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Electrolyte Engineering in Aqueous Zinc-Ion Batteries: From Solvation Chemistry to Interface Stabilization

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Received: 28 April 2026 | **Revised:** 26 May 2026 | **Accepted:** 28 May 2026

Keywords: aqueous zinc-ion batteries | dendrite inhibition | electrolyte engineering | interface regulation | solvation

ABSTRACT

Aqueous zinc-ion batteries (AZIBs) are widely regarded as promising next-generation energy storage systems due to the low cost, inherent safety, and environmental compatibility. However, the practical application of AZIBs is still hindered by uncontrolled zinc dendrite growth, hydrogen evolution reactions, electrode corrosion, cathode dissolution, and poor adaptability under extreme temperatures. These challenges are closely related to the strong solvation structure of Zn²⁺ and the high activity of water molecules in aqueous environments. Therefore, electrolyte engineering has emerged as a key strategy to regulate ion transport, stabilize electrode/electrolyte interfaces, and improve overall battery performance. This review provides a systematic overview of recent advances in electrolyte design for AZIBs. First, the working principles of AZIBs and the key electrolyte-related challenges are discussed. Subsequently, recent advances in electrolyte regulation strategies are comprehensively reviewed, including solvent engineering, electrolyte salt optimization, functional electrolyte additives, and composite electrolyte design. Finally, the critical challenges and future research directions are discussed from the perspectives of electrolyte composition design, advanced characterization techniques, and mechanistic understanding. This review aims to provide fundamental insights and design guidelines for developing high-performance electrolytes and promoting the practical application of AZIBs technologies in large-scale energy storage systems.

1 | Introduction

The rapid growth of renewable energy and electrified transportation has intensified the demand for safe, sustainable, and cost-effective electrochemical energy storage systems [1–6]. Although lithium-ion batteries (LIBs) are widely used in portable electronics and electric vehicles owing to their high energy density and mature industrial ecosystem, their large-scale deployment for grid storage faces increasing concerns [7–13]. The uneven geographical distribution of lithium resources, rising raw material costs, and dependence on critical elements such as cobalt raise sustainability and supply-chain issues [14–18]. In addition, the

use of flammable organic electrolytes poses inherent safety risks, particularly under abusive or large-format operating conditions [19–21]. These challenges motivate the exploration of alternative battery chemistries and intrinsically safer electrolytes [22].

Aqueous zinc-ion batteries (AZIBs) have attracted considerable attention as a viable candidate owing to the merits of metallic zinc, including high theoretical capacity, low redox potential, natural abundance, and environmental benignity [23–33]. More importantly, the adaptation of aqueous electrolytes endows AZIBs with high ionic conductivity and improved safety, which makes them well suited for large-scale stationary energy storage

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[24, 25, 27, 34–38]. However, on the anode side, uneven Zn deposition induces dendrite growth, leading to short circuit and limited cycling life. Concurrent parasitic reactions, including hydrogen evolution and surface by-product formation, further deteriorate Coulombic efficiency and interfacial stability. On the cathode side, structural instability, dissolution, and sluggish Zn^{2+} diffusion often compromise capacity retention and rate capability [39–47]. Notably, many of these issues originate from the intrinsic properties of the aqueous electrolyte, where the high activity of water and the strong solvation of Zn^{2+} fundamentally govern interfacial chemistry, deposition behavior, and reaction reversibility [24, 48–51].

In response to these challenges, numerous approaches have been explored, including cathode structure engineering, artificial interphase construction on Zn anodes, and electrolyte optimization [27, 52–54]. Among them, electrolyte engineering plays a central and decisive role, as it simultaneously regulates Zn^{2+} solvation structure, interfacial reaction pathways, and electrochemical stability window [36, 52, 53]. Recent advances in high-concentration electrolytes, solvation-structure modulation, functional additives, hybrid aqueous systems, and gel or quasi-solid-state electrolytes have demonstrated significant effectiveness in suppressing dendrite formation, mitigating side reactions, and enhancing cycling durability [28, 55, 56].

Given the fundamental influence of the electrolyte on both electrode interfaces and ion transport, a thorough understanding of the principles governing electrolyte engineering is essential for advancing AZIBs technology. In this review, we systematically summarize recent advances in electrolyte engineering for AZIBs from the perspectives of solvation chemistry regulation and interfacial stabilization. First, the operating principles of AZIBs and the major challenges arising from electrolyte chemistry are presented, with particular emphasis on the origins of zinc dendrite growth, hydrogen evolution, anode corrosion, cathode dissolution, and uneven zinc-ion deposition. Subsequently, recent progress in electrolyte regulation strategies is comprehensively discussed, including solvent engineering, electrolyte salt optimization, functional additives, and composite electrolyte design, with focus on how these approaches regulate Zn^{2+} solvation structures, suppress water activity, and stabilize electrode/electrolyte interfaces. Finally, the unresolved

issues and future directions are outlined to guide the rational development of high-performance and practically viable AZIBs. This review seeks to clarify how electrolyte chemistry governs battery performance, offer guidance for the rational design of advanced electrolyte systems, and contribute to the fundamental understanding and practical implementation of AZIBs in safe and sustainable energy storage.

2 | Working Principle of AZIBs

Typical AZIBs consist of four main components [57]: (1) a cathode material for storing Zn^{2+} , (2) a separator that provides an ion transport channel, (3) a zinc anode, and (4) an aqueous electrolyte. The battery energy storage process is strongly influenced by the choice of cathode material and electrolyte. Figure 1 presents the basic configuration of AZIBs and highlights the electrolyte-mediated charge storage pathways. In addition to Zn^{2+} insertion, H^+ insertion and $\text{Zn}^{2+}/\text{H}^+$ coinsertion may also occur at the cathode side, depending on the cathode structure and electrolyte environment. Meanwhile, the aqueous electrolyte governs Zn^{2+} solvation/desolvation, ion transport, and interfacial reactions at both electrodes. Therefore, the working mechanism of AZIBs should be understood not only as Zn^{2+} shuttling between two electrodes but also as a coupled process involving solvation chemistry and electrode/electrolyte interfacial behavior. During discharge, the Zn anode releases Zn^{2+} through oxidation, and the generated ions migrate across the electrolyte to the cathode and are stored in the cathode host, while electrons are transferred through the external circuit to provide electrical output. During charging, Zn^{2+} is released from the cathode, transported back to the anode, and reduced to Zn metal. This reversible Zn deposition/stripping and insertion/extraction process constitutes the fundamental working principle of AZIBs.

AZIBs employ metallic zinc directly as the anode active material, featuring a simple electrode design and a relatively high theoretical capacity [58, 59]. However, in aqueous environments, Zn^{2+} commonly exists as hydrated ions (e.g., $[\text{Zn}(\text{H}_2\text{O})_6]^{2+}$), and their solvation structures, desolvation process, and interfacial reaction kinetics can significantly influence the deposition morphology, interfacial stability, and cycle life of the battery [60–63].

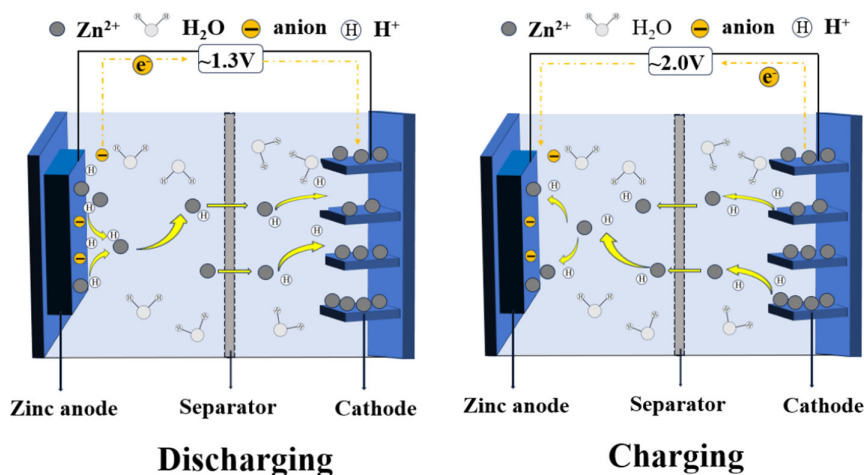
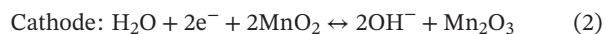
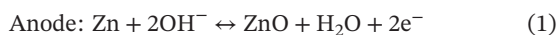


FIGURE 1 | Schematic illustration of electrolyte-mediated charge storage processes in AZIBs.

Furthermore, water can also promote hydrogen evolution and the generation of interfacial by-products, making the electrolyte a crucial factor in regulating electrochemical behavior.

In terms of electrolytes, aqueous zinc-based batteries are typically categorized into alkaline systems and neutral/mildly acidic zinc-ion systems [64, 65]. These two systems exhibit distinct electrochemical reaction mechanisms.

In alkaline electrolytes, the zinc anode mainly undergoes Zn/ZnO conversion reactions, while cathode materials typically involve hydroxide-related conversion reactions. Although such systems have long been used in zinc batteries, their limited reversibility often hinders rechargeable applications. Taking alkaline zinc-manganese batteries as a representative example, alkaline electrolytes are commonly used because their high OH⁻ mobility and fast ion transport can effectively reduce internal resistance and energy loss during operation. As a result, such alkaline battery systems usually exhibit relatively high energy density, good storage stability, and steady discharge performance [31, 34, 35, 66]. In alkaline zinc-manganese batteries, the anode Zn is oxidized to ZnO during discharge, while MnO₂ at the cathode is reduced to Mn₂O₃. Electrons pass through the external circuit to provide electrical output. The typical reaction mechanism is as follows:

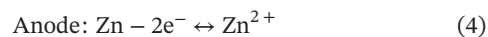


However, alkaline zinc ion batteries have limitations, with forced charging risking leakage or explosion, leading to low resource utilization efficiency [67]. Additionally, irreversible side reactions on the MnO₂ cathode and Zn anode lower the Coulombic efficiency and accelerate capacity fading [68].

In contrast, modern rechargeable AZIBs are mainly operated in neutral or mildly acidic electrolytes containing zinc salts [69]. Taking the rechargeable zinc-ion battery employing an α -MnO₂ cathode as an example, it typically uses a neutral aqueous solution containing Zn²⁺ as the electrolyte. During discharge, zinc at the anode undergoes oxidation to form soluble Zn²⁺, which enters the electrolyte and then inserts into the cathode [70, 71]. During charging, Zn²⁺ is extracted from the cathode, migrates through the electrolyte, and finally deposits on the zinc anode surface after gaining electrons [72]. Mildly acidic electrolytes allow reversible Zn plating/stripping and suppress hydrogen evolution [73].

In this system, Zn²⁺ can rapidly and reversibly deposit/dissolve on the zinc anode surface and insert/extract from the α -MnO₂ tunnels [74]. The electrochemical principle of Zn-ion batteries is unique: during charging, Zn²⁺ detaches from the α -MnO₂ tunnel structure and deposits on the zinc anode; during discharge, zinc loses electrons to form Zn²⁺, which inserts into the α -MnO₂ tunnels. This working mode resembles a “rocking chair battery,”

with Zn²⁺ shuttling between the positive and negative electrodes [75]. The typical reaction principle is as follows:



Besides intercalation-type cathodes, conversion-based cathodes have also attracted increasing attention in aqueous zinc-based batteries because of their multielectron redox chemistry. Typical examples include I⁻- and Br⁻-based cathodes, whose charge storage process relies on the reversible conversion of redox active species rather than the insertion of Zn²⁺ into a rigid host framework. In Zn-iodine batteries, the I⁻/I₂ redox couple provides considerable redox activity, while further I⁰/I⁺ conversion can potentially increase energy output [76, 77]. However, this conversion process is usually accompanied by the formation of soluble polyiodide intermediates, such as I₃⁻ and I₅⁻. These species can dissolve into aqueous electrolyte and migrate across the separator, leading to active material loss, self-discharge, and reduced cathode reversibility. More importantly, the crossover of polyiodides to the Zn anode can induce chemical corrosion, intensify water-related side reactions, and disturb uniform Zn deposition [78]. Similar concerns may also exist in bromine-based systems because soluble bromine species can participate in shuttle behavior and parasitic interfacial reactions. Therefore, for conversion-based cathodes, electrolyte design should not only focus on Zn²⁺ transport and anode stability but also regulate the dissolution, confinement, adsorption, and interfacial conversion of cathode-derived redox intermediates.

Overall, both intercalation-type and conversion-based AZIBs are strongly influenced by electrolyte. The electrolyte not only governs Zn²⁺ transport, solvation structure, and Zn anode reversibility but also affects cathode/electrolyte interfacial reactions, redox intermediate migration, and the stability of active materials. Therefore, understanding the relationship among electrolyte composition, charge storage mechanism, and interfacial behavior is essential for designing high performance AZIBs. The following sections will discuss how different electrolyte systems improve the electrochemical performance of AZIBs by regulating Zn²⁺ solvation, water activity, ion transport, and electrode/electrolyte interfaces [79–81].

