ISSN: 2710-0723

Recent Advances in Molybdenum Disulfide Coatings for Stabilizing Zinc Anodes in Aqueous Zinc-Ion Batteries

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Abstract

Aqueous zinc-ion batteries (AZIBs) have emerged as one of the most promising next-generation battery technologies for large-scale energy storage due to their high safety, low cost, and environmental friendliness. However, the practical application of zinc metal anodes faces severe challenges such as uncontrollable dendrite growth, hydrogen evolution reaction, corrosion, and passivation, which significantly restrict the cycling lifespan and Coulombic efficiency of AZIBs. Interface engineering, particularly the construction of artificial protective layers, is considered an effective strategy to address these issues. The two-dimensional layered material molybdenum disulfide (MoS_2), with its unique physicochemical properties-including high mechanical strength, excellent chemical stability, two- dimensional ion diffusion channels, and tunable electronic structure-has become an ideal protective coating material for zinc anodes.This paper systematically reviews the recent research progress of MoS_2 coatings in stabilizing zinc anodes. First, the failure mechanisms of zinc anodes and the multiple protection mechanisms of MoS_2 coatings are elaborated, including acting as a physical barrier, regulating Zn^{2+} ion flux to achieve unInterface deposition, reducing the nucleation overpotential, and inhibiting side reactions. Second, the current mainstream preparation strategies for MoS_2 coatings are detailed, including binder-free electrochemical deposition and scalable spray coating methods. The influence of coating thickness, uniformity, and phase (e.g., 2H phase vs. 1T′ phase) on performance is discussed.Subsequently, the enhanced electrochemical performance of MoS_2@Zn anodes in symmetric cells and full cells is summarized. For example, it can achieve stable cycling for over 1200 hours at 0.5 mA cm-2, and when paired with a MnO_2 cathode, it exhibits a high reversible capacity of 225.8 mAh g-1 with excellent capacity retention. Furthermore, MoS_2 composite coatings (e.g., PEDOT:PSS/MoS_2) and emerging applications of MoS_2 in cathode protection are introduced. Finally, the challenges and future development directions for MoS_2 coatings are prospected, including performance validation at high depths of discharge, optimization of large-scale preparation processes, and the design of multifunctional synergistic coatings. The aim is to provide theoretical guidance and practical references for the development of high-performance aqueous zinc-ion batteries.

Keywords

Aqueous Zinc-Ion Batteries; Zinc Anode; Molybdenum Disulfide (MoS_2); Interface Engineering; Dendrite Inhibition; Artificial Coating.

1. Introduction:

With the growing global demand for renewable energy sources (such as solar and wind power) and the increasingly prominent... In light of the environmental issues associated with traditional fossil fuels, the development of efficient, safe, and low-cost large-scale energy storage technologies has become an urgent need in the energy sector. Although lithium-ion batteries dominate the fields of portable electronic devices and electric vehicles, their high cost, limited lithium resources, and safety concerns stemming from organic electrolytes restrict their further application in areas like grid-scale energy storage[1].

Against this backdrop, aqueous zinc-ion batteries (AZIBs) have re-entered the research spotlight due to their unique advantages: zinc metal anodes possess a high theoretical capacity (820 mAh g^{-1} or 5855 mAh cm^{-3}), a low redox potential (-0.76 V vs. SHE), excellent compatibility with aqueous electrolytes, inherent safety, and abundant reserves on Earth[2]. These characteristics endow AZIBs with broad application prospects in large-scale energy storage, flexible electronics, and wearable devices.

However, the commercialization of AZIBs is severely hindered by inherent problems associated with the zinc anode itself. In aqueous electrolytes, the zinc anode faces two core challenges: First, uncontrolled zinc dendrite growth. During repeated plating/stripping processes, due to non-uniform electric field distribution and Zn^{2+} concentration gradients, Zn^{2+} tends to preferentially deposit on protrusions on the electrode surface, forming dendritic or mossy structures. These dendrites can pierce the separator, leaFirst, internal short-circuits and battery failure can be caused by dendrite growth[3]. Second, there are severe parasitic side reactions. The electrochemical stability window of water is narrow (approximately 1.23 V), making it prone to the hydrogen evolution reaction (HER) near the zinc deposition potential. This not only reduces Coulombic efficiency and consumes the electrolyte but also generates hydrogen gas, increasing the internal battery pressure.

