

Research progress and perspectives on interface engineering of zinc metal anodes in aqueous zinc-ion batteries

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Aqueous zinc-ion batteries (AZIBs) have garnered increasing interest due to their high safety, low cost, environmental friendliness, and the natural abundance of zinc metal. However, practical operation is severely hindered by dendrite growth, hydrogen evolution, and interfacial side reactions at the zinc anode, which lead to poor reversibility and limited cycling stability. Interface engineering has emerged as an effective strategy to directly regulate interfacial reactions and optimize Zn plating/stripping behavior, thereby enhancing anode stability. This review systematically summarizes recent progress in zinc anode interface engineering, with emphasis on artificial solid electrolyte interphases (SEI), electrolyte/interfacial regulation, and separator modification. The design principles and underlying mechanisms of these strategies in suppressing dendrites, mitigating hydrogen evolution, and reducing interfacial side reactions are critically discussed. Finally, current challenges and future perspectives for interfacial design toward the practical deployment of AZIBs are outlined.

interface engineering; interfacial stability; aqueous zinc-ion batteries; zinc metal anode

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1. Introduction

In recent years, the continuous depletion of nonrenewable energy resources and the escalating environmental and ecological challenges have accelerated the global transition toward a low-carbon energy system dominated by renewable sources. Electrochemical energy storage systems, characterized by their operational convenience and high energy conversion efficiency, have emerged as one of the primary energy storage technologies in contemporary energy infrastructures^[1]. In this context, the development of next-generation energy storage technologies featuring high safety, low cost, and scalability has become a critical technological pathway for achieving the “dual-carbon” strategic goals. Among existing energy storage systems, lithium-ion batteries, as the most mature and extensively commercialized technology, currently dominate the markets of portable electronics and electric transportation^[2-4]. However, their

large-scale deployment in grid-level energy storage is increasingly hindered by high material costs, intrinsic safety concerns, and the uneven global distribution of lithium resources, which collectively limit their further expansion^[5-7]. Against this background, aqueous zinc-ion batteries (AZIBs) have emerged as a promising alternative and have attracted considerable attention from both academia and industry. Owing to the use of aqueous electrolytes, AZIBs inherently eliminate the flammability and explosion risks associated with organic electrolyte systems. Moreover, as summarized in Table 1, zinc metal anodes exhibit a high theoretical specific capacity (C_t) of $820 \text{ mAh}\cdot\text{g}^{-1}$, a relatively low redox potential ($\varphi^\ominus$) of -0.76 V (vs. SHE), as well as abundant reserves, low cost, and environmental benignity. These intrinsic advantages render zinc-based batteries one of the most competitive candidates for next-

Table 1 Comparison of key parameters between zinc and lithium

Element	Zinc	Lithium	References
$\varphi^\ominus/\text{V}$ (vs. SHE)	−0.76	−3.04	[18]
Crustal abundance/($\text{mg}\cdot\text{kg}^{-1}$)	52–83	22–32	[19]
$C_t/(\text{mAh}\cdot\text{g}^{-1})$	820	3860	[19]

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generation rechargeable energy storage systems^[8-11].

Despite these merits, the electrochemical performance of AZIBs under practical operating conditions remains far from ideal. The primary bottlenecks originate from the insufficient thermodynamic stability and poor electrochemical reversibility at the interface between the zinc metal anode and the aqueous electrolyte. Specifically, during repeated plating/stripping processes, zinc anodes are susceptible to a series of detrimental phenomena, including dendritic growth, hydrogen evolution reaction (HER), corrosion, and interfacial instability. These issues collectively lead to low Coulombic efficiency, severe polarization, and rapid capacity decay, ultimately resulting in a shortened cycling lifespan^[12-14]. Notably, these parasitic reactions are not independent but are intrinsically coupled and mutually reinforcing, thereby accelerating the degradation and failure of zinc metal anodes. To overcome these critical challenges, interface-engineering-based strategies have been extensively explored. By directly targeting the electrode/electrolyte interface—the central “electrochemical battlefield”—interface engineering is widely recognized as one of the most effective approaches to fundamentally regulate zinc deposition behavior and suppress undesirable side reactions^[15-16]. Through the construction of artificial solid electrolyte interphases (SEI), the introduction of functional protective layers, or the modulation of interfacial ion transport and local electrochemical microenvironments, zinc deposition can be homogenized, while dendrite growth and HER can be effectively suppressed. Consequently, the overall electrochemical performance, stability, and durability of AZIBs can be significantly improved^[17].