3 | Key Challenges of Electrolytes in AZIBs

Although aqueous electrolytes endow AZIBs with rapid ionic transport and intrinsic safety, they also introduce a series of critical challenges that hinder the practical use of AZIBs. Due to the strong solvation of Zn²⁺ and the intrinsically high reactivity of water molecules, aqueous systems are commonly accompanied by uncontrolled Zn dendrite growth, hydrogen evolution reaction (HER), anode corrosion, cathode dissolution, and uneven zinc-ion deposition behavior. These electrolyte-related issues are closely interconnected and collectively lead

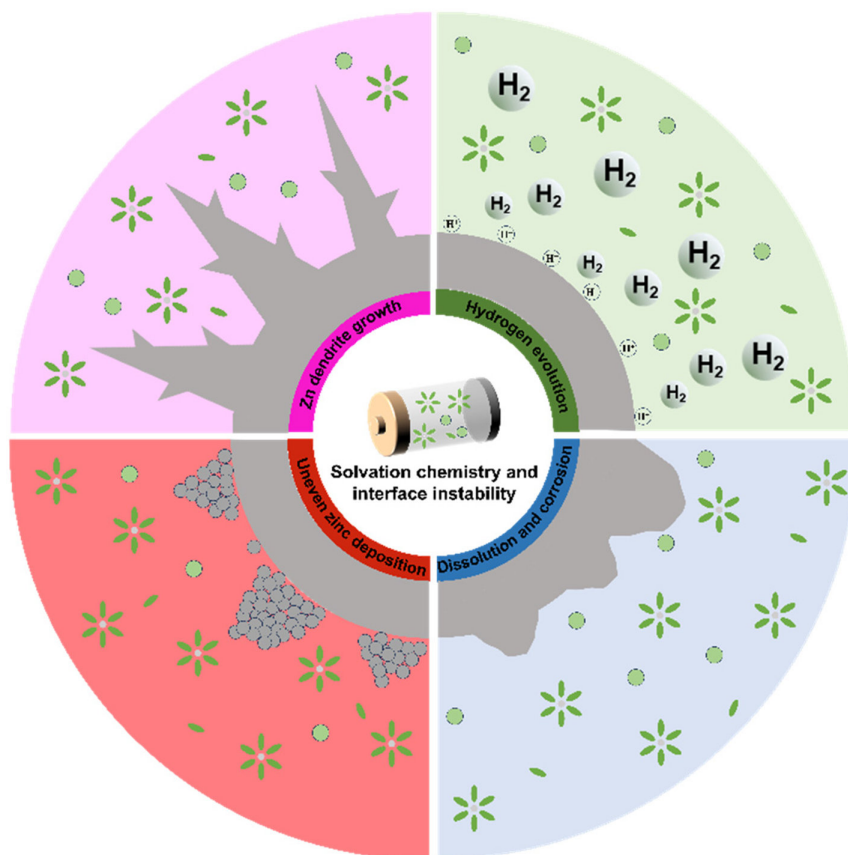


FIGURE 2 | Electrolyte-related challenges and interfacial degradation mechanisms in AZIBs.

to deteriorated reversibility, unstable interfacial chemistry, and shortened cycling life. The major electrolyte-related challenges in AZIBs are schematically summarized in Figure 2. These challenges are not isolated failure modes, but are closely related to strong Zn^{2+} solvation, high water activity, unstable hydrogen bond networks, and nonuniform interfacial ion flux. These electrolyte derived factors collectively lead to HER, corrosion, cathode dissolution, uneven Zn deposition, and dendrite growth.

3.1 | Uncontrolled Zn Dendrite Growth

Uncontrolled Zn dendrite growth is one of the major obstacles in AZIBs during repeated cycling [32, 82]. In aqueous electrolytes, Zn^{2+} is strongly solvated by water molecules, and its deposition process is highly sensitive to interfacial electric-field distribution, ion concentration gradients, and the heterogeneous surface of the Zn anode. Under such conditions, Zn tends to preferentially nucleate and grow at high-energy or high-curvature regions, gradually forming protrusions and dendritic structures. As dendrites continue to grow, they may pierce the separator and trigger internal short circuits, thereby seriously undermining battery safety and cycling stability. In addition, dendrite growth also leads to low Zn utilization and poor Coulombic efficiency because part of the deposited Zn becomes electrically isolated during repeated plating/stripping processes. Therefore, suppressing dendrite formation is a fundamental prerequisite for achieving stable and durable AZIBs.

3.2 | Hydrogen Evolution

In aqueous electrolytes, HER is one of the most representative parasitic reactions in AZIBs and is closely associated with the intrinsically high activity of water molecules [50, 83, 84]. Zn^{2+} in aqueous media is usually coordinated by water molecules to form hydrated complexes. Water is not only the ion-transport medium but also an active participant in interfacial electrochemical reactions. During Zn plating, once the local electrode potential becomes sufficiently negative, water molecules at the Zn/electrolyte interface can undergo reduction to generate H_2 . The process is further aggravated by the strong coupling between solvated Zn^{2+} and surrounding water, which continuously brings reducible water molecules to the anode interface. As a result, HER competes directly with reversible Zn deposition/stripping, consuming electrons and electrolyte simultaneously and thereby lowering Coulombic efficiency [85].

The adverse impact of HER in AZIBs extends beyond hydrogen generation itself. It not only consumes electrolyte and induces local pH fluctuations but also promotes the formation of passivating by-products, disrupts interfacial contact, aggravates polarization, and accelerates Zn corrosion and interfacial degradation. These parasitic effects become more pronounced under prolonged cycling or high-current operation, ultimately undermining battery reversibility and stability. Mechanistically, HER is closely governed by electrolyte chemistry, including water activity, Zn^{2+} solvation structures, pH, anion species, and the hydrogen bond network. Therefore, suppressing HER

fundamentally requires electrolyte regulation at both the molecular and interfacial levels [86].

3.3 | Cathode Dissolution and Anode Corrosion

The incompatibility between aqueous electrolytes and electrode materials can easily induce cathode dissolution and anode corrosion, both of which are major causes of performance decay in AZIBs [87, 88]. On the cathode side, active material dissolution is particularly common in manganese- and vanadium-based cathodes, where long-term exposure to aqueous electrolytes may destabilize the host structure and continuously release active species into the electrolyte. Recent studies on $V_2O_5/V_3O_7 \cdot H_2O$ heterostructures further indicate that rational heterostructure design can improve aqueous Zn-ion storage by enhancing cathode structural stability and reaction kinetics, highlighting the close relationship between cathode stability and electrolyte compatibility [89]. On the anode side, metallic Zn is thermodynamically unstable in aqueous environments and is therefore vulnerable to spontaneous corrosion and by-product formation. These two degradation pathways are closely coupled through the electrolyte and together accelerate capacity fading, interfacial instability, and poor cycling reversibility.

Cathode dissolution in AZIBs mainly arises from structural instability and chemical incompatibility between cathode materials and aqueous electrolytes. During repeated cycling, Zn^{2+} insertion/extraction can induce lattice distortion, local stress accumulation, and even partial framework collapse, thereby weakening the confinement of active species in the host structure. In addition, the coparticipation of protons and water molecules, together with high water activity and local pH fluctuations, can further destabilize the cathode framework and accelerate the dissolution of active components, particularly in Mn- and V-based cathodes. Once dissolved into the electrolyte, these species may migrate to the Zn anode and trigger undesirable interfacial side reactions, while the continuous loss of active materials simultaneously reduces the number of available redox centers and deteriorates cathode structural integrity. Meanwhile, Zn anode corrosion in aqueous electrolytes also consumes active Zn and produces passivation by-products, further increasing interfacial resistance and disturbing Zn^{2+} transport. Therefore, cathode dissolution and anode corrosion should be regarded as coupled electrolyte-mediated degradation processes, highlighting the critical role of electrolyte engineering in maintaining stable electrode/electrolyte interfaces and prolonging the lifespan of AZIBs.

3.4 | Uneven Zinc-Ion Deposition Behavior

Uneven zinc-ion deposition is another key challenge in aqueous electrolyte systems. In practical cycling, the transport, desolvation, and interfacial reduction of Zn^{2+} are often spatially non-uniform due to concentration gradients, interfacial electric-field inhomogeneity, and heterogeneous solvation environments. As a consequence, Zn^{2+} tends to preferentially deposit at localized regions on the anode, leading to uneven nucleation, surface roughening, and the gradual development of nonuniform Zn deposition morphologies. This process not only aggravates local current concentration and promotes dendrite formation but also

accelerates interfacial side reactions and by-product accumulation. Therefore, achieving homogeneous Zn^{2+} transport and deposition through electrolyte regulation is essential for improving the electrochemical reversibility of the Zn anode and the long-term cycling durability of AZIBs [24, 90–92].

Overall, dendrite growth, HER, corrosion, cathode dissolution, and uneven Zn^{2+} deposition all originate from the fundamental solvation structure of Zn^{2+} and high reactivity of water in electrolytes. Therefore, electrolyte engineering through modulation of solvation chemistry, suppression of water activity, and stabilization of interfacial reactions emerges as the central strategy to address these interconnected challenges.

4 | Strategies for Regulating Electrolytes

The electrolyte of AZIBs is not merely an ion-conducting medium, but a key component that governs Zn^{2+} solvation chemistry, water activity, interfacial reactions, and battery reversibility [80]. In addition to basic solvents and zinc salts, modern electrolyte systems often incorporate functional additives or polymer/inorganic frameworks to achieve more precise interfacial and transport regulation. Accordingly, electrolyte engineering in AZIBs can be generally categorized into four aspects: solvent regulation, electrolyte salt optimization, functional additive engineering, and composite electrolyte design. In the following sections, these representative strategies are discussed in detail to illustrate how rational electrolyte design can suppress side reactions, stabilize Zn deposition/stripping, and improve the overall electrochemical behavior of AZIBs.

To further connect these diverse electrolyte strategies with a unified mechanism, the relationship among Zn^{2+} solvation, interfacial desolvation, nucleation, and deposition should be clarified. From a mechanistic perspective, most electrolyte engineering strategies in AZIBs can be understood through a continuous “solvation–desolvation–nucleation–deposition” framework. In the bulk electrolyte, Zn^{2+} is usually coordinated by water molecules to form hydrated species such as $[Zn(H_2O)_6]^{2+}$. Different solvents, anions, and additives can modify the first solvation shell of Zn^{2+} by partially replacing coordinated water molecules, tuning Zn^{2+} -solvent interactions, or promoting ion pair formation between Zn^{2+} and anions. For example, highly concentrated electrolytes and anion regulated systems usually increase anion participation in the primary solvation sheath. Organic aqueous hybrid electrolytes and functional additives can reduce the amount of highly active coordinated water. These changes directly affect the stability of solvated Zn^{2+} and the number of water molecules transported to the electrode interface.

When solvated Zn^{2+} approaches the Zn anode, its first solvation shell must be partially or completely removed before electron transfer and Zn deposition can occur. Therefore, the evolution of the solvation structure strongly influences the interfacial desolvation barrier and charge transfer kinetics. A water-rich and strongly hydrated Zn^{2+} solvation sheath usually requires a higher desolvation energy. This slows interfacial charge transfer, increases local polarization, and brings more active water to the Zn surface, thereby promoting HER, corrosion, and by-product

formation. By contrast, an anion or additive regulated solvation sheath with reduced active water can lower the desolvation barrier, suppress parasitic reactions, and facilitate faster Zn^{2+} reduction. Thus, bulk solvation regulation can be regarded as the upstream step that controls interfacial desolvation behavior. It should also be noted that interfacial desolvation is a dynamic process coupled with local electric-field evolution, ion redistribution, and interfacial water reorientation, rather than a simple transition determined only by the static bulk solvation structure.

The desolvation kinetics further determine Zn nucleation and deposition behavior. Sluggish desolvation and uneven interfacial Zn^{2+} supply tend to increase nucleation overpotential, generate nonuniform nucleation sites, and amplify local electric-field gradients. These effects favor vertical Zn growth and dendrite formation. In contrast, fast and homogeneous desolvation can reduce concentration polarization, regulate nucleation density, homogenize Zn^{2+} flux, and guide compact or lateral Zn deposition. Therefore, different electrolyte systems, although diverse in composition, can be compared within the same mechanistic framework. They regulate the first solvation shell of Zn^{2+} , alter the interfacial desolvation process, and finally control Zn nucleation and deposition morphology.

4.1 | Solvent Regulation

In AZIB research, electrolyte solvation structures and interfacial stability are critical for addressing challenges like HER, passivation, and morphological changes of zinc anodes [32, 42]. Rational regulation of solvation structures can improve electrodelectrolyte interfacial stability, thereby enhancing battery performance and lifespan, which is of great significance in both academic research and practical applications [93, 94].

4.1.1 | Pure-Water Solvents

Water is the most commonly used solvent in AZIB electrolytes owing to its low cost, high safety, nonflammability, and intrinsically high ionic conductivity [28, 95, 96]. However, pure aqueous electrolytes also suffer from several inherent drawbacks, including strong Zn^{2+} solvation, high water activity, unstable Zn/electrolyte interfaces, hydrogen evolution, and dendrite growth [97]. Recent studies have shown that even within a nominally pure aqueous system, rational regulation of water molecules themselves can effectively improve Zn electrochemistry [98].

Wang et al. reported a pure-water electrolyte strategy based on controlling the O–H bonds of water molecules to regulate the solvation environment of Zn^{2+} and stabilize the Zn/electrolyte interface. By tailoring the network of hydrogen bonds, the electrolyte can weaken the reactivity of free water, modify the Zn^{2+} solvation structure, and reduce parasitic water-induced side reactions. Consequently, the interfacial reversibility of Zn plating/stripping is significantly improved, while the growth of dendrites and interfacial corrosion can be markedly suppressed. This work demonstrates that pure-water solvents are not merely a baseline system for comparison, but can also serve as an active platform for solvent regulation. More importantly, it highlights that direct manipulation of water chemistry itself is a promising

route to address key challenges of AZIBs, including Zn^{2+} solvation imbalance, poor interfacial stability, and low reversibility.

Despite these advances, the regulation space of pure-water solvents remains relatively limited compared with hybrid solvents or highly concentrated systems. The strong coordination between Zn^{2+} and coordinated water molecules and the intrinsically high reactivity of water still make it difficult to completely suppress HER, corrosion, and nonuniform Zn deposition. Therefore, pure-water solvent regulation is better understood as a fundamental strategy for probing water chemistry and interfacial mechanisms while also providing important design inspiration for the development of more advanced electrolyte systems.