Concurrently, HER causes a local pH increase at the electrode interface, inducing the formation and accumulation of insulating by-products like zinc hydroxide sulfate (e.g., $Zn_4SO_4(OH)_6{\cdot}xH_2O$). These by-products cover the electrode surface, hindering ion transport and accelerating battery capacity decay. Additionally, the zinc anode itself is subject to spontaneous corrosion in aqueous electrolytes.

To address these issues, researchers have developed various strategies, including electrolyte optimization (e.g., high-concentration salts, additives), electrode structure design (e.g., 3D frameworks), and interface engineering. Among these, interface engineering-specifically, constructing an artificial protective layer on the zinc anode surface-is considered one of the most direct and effective methods.

An ideal protective layer should possess the following characteristics: good electronic insulation and ionic conductivity to block electrons while allowing rapid Zn^{2+} transport; high mechanical strength to physically suppress dendrite penetration; excellent chemical and electro-Chemical stability, preventing side reactions with the electrolyte; and zincophilicity to guide uniform Zn^{2+} nucleation and deposition.

In recent years, two-dimensional (2D) nanomaterials have shown great potential for constructing zinc anode protective layers due to their unique layered structures and tunable physicochemical properties. Materials such as graphene, MXenes, hexagonal boron nitride (h-BN), and molybdenum disulfide (MoS_2) have been extensively studied. Among them, molybdenum disulfide (MoS_2), a typical transition metal dichalcogenide, has garnered particular attention[3].

MoS_2 possesses a layered structure similar to graphite, with layers held together by weak van der Waals forces, creating 2D ion diffusion channels that facilitate rapid Zn^{2+} transport[4]. Its high mechanical strength can physically impede dendrite growth. Furthermore, MoS_2 exhibits good chemical stability and zincophilic properties, which can induce uniform Zn^{2+} nucleation and reduce the nucleation overpotential. More importantly, by tuning its crystal phase (e.g., semiconducting 2H phase and metallic 1T′ phase), its electronic conductivity and electrochemical performance can be further optimized.

This paper aims to systematically review the research progress of MoS_2 coatings in stabilizing zinc anodes and enhancing the performance of aqueous zinc-ion batteries. It will first delve into the mechanisms of action of MoS_2 coatings, then introduce their mainstream preparation methods, summarize their electrElectrochemical performance in symmetric and full cells, and finally discuss current challenges and future development directions.

2. Multiple Protection Mechanisms of MoS_2 Coating

2.1. Analysis of Zinc Anode Failure Mechanism

The dense and uniform MoS_2 layer covering the surface of the zinc anode mainly serves as a physical barrier. This interfacial layer effectively isolates free water molecules, dissolved oxygen, and corrosive anions in the electrolyte, greatly reducing their direct contact with the underlying zinc metal. As a result, it significantly inhibits the hydrogen evolution reaction and chemical corrosion. By blocking the interfacial contact between water and zinc, this protective layer curbs the generation of insulating by-products such as basic zinc sulfate at the source, helping to maintain a clean electrode interface and ensure unobstructed ion transport channels. Research shows that, compared with bare zinc anodes, the surface of zinc anodes protected by the binder-free MoS_2 layer has significantly reduced corrosion by-products, confirming the effectiveness of this physical barrier in stabilizing the electrode interface[5].

2.2. Multiple Protection Mechanisms of MoS_2 Coating

2.2.1. Physical Barrier Effect Suppresses Side Reactions

The dense and uniform MoS_2 layer covering the surface of the zinc anode primarily acts as a physical barrier. It effectively reduces the direct contact between free water molecules, dissolved oxygen, and corrosive anions in the electrolyte. It blocks the contact between the electrolyte and the underlying zinc metal, thereby significantly inhibiting the occurrence of hydrogen evolution reactions and chemical corrosion. By blocking the contact between water and zinc, it fundamentally curbs the formation of insulating by-products such as zinc hydroxide sulfate, maintaining a clean electrode interface and unobstructed ion transport channels. Studies have shown that the surface of zinc anodes protected by binder-free MoS_2 exhibits significantly fewer corrosion by-products compared to bare zinc anodes.

