteries featuring a "brick-and-mortar" structure of porous Ti3C2Tx and aramid nanofibers (ANFs). This design enhances lithium-ion transport and reduces the polysulfide shuttle effect, thereby improving stability. The separator showed high ionic conductivity (0.42 mS·cm-1), exceptional mechanical strength (Young's modulus of 5.6 GPa), and supported over 3,500 cycles with minimal capacity decay (0.013% per cycle). Its structure optimizes ion transport and provides crucial mechanical and thermal stability for high-performance batteries (Figure 9).[51]

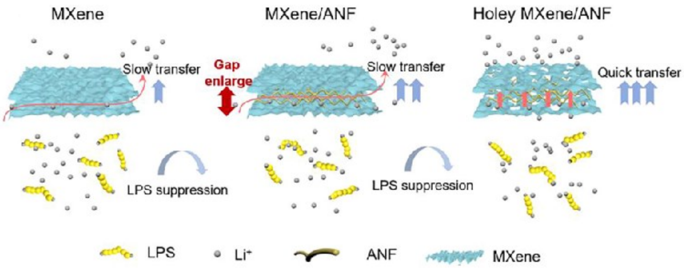

Figure 9. Schematic configuration of the Li–S batteries using PP and MPP separators.[51] Copyright 2023, Elsevier.

Another key application of MXene in separator is as an intermediate layer, which plays a crucial role in improving the efficiency and stability of the battery. MXene intermediate layer can not only provide a physical barrier, but also provide chemical capture and catalytic activity to effectively inhibit the shuttle of polysulfide lithium. For example, Zhang et al. developed MXene/CNT/MXene sandwich structures that significantly improve lithium polysulfide confinement under high sulfur loads, demonstrating significant advances in the design and function of Li-S batteries (Figure 10).[52]

Dong et al. demonstrated the effectiveness of MXene nanosheets as the middle layer of an all-MXene integrated electrode designed for high-energy-density Li-S batteries. In This study, delaminated TiC MXene nanosheets were used as an intermediate layer on a polypropylene separator, showing the great potential of MXene to improve the performance of Li-S batteries (Figure 11).[53]

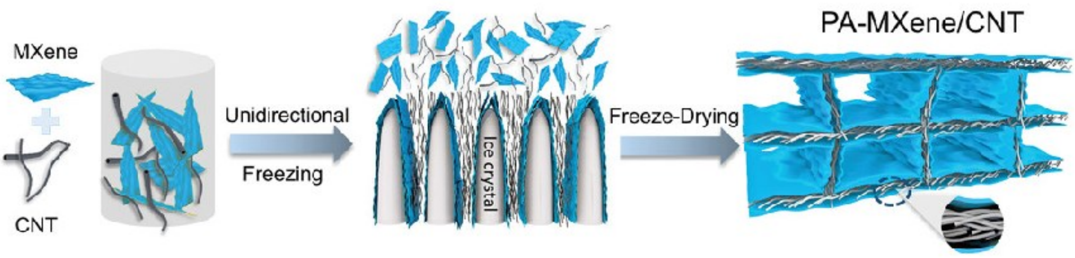

Figure 10. Schematic of the assembly process of the PA-MXene/CNT aerogel by the unidirectional freeze-drying.[52] Copyright 2021, John Wiley and Sons.