4.1.2 | Organic Solvents

To balance capacity and stability, researchers have proposed to tailor the electrolytes of AZIBs by introducing organic solvents, in order to suppress the adverse influence of water while still allowing sufficient proton insertion. For instance, Naveed et al. introduced triethyl phosphate (TEP) as the major organic solvent and further developed a mixed TEP/ H_2O electrolyte system to address zinc dendrite growth and low Coulombic efficiency in conventional aqueous electrolytes [99]. The authors designed a TEP-based electrolyte using $\text{Zn}(\text{CF}_3\text{SO}_3)_2$, and optimized a mixed system of TEP: H_2O (7:3) for full cells. This electrolyte features high safety and a broad electrochemical stability window (2.25 V). The in situ formed $\text{Zn}_3(\text{PO}_4)_2$ interfacial layer acts as a template, guiding uniform zinc nucleation and forming a unique highly interconnected porous structure instead of dendrites, thereby markedly improving the reversibility and stability of the anode. The symmetric cells achieved stable cycling for over 3000 h. In pure TEP, the zinc anode CE reached as high as 99.68%, and in the mixed system, it remained around 93.7%, far surpassing aqueous electrolytes.

Geng et al. proposed a hybrid aqueous electrolyte design strategy to address the issue of slow desolvation kinetics caused by strong binding between organic solvents and Zn^{2+} in conventional mixed electrolytes at low temperatures [100]. As shown in Figure 3a, protic solvents and dipolar aprotic solvents exhibit distinct dielectric constants and molecular characteristics. These differences strongly influence their interaction with Zn^{2+} and thus affect the local solvation environment in mixed electrolytes. In conventional hybrid systems, organic solvent molecules can directly enter the primary solvation sheath of Zn^{2+} , as illustrated in Figure 3b. Although such solvent participation may improve low-temperature fluidity, the coordination between Zn^{2+} and organic solvent molecules usually makes interfacial desolvation more difficult. As a result, sluggish charge transfer, large polarization, low Coulombic efficiency, and dendrite accumulation may occur. To tackle this issue, Geng et al. designed a zinc acetate/DMSO-based electrolyte in which the primary solvation structure is deliberately reconstructed. In the optimized system, acetate ions strongly interact with water molecules through hydrogen bonding, which blocks DMSO from participating in the Zn^{2+} coordination environment. Consequently, an organic solvent-free primary solvation structure is formed, as illustrated in Figure 3c. In this configuration, Zn^{2+} is mainly coordinated by acetate anions and water molecules, which facilitates interfacial

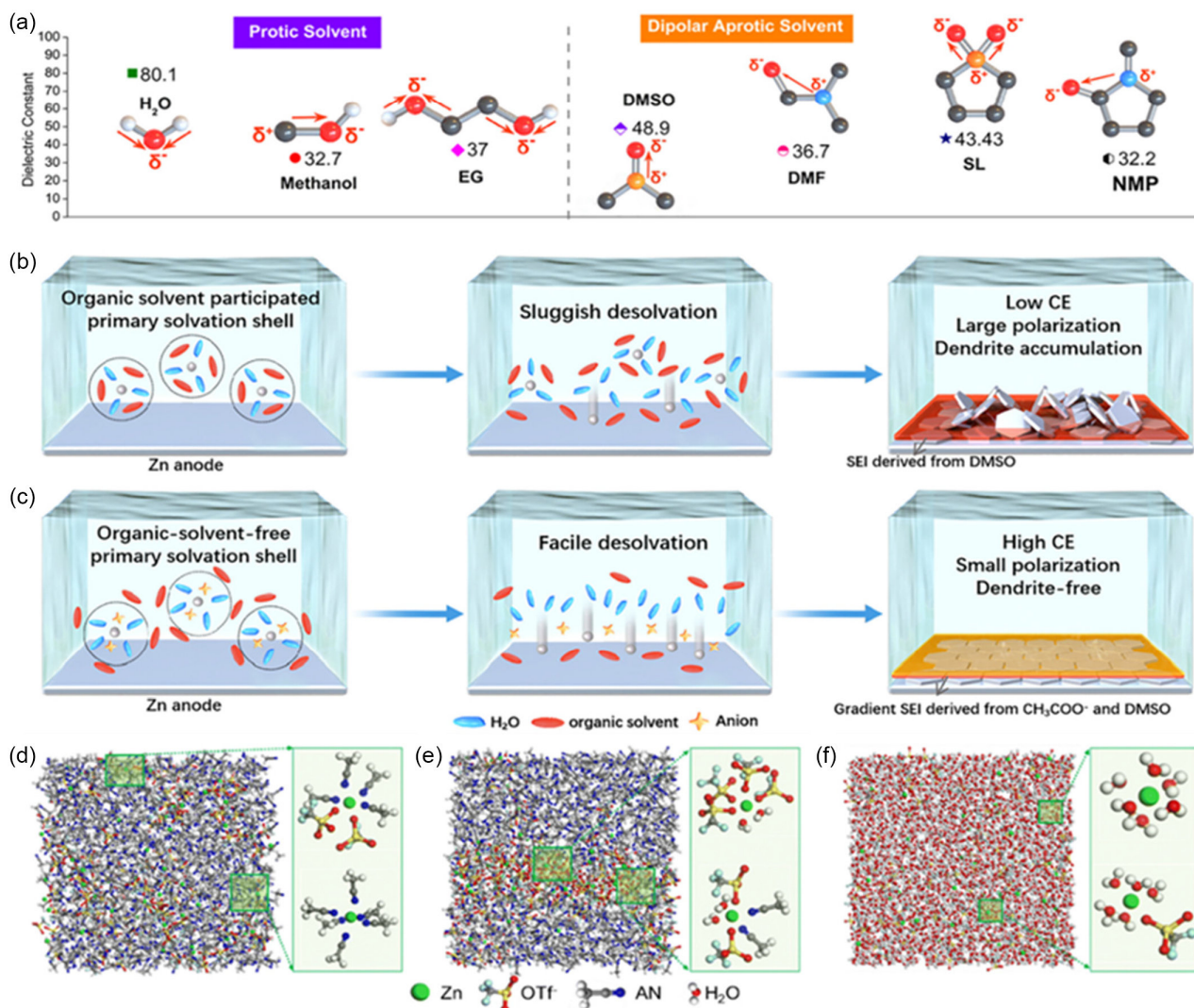


FIGURE 3 | (a) Dielectric constant and structural formula of the protic solvents of H₂O, methanol, and ethylene glycol (EG) and the dipolar aprotic solvents of dimethyl sulfoxide (DMSO), *N,N*-dimethylformamide (DMF), sulfolane (SL), and *N*-methyl pyrrolidone (NMP). (b,c) Schematic illustration showing the effects of primary solvation shell of (b) ZSD₂ and (c) ZAD₄ on the Zn anode at low temperature [100]. Reproduced with permission. Copyright 2025, Elsevier. Comparison of Zn(OTf)₂ electrolytes in AN with various H₂O contents. MD simulation snapshots of (d) E-0% H₂O, (e) E-8% H₂O, and (f) E-100% H₂O [101]. Reproduced with permission. Copyright 2024, Royal Society of Chemistry.

desolvation and accelerates charge transfer. This weakly solvated structure also helps form a gradient solid electrolyte interphase, thereby promoting uniform and dendrite free Zn deposition. Owing to these effects, the electrolyte exhibits excellent low-temperature electrochemical performance. It enables stable Zn deposition/stripping for up to 5600 h at -20°C and achieves a Zn utilization as high as 51%. In hybrid electrolytes, the key issue is not simply whether organic solvents are introduced, but whether their participation in the primary solvation sheath can be properly controlled. This case shows that solvent regulation affects Zn deposition not only by changing water activity but also by determining whether organic solvent molecules enter the first solvation shell of Zn²⁺. Such regulation directly changes the interfacial desolvation barrier, charge transfer kinetics, and subsequent Zn nucleation/deposition behavior.

Jiang et al. proposed a hybrid-solvent electrolyte approach by introducing a small amount of water into acetonitrile, in order to

balance capacity and stability in vanadium-based AZIBs [101]. In this design, the key point is not simply the use of mixed solvents, but the deliberate control of Zn²⁺ solvation through trace water regulation. A trace amount of water can provide sufficient proton participation for the cathode reaction, while excessive side reactions triggered by water are simultaneously suppressed. As shown in Figure 3d-f, local solvation structure changes markedly with water content. In the anhydrous system, Zn²⁺ is mainly coordinated in an organic-solvent-dominated environment (Figure 3d). After introducing 8% H₂O, a more balanced coordination structure is formed, in which water participates in the solvation sheath without overwhelming the whole electrolyte environment (Figure 3e). In the fully aqueous system, water becomes the dominant coordinating component around Zn²⁺ (Figure 3f), which is more likely to aggravate side reactions, cathode dissolution, and Zn anode corrosion. Therefore, the 8% H₂O containing electrolyte achieves an effective compromise. It reduces overall water activity while retaining sufficient ionic conductivity and

proton availability. As a result, both high capacity and improved stability can be achieved in the vanadium-based battery system. In addition, this electrolyte also exhibits excellent low-temperature adaptability and can support battery operation at -50°C . These results indicate that rational control of trace water content in organic and aqueous hybrid electrolytes is an effective way to tune Zn^{2+} solvation structure and interfacial stability simultaneously.

4.1.3 | Deep Eutectic Solvents

Deep eutectic solvents (DES) serve as the core modification system for AZIBs. By restructuring hydrogen bond networks and regulating solvation chemistry, they fundamentally address major issues of traditional aqueous electrolytes. The core improvement strategy is to retain the advantages of aqueous electrolytes, including high ionic conductivity and high safety, while eliminating or mitigating the negative effects of free water. Meanwhile, molecular-level coordination is used to optimize Zn^{2+} transport and electrode interface behavior, achieving a synergistic enhancement of battery cycling stability, Coulombic efficiency, and wide-temperature performance.

However, DES-based and hydrated eutectic electrolytes still face several practical limitations. Their strong hydrogen bond and coordination networks are beneficial for reducing free water activity and stabilizing the Zn/electrolyte interface, but they usually lead to relatively high viscosity and sluggish Zn^{2+} transport. This may increase polarization and limit rate capability, especially under high current density, low-temperature, or high-loading cathode conditions. In addition, the preparation of some eutectic systems requires precise component ratios and thermal treatment, which may increase processing complexity. Therefore, future DES-based electrolyte design should balance water activity suppression with fast ion transport, interfacial compatibility, and scalable preparation.

In order to mitigate the high water activity and interfacial instability commonly encountered in aqueous electrolytes, Thorat et al. prepared an electrolyte “ZE DES” by mixing ZnCl_2 with EG at a molar ratio of 1:4 at 100°C [102]. In this system, EG acts as a hydrogen bond donor and interacts with ZnCl_2 to form a stable eutectic liquid structure, which endows the electrolyte with a relatively wide electrochemical window of about 2V and good thermal stability. Such a design was proposed to address the Zn dendrite growth and interfacial instability issues in conventional AZIBs. To evaluate its effectiveness, the authors compared the Zn deposition morphology and stripping/plating behavior in ZE DES and conventional ZnSO_4 electrolyte. After cycling, the Zn anode in 2m ZnSO_4 shows a damaged surface with obvious non-uniform deposition features (Figure 4a,b), whereas the Zn anode cycled in ZE DES presents a much flatter and denser morphology without obvious dendrites (Figure 4c,d). Consistently, the Zn||Zn symmetric cell with ZE DES delivers stable cycling for 300h at 0.2mAh cm^{-2} (Figure 4e), with a cycle life approximately twice that of the conventional aqueous electrolyte, while the ZnSO_4 -based cell exhibits earlier polarization fluctuation and poorer reversibility (Figure 4f). Based on these experimental results, the authors attributed the improved electrochemical stability to the unique eutectic coordination environment formed between EG and ZnCl_2 . Through hydrogen bond interactions and Zn—O

coordination, EG was proposed to regulate the local coordination structure, promote salt dissociation, and form a compact molecular arrangement, thereby reducing solvent induced side reactions and enabling more uniform and reversible Zn deposition.

In addition to conventional DES, hydrated eutectic electrolytes have also emerged as an important related category for regulating Zn^{2+} solvation chemistry and interfacial behavior in aqueous systems. To further regulate Zn^{2+} solvation and interfacial stability, Liu et al. designed a hydrated eutectic electrolyte, termed ZU-8, by mixing ZnCl_2 , urea, and H_2O at a molar ratio of 1:3:8 [103]. In this electrolyte, urea serves as a key hydrogen bonding and coordination component, enabling the formation of a hydrated eutectic structure with reduced water activity and regulated Zn^{2+} solvation behavior. Such a design was proposed to address the interfacial side reactions, dendrite growth, and poor Zn reversibility commonly encountered in conventional aqueous ZnSO_4 electrolytes.

To evaluate its effect, the authors compared the Zn deposition morphology in ZU-8 and ZnSO_4 electrolytes. As shown in Figure 4g,h, Zn deposited in ZU-8 exhibits a dense and uniform morphology at different magnifications, indicating homogeneous nucleation and growth. In contrast, the Zn deposits formed in ZnSO_4 electrolyte display large, irregular, and dendritic structures (Figure 4i,j), reflecting severe deposition inhomogeneity. To explain this difference, the authors proposed that the superior interfacial behavior of ZU-8 originates from its reconstructed solvation and hydrogen bonding environment. As schematically illustrated in Figure 4k, urea molecules are incorporated into Zn^{2+} solvation sheath and disrupt original hydrogen bond network of water, while simultaneously forming a stabilizing interfacial layer that promotes uniform Zn deposition. By comparison, the conventional ZnSO_4 electrolyte retains a water-dominated solvation structure and strong hydrogen bond network, which favor dendrite growth and by-product formation (Figure 4l). Therefore, the work demonstrates that hydrated eutectic electrolytes such as ZU-8 can effectively couple solvation-structure regulation with interface stabilization, thereby enabling more uniform and reversible Zn deposition.