2.2.2. Regulating Zn^{2+} Ion Flow and Uniform Nucleation

The MoS_2 coating itself possesses a certain degree of conductivity (especially the metallic 1T′ phase MoS_2), which helps to make the electric field distribution on the electrode surface more uniform, eliminating local "hot spots" and preventing the excessive accumulation of Zn^{2+} in specific areas. MoS_2 exhibits zincophilic properties, and its surface or edge sites can serve as preferential nucleation centers for Zn^{2+}. These uniformly distributed nucleation sites lower the nucleation overpotential of zinc, guiding Zn^{2+} to nucleate simultaneously and uniformly, rather than preferentially nucleating at a few tips. Research from Northwest Normal University confirms that MoS_2-protected zinc anodes exhibit a lower nucleation overpotential. The two-dimensional interlayer channels of MoS_2 can confine and guide the diffusion pathway of Zn^{2+}, making its flux more uniformly distributed across the electrode surface.

2.2.3. Promote Zn^{2+} Transport Kinetics

Furthermore, the use of the metallic 1T′ phase MoS_2 not only facilitates ion transport but also simultaneously improves the electron transfer process at the interface. This significantly reduces interfacial impedance and voltage hysteresis, ultimately enhancing the rate performance of the battery. Building on this, a PEPM coating constructed by compositing MoS_2 with the highly conductive polymer PEDOT:PSS has successfully established rapid ion/electron transport channels. Experimental results demonstrate that symmetric batteries employing this coating can cycle stably for up to 2000 hours at an extremely low overpotential (<50 mV), fully

ISSN: 2710-0723

showcasing the significant potential of binder-free MoS_2-based interfacial layers in optimizing the kinetics and stability of zinc anodes.

2.2.4. Guided Crystal Epitaxial Growth (Frontier Mechanisms)

The latest research has revealed a deeper role of MoS_2 coatings-inducing the epitaxial electrodeposition of zinc. Highly oriented single-crystal MoS_2 thin films can serve as a substrate, utilizing the lattice matching relationship between its crystal lattice and that of zinc to induce zinc atoms to grow epitaxially along a specific crystallographic orientation (e.g., along the Zn(002) plane). This growth mode results in a highly dense, flat, and dendrite-free zinc deposition layer, fundamentally eliminating the thermodynamic and kinetic conditions for dendrite formation from a crystallographic perspective (Fig. 1).

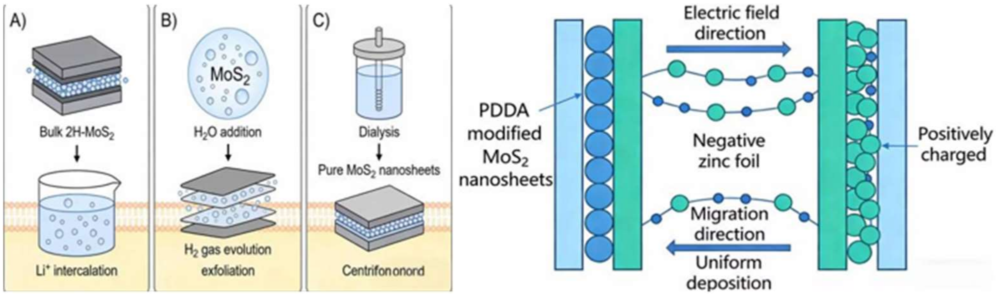

Fig. 1 Schematic illustration of MoS_2 nanosheet preparation and zinc anode interface regulation. (A)Li^+ intercalation of bulk 2H-MoS_2. (B) Aqueous-phase exfoliation of Li^+-intercalated MoS_2 via H_2 gas evolution. (C) Purification of MoS_2 nanosheets by dialysis and centrifugation. (D) PDDA-modified MoS_2 nanosheets guiding uniform Zn^{2+} deposition to suppress zinc dendrite growth. Positively charged PDDA-modified MoS_2 nanosheets regulate Zn^{2+} migration via electrostatic attraction under an electric field, enabling uniform zinc deposition and enhanced cycling stability of zinc-ion batteries.

3. Preparation Methods and Structural Regulation of MoS_2 Coatings

Constructing a high-quality, controllable MoS_2 coating on the surface of the zinc anode is a prerequisite for realizing its protective function. Currently, researchers have developed various preparation strategies, mainly including binder-free electrochemical deposition and scalable solution spraying methods.

3.1. Binder-free Electrochemical Deposition Method

Electrochemical deposition is an effective strategy for directly depositing functional materials onto a conductive substrate. This method eliminates the need for insulating polymer binders, resulting in a coating that is tightly bonded to the substrate. This not only lowers the interfacial resistance but also fully exposes the active sites. Taking the work of the research team at Northwest Normal University as an example, they systematically reported a complete process for constructing a MoS_2@Zn composite anode via electrochemical deposition.