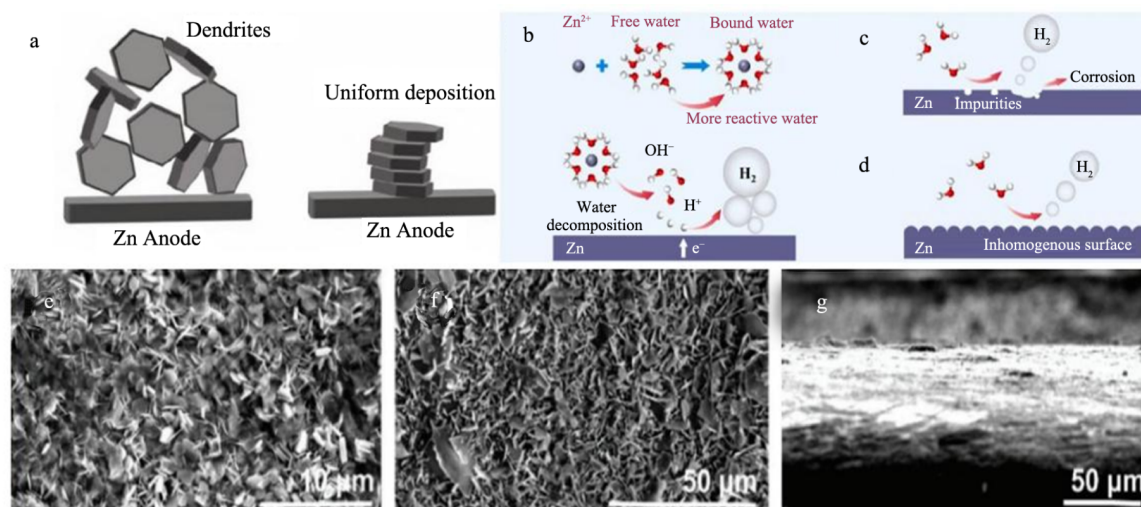
Based on these considerations, this review focuses on interface engineering strategies for zinc metal anodes and systematically summarizes recent representative advances in this rapidly evolving field. First, the fundamental failure mechanisms of zinc anodes in AZIBs are briefly outlined.

Subsequently, various interface engineering strategies are discussed in detail, with particular emphasis on their design principles, underlying mechanisms, and recent progress, supported by representative case studies. Finally, a comparative assessment of different interface engineering approaches is provided, followed by an outlook on the remaining scientific challenges and future research directions, aiming to offer theoretical insights and practical guidance for the development of high-performance, long-lifespan aqueous zinc-ion battery systems.

2. Overview of the major challenges faced by zinc anodes

2.1 Dendrite growth

Dendrite growth represents a critical challenge commonly encountered in most metal-ion battery systems^[20]. Under practical conditions, it is difficult for zinc metal anodes to maintain an ideally smooth surface. Instead, various microscopic inhomogeneities, such as surface roughness, scratches, and structural defects, are inevitably present. These local concavities or imperfections tend to induce nonuniform current distribution, leading to localized current density amplification. As a result, Zn^{2+} preferentially undergo reduction at these sites during deposition, thereby initiating heterogeneous zinc plating behavior^[21]. As illustrated in Figure 1a, during the early stage of electrodeposition, minor surface irregularities on the electrode are progressively amplified under the influence of electric field concentration effects. The protruding tips exhibit significantly enhanced local electric field intensity, which promotes the preferential migration and accumulation of Zn^{2+} , consequently resulting in higher deposition rates at these locations. With continued deposition, such nonuniform growth is continuously exacerbated and ultimately evolves



Schematic illustration of uniform and non-uniform zinc deposition (a), mechanistic illustration of the HER on metallic zinc in aqueous electrolytes (b–d), and FE-SEM images of the Zn metal electrode after charge-discharge in ZnSO_4 (e–g)

Figure 1 Schematic illustration of major interfacial challenges at zinc metal anodes in AZIBs

into elongated, sharp, dendritic zinc structures^[22]. The persistent growth of zinc dendrites not only severely deteriorates the electrochemical reversibility of the zinc anode but may also penetrate the separator under extreme conditions, triggering internal short circuits and posing serious safety hazards. Consequently, dendrite formation is recognized as one of the primary factors limiting the long-term stability and reliable operation of AZIBs^[23].

2.2 Parasitic reactions (hydrogen evolution and corrosion)

In aqueous electrolyte systems, owing to the fact that the standard electrode potential of metallic zinc (-0.76 V vs. SHE) is significantly more negative than the hydrogen evolution potential of water, HER is thermodynamically inevitable and constitutes one of the primary competing parasitic reactions during zinc deposition^[9]. The HER mechanism is schematically illustrated in Figures 1b–1d^[13]. As shown in Figure 1b, Zn^{2+} in the electrolyte strongly interact with water molecules to form stable solvation structures, which, upon migration to the zinc metal surface, readily induce the reduction of coordinated water molecules. As depicted in Figure 1c, free water molecules can directly react with zinc metal containing impurities or surface defects, thereby facilitating the occurrence of HER. Furthermore, as illustrated in Figure 1d, the microscopic surface roughness or structural inhomogeneity of zinc electrodes can intensify local electric fields and increase the density of electrochemically active sites, thus further accelerating water reduction and exacerbating hydrogen evolution. The occurrence of HER not only continuously consumes electrons involved in the electrochemical reactions, leading to a pronounced decrease in Coulombic efficiency, but also induces a localized increase in pH at the electrode/electrolyte interface, thereby disrupting the chemical stability of the system. With the sustained generation of hydrogen gas, its evolution and accumulation at the electrode surface may result in internal gas buildup, causing cell swelling and even mechanical failure^[22–23]. Moreover, hydrogen is a highly flammable gas, and its continuous generation and accumulation in sealed battery systems can substantially increase the risk of explosion, posing serious safety concerns for the operation of AZIBs^[24].