4.2 | Electrolyte Salts

Electrolyte salts are the key functional components of electrolytes. Their type, concentration, and ionic properties directly determine the electrolyte's ionic conductivity, the solvation structure of Zn^{2+} , water activity, and electrode interface behavior, while also collectively influencing the electrolyte's electrochemical stability window, wide-temperature performance, and battery cycle life. As the primary variable for regulating electrolyte performance, controlling the salts is therefore critically important.

4.2.1 | Sulfate Systems

AZIBs often use ZnSO_4 as the main solute due to its high solubility and ionic conductivity, providing efficient ion transport channels [28]. However, pure ZnSO_4 electrolytes suffer from Zn dendrite growth and HER, degrading performance and lifespan [104]. Therefore, traditional zinc salts still need to be improved. Xu et al. reported AZIBs operating in a mild electrolyte environment [73].

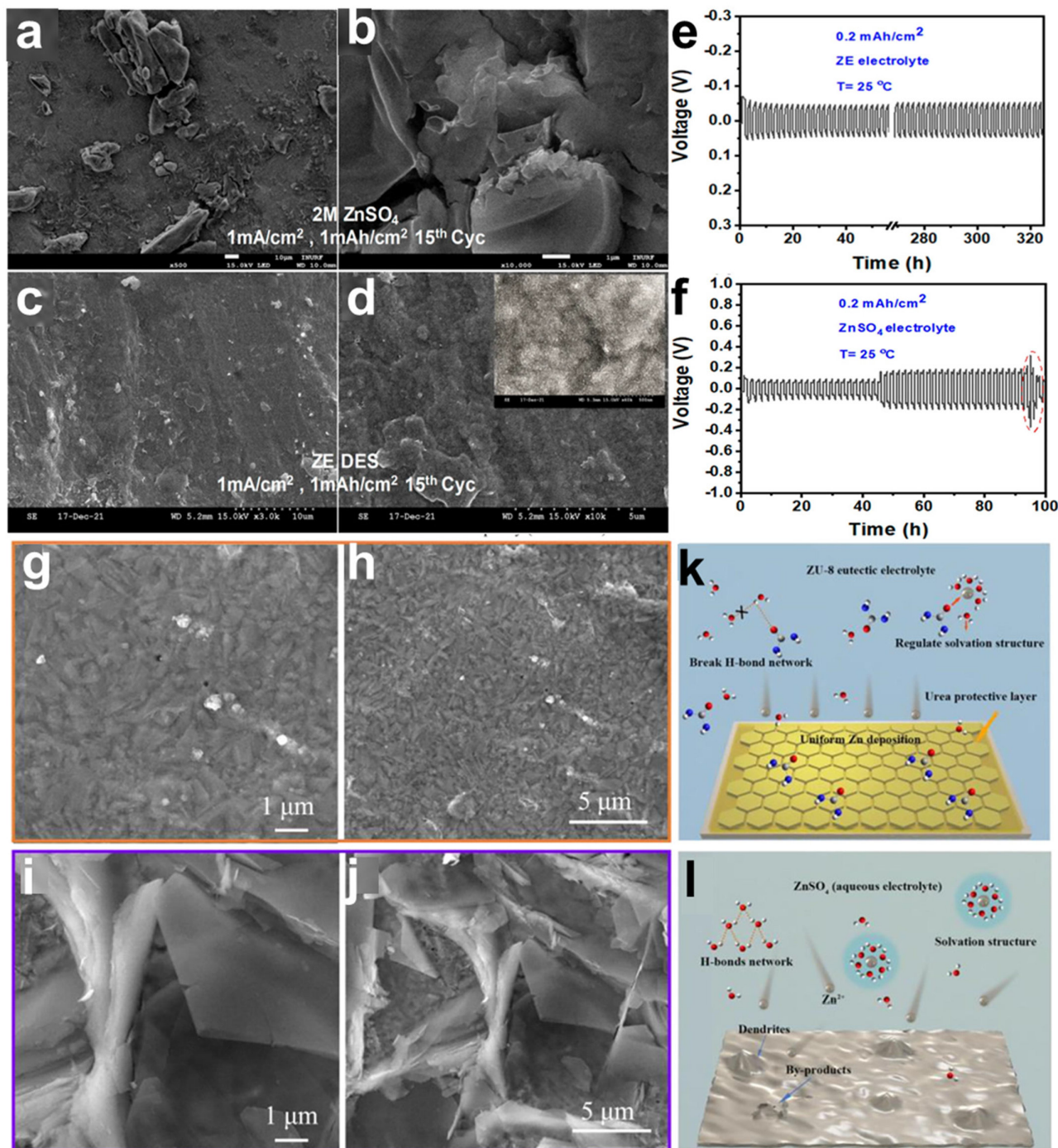


FIGURE 4 | (a,b) 2M ZnSO₄ electrolyte ZE electrolyte [102]; (c,d) ZE electrolyte after 15 cycles at 1 mA h cm⁻² [102]. Long-term stripping/plating performance of the Zn metal anode at 0.2 mA h cm⁻² in (e) ZE electrolyte and (f) aqueous 1 M ZnSO₄ [102]. Reproduced with permission. Copyright 2022, Frontiers. SEM images of Zn nuclei deposited on the Cu collector at 0.1 mA cm⁻² with 0.25 mA h cm⁻² in (g,h) ZU-8 and (i,j) ZnSO₄ electrolytes [103]. Schematic illustration of the solvation structure and interface chemistry in (k) ZU-8 and (l) ZnSO₄ electrolytes [103]. Reproduced with permission. Copyright 2019, Elsevier.

Their system employed an α -MnO₂ cathode and a mild aqueous electrolyte based on ZnSO₄ or Zn(NO₃)₂. Under such conditions, the battery chemistry was interpreted in terms of reversible Zn²⁺ migration between cathode and anode, which is distinct from the largely irreversible reaction pathways typically associated with conventional alkaline Zn||MnO₂ systems. This work provided an important basis for understanding the configuration and electrochemical mechanism of mild AZIBs, and it also offered

useful guidance for the subsequent development of rechargeable AZIB systems.

Pan et al. constructed a mildly acidic aqueous ZnSO₄ electrolyte system by rationally selecting the electrolyte salt [105]. The purpose of this design was to overcome the poor reversibility of conventional alkaline zinc–manganese batteries, in which cathode structural collapse and Zn anode passivation usually

shorten cycle life. The weakly acidic environment created by ZnSO_4 significantly improved the reversibility of battery chemistry. Consequently, the system delivered a high operating voltage and a high specific capacity of 285 mAh g^{-1} . It also achieved excellent long-term stability, retaining 92% of its capacity after 5000 cycles, with the Zn anode remaining highly stable during cycling. Mechanistically, the ZnSO_4 electrolyte provided a specific proton environment that changed the reaction pathway of the $\alpha\text{-MnO}_2$ cathode. Instead of undergoing irreversible phase transformation, the cathode followed a more reversible reaction process mainly involving H^+ . This work demonstrated the important role of electrolyte chemistry in improving reversibility and established a useful strategy for developing low cost aqueous zinc-based batteries.

4.2.2 | Chloride Systems

Single component aqueous zinc chloride solutions are often used as electrolytes due to their high solubility [24]. In aqueous solution, the high mobility of Zn^{2+} and Cl^- is beneficial for rapid ion transport and may, in principle, favor good rate performance. From this perspective, chloride-based electrolytes represent an important class of zinc salt systems in AZIBs. However, dilute ZnCl_2 electrolytes still suffer from poor interfacial stability. The high reactivity of free water and the insufficient regulation of Zn^{2+} solvation often lead to severe hydrogen evolution, by-product formation, and non-uniform Zn deposition. These parasitic processes reduce reversibility and accelerate cell failure. In addition, the electrochemical stability window of conventional dilute chloride electrolytes remains limited, which further restricts their practical application [24].

Therefore, although ZnCl_2 -based electrolytes possess intrinsic advantages in ionic transport, their interfacial chemistry must be carefully regulated before they can be used in stable AZIB systems. In particular, strategies based on solvation restructuring and water activity suppression have proven especially important for improving the reversibility of chloride electrolytes. Representative progress in this direction, especially through high-concentration ZnCl_2 systems, will be discussed in the following section [106].

4.2.3 | High-Concentration Electrolytes

High-Concentration electrolytes significantly expand the electrochemical operating window of AZIBs by forming a solvation structure with reduced free water, suppress zinc dendrite growth and HER at the anode, alleviate cathode dissolution and structural degradation, and enable reliable operation over a wide temperature range.

To suppress water decomposition and stabilize the Zn anode interface, Wang et al. developed an ultrahigh-concentration water-in-salt electrolyte based on 1 m $\text{Zn}(\text{TFSI})_2$ and 20 m LiTFSI [107]. As shown in Figure 5a, the molecular dynamics simulation snapshot of the high-concentration zinc electrolyte (HCZE) illustrates the spatial organization of ions and water molecules in the ultrahigh-concentration electrolyte. This highly crowded configuration suggests a fundamentally reconstructed solution structure, in which free water is greatly restricted. Figure 5b further shows that the Zn^{2+} coordination environment gradually

changes with increasing LiTFSI concentration. In the dilute system, Zn^{2+} is mainly coordinated by water molecules. In the concentrated system, TFSI^- anions increasingly enter the primary solvation sheath. This trend is quantitatively confirmed by Figure 5c,d. The $\text{Zn}^{2+}\text{-O}(\text{TFSI})$ coordination number increases, whereas the $\text{Zn}^{2+}\text{-O}(\text{water})$ coordination number decreases as the salt concentration rises. Figure 5e further supports the presence of a distinct local aggregation structure in the concentrated electrolyte. Meanwhile, the pH change shown in Figure 5f reflects the concentration-dependent evolution of the electrolyte chemistry. Figure 5g provides additional evidence from ^{17}O NMR. The chemical shift change indicates that water molecules exhibit stronger interactions with the surrounding ionic species in the highly concentrated system. Consequently, the activity of free water is markedly reduced and electrochemical stability window is broadened. Based on these results, the authors proposed that the anion-rich solvation environment promotes preferential interfacial decomposition of TFSI^- derived species. This process leads to the in situ formation of a dense ZnF_2 rich interphase on the Zn surface. Such an interphase effectively suppresses HER, corrosion, and dendrite growth. Consequently, the electrolyte enables highly reversible Zn plating/stripping. This interfacial stabilization is also reflected in full cell performance. As shown in Figure 5h, $\text{Zn}||\text{LiMn}_2\text{O}_4$ cell maintains stable cycling over 4000 cycles with high Coulombic efficiency. These results demonstrate that the water-in-salt strategy is effective for simultaneously regulating solvation chemistry and interfacial behavior.

Zhang et al. employed low cost ZnCl_2 as the sole solute and constructed a highly concentrated “ ZnCl_2 water-in-salt” electrolyte by increasing the salt concentration to 30 m [106]. This strategy was designed to overcome the excessively high-water activity in conventional dilute aqueous electrolytes. In the concentrated system, the local liquid structure is fundamentally reconstructed. As shown in Figure 6a, the Raman spectra of ZnCl_2 electrolytes change markedly with increasing concentration, indicating a significant evolution of the ionic environment. Figure 6b,c further reveal that, in the 20 m and especially the 30 m electrolyte, chloride-rich Zn coordination species become dominant. In particular, the 30 m ZnCl_2 electrolyte is characterized by the formation of $[\text{ZnCl}_4]^{2-}$ dominated ionic clusters, while the proportion of water-coordinated species is greatly reduced. This solvation structure effectively confines most water molecules within the ionic coordination environment and markedly lowers the activity of free water. As a result, HER and the generation of by-products, including ZnO and $\text{Zn}(\text{OH})_2$ are significantly suppressed. More importantly, the altered coordination chemistry also changes the nucleation and deposition behavior of Zn, enabling more compact and uniform Zn deposition. The improved reversibility is clearly reflected in the electrochemical results. As shown in Figure 6d,e, the average CE increases from 73.2% in the 5 m electrolyte to 95.4% in the 30 m electrolyte. These results demonstrate that concentration induced solvation restructuring is highly effective for regulating water activity and stabilizing Zn deposition behavior in chloride-based electrolytes.