The preparation process primarily consists of three key steps. First, the commercial MoS_2 powder requires exfoliation to obtain few-layer or monolayer nanosheets, often achieved through a lithium-ion intercalation method: the MoS_2 powder is immersed in a solution containing n-butyllithium, allowing Li^+ to insert between the MoS_2 layers; subsequently, the violent reaction of the lithiated product with water generates hydrogen gas, which exfoliates the bulk MoS_2 into nanosheets. The exfoliated suspension must then undergo further. Further centrifugation and dialysis are carried out to remove residual lithium salts and impurities. Second, to enhance the efficiency of MoS_2 nanosheets migrating towards the anode and depositing uniformly under the influence of an electric field, a small amount of cationic polymer, such as polydiallyldimethylammonium chloride (PDDA), is typically introduced into the anodic suspension prior to electrodeposition. The adsorption of PDDA renders the surface of the originally negatively charged MoS_2 nanosheets positively charged. This not only reduces electrostatic repulsion between the nanosheets but also facilitates their uniform deposition on the surface of the cathode (zinc foil). Finally, using the pre-treated zinc foil as the working electrode, electrochemical deposition is carried out in the suspension containing PDDA-MoS_2. The deposition process can employ cyclic voltammetry (CV) or a potentiostatic mode, allowing for precise control over the thickness and morphology of the MoS_2 coating by adjusting parameters such as deposition potential, number of cycles, or deposition time. Upon completion of the deposition, a MoS_2@Zn composite anode with a uniformly coated MoS_2 layer on its surface is obtained.

3.2. Scalable Solution Spray Coating Method

In view of this, to realize the practical application and large-scale preparation of MoS_2 protective layers, researchers are also devoting efforts to developing scalable technologies that feature simple processes, controllable costs, and industrial coCompatibility. A research team at Chungbuk National University in South Korea reported a scalable spray coating process based on a solution method for constructing MoS_2@Zn composite anodes. The implementation of this method typically involves the following steps: First, prepare a high-quality dispersion liquid of two-dimensional MoS_2 nanosheets. Subsequently, using zinc foil as the substrate, the dispersion is uniformly sprayed onto the surface of the heated substrate using equipment such as a spray gun. During this process, the thickness and uniformity of the MoS_2 protective layer can be effectively controlled by adjusting the concentration of the sprayed solution, the number of spraying cycles, and the substrate temperature.

Compared to other film-forming methods, the spray coating technique demonstrates significant potential for large-scale application due to its advantages such as simple operation, low cost, and ease of achieving large-area preparation. Research indicates that the MoS_2@Zn anode obtained through this strategy can also effectively inhibit dendrite growth and interfacial side reactions, maintaining stable long-cycle performance even under harsh electrochemical testing conditions. This further validates the multi-route feasibility and practical application prospects of the MoS_2 interface modification strategy.

3.3. The Effect of Coating Parameters on Performance

Research on the structure-property relationship of MoS_2 protective layers indicates that their intIntrinsic properties-particularly thickness, uniformity, and crystal phase-have a decisive impact on the ultimate electrochemical performance of the zinc anode. Deeply understanding and precisely controlling these parameters is central to optimizing the interfacial functionality of MoS_2@Zn composite anodes.Firstly, the coating thickness directly affects the balance between protective effectiveness and ion transport kinetics and must be precisely optimized. If the coating is too thin, it struggles to completely cover the zinc substrate surface; residual defect sites allow electrolyte penetration, weakening its physical barrier function and limiting the protective effect. Conversely, if the coating is too thick, it significantly prolongs the diffusion path for Zn^{2+}, increases interfacial transport resistance, and consequently leads to a higher polarization voltage and diminished rate performance. Therefore, an ideal protective layer should be as thin as possible while ensuring adequate substrate coverage, seeking an optimal balance between suppressing side reactions and facilitating ion transport.

Secondly, coating uniformity is a critical factor determining interfacial stability. A large-area, defect-free coating with uniform thickness ensures homogeneity in electric field distribution and ion flux, preventing the formation of local "hot spots." Conversely, a non-uniform coating can cause distortion of the interfacial electric field and local variations in ion flux, which may... Teardrops induce preferential nucleation and growth of zinc dendrites. Whether it is the charge modification of nanosheets in electrochemical deposition or the fine control of process parameters in spray coating, the core objective is to achieve highly uniform coating coverage on a macroscopic scale.