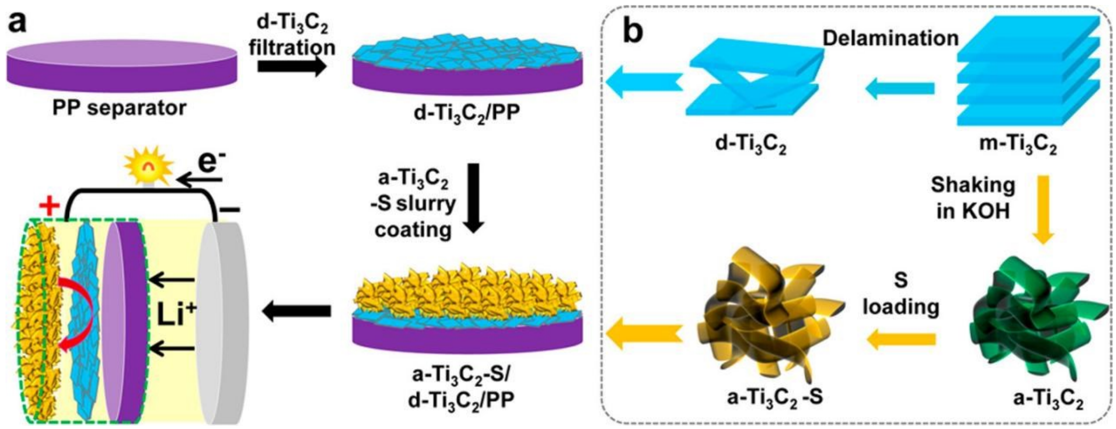

Figure 11. (a) Schematic of the fabrication of the flexible and integrated a-Ti3C2–S/d-Ti3C2/PP electrode for Li–S batteries. (b) Schematic of the preparation of d-Ti3C2 nanosheets and a-Ti3C2–S hybrid.[53] Copyright 2018, American Chemical Society.

Zhang et al. used a self-assembled KB@Ti3C2Tx composite as an intermediate layer on a separator to slow the cross-diffusion of polysulfides in Li-S batteries (Figure 12). This approach provides a comprehensive design strategy that takes advantage of the unique properties of MXene nanosheets to enhance the sulfur load and overall stability of Li-S batteries.[54]

3.3. Applications in anodes

With excellent electrical conductivity, mechanical strength, and ion transport capabilities, MXenes show great potential in anode design for Li-S batteries.[55] These properties enable MXenes to support highly conductive pathways of electrons and ions, resist mechanical stress, and maintain structural integrity

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over the battery operating cycle, making them ideal for improving battery anode durability and efficiency.[56]

Li et al. developed a flexible Ti3C2 MXene-lithium thin-film anode with a layered structure. By strategically embedding lithium within the nanoscaled gaps of MXene layers, they were able to confine the plating of lithium effectively, achieving very low overpotential (32 mV at 1.0 mA·cm-2) and maintaining stable cycling performance over 200 cycles with only a 1.5% increase in overpotential.[57]

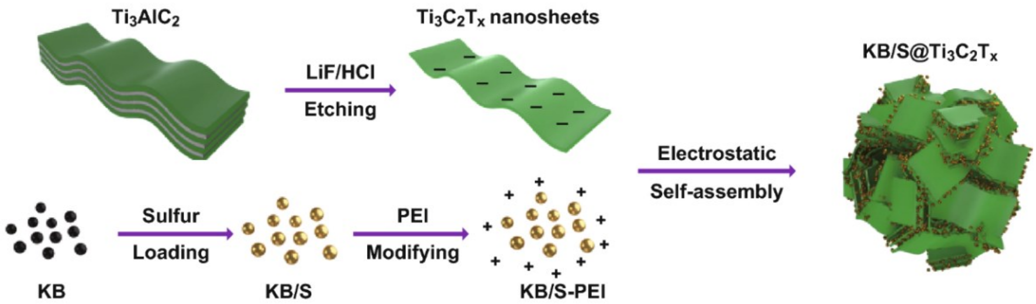

Figure 12. Schematic illustration of the fabrication of the KB/S@Ti3C2Tx composite.[54] Copyright 2020, Springer Nature.