Fenta et al. further examined an often overlooked factor in water-in-salt electrolytes (WiSEs), namely the Zn salt ratio in mixed-cation systems [108]. By systematically tuning the mole ratio of $\text{Zn}(\text{TFSI})_2$ to LiTFSI while maintaining a low water content, they constructed a series of WiSEs with varying Zn contents. As shown

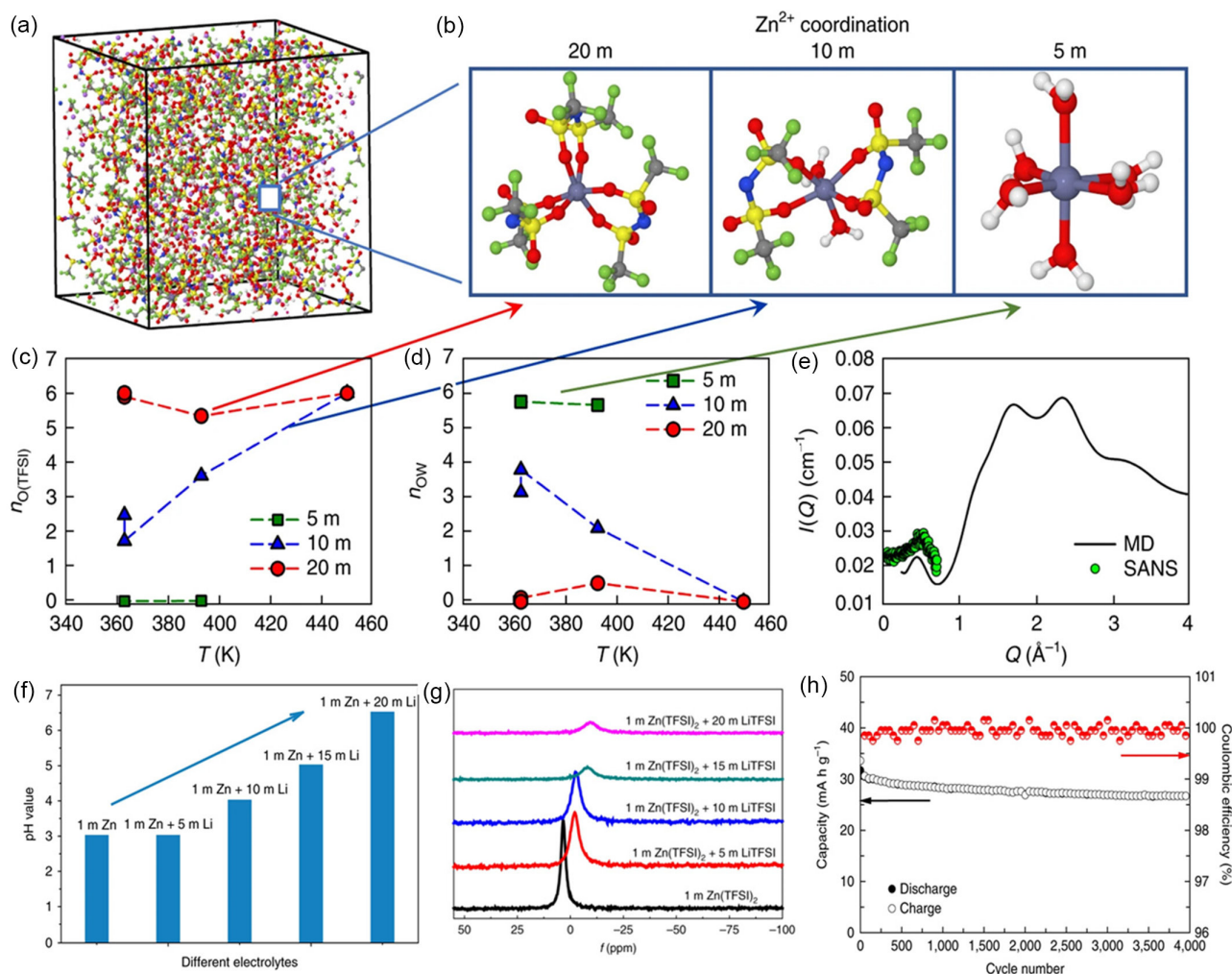


FIGURE 5 | (a) Snapshot of the MD simulation cell for HCZE (1 m Zn(TFSI)₂ + 20 m LiTFSI) [107]. (b) Representative Zn²⁺ solvation structures in the electrolytes with 1 m Zn(TFSI)₂ and three concentrations of LiTFSI (5, 10, and 20 m) [107]. (c,d) Zn²⁺-O(TFSI) [107] (c) and Zn²⁺-O(water) (d) coordination numbers for aqueous electrolytes with 1 m Zn(TFSI)₂ and three concentrations of LiTFSI (5, 10, and 20 m). (e) The experimental SANS curve (green circles) and the simulated form at 298 K for 1 m Zn(TFSI)₂ + 20 m LiTFSI electrolyte in D₂O [107]. (f) The pH values of the electrolytes with varying LiTFSI concentrations [107]. (g) The change with salt concentration of chemical shifts for ¹⁷O nuclei in solvent (water) [107]. (h) The cycling stability and CE of the Zn/LiMn₂O₄ full cell in HCZE [107]. Reproduced with permission. Copyright 2025, Springer Nature.

in Figure 6i, increasing the Zn ratio markedly enhances the Zn²⁺ transference number, indicating a greater contribution of active Zn²⁺ ions to interfacial transport. This effect is closely linked to the nucleation and diffusion process of Zn during deposition. In low Zn ratio electrolytes, Zn deposition is dominated by two dimensional diffusion, which tends to cause ion accumulation and localized growth (Figure 6g). By contrast, high Zn ratio electrolytes promote a three dimensional diffusion mode (Figure 6h), which is more conducive to uniform Zn deposition. The corresponding electrochemical response is shown in Figure 6f. With increasing Zn ratio, the nucleation overpotential is significantly reduced, suggesting facilitated Zn nucleation kinetics and alleviated concentration polarization. Consequently, the high Zn content electrolyte exhibits much better Zn reversibility and cycling stability than the low Zn content counterparts. These results demonstrate that, in addition to reducing water activity, rational optimization of the Zn salt ratio is also crucial for improving ion transport, modulating Zn deposition behavior, and stabilizing water-in-salt electrolyte systems.

From the solvation–desolvation viewpoint, high concentration electrolytes mainly shift Zn²⁺ coordination from a water dominated solvation shell to an anion-rich or ion clustered structure. This reconstruction reduces the number of active water molecules around Zn²⁺, lowers the probability of water being delivered to the Zn surface, and promotes more stable interfacial desolvation and deposition kinetics.

Despite these advantages, highly concentrated electrolytes still face several practical limitations. Their large salt consumption inevitably increases material cost, especially when expensive fluorinated salts such as Zn(TFSI)₂ or LiTFSI are used. The high salt concentration also increases electrolyte viscosity, which can slow ion transport, reduce electrolyte wettability, and aggravate polarization under high rate or low-temperature conditions. In addition, concentrated electrolytes may show limited compatibility with thick electrodes and lean electrolyte cell configurations because sufficient electrolyte infiltration becomes more difficult. Therefore, although highly concentrated electrolytes are effective

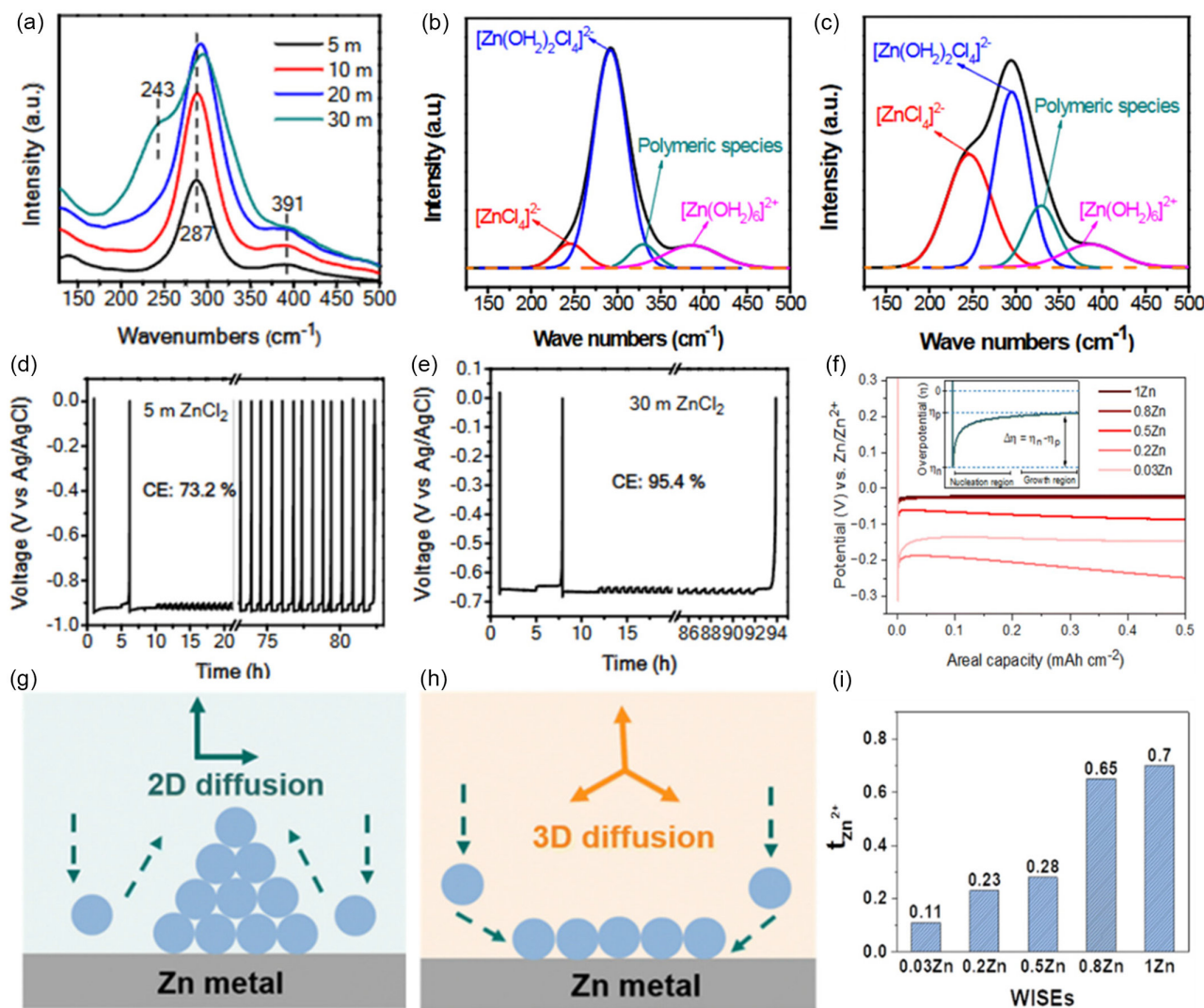


FIGURE 6 | (a) Raman spectra of ZnCl_2 solutions of different concentrations. Raman spectra of ZnCl_2 electrolytes with the concentrations of (b) 20 and (c) 30 m [106]. CE measurements of Zn plating/stripping in asymmetric Zn||Zn cells in (d) 5 m and (e) 30 m ZnCl_2 electrolytes [106]. Reproduced with permission. Copyright 2025, Royal Society of Chemistry. (f) Galvanostatic Zn electrode position on Ti in Zn||Ti half cells at 1 mA cm^{-2} and 0.5 mAh cm^{-2} using different WiSEs [106]. Schematics showing (g) 2D and (h) 3D diffusion processes [108]. (i) The transference number in different electrolytes [108]. Reproduced with permission. Copyright 2025, Royal Society of Chemistry.

in regulating water activity and Zn^{2+} solvation, their cost, viscosity, transport kinetics, and scalability should be carefully evaluated for practical AZIB applications.

4.2.4 | Other Solute Systems

In addition to conventional sulfate and chloride-based electrolytes, other types of zinc salts have also been explored to further regulate solvation and interfacial behavior in AZIBs. These alternative zinc salts are not only ion sources, but may also provide additional chemical functionalities through their intrinsic anion structures. In particular, biomass derived organic zinc salts and fluorinated organic zinc salts have attracted increasing attention. Their distinct coordination characteristics can influence Zn^{2+} solvation environments, interfacial reactions, and Zn deposition behavior, thereby offering new opportunities for improving battery reversibility and stability.

Xu et al. explored a biomass derived organic zinc salt as an alternative electrolyte and developed a dual functional system using zinc gluconate (Zn Glu) as the sole solute, without introducing any additional additives [109]. This design takes advantage of the hydroxyl and carboxyl rich structure of the gluconate anion, which can simultaneously regulate Zn^{2+} solvation and interfacial deposition behavior. As shown in Figure 7a, molecular dynamics simulations reveal that the Zn-Glu electrolyte structure evolves with concentration, indicating that the local coordination environment of Zn^{2+} is effectively tuned by gluconate species. Based on this solvation regulation, the authors proposed that Glu anions could adsorb preferentially on the Zn surface and form a molecular shielding layer. Such an interfacial layer helps suppress Zn^{2+} accumulation at high energy tip regions through electrostatic repulsion and steric hindrance. The corresponding deposition process is illustrated in Figure 7b,c. At lower zinc gluconate concentration, Zn deposition remains relatively uneven, whereas a higher concentration of Zn Glu promotes a much smoother and

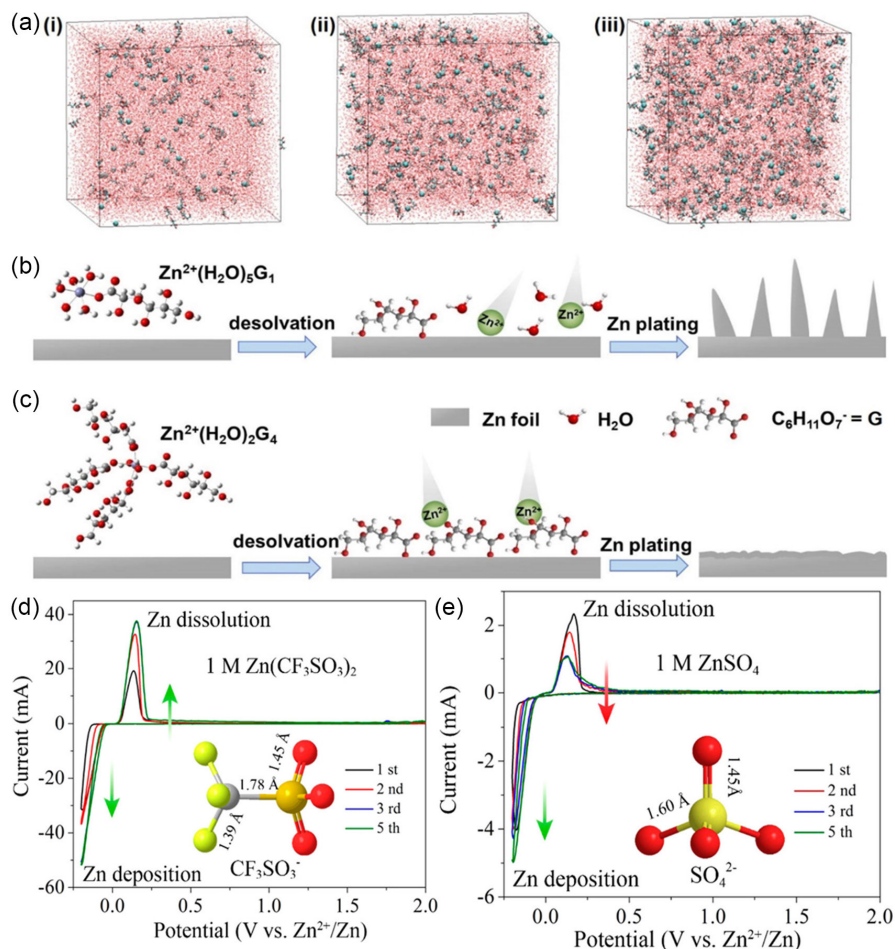


FIGURE 7 | (a) 3D snapshot of zinc gluconate system obtained from MD simulations: (i) 0.1, (ii) 0.2, and (iii) 0.3 m [109]. Comparison of Zn plating process in (b) 0.1 and (c) 0.3 m zinc gluconate electrolyte [109]. Reproduced with permission. Copyright 2023, Elsevier. Cyclic voltammograms of Zn electrode in aqueous electrolyte of (d) 1 m $\text{Zn}(\text{CF}_3\text{SO}_3)_2$ and (e) 1 m ZnSO_4 at the scan rate of 0.5 mV s^{-1} between -0.2 and 2.0 V [110]. Reproduced with permission. Copyright 2024, American Chemical Society.

more uniform deposition morphology. Consequently, the reversibility of the Zn anode is significantly improved. The symmetric cell with Zn Glu electrolyte achieved stable cycling for 500h, which was much longer than that of the ZnSO_4 -based control system. This work demonstrates that biomass derived zinc salts can function not only as charge carriers, but also as intrinsic solvation and interfacial regulators, thereby offering an attractive strategy for stabilizing Zn anodes in AZIBs.