Furthermore, crystal phase engineering offers a new dimension for enhancing the intrinsic conductivity of MoS_2 protective layers. Common crystal phases of MoS_2 include the stable semiconducting phase (2H phase) and the metastable metallic phase (1T′ phase). Among these, the 1T′ phase MoS_2 has attracted significant attention due to its higher intrinsic electronic conductivity. As an interfacial layer, 1T′ phase MoS_2 can more effectively promote interfacial charge transfer and reduce interfacial impedance, thereby enhancing the battery's rate performance. Existing research has precisely constructed atomically thin 1T′ MoS_2 at the cathode-electrolyte interface, significantly improving the electronic transport and structural stability of cathode materials. This strategy suggests that applying the highly conductive 1T′ phase MoS_2 to the protection of zinc anodes in the future holds promise for further breaking through interfacial kinetic bottlenecks and achieving superior overall electrochemical performance.

4. Electrochemical Performance of MoS_2@Zn Anode

The stabilizing effect of the MoS_2 coating on the zinc anode ultimately manifests in significantly enhanced electrochemical performance.e, including longer cycle life and lower voltage hysteresis in symmetric cells, as well as higher capacity retention and rate capability in full cells.

4.1. Symmetric Battery Performance

Long-term cycling stability: Research from Northwest Normal University showed that MoS_2@Zn symmetric cells prepared by electrochemical deposition achieved stable long-term cycling for over 1200 hours at a current density of 0.5 mA cm^{-2} and an areal capacity of 0.1 mAh cm^{-2}, while bare zinc symmetric cells short-circuited within a short time. MoS_2@Zn anodes prepared by South Korean scholars via spray coating also achieved stable cycling for over 1000 hours under more stringent conditions of 1 mA cm^{-2} and 1 mAh cm^{-2}. Low voltage hysteresis: voltage hysteresis (overpotential) It is an important indicator reflecting the kinetics of interfacial reactions. The MoS_2 coating, by lowering the nucleation barrier and accelerating ion transport, significantly reduces voltage hysteresis during cycling. For example, researchers at the University of Tokyo constructed a PEDOT:PSS/MoS_2 composite coating that enabled symmetric cells to maintain an overpotential consistently below 50 mV for over 2000 hours of cycling, demonstrating an extremely stable interface and fast kinetic characteristics.

4.2. Full Battery Performance

Assembling full cells by pairing the MoS_2@Zn composite anode with different cathode materials is a critical validation for assessing its practical application potential. Among these, the configuration paired with the α-MnO_2 cathode has been the most extensively studied. Experimental results demonstrate that the MoS_2@Zn||α-MnO_2 full cell exhibits excellent electrochemical performance: at a 1C rate, it delivers an initial reversible capacity of 225.8 mAh g^{-1} and maintains a high capacity retention rate after 300 charge-discharge cycles. Morphological characterization of the zinc anode after cycling revealed no significant dendrite growth on the surface, confirming the effectiveness of the MoS_2 protective layer during full cell operation. Notably, some studies have reported that this system achieves a capacity retention rate of nearly 100% after 100 cycles, further highlighting its exceptional cycling stability.

In addition to α-MnO_2, the MoS_2@Zn anode has also proven suitable for other cathode systems. For instance, when paired with a zinc vanadium oxide cathode, the full cell similarly demonstrates more stable cycling performance compared to the control group using a bare zinc anode. These findings indicate that the MoS_2 interface modification strategy exhibits good compatibility with a variety of cathode material systems, providing strong support for the practical development of high-performance aqueous zinc-based batteries.

4.3. Expanded Applications of MoS_2 in Cathode Protection

Interestingly, MoS_2 can not only protect the zinc anode but can also serve as a protective layer for the cathode through rational structural design. A research team at Northwestern Polytechnical University coated atomic-layer 1T′ MoS_2 onto the surface of an amorphous $Zn_{0.25}V_2O_5$ (aZVO) cathode, constructing an aZVO@MoS_2 core-shell heterostructure. This design utilizes the 1T′ MoS_2 coating as an artificial cathode-electrolyte interphase, effectively inhibiting vanadium dissolution and improving electron transport, thereby enabling the cathode to exhibit excellent cycling stability even at an ultra-low current density (0.05 A g^{-1}). This indicates that MoS_2, as amultifunctional interface material, has broader applicability in AZIBs.