In addition, in commonly used zinc sulfate-based aqueous electrolytes, the HER continuously produces OH^- ions, leading to an elevated local pH at the zinc anode interface. Under alkaline conditions, Zn^{2+} readily reacts with OH^- to form $\text{Zn}(\text{OH})_2$, which can further transform into ZnO and other by-products. These electrochemically inert species gradually accumulate on the anode surface, forming passivation layers that impede the reversible plating/stripping of zinc. Meanwhile, OH^- ions can also cooperatively react with Zn^{2+} and SO_4^{2-} to generate basic zinc sulfate and related compounds. The progressive accumulation of these by-products markedly increases interfacial resistance, under-

mines the structural integrity of the zinc anode, and accelerates battery performance degradation. Liu et al.^[25] employed transmission electron microscopy (TEM) to reveal that zinc dendritic structures are not composed solely of metallic zinc but also contain various by-products generated from interfacial parasitic reactions, indicating that continuous side reactions can further promote dendrite formation and growth. As shown in Figures 1e–1g, these parasitic products often include electrically insulating species such as $\text{Zn}_4\text{SO}_4(\text{OH})_6 \cdot \text{H}_2\text{O}$ ^[26]. The deposition of such insulating by-products on the electrode surface severely hinders Zn^{2+} transport and interfacial charge-transfer processes, leading to aggravated polarization and ultimately suppressing normal electrochemical reactions or even causing battery failure^[27].

2.3 Impact of interfacial instability on battery performance

During practical operation of AZIBs, dendrite growth, hydrogen evolution, and various interfacial side reactions do not occur independently; rather, they constitute a complex, mutually reinforcing, and co-evolving process^[28]. As illustrated in Figure 2, there exists a significant interplay among these failure phenomena. Specifically, the continuous growth of zinc dendrites markedly increases the specific surface area of the metallic zinc anode, thereby exposing additional electrochemically active sites, which further accelerates hydrogen evolution and other interfacial parasitic reactions^[18]. Simultaneously, the gaseous by-products and insoluble compounds generated from side reactions can in turn disrupt the structural integrity of the interface, exacerbating the heterogeneity of zinc deposition. This series of coupled effects ultimately leads to a pronounced decrease in Coulombic efficiency, aggravated electrode polarization, and progressively rising interfacial impedance, thereby accelerating capacity fading and significantly shortening the cycling lifespan of the battery^[9,29]. Therefore, fundamentally regulating and stabilizing the zinc anode/electrolyte interface to suppress the co-evolution of dendrite growth and parasitic reactions is pivotal for enhancing the overall

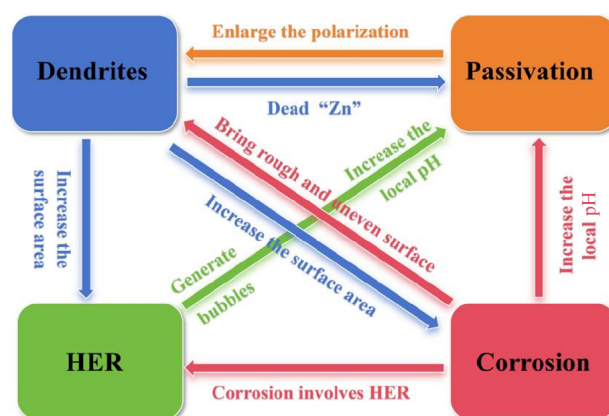


Figure 2 Interconnection of failure phenomena in zinc metal anodes

electrochemical performance and long-term stability of AZIBs.

3. Interface engineering strategies for zinc metal anodes

The failure of zinc metal anodes fundamentally arises from the highly unstable chemical and physical environment at the electrode/electrolyte interface. This interfacial instability directly triggers dendrite growth, hydrogen evolution, and various parasitic side reactions. Consequently, the stability of the zinc anode interface is widely recognized as a critical factor limiting the electrochemical performance and cycling lifespan of AZIBs. In recent years, researchers have proposed and developed a range of interface engineering strategies targeting the zinc anode, approaching the problem from multiple scales and dimensions. The objective of these strategies is to construct a stable and controllable electrode-electrolyte interface, thereby promoting uniform zinc plating/stripping and effectively suppressing interfacial side reactions. As illustrated in Figure 3, current interface engineering approaches for zinc anodes can be broadly categorized into three main types: (i) construction of artificial SEI, (ii) electrolyte optimization and interfacial regulation, and (iii) separator modification and interface-induced control strategies. These strategies collectively aim to modulate the local chemical environment, homogenize ion flux, and enhance interfacial structural integrity, providing a solid foundation for the rational design of high-performance, durable AZIBs.

3.1 Construction of artificial SEI

The concept of constructing an artificial SEI is inspired by the successful experience in lithium-ion batteries. By introducing a physically and chemically stable protective layer on the zinc anode surface, direct contact between the electrode and water in the electrolyte can be effectively pre-

vented, thereby mitigating the occurrence of HER^[30-31]. Artificial SEI can be broadly categorized into three types based on the composition of the interfacial layer: inorganic coatings, organic coatings, and composite SEI layers.

3.1.1 Inorganic coatings

Inorganic coatings are typically composed of metal oxides or metal alloys, such as Sc_2O_3 ^[32], Na_2TiO_3 ^[33], and Zn-Al ^[34]. These coatings are generally constructed directly on the zinc anode surface to form a stable interfacial layer, which facilitates uniform zinc deposition and effectively suppresses dendrite growth and interfacial side reactions. As shown in Figure 4a, certain porous inorganic coatings, such as TiO_2 , can selectively regulate ion transport, further inhibiting undesirable side reactions^[35]. In addition, Lv et al. employed atomic layer deposition to construct an ultrathin In_2O_3 inorganic interfacial layer on zinc anodes, achieving dendrite-free deposition and long cycle life in aqueous batteries, as illustrated in Figure 4b^[36]. In summary, inorganic coatings provide a stable and efficient deposition environment for zinc anodes by modulating interfacial chemistry and crystallographic growth behavior. They represent one of the key strategies for achieving long lifespan and high-performance AZIBs. Nevertheless, the rigidity of inorganic coatings may lead to cracking under long-term cycling, motivating the development of more mechanically adaptable alternatives.