Zhang et al. reported a high-performance aqueous electrolyte using zinc trifluoromethanesulfonate ($\text{Zn}(\text{CF}_3\text{SO}_3)_2$) as the sole zinc salt [110]. This electrolyte design takes advantage of the bulky and weakly coordinating nature of the CF_3SO_3^- anion. Compared with conventional ZnSO_4 -based systems, such an anion environment can weaken the strong electrostatic interaction around Zn^{2+} and facilitate its interfacial reaction kinetics. It was proposed that the large CF_3SO_3^- anions reduce the number of water molecules coordinated to Zn^{2+} , thereby lowering the desolvation barrier and promoting faster Zn^{2+} transport. The electrochemical behavior shown in Figure 7d,e supports this effect. In the $\text{Zn}(\text{CF}_3\text{SO}_3)_2$ electrolyte, the Zn electrode exhibits more favorable deposition/dissolution characteristics and larger current response (Figure 7d), whereas the ZnSO_4 electrolyte shows relatively sluggish behavior

under similar conditions (Figure 7e). This difference indicates that the fluorinated organic anion can effectively improve Zn reaction kinetics. In addition, the modified solvation environment was reported to suppress the disproportionation of manganese-based cathodes, which is beneficial for improving cathode stability. These results suggest that fluorinated organic zinc salts can serve not only as Zn^{2+} sources, but also as effective regulators of solvation chemistry and electrode reaction behavior in AZIBs.

4.3 | Functional Additives

The core development trend of AZIBs electrolytes lies in controlling water activity and optimizing ion transport [23, 24]. From the early pure inorganic salt ($\text{ZnSO}_4/\text{ZnCl}_2$) systems to the later water-in-salt and DES systems, electrolyte stability has been greatly improved. However, practical issues such as material cost, viscosity, ion transport, and scalable preparation should also be considered. In comparison, the “base electrolyte + trace multifunctional additives” strategy combines low cost, high ionic conductivity, and excellent interfacial stability, making it currently the most balanced and promising technical route for advancing AZIBs toward practical applications.

Different from solvent or salt regulation, functional additives often work at low concentrations but exert strong effects at the electrode/electrolyte interface. Some additives participate in Zn^{2+} coordination and weaken the original hydration shell, while others preferentially adsorb on the Zn surface and regulate local electric-field distribution or nucleation sites. Therefore, additive engineering can couple solvation shell modification with interfacial nucleation control, providing a direct route to homogeneous Zn deposition.

4.3.1 | Cationic Electrolyte Additives

Cationic additives, especially metal cations, can regulate Zn deposition behavior mainly through electrostatic shielding and competitive adsorption effects. Because these cations are not readily reduced under the operating conditions of AZIBs, they tend to accumulate preferentially near the Zn anode/electrolyte interface, particularly at high energy protrusions or tip regions where the local electric-field is strongest. This interfacial enrichment forms a positively charged shielding layer, which can redistribute electric-field and repel further accumulation of Zn^{2+} at these sites. As a result, Zn^{2+} deposition is suppressed at tip regions and becomes more uniform over the electrode surface. In addition, some cationic additives can compete with Zn^{2+} for interfacial adsorption sites, thereby reducing localized Zn nucleation and mitigating dendritic growth. Through these combined effects, cationic additives help homogenize Zn-ion flux, stabilize interfacial deposition behavior, and improve the reversibility of Zn plating/stripping.

For instance, Liu et al. introduced a Sodium Citrate (SC) additive into a 2 m $ZnSO_4$ electrolyte to regulate Zn deposition behavior through a combined effect of electrostatic shielding and solvation modulation [111]. In this system, Na^+ and citrate species play different but complementary roles. As schematically illustrated in Figure 8a, the blank $ZnSO_4$ electrolyte tends to induce severe side reactions, by-product formation, and dendritic Zn growth on the Zn anode. After adding SC, interfacial chemistry changes markedly (Figure 8b). The Na^+ cations can accumulate preferentially near high-energy protrusions on the Zn surface and form a positively charged shielding layer. This layer helps redistribute the local electric-field and suppresses further Zn^{2+} enrichment at tip regions, thereby promoting more uniform Zn deposition. Meanwhile, the citrate anion, which contains abundant functional groups, can participate in modulating the Zn^{2+} solvation environment and weaken water activity. Such a synergistic effect reduces parasitic reactions and facilitates more stable interfacial deposition. Consequently, the optimized electrolyte enables the Zn||Zn symmetric cell to cycle stably for over 760 h at 1 mA cm^{-2} . The assembled Zn||NVO full cell maintains a capacity retention of 40.0% after 400 cycles at 1 A g^{-1} (significantly better than the 23.8% of the blank group), demonstrating the effectiveness of this strategy.

Cao et al. introduced trace amounts of tetramethylammonium sulfate (TMA_2SO_4) into an aqueous $ZnSO_4$ electrolyte to regulate Zn deposition behavior and improve interfacial stability [112]. In this system, the TMA^+ cations were proposed to play a key role through electrostatic shielding and deposition-orientation regulation. As illustrated in Figure 8c, TMA^+ tends to adsorb

preferentially at high-energy protrusions on the Zn surface. This interfacial adsorption forms a positively charged shielding layer, which suppresses localized Zn^{2+} accumulation at tip regions and promotes deposition in relatively recessed areas. As a result, Zn growth becomes more uniform and the electrode surface is gradually smoothed. The corresponding morphological comparison is shown in Figure 8d–f. The bare Zn foil exhibits its initial surface state (Figure 8d). After soaking in 2 m $ZnSO_4$, Zn surface becomes much rougher and shows obvious corrosion-related features, as confirmed by the SEM image and elemental mapping in Figure 8e. By contrast, after soaking in the $ZnSO_4$ electrolyte containing TMA_2SO_4 , Zn surface remains much smoother and more uniform (Figure 8f), indicating that the additive effectively suppresses interfacial deterioration. In addition, the adsorption of TMA^+ was reported to guide lateral Zn growth along the foil surface, which helps inhibit vertical dendrite propagation. Owing to these effects, the modified electrolyte significantly enhances battery stability. The symmetric cells exhibited a long cycling life of over 1800 h, and Zn|| MnO_2 full cells retained 98.72% of capacity after 200 cycles. Therefore, this work further demonstrates that cationic electrolyte additives can stabilize Zn anode by simultaneously regulating local electric-field distribution and Zn deposition behavior.

Chen et al. introduced a trace amount of ammonia monohydrate ($NH_3 \cdot H_2O$) into conventional $ZnSO_4$ electrolyte to regulate Zn anode through simultaneous pH adjustment and cation modulation [113]. This additive slightly increases the electrolyte pH from 4.1 to 5.2 and introduces NH_4^+ species into the interfacial environment. As illustrated in Figure 8g, in the blank electrolyte with a lower pH, severe parasitic reactions occur during cycling. HER is easily triggered, the Zn surface becomes corroded, and an uneven zinc hydroxy sulfate layer forms. These effects lead to interfacial heterogeneity, tip amplification, and eventually dendritic growth. After adding $NH_3 \cdot H_2O$, the interfacial chemistry changes significantly, as shown in Figure 8h. First, the increase in pH lowers the thermodynamic tendency for HER, which helps suppress water reduction at the Zn surface. Second, the modified electrolyte promotes the in situ formation of a more uniform and dense zinc hydroxy sulfate-based interphase. Such a layer can isolate Zn anode from direct water attack and facilitate more stable Zn^{2+} transport. Third, NH_4^+ is preferentially adsorbed near high-energy protrusions on the Zn surface. This adsorption redistributes local electric-field through a shielding effect and suppresses localized Zn growth. Therefore, the beneficial role of $NH_3 \cdot H_2O$ does not come from a single factor, but from the combined regulation of HER, interphase formation, and interfacial ion distribution. Owing to these synergistic effects, the symmetric cell achieved stable cycling for 1500 h and operated for 250 h at a 5 mA cm^{-2} . The Zn|| MnO_2 full cell likewise exhibited excellent rate capability and capacity retention.

4.3.2 | Anionic Electrolyte Additives

Anionic electrolyte additives have become an effective means of improving the performance of AZIBs [114, 115]. These additives can modify Zn deposition behavior, suppress side reactions, and improve interfacial stability, thereby enhancing cycling performance and extending battery lifespan. Recent years have witnessed substantial advances in anionic additives for high performance AZIBs.

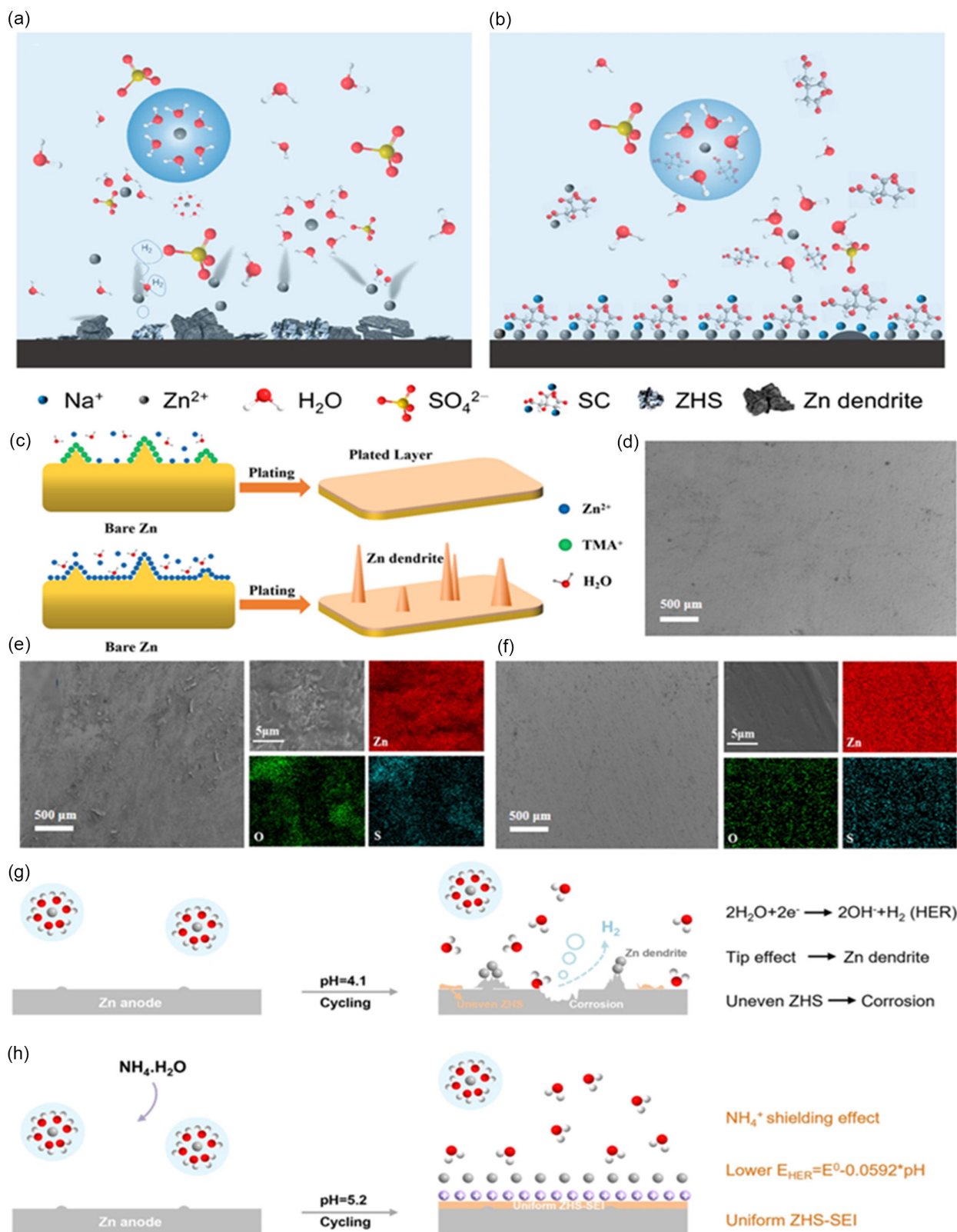


FIGURE 8 | Schematic illustration of electrochemical behaviors at the anode/electrolyte interface in different electrolyte systems: (a) pure ZnSO₄ electrolyte and (b) ZnSO₄ electrolyte containing sodium citrate additive [111]. Reproduced with permission. Copyright 2024, MDPI. (c) Schematic illustration of the stripping/plating of Zn metal in 2 m ZnSO₄ with/without electrolyte additives [112]. (d) SEM image of bare Zn foil [112]. (e) SEM image and EDS mapping of bare zinc foil after soaking in ZnSO₄ (2 m) for 10 days [112]. (f) SEM image and EDS mapping of bare zinc foil after soaking in ZnSO₄ (2 m) + TMA₂SO₄ for 10 days [112]. Reproduced with permission. Copyright 2022, Elsevier. Schematic illustration of the Zn plating process in (g) BE and (h) DE [113]. Reproduced with permission. Copyright 2023, Springer Nature.