Cao et al. reported that perpendicular MXene arrays enabled a high-rate capability of 20 mA·cm−2 and a low overpotential of 25 mV. These arrays, structured to optimize ionic flow and electron transport, delivered a high specific capacity of 2056 mAh·g−1 and demonstrated excellent cycle stability for up to 1700 hours (Figure 13). The vertical alignment of these MXene arrays played a crucial role in inhibiting volume changes and dendrite growth, thereby solidifying their utility in the construction of high-performance lithium metal anodes with prolonged operational life.[58]

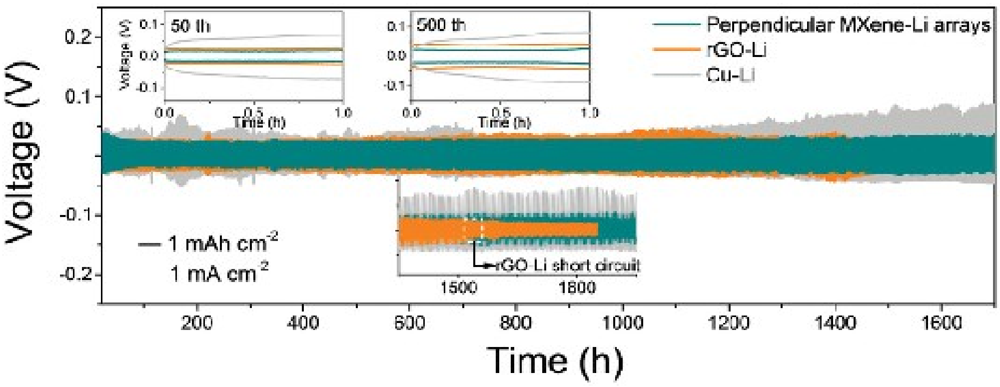

Figure 13. Cycling performances of symmetric cells with perpendicular MXene–Li arrays, rGO–Li, and Cu–Li arrays at 1 mA·cm−2, 1 mAh·cm−2, showing a long cycle life up to 1600 h with low overpotentials of ≈25 mV, superior to rGO–Li and Cu–Li arrays.[58] Copyright 2019, John Wiley and Sons.

Zhang et al. showed that parallelly aligned MXene layers facilitated a uniform and dense solid electrolyte interface with lithium fluoride, which effectively uniformized the electromigration of lithium ions. This arrangement optimized ion transport and minimized the formation of non-uniform lithium deposits. The hybrid anode achieved a cycle life of up to 900 hours and demonstrated exceptional deep stripping–plating capacities of up to 35 mAh·cm−2, crucial for high-density energy storage applications (Figure 14).[59]

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Li et al. found that layered MXene protected lithium metal anodes showed significant improvement in inhibiting the polysulfide shuttle effect, optimizing overall sulfur utilization and maintaining a high coulombic efficiency above 99% over 500 cycles (Figure 15). The strategic implementation of MXene layers served to mitigate sulfur loss and electrolyte degradation, key factors in extending the life and efficiency of Li-S batteries.[60]

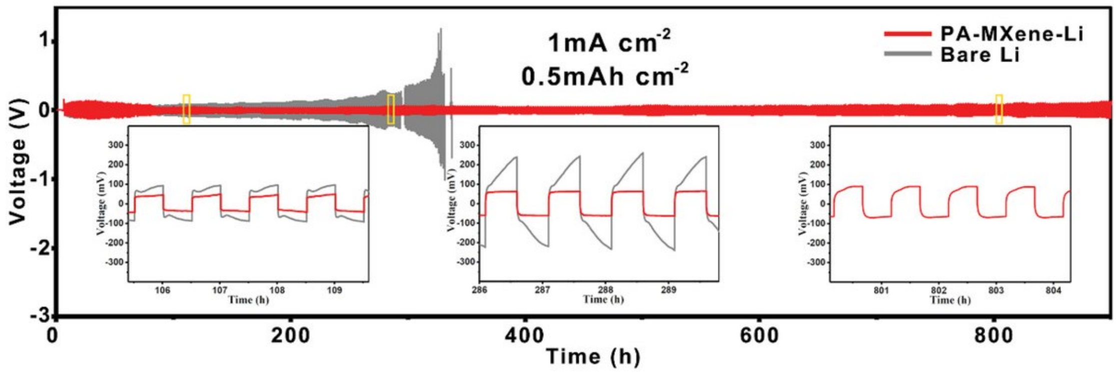

Figure 14. Galvanostatic cycling of symmetric cell with PA-MXene-Li anodes at 1.0 and 0.5 mAh·cm−2, exhibiting a long life up to 900 h.[59] Copyright 2019, John Wiley and Sons.