3.1.2 Organic coatings

To address the mechanical rigidity and interfacial durability issues associated with inorganic coatings, organic coatings based on polymeric materials have emerged as an effective and flexible alternative. Organic coatings are primarily composed of polymers, such as polyamide^[37] and polyimide^[38]. Typical polymer layers include polyacrylic acid (PAA) and polyaspartic acid (PASP), whose molecular structures are rich in polar functional groups such as carboxyl ($-\text{COOH}$) and hydroxyl ($-\text{OH}$) groups. These functional groups can readily coordinate with metallic zinc or Zn^{2+} , allowing the polymer to firmly adsorb onto the metal surface. In addition, polymeric materials exhibit excellent mechanical flexibility, enabling them to accommodate the volumetric changes of the zinc anode during repeated charge-discharge cycles. This flexibility prevents cracking that is commonly observed in traditional rigid inorganic coatings, providing continuous and stable interfacial protection for the zinc anode^[39-40].

As shown in Figures 4c and 4d, in AZIBs, a PVB/Cu-Zn nanoparticle composite protective layer can be constructed on the zinc anode surface to form SEI, achieving multifaceted regulation of zinc deposition behavior^[41]. This protective layer exhibits pronounced hydrophobicity, effectively reducing the affinity of the zinc surface for water molecules, thereby suppressing hydrogen evolution and other interfacial side reactions. Consequently, compared with bare zinc, during repeated plating/stripping processes, zinc deposits uniformly along the substrate, forming dense

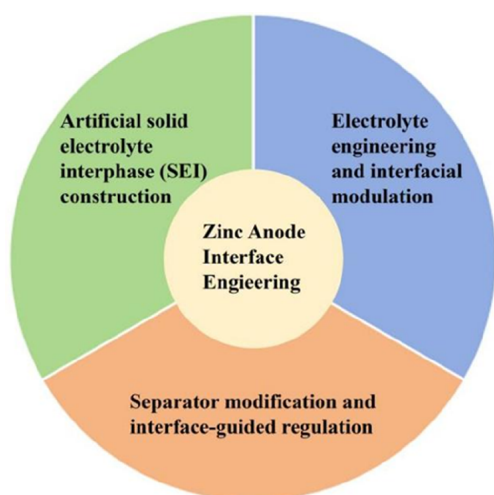
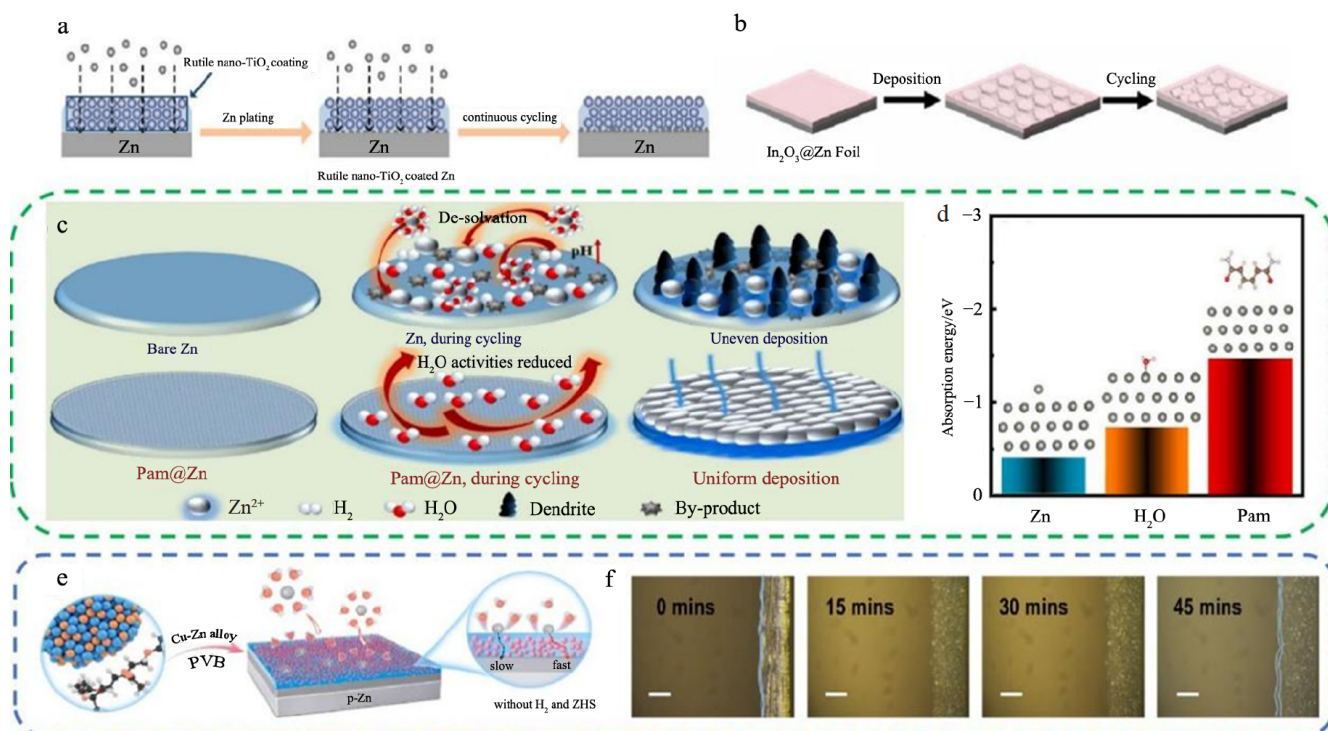