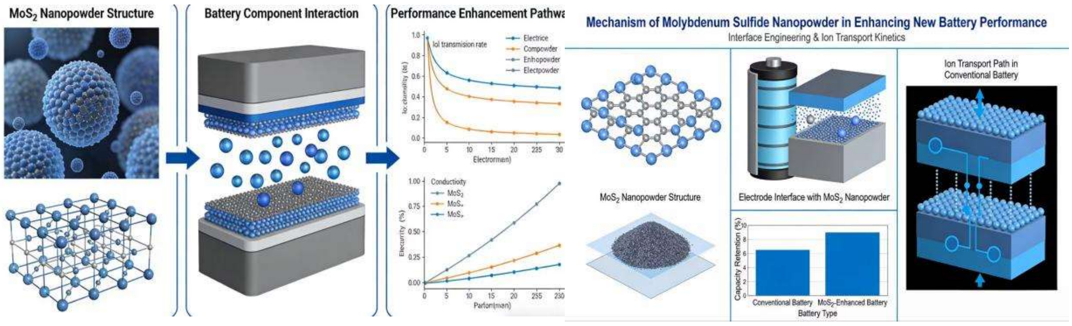

Fig. 2 Mechanism of MoS_2 nanopowder for enhanced new battery performance

Fig.2 shows schematic of MoS_2 nanopowder structure, electrode interaction, performance pathways, and interface engineering, demonstrating improved ion transport, conductivity, and capacity retention in MoS_2-modified batteries.

5. Conclusion and Outlook

As a typical two-dimensional layered material, MoS_2 has demonstrated significant potential in stabilizing zinc anodes for aqueous zinc-ion batteries through interface engineering strategies. Its multiple protection mechanisms-including physically isolating side reactions, regulating Zn^{2+} ion flux to achieve uniform deposition/nucleation, promoting interfacial reaction kinetics, and even guiding epitaxial crystal growth-work synergistically to effectively inhibit dendrite growth and parasitic reactions, significantly enhancing the cycle life and Coulombic efficiency of the batteries. Currently, researchers have developed various methods such as electrochemical deposition and solution spraying to achieve controllable preparation of MoS_2 coatings, and have verified their excellent electrochemical performance in both symmetric cells and full cells, for instance, achieving stable cycling for over 1000 hours and high capacity retention.

Despite significant progress, the field still faces a series of challenges, and future research should focus on the following aspects:Performance validation under high depth of discharge (DOD): Currently, most of the excellent performance is achieved under conditions of low depth of discharge (DOD < 1%) and low areal capacity. Future research must shift focus to evaluating the effectiveness of MoS_2 coatings under practical conditions with high depth of discharge (e.g., 50%, 80%) and high areal capacity (>10 mAh cm^{-2}) to prove their application value in high-energy-density batteries.

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Optimization of Large-Scale, Low-Cost Preparation Processes:Although the spraying method shows certain scalability, to achieve truly industrial application, it is still necessary to develop simpler, more efficient, lower-cost, and more reproducible coating techniques, such as roll-to-roll coating, and to ensure the uniformity and defect-free nature of large-area coatings.

Design of Multifunctional Synergistic Coatings: A single MoS_2 coating may not meet all requirements. Future exploration can focus on composite coatings of MoS_2 with highly conductive polymers (e.g., PEDOT:PSS), carbon materials (graphene), or inorganic nanoparticles with specific functions, achieving a "1+1>2" effect through synergy. For example, the PEDOT:PSS/MoS_2 composite coating has already demonstrated excellent performance.

Crystal Phase and Defect Engineering: In-depth study of the influence of different crystal phases (2H vs. 1T′) and the introduction of defects (e.g., sulfur vacancies) on the zincophilicity, ionic conductivity, and catalytic activity of MoS_2 coatings. Further optimizing their protective performance through precise atomic-level regulation. Drawing on successful experiences in photo-responsive batteries and cathode protection may reveal new directions for optimization.

In-depth Characterization of Failure Mechanisms: Combining advanced in-situ/ex-situ characterization techniques (e.g., in-situ optical microscopy, in-situ Raman spectroscopy, synchrotron radiation X-ray imaging) to deeply investigate the structural evolution, interfacial chemical changes, and eventual failure mechanisms of MoS_2 coatings during long-term cycling. This will provide theoretical guidance for designing more stable next-generation protective layers.

In summary, MoS_2-based coatings offer a promising pathway for developing high-performance, long-life aqueous zinc-ion batteries. Through continued fundamental research and process innovation, it is expected to overcome existing challenges and promote the practical application of this advanced energy storage technology.

Acknowledgments

This work was supported by the College Students' Innovative Entrepreneurial Training Plan Program.

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