Kim et al. introduced ZnBr_2 into a conventional 1 M ZnSO_4 electrolyte as an anionic additive to regulate Zn deposition behavior and suppress dendrite growth [116]. In the blank ZnSO_4 electrolyte, dendritic growth readily develops during cycling because of nonuniform ion distribution and local electric-field amplification, as schematically shown in Figure 9a. After the addition of ZnBr_2 , the interfacial behavior changes markedly. As illustrated in Figure 9b, the bromide containing electrolyte can effectively inhibit the formation of sharp Zn dendrites and promote a much smoother deposition morphology. The underlying mechanism is summarized in Figure 9c. Br^- anions were proposed to exhibit a chemical polishing effect by selectively reacting with high energy protrusions on the Zn surface. The dendrite tips are more likely to be etched than the flatter regions because of their higher surface energy and stronger local reactivity. This selective dissolution smooths the rough Zn surface, weakens tip amplification, and helps homogenize the interfacial electric-field. As a result, the subsequent Zn deposition becomes more uniform, and the risk of dendrite induced short circuits is significantly reduced. Owing

to this dynamic interfacial regulation, the electrolyte containing 10 wt% ZnBr_2 enabled symmetric cells to cycle stably for 2000 h, demonstrating markedly improved long-term Zn anode stability. This work shows that anionic additives can stabilize Zn anodes not only by modifying solvation chemistry, but also by directly regulating surface evolution through selective interfacial reactions.

Lin et al. employed 3-mercapto-1-propanesulfonic acid sodium (MPS) as an additive to regulate Zn deposition at the molecular level [117]. MPS contains both sulfonate and mercapto functional groups. This bifunctional structure allows it to interact with the Zn anode surface and influence Zn^{2+} interfacial behavior. According to the theoretical analysis shown in Figure 9d, the MPS anion exhibits stronger adsorption on the Zn (100) and (101) crystal planes than on the (002) plane. Such preferential adsorption can selectively occupy high-energy deposition sites and thus alter the exposed crystallographic orientation of the Zn surface. The relative growth of the Zn (002) plane is promoted. Figure 9e further shows that, in the presence of MPS, the adsorption energy

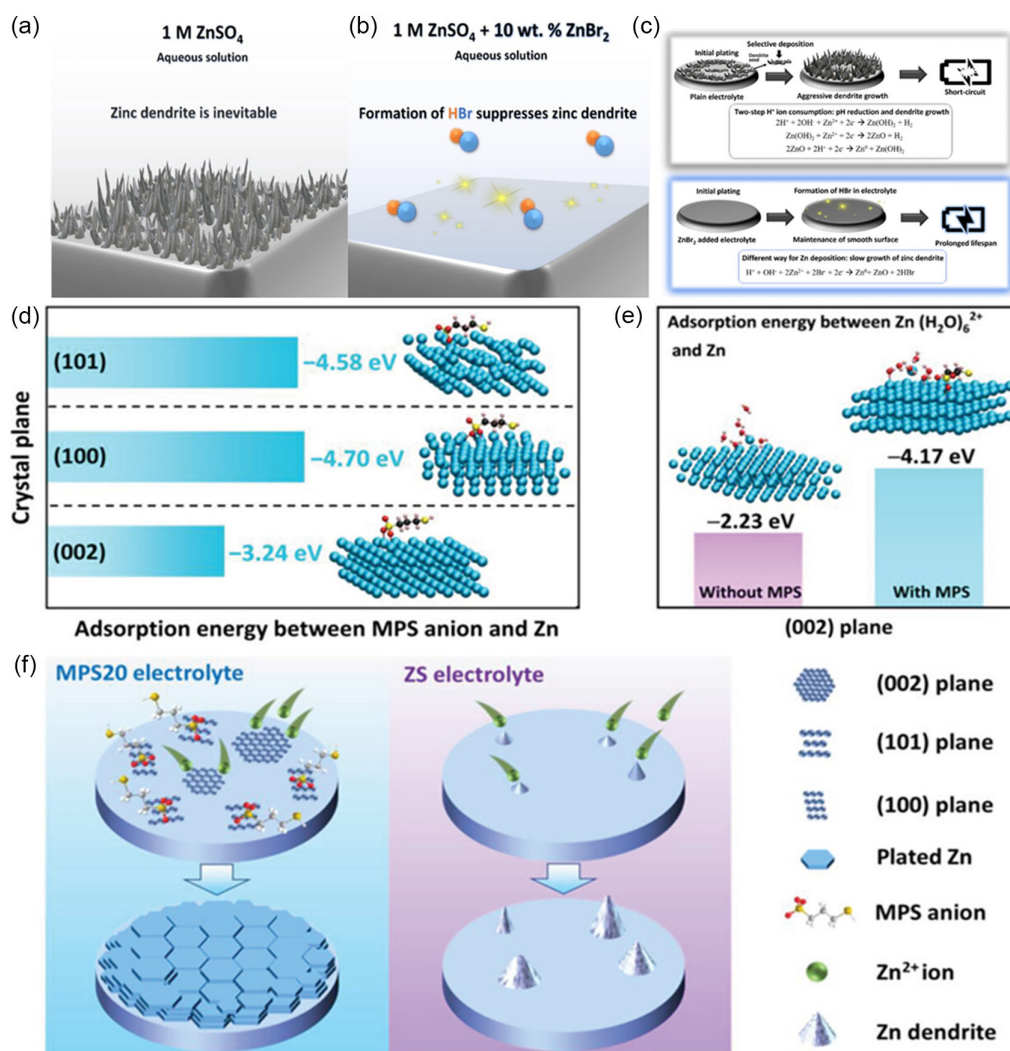


FIGURE 9 | Schematic illustration of without electrolyte additives (a) and with additives (b). (c) Schematic illustration summarizing the reaction mechanism of ZnBr_2 electrolyte additive on effectiveness of zinc dendrite suppression [116]. Reproduced with permission. Copyright 2025, Elsevier. (d) The adsorption energy between MPS anions and different crystal planes of Zn anode [117]. (e) The adsorption energy between zinc hexahydrate ion and (002) plane with or without MPS [117]. (f) The schematic illustration of the Zn electrodeposition process with or without MPS additive [117]. Reproduced with permission. Copyright 2023, Wiley-VCH.

between hydrated Zn^{2+} species and the Zn (002) plane becomes much stronger. This indicates that the additive not only modifies the surface structure, but also facilitates the subsequent interfacial adsorption of Zn^{2+} on the preferred crystal plane. The overall effect is illustrated in Figure 9f. In the MPS containing electrolyte, Zn deposition tends to proceed in a more ordered and planar manner, leading to a compact (002) textured Zn layer. By contrast, the blank electrolyte favors localized tip growth and dendrite formation. Therefore, the role of MPS is not simply to block side reactions, but to actively redirect Zn nucleation and growth through facet selective interfacial adsorption. Owing to this regulation, the cycling lifespan of the Zn||Zn symmetric cell increased from 130 h to 4500 h at 1 mA cm^{-2} , and the system maintained excellent stability at 50%–80% depth of discharge. These findings suggest that anionic additives are effective in stabilizing Zn anodes by coupling interfacial adsorption chemistry with crystallographic growth regulation.

4.3.3 | Organic Molecular Additives

Organic molecules electrolyte additives have become an efficient strategy to inhibit zinc dendrite growth and side reactions owing to their abundant functional groups and structural diversity. Unlike simple inorganic ions, these molecules can interact strongly with water, Zn^{2+} , and the Zn surface at the same time. As a result, they are particularly useful for reducing side reactions caused by water, guiding Zn deposition, and improving interfacial stability.

Wang et al. reported a representative case in which 1,3,6-hexanetricarbonitrile (HTCN-*x*) was introduced into $\text{Zn}(\text{OTf})_2$ electrolytes [118]. The strong interaction between HTCN and H_2O lowers the amount of highly reactive solvated water molecules outside the Zn^{2+} solvation sheath. It helps suppress side reactions at the Zn interface. Meanwhile, HTCN was found to promote the formation of a strong and stable interphase on the Zn anode, which further inhibits dendrite growth and corrosion. Therefore, the beneficial role of HTCN originates from the combined regulation of water activity and interfacial protection, rather than from a single mechanism. Owing to these effects, the symmetric cell cycled stably for 950 h at 1 mA cm^{-2} . The Zn||ZVO full cell also delivered an initial capacity of 355.6 mAh g^{-1} at 0.1 A g^{-1} and retained 330 mAh g^{-1} after 465 cycles at 1 A g^{-1} .

Building on this idea, Xia et al. further emphasized the importance of molecular coordination strength in electrolyte design [119]. They introduced diethylenetriaminepentaacetic acid pentasodium salt as an additive to dynamically regulate the anode electrolyte interface. Different from additives that mainly reduce water activity, this molecule was designed to tune Zn^{2+} interfacial behavior through a chelation-strength effect. Such regulation helps suppress water-related side reactions and reduce dendritic growth during repeated plating/stripping. The additive works by balancing interfacial coordination and Zn deposition kinetics, thereby stabilizing the Zn anode over long-term cycling. As a result, the modified electrolyte enabled the symmetric cell to operate stably for more than 3500 h at 1 mA cm^{-2} . The Zn|| $\text{NH}_4\text{V}_4\text{O}_{10}$ full cell also retained 84.6% of its capacity after 500 cycles at 1 A g^{-1} .

Taken together, these studies show that organic molecular additives can stabilize AZIBs through multiple complementary pathways, including water activity regulation, interphase construction, and coordination-mediated interfacial control. This makes them a highly versatile class of electrolyte additives for improving Zn reversibility and long-term cycling durability.

4.3.4 | Other Novel Functional Additives

In addition to conventional ions and organic molecules, there has been widespread interest in developing novel multifunctional electrolyte additives such as biomass derivatives and two-dimensional nanomaterials. For example, lignin derivatives, with their abundant polar functional groups, can be strategically introduced into electrolytes as cationic surfactants. These macromolecular additives can restructure the Zn^{2+} solvation shell and preferentially adsorb onto the anode surface to form a protective layer, effectively eliminating dendrites. Graphene oxide (GO), as a two-dimensional additive, self-assembles on the zinc surface through electrostatic interactions to form an “ion rectifying shield”. This structure possesses high mechanical strength and excellent ion sieving capabilities, and it can also induce preferential zinc growth along the (002) crystal plane by regulating interface electric-field distribution and ion flux.

Xu et al. synthesized quaternized lignin (QL(80)) from abundant, low cost kraft lignin as an electrolyte additive for Zn|| α - MnO_2 batteries [120]. 1 wt% QL(80) minimized Zn corrosion and promoted uniform deposition, yielding the highest open-circuit voltage after full charge storage and optimal rate performance. Zn||Zn symmetric batteries cycled 200 h at 5 mA cm^{-2} .

Wang et al. adopted a strategy of using two-dimensional nanomaterials as electrolyte additives, introducing a trace amount of graphene oxide (GO) into the conventional 2 M ZnSO_4 electrolyte [121]. This novel composite electrolyte structure leverages GO's amphiphilic and negatively charged properties to solve the issues of disordered dendrite growth caused by nonuniform electric-field distribution on the Zn anode surface and hydrogen evolution corrosion triggered by direct contact with water molecules. Taking advantage of the reductive nature of zinc metal, GO is partially reduced to rGO upon contact with the anode and assembles into a conductive two-dimensional protective layer. The two-dimensional planar structure of the rGO layer restricts vertical diffusion of Zn^{2+} ions, forcing them to migrate horizontally, thereby homogenizing the ion flux. The results suggest that GO-added system significantly reduces the nucleation overpotential, enabling symmetric cells to achieve an ultralong cycling lifespan of over 2500 h (compared to $\approx 100 \text{ h}$ for the blank group) and demonstrating higher capacity retention in full-cell tests.

Nevertheless, additive-based strategies also have limitations. Because most additives are used at low concentrations, their effectiveness can be sensitive to dosage, electrolyte composition, and cycling conditions. Some additives may be gradually consumed, decomposed, or incorporated into interfacial by-products during long-term cycling, which can weaken their protective effect. In addition, excessive additive concentration may reduce ionic conductivity, increase viscosity, or introduce unexpected side reactions. Therefore, future additive design should consider

not only molecular functionality, but also long-term stability, concentration tolerance, compatibility with cathode materials, and practical electrolyte formulation.

4.4 | Composite Electrolyte Design

With the increasing demand for higher safety and energy density of AZIBs, electrolyte systems are gradually evolving from conventional liquid electrolytes toward quasi-solid-state and even all-solid-state configurations. The core principle of composite electrolyte design is to achieve an optimal balance between ionic conductivity and mechanical robustness. By integrating polymer frameworks with functional additives or inorganic components, composite electrolytes can simultaneously regulate ion transport, stabilize electrode/electrolyte interfaces, and suppress parasitic reactions. Recent separator/electrolyte interface designs, such as boron-integrated cellulose nanofiber separators with dynamic Zn^{2+} coordinating oxygen sites, further demonstrate that polymer-based frameworks can regulate interfacial electric-field distribution and Zn^{2+} flux, thereby improving Zn deposition stability [122]. In recent years, various composite electrolyte systems have been proposed, generally including quasi-solid hydrogel electrolytes, organic–inorganic hybrid electrolytes, and all-solid composite electrolytes. Hydrogel-based systems utilize polymer networks to confine water molecules and construct continuous Zn^{2+} transport pathways, thereby improving mechanical stability and mitigating dendrite penetration. Organic–inorganic composite electrolytes introduce inorganic fillers or frameworks to enhance mechanical strength, regulate Zn^{2+} solvation structure, and homogenize ion flux. Meanwhile, all-solid composite electrolytes aim to eliminate free water and further improve safety and temperature tolerance while maintaining reasonable ionic conductivity. These emerging composite electrolyte architectures have demonstrated promising potential in addressing critical challenges in AZIBs, including electrolyte leakage, Zn dendrite growth, hydrogen evolution, and unstable interfacial reactions, thereby enabling improved cycling stability and operational safety. Consequently, composite electrolyte engineering has become an important direction for advancing practical AZIB technologies.