Figure 3 Major strategies for interface engineering of zinc metal anodes



Schematic illustrations of morphology evolution for TiO₂ coated Zn foils during Zn stripping/plating cycling (a), Zn plating on In₂O₃@Zn foil (b), the deposit Zn and Pam@Zn anodes (c) and absorption energies (d); the fabrication of inorganic-organic composite surface layer on Zn metal surface (e), and *in-situ* optical microscopy images of zinc metal deposition on p-Zn under a current density of 1 mA·cm⁻² (f)

Figure 4 Representative SEI for zinc metal anodes in AZIBs

layers that effectively inhibit dendrite growth and surface hydrogen evolution. These findings demonstrate that organic polymer-based interface engineering can synergistically regulate zinc anode behavior at both the molecular and macroscopic morphological levels, representing an effective strategy to enhance the cycling stability and safety of AZIBs. However, the durability, uniformity, and scalability of these polymer-nanoparticle SEI layers under realistic cycling conditions require further investigation. While organic coatings provide excellent flexibility and interfacial stability, combining them with inorganic components can further enhance mechanical robustness and ionic transport, paving the way for composite SEI designs.

3.1.3 Composite SEI

Building on the complementary advantages of organic and inorganic layers, composite SEI coatings have emerged as a promising strategy to simultaneously leverage the flexibility of polymers and the mechanical and ionic benefits of inorganic materials. Composite SEI coatings integrate organic and inorganic materials to leverage the advantages of both types. For instance, a composite layer composed of polyvinyl butyral (PVB) and Cu-Zn alloy nanoparticles combines the flexibility and functional group benefits of polymers with the mechanical robustness and ionic selectivity of inorganic components. As shown in Figure 4e, Zhang et al.^[42] constructed an organic-inorganic composite protective layer on the zinc anode surface, providing fast Zn²⁺ diffusion channels. This composite layer, functioning

as an artificial SEI, simultaneously offers surface hydrophobicity, uniform Zn²⁺ flux, and low nucleation energy. In addition, as shown in Figure 4f, during repeated zinc plating/stripping, it effectively suppresses dendrite growth and surface hydrogen evolution, thereby stabilizing zinc deposition and enhancing the electrochemical performance of AZIBs. Collectively, these studies indicate that interfacial coatings for Zn anodes should not be regarded as static protective layers, but as dynamically evolving interphases whose effectiveness depends on their ability to accommodate continuous Zn deposition, dissolution, and interfacial reconstruction. From this perspective, the rational design of SEI coatings should prioritize not only initial interfacial properties, but also their long-term adaptive behavior under practical electrochemical conditions. Future progress will therefore rely on application-oriented and mechanism-guided integration of organic and inorganic components, rather than incremental material substitution.

3.2 Electrolyte optimization and interfacial regulation

3.2.1 High-concentration electrolytes (water-in-salt)

Increasing the electrolyte concentration is considered an effective strategy for regulating interfacial reaction kinetics in AZIBs, primarily by reducing the activity of free water molecules. As the electrolyte concentration rises, water molecules increasingly participate in the solvation structures of Zn²⁺ and coordinating anions, resulting in a substantial reduction of free water content. This diminishes the kinetics

of water-involved HER and effectively suppresses hydrogen evolution at the zinc anode surface^[43].

Simultaneously, in high-concentration electrolytes, Zn^{2+} forms more stable coordination structures with anions, leading to a more uniform distribution of zinc ions in the electrolyte. This mitigates flux inhomogeneities at the interface and promotes uniform zinc deposition on the electrode, significantly reducing the likelihood of dendrite nucleation and growth^[44]. For example, Yang et al.^[45] introduced a unique bisphenylsulfonamide anion (BBI^-) that coordinates with Zn^{2+} , weakening the ion-dipole interactions between Zn^{2+} and H_2O and thereby suppressing water decomposition. The resulting Zn^{2+} - BBI^- solvation structure increases the nucleation overpotential for zinc, promoting uniform deposition and effectively inhibiting dendrite growth. Furthermore, the hydrogen-bonding network formed between BBI^- and H_2O can restrict water molecule diffusion, thereby reducing the dissolution of water-soluble cathode materials into the electrolyte. From the perspective of solvation structure modulation, this strategy simultaneously suppresses HER and dendrite growth at the anode while mitigating active material dissolution at the cathode, substantially broadening the electrochemical stability window of the electrolyte. Although high-concentration electrolytes effectively modulate Zn^{2+} solvation and suppress HER, additional strategies are often required to further fine-tune interfacial reactions and enhance zinc deposition uniformity under practical operating conditions.