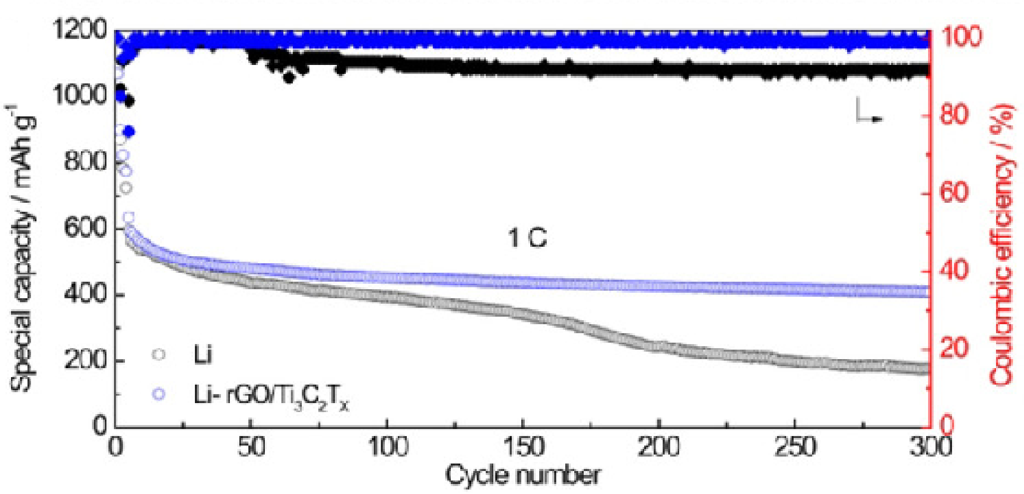

Figure 15. Cycling performance of Li−S cells with bare Li metal and Li-rGO/Ti3C2Tx anodes.[60] Copyright 2020, John Wiley and Sons.

Wang et al. tested MXene derivative skeletons across a wide temperature range. They maintained high conductivity and mechanical integrity from -40 to 60 °C, enabling stable lithium ion flux under varying environmental conditions and thus broadening the practical applications of lithium metal anodes (Figure 16).[61]

Wei et al. discussed the development of flexible and stable 3D lithium metal anodes using self-standing MXene/COF frameworks. They reported that lithiophilic COF-LZU1 microspheres distributed among the MXene film framework promoted uniform lithium nucleation by homogenizing the Li+ flux and lowering the nucleation barrier. This novel design led to dense and dendrite-free lithium deposition, significantly enhancing the coulombic efficiency and electrochemical stability of the symmetric cells, with Li-S full cells exhibiting superior electrochemical performance.[62]

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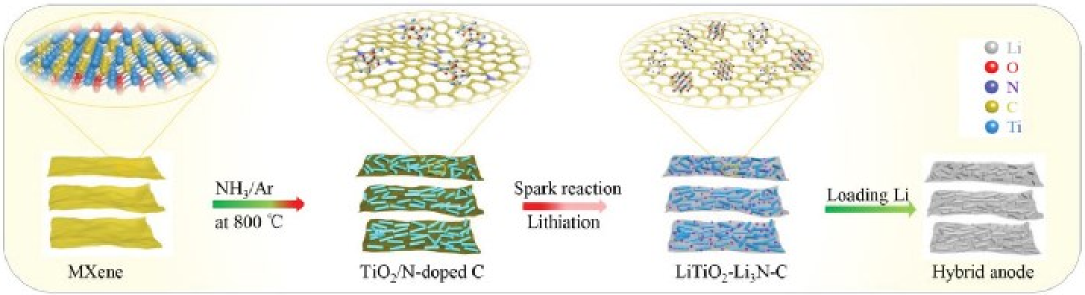

Figure 16. Schematic of the fabrication process of the LiTiO2-Li3N-C electrode.[61] Copyright 2021, John Wiley and Sons.