3.2.2 Functional additives

Building on this principle, functional electrolyte additives have been introduced to selectively regulate Zn^{2+} solvation structures and interfacial kinetics, providing a complementary approach to high-concentration electrolytes for mitigating HER and dendrite formation. During HER, the water molecules consumed in hydrogen generation mainly originate from strongly coordinated water in the Zn^{2+} solvation shell, rather than from free water molecules in the electrolyte. Therefore, weakening the coordination between Zn^{2+} and water, thereby reducing the number of bound water molecules, is considered an effective strategy to suppress HER^[13]. Based on this principle, the introduction of functional electrolyte additives capable of regulating Zn^{2+} solvation structure and interfacial reactions has attracted extensive attention.

Recently, a variety of functional additives have been reported to further optimize zinc anode interfacial performance. Wang et al.^[46] proposed a zwitterionic additive (See Figure 5a), which forms a dynamically adsorbed layer on the zinc surface. This layer modulates the interfacial Zn^{2+} solvation structure and local electric field distribution while reducing the activity of bound water, resulting in highly reversible zinc deposition and long cycle life in symmetric $\text{Zn}||\text{Zn}$ cells. Shang et al.^[47] investigated ethylenediamine-tetraacetic acid zinc disodium salt (EDTA-ZnNa_2 , EZN) as an electrolyte additive, demonstrating that the chelating agent stabilizes the Zn^{2+} solvation structure (See Figure 5b) and forms a protective layer on the anode surface, effec-

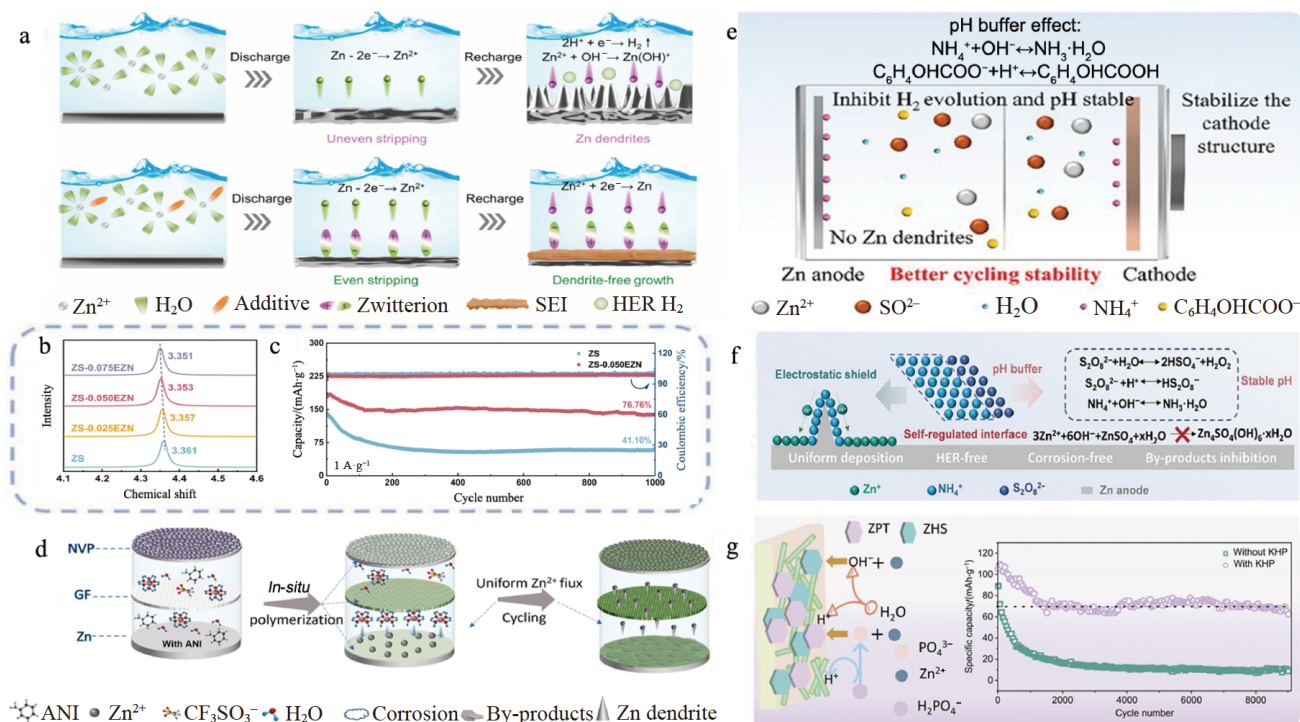


Illustration of dendritic Zn growth and side reactions in AZIBs and effect of dynamic adsorption mechanism on Zn stripping and plating (a), ¹H NMR spectra of different electrolytes in the EZN system (b) and corresponding cycling performance (c), schematic diagram of the multi-interface optimization strategy (d) and Zn anode surface deposition process in ZnSO_4 -AS electrolyte (e), the schematic diagram of zinc plating processes in the ZnSO_4 -APS electrolyte (f), and cycle performance in the KHP electrolyte at a high current density of $5 \text{ A} \cdot \text{g}^{-1}$ (g)

Figure 5 Electrolyte additive-driven interfacial engineering for stabilized zinc metal anodes

tively suppressing HER and dendrite growth, thereby enhancing Coulombic efficiency and cycling stability (See Figure 5c). Wang et al.^[48] applied aniline as an electrolyte additive, which *in-situ* polymerizes on the electrode and separator surfaces (See Figure 5d), optimizing multiple interfaces simultaneously and improving both cycle life and rate performance of AZIBs. Despite the effectiveness of functional additives in regulating Zn^{2+} solvation and interfacial reactions, the evolution of local electrolyte pH during cycling remains a critical factor governing HER kinetics and zinc anode stability.

3.2.3 pH buffering systems

Beyond solvation structure regulation, controlling the local pH environment at the electrode/electrolyte interface has emerged as another crucial strategy for suppressing HER and stabilizing zinc deposition. The occurrence of HER is strongly influenced by the pH of the electrolyte. While an elevated pH can effectively suppress HER, excessively high pH levels may induce parasitic reactions at the zinc anode. Therefore, precise regulation of electrolyte pH is critical for minimizing both HER and interfacial side reactions^[49].

To achieve fine pH control, Liu et al.^[50] introduced ammonium bicarbonate as a buffering agent. The $\text{NH}_4^+/\text{HCO}_3^-$ buffer system stabilizes the electrolyte environment by preferentially interacting with water molecules, thereby suppressing side reactions and enabling dendrite-free zinc anodes. Lv et al.^[51] proposed ammonium salicylate (AS) as a bidirectional pH buffering additive, in which amino and carboxyl functional groups not only modulate the local pH but also guide uniform Zn^{2+} deposition on the zinc anode (See Figure 5e), improving deposition morphology and cycling stability. Furthermore, the application of ammonium persulfate (APS) as a multifunctional pH buffering agent has been reported^[52-53]. APS self-regulates the interfacial environment to form a stable protective layer, thereby suppressing HER and dendrite growth (See Figure 5f), and achieving highly reversible zinc anodes with extended cycle life. Liu et al.^[54] also investigated a KH_2PO_4 based buffering system, which not only stabilizes the electrolyte pH but also mitigates issues arising from low proton concentration by introducing KH_2PO_4 into the electrolyte. As shown in Figure 5g, this approach improves zinc deposition behavior and overall electrochemical performance. Overall, pH buffering strategies provide a simple yet effective means of stabilizing the zinc/electrolyte interface, simultaneously suppressing hydrogen evolution and dendrite formation, and thereby enhancing the cycling stability and reversibility of AZIBs.

3.2.4 Electric double layer (EDL) engineering

In addition to conventional strategies such as high-concentration electrolytes and functional additives, direct modulation of the EDL at the Zn/electrolyte interface has emerged as a powerful approach to regulate Zn deposition and suppress parasitic reactions. By tailoring the local dis-

tribution of ions and solvent molecules near the Zn surface, EDL engineering can effectively reduce the activity of free water and alter the desolvation kinetics of Zn^{2+} . Tang et al.^[55] demonstrated that sodium difluoro(oxalato)borate (NaDFOB) anions act as Zn^{2+} trapping agents, preferentially depositing on the Zn(100) crystal facets to create a Zn^{2+} -rich, water-poor interfacial layer, thereby inhibiting hydrogen evolution and dendrite formation and enhancing the uniformity and kinetics of Zn plating/stripping. Similarly, Peng et al.^[56] reported that a nonionic amphiphilic polysorbate (PS) additive spontaneously adsorbs on the Zn surface, forming a water-shielding EDL that blocks H_2O access, suppresses side reactions, and stabilizes the SEI. This synergistic EDL-SEI effect promotes planar Zn deposition, improves desolvation and Zn^{2+} transport, and enables highly reversible plating/stripping over extended cycles. Collectively, these studies highlight that EDL-focused electrolyte design—whether through anion-mediated preferential adsorption or amphiphilic surface modifiers—provides a mechanistic route to regulate interfacial solvation structure and electrochemical performance, representing a promising frontier for engineering durable and high-performance aqueous Zn metal anodes.

3.3 Separator modification and interface-induced regulation

The separator plays a critical role in AZIBs, not only providing ion transport pathways but also effectively suppressing dendrite growth and interfacial side reactions. As illustrated in Figure 6a, Yu et al.^[57] chemically modified a chitosan-based matrix *via* a Schiff-base reaction, transforming its surface charge from positive to negative. In addition, NbN with strong zinc affinity was coated onto the chitosan matrix to form a Janus separator with a thickness of only 19 μm . This separator simultaneously facilitates Zn^{2+} transport while limiting two-dimensional Zn^{2+} diffusion, thereby significantly suppressing dendrite growth, by-product formation, and hydrogen evolution. Zhao et al.^[58] reported a polyaniline-functionalized glass fiber separator (PANI-GF), as shown in Figure 6c. Its porous structure restricts zinc ion flux, while abundant nitrogen-containing functional groups adsorb water molecules to reduce interfacial side reactions. Moreover, by modulating the local pH, the separator mitigates MnO_2 cathode dissolution (See Figure 6b), leading to markedly improved full-cell cycling performance. Luo et al.^[59] further proposed a multifunctional separator (MFS) design (See Figure 6f), where a metal-organic framework (MOF) coating is *in-situ* grown on glass fiber. This approach homogenizes interfacial ion flux and increases the Zn^{2+} transference number (See Figures 6d and 6e), resulting in outstanding electrochemical performance. Taken together, these separator engineering strategies highlight a paradigm shift from passive ion-conducting membranes toward multifunctional interfacial regulators. Rather than serving solely as physical barriers, advanced