4. Summary and outlook

Li-S batteries are known for their high theoretical energy density of about 2,600 Wh·kg-1, which is substantially higher than conventional lithium-ion batteries, but theyt face several challenges that hinder their commercialization. These challenges include poor electrical conductivity of sulfur, volume expansion, and polysulfide shuttle. MXenes have become a game changer in the field of Li-S batteries. They have a unique accordion-like form, high electrical conductivity and excellent mechanical properties. These characteristics make MXenes particularly suitable for energy storage applications where high electrical conductivity and structural stability are critical. This detailed analysis focuses on how MXenes can address current challenges and enable next-generation energy storage solutions and explores the future prospects of MXenes' applications in revolutionizing Li-S battery technology.

One of the most promising applications for MXenes is the modification of cathodes for Li-S batteries. By integrating MXenes and sulfur in the cathode, the polysulfide shuttle effect can be mitigated. Due to their high surface area and strong adsorption capacity, MXenes help to confine sulfur and lithium polysulfides within the cathode. This improves the cycle stability and efficiency of the batteries. In addition, MXenes can act as a conductive adhesive, improving the overall conductivity of the cathode. This is crucial because the insulating nature of sulfur limits battery performance. With MXenes, the transfer of electrons between sulfur species is facilitated, thus improving the charge and discharge rate and overall performance of batteries.

In addition to cathodes, MXenes are also used for anode and separator enhancement. The addition of MXenes to the anode helps regulate the deposition of lithium and promotes a uniform lithium ion flux on the anode surface, thereby minimizing the formation of dendrites and maintaining structural integrity. This is essential to improve battery safety and longevity.

In the separator, MXenes improve ionic conductivity and mechanical strength. They can be integrated into the separator matrix to form a stronger barrier that can withstand physical stress and inhibit the growth of dendrites through the separator, further improving the safety of batteries.

In order for MXenes to have a broader impact on Li-S battery technology, scalable and cost-effective synthesis methods need to be developed. Research into more sustainable and efficient production techniques, possibly through green chemical methods or continuous manufacturing processes, will be essential.

Future development of MXenes in Li-S batteries could also utilize the synthesis of mixed materials. Combining MXenes with other functional materials such as carbon nanotubes, graphene or conductive polymers can further improve the performance of batteries. This hybrid battery can take advantage of the complementary properties of each component, such as increased mechanical strength, enhanced electrical conductivity and superior chemical stability, enabling next-generation battery designs with higher performance metrics.

The functionalization of MXenes through surface modification allows their performance to be tuned to better meet specific battery requirements. For example, surface functional groups can be designed to enhance the adsorption of polysulfides or to improve compatibility with different electrolytes. This customization is critical to optimizing the integration of MXenes into a variety of battery components and can open new avenues for the design of high-performance Li-S batteries.

Under practical conditions, comprehensive system-level testing and integration of MXenes in lithium batteries will be critical. This includes evaluating battery performance enhanced by MXenes in terms of energy density, charge and discharge rate, temperature stability and life cycle under various operating pressures. Such an assessment will help identify practical challenges and enable iterative enhancements based on performance feedback.

As the use of MXenes in Li-S batteries continues to advance, compliance with regulatory and safety standards will become increasingly important. Establishing guidelines for the safe handling, disposal and recycling of MXenes and Mxene-enhanced batteries is critical, especially as these materials move towards commercialization. Developing standards and best practices will ensure environmental compatibility and user safety, promoting wider adoption and market acceptance.

In conclusion, MXenes, with its excellent material properties and versatility of battery component design, holds considerable promise for addressing the limitations of current Li-S batteries. MXenes has potential breakthroughs in scalable production, hybrid material development, advanced functionalization, and comprehensive system-level integration of Li-S batteries. By overcoming existing obstacles, MXenes is at the forefront of advancing next-generation battery technology, heralding a new era of energy storage solutions.

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