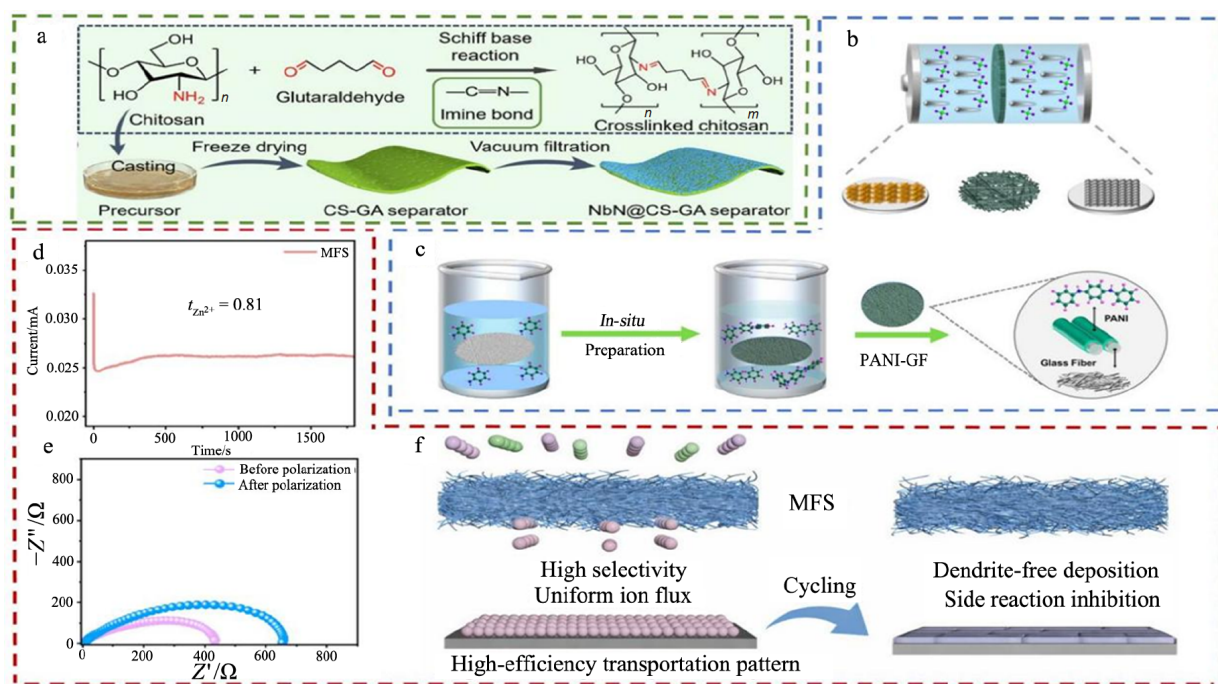


Illustration of the crosslinking process of chitosan and schematic diagram of the preparation process of NbN@CS-GA separator (a), schematic illustration of Zn anode, separators, and cathode for Zn|PANI-GF|MnO₂ (b), the *in-situ* preparation of PANI-GF (c), current variation with time during polarization and associate EIS curves fitting (d and e), and schematic illustration of ion transportation pattern for MFS separator (f)

Figure 6 Design and interfacial regulation mechanisms of functional separators in aqueous zinc-based batteries

separators actively modulate ion flux distribution, interfacial chemistry, and even local electrochemical environments across both electrodes. From this perspective, future separator design should be guided by system-level considerations, balancing ionic selectivity, interfacial stability, and compatibility with both Zn anodes and cathode chemistries under practical operating conditions.

4. Conclusion and perspectives

In recent years, considerable progress has been made in interface engineering strategies for stabilizing zinc metal anodes in AZIBs, encompassing artificial SEI construction, electrolyte regulation, and separator functionalization. Through deliberate modulation of interfacial structures, solvation environments, and ion-transport pathways, these approaches effectively mitigate dendrite formation and parasitic reactions, thereby enhancing the reversibility and cycling stability of Zn plating/stripping. Future research on zinc anode interface engineering should move beyond empirical optimization toward mechanism-driven design. Achieving a fundamental understanding of the dynamic evolution of the Zn/electrolyte interface under practical operating conditions—such as high current densities and prolonged cycling—will require the combined use of advanced *in-situ*/operando characterization techniques and multiscale theoretical modeling. In parallel, interfacial architectures with durable structural integrity and chemical stability should be rationally developed and assessed from a system-level perspective, accounting for their compatibility

with electrolytes, separators, and cathode chemistries, as well as scalability and manufacturing feasibility for practical AZIBs.

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水系锌离子电池中锌金属负极界面工程的研究进展与展望

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摘要 水系锌离子电池 (AZIBs) 凭借其高安全性、低成本、环保特性以及锌金属的天然储量, 近年来备受关注。然而, 枝晶生长、氢气析出以及锌负极界面副反应等问题严重制约了其实际应用, 导致电池可逆性差、循环稳定性有限。界面工程作为有效策略, 通过直接调控界面反应并优化锌沉积/剥离行为, 显著提升了负极稳定性。本文系统综述了锌负极界面工程的最新进展, 重点探讨人工固态电解质界面、电极/电解液界面调控及隔膜改性等研究方向。解析了这些策略在抑制枝晶、减少氢气析出及降低界面副反应方面的设计原理与作用机制。最后, 本文概述了当前界面设计面临的挑战及未来发展方向, 为AZIBs的实际应用提供思路。

关键词 界面工程; 界面稳定性; 水系锌离子电池; 锌金属负极