Within the solvation–desolvation–deposition framework, composite electrolytes regulate Zn deposition mainly by confining free water, constructing continuous Zn^{2+} transport pathways, and homogenizing interfacial ion flux. Polymer chains and inorganic fillers can also interact with Zn^{2+} or anions, thereby influencing local desolvation behavior and suppressing concentrated Zn^{2+} accumulation at high energy sites.

Nevertheless, composite electrolytes also face several challenges in practical implementation. Polymer networks can improve mechanical stability and suppress electrolyte leakage, but excessive cross-linking or overly dense gel structures may decrease ionic conductivity and slow Zn^{2+} migration. The incorporation of inorganic fillers can enhance mechanical strength and homogenize ion flux, yet poor filler dispersion or weak polymer–filler compatibility may generate interfacial defects and uneven ion transport pathways. Moreover, the interfacial contact between solid or quasi-solid electrolytes and electrodes may degrade during repeated cycling, particularly under high areal capacity or

flexible deformation conditions. Therefore, composite electrolyte design should further balance mechanical robustness, ionic conductivity, electrode compatibility, and scalable fabrication.

4.4.1 | Quasi-Solid Hydrogel Electrolytes

Hydrogel electrolytes are currently the most widely used composite systems, essentially consisting of a polymer three-dimensional network that confines liquid water/zinc salts. They mainly utilize highly hydrophilic polymers (such as PVA, PAM, cellulose) to construct the framework. Current research on hydrogel electrolytes mainly centers on the rational construction of polymer networks and the resulting regulation of interfacial and transport properties. One important direction is to build physically cross-linked networks, in which dynamic noncovalent interactions endow the electrolyte with flexibility, self-recovery capability, and structural adaptability during repeated deformation or cycling. Another common strategy is to introduce chemical cross-linking to reinforce the polymer framework and improve mechanical robustness, which is particularly important for resisting deformation and suppressing dendrite penetration. Beyond network construction itself, the functional groups on polymer chains also play a crucial role in electrolyte regulation. Their strong interactions with water molecules can effectively restrict free water activity and thereby mitigate HER and other water-induced side reactions. Meanwhile, these polar sites can interact with Zn^{2+} and provide continuous ion transport pathways within the polymer matrix. In this way, hydrogel electrolytes are able to combine intimate interfacial contact with efficient ionic conduction, making them promising platforms for stabilizing Zn electrochemistry in AZIBs.

Following the general design principle of hydrogel electrolytes, Zhang et al. developed a biomass-derived gel electrolyte by introducing sodium carboxymethyl cellulose into an aqueous ZnSO_4 solution [123]. They introduced carboxymethyl cellulose sodium (CMC) as a gelling agent into an aqueous zinc sulfate solution to design a CMC-based reference solid-state hydrogel electrolyte. This structure possesses mechanical stability, addressing the challenges of traditional liquid electrolytes, which are difficult to encapsulate and prone to leakage in batteries. CMC long-chain molecules form a dense three-dimensional network skeleton in the aqueous solution through hydrogen bonding and intermolecular interactions, effectively immobilizing free water molecules and ions. This not only prevents the electrolyte from leaking but also retains fast Zn^{2+} transport channels. Meanwhile, this gel layer acts as a flexible separator, providing mechanical cushioning when the battery is bent or twisted, ensuring close contact between electrodes and preventing short circuits. Thanks to the support of this gel electrolyte, the assembled coaxial fiber AZIB exhibits excellent mechanical flexibility and electrochemical stability. The battery achieves an ultrahigh volumetric energy density, and after 3,000 bending cycles, the capacity retention remains as high as 93.2%, demonstrating the practicality of this gel-based strategy. Building on this concept, later studies further explored hydrogel electrolytes with enhanced structural functionality and more sophisticated network design.

Building on the development of hydrogel electrolytes, Huang et al. further designed a supramolecular hydrogel electrolyte

based on polyvinyl alcohol (PVA) and $\text{Zn}(\text{CF}_3\text{SO}_3)_2$ through a physical cross-linking strategy [124]. This system was developed to address mechanical damage and structural failure in flexible AZIBs. In this hydrogel, the PVA chains are rich in hydroxyl groups and can form a dynamic physically cross-linked network through hydrogen bond interactions. As illustrated in Figure 10a, when the electrolyte is cut or fractured, the broken polymer chains can reconnect through the reorganization of interfacial hydrogen bonds. This dynamic reconstruction enables rapid self-healing of the hydrogel network without external stimulation. The electrolyte can recover both structural integrity and ionic conduction after mechanical damage. Benefiting from this self-healing behavior, the integrated $\text{Zn}||\text{MnO}_2$ battery showed full recovery of charge/discharge performance after being cut and rejoined. The device also maintained stable electrochemical output under repeated bending and twisting. These results indicate that physically cross-linked supramolecular hydrogels can effectively combine mechanical flexibility with structural recoverability, thus offering a promising electrolyte design for durable flexible AZIBs devices. While self-healing hydrogels improve the tolerance of AZIBs to mechanical damage, further optimization of the polymer network is still required to balance mechanical robustness, electrolyte uptake, and ion transport.

Building on earlier hydrogel designs that mainly addressed electrolyte leakage, flexibility, and self-recovery, Yu et al. further improved the polymer network (IPN) architecture by constructing an interpenetrating polymer network hydrogel membrane, denoted as M-PCMIH [125]. This material was prepared through a rapid dual cross-linking strategy that combines covalent and non-covalent interactions. As illustrated in Figure 10b, a homogeneous dispersion containing PVA and carboxymethyl chitosan (CMCS) was cross-linked in the presence of glutaraldehyde under

acidic conditions, leading to the formation of an IPN hydrogel. In this structure, the PVA component forms one network through covalent acetal and hemiacetal linkages, while the CMCS component builds the second network through ionic and hydrogen bond interactions. The corresponding molecular interactions are further illustrated in Figure 10c, which shows that the coexistence of multiple hydrophilic and interactive sites provides a stable and highly absorbent polymer framework. Consequently, the hydrogel membrane can retain a large amount of electrolyte while maintaining structural integrity. Figure 10d further presents the integration concept of this membrane in a flexible energy storage device, highlighting its suitability as a functional gel electrolyte platform. Compared with earlier hydrogel systems that mainly emphasized mechanical adaptability, this work emphasizes the significance of network engineering. Such a design provides a better balance between electrolyte uptake, mechanical robustness, and ion-transport continuity, and therefore offers an important basis for the subsequent development of more multifunctional hydrogel electrolytes for AZIBs. This work shifted the focus of hydrogel electrolyte design from simple functional introduction to more refined polymer network engineering, thereby providing a structural basis for later multifunctional hydrogel systems.

Building on previous hydrogel designs that emphasized flexibility, self-healing, and network engineering, Ling et al. further developed a highly stretchable, self-healing hydrogel electrolyte (SHE) for stabilizing Zn electrochemistry in flexible mild AZIBs [126]. This electrolyte was constructed from a dual cross-linked agarose/polyacrylamide (PAM) network reinforced with carboxymethyl cellulose (CMS). As illustrated in Figure 10e, the SHE is formed through a stepwise network-construction process, which gives the hydrogel both structural robustness and damage tolerance. The corresponding self-healing mechanism is shown

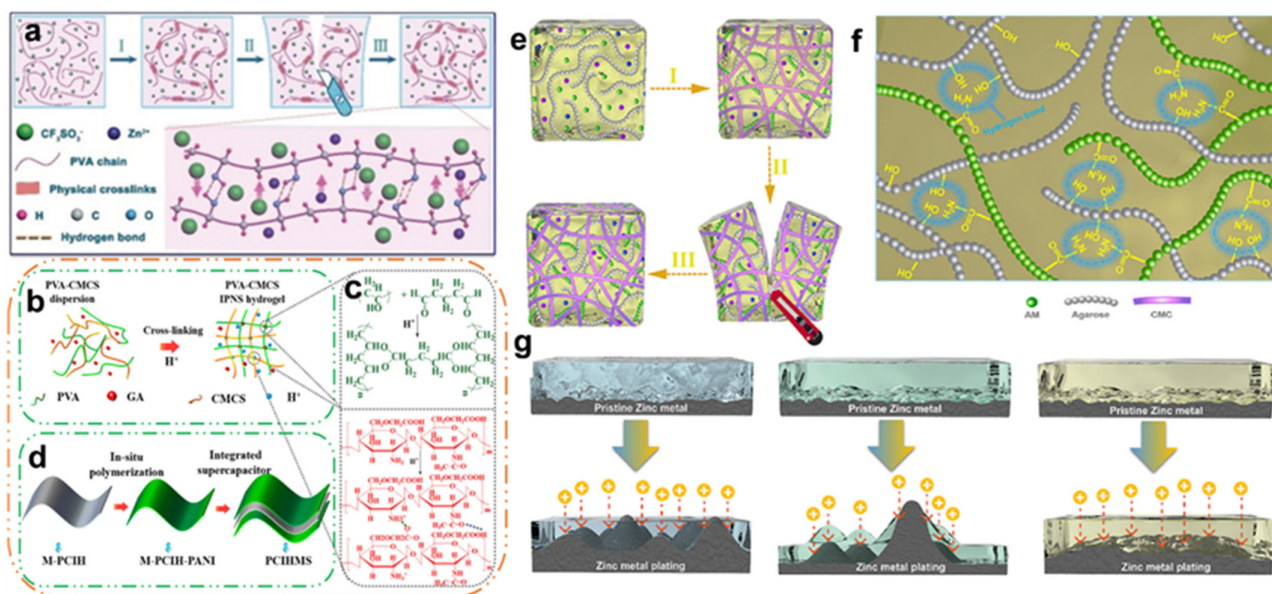


FIGURE 10 | (a) Process of fabricating self-healing electrolyte and its self-healing illustration. I freezing 15 h at -18°C and thawing 24 h at room temperature; II cutting; III self-healing [124]. Reproduced with permission. Copyright 2019, Wiley-VCH. (b,c) the interpenetrating polymeric networks of bi-cross-linked hydrogel macromolecules membrane, and (d) the assembled process [125]. Reproduced with permission. Copyright 2021, Elsevier. (e) Composition of SHEs and their self-healing illustration [126]. (f) The origin of self-healability (hydrogen bond) [126]. (g) Schematic illustrations of morphology evolution for Zn foils with LEs (i), pure PAMs (ii) and SHEs (iii) during Zn stripping/plating cycling [126]. Reproduced with permission. Copyright 2021, Elsevier.

in Figure 10f. Multiple hydrogen bond interactions among agarose, PAM, and CMC enable the fractured network to reconnect dynamically after mechanical damage. In this way, the electrolyte can recover its structural integrity while maintaining ionic transport pathways. Beyond mechanical recovery, the SHE also plays a key role in modulating the Zn deposition process. As shown in Figure 10g, compared with liquid electrolytes and pure PAM hydrogel, the SHE promotes a much more uniform Zn ion flux and deposition process on the Zn metal surface. This effect can be attributed to its porous network structure and more homogeneous interfacial environment, which help reduce local current concentration and suppress irregular Zn growth. Zn symmetric cells using the SHE exhibit markedly improved cycling stability for 300 h at 5 mA cm^{-2} , together with a low voltage hysteresis of about 80 mV. These results indicate that multifunctional hydrogel electrolytes can simultaneously improve mechanical durability, self-healing capability, and Zn deposition stability, and therefore represent an important direction for advanced flexible AZIB electrolytes.

4.4.2 | Organic–Inorganic Composite Electrolytes

To address the issues of low mechanical modulus in pure hydrogel and its difficulty in physically blocking zinc dendrite penetration, inorganic fillers are introduced for reinforcement, mainly by in situ doping or blending inorganic nanoparticles, two-dimensional materials (e.g., MXene and GO), or porous materials (e.g., MOFs) into the polymer matrix [127, 128]. Inorganic fillers can act as a “skeleton,” significantly increasing the Young’s modulus and physically hindering dendrite growth; the acidic/basic sites on their surfaces can anchor SO_4^{2-} , improving Zn^{2+} transference number. Additionally, two-dimensional fillers can construct directional channels to homogenize ion flux.

Liu et al. adopted the ionotronic dual-network design strategy, using one-dimensional cellulose nanofibers (CNF) as cross-linkers and scaffolds, assembled with two-dimensional MXene nanosheets to create a composite hydrogel (MXene–CNF) with ultrahigh toughness [129]. This novel electrolyte interface structure exhibits mechanical properties that combine rigidity and flexibility, as well as hybrid conductivity, while also possessing excellent zinc affinity. The design aims to address dendrite growth on the zinc anode caused by uneven stress distribution and volume expansion, as well as the side HER triggered by high interfacial reactivity. The tough CNF network can effectively withstand and constrain the stress from zinc deposition-induced volume expansion, physically suppressing the vertical growth of dendrites. The high conductivity of MXene, combined with zinc-affinity sites, homogenizes the electric-field and Zn^{2+} ion flux at the interface, reducing the nucleation barrier of zinc and achieving dense and smooth deposition. Experimental results show that this composite interfacial layer enables the full cell to maintain a high specific capacity even under high loading, significantly enhancing the device’s durability.

Xiong et al. employed a physical doping strategy, introducing inorganic fumed silica as a functional filler into a biomass hydrogel matrix to prepare a novel composite hydrogel electrolyte (CHE) [130]. This new electrolyte structure features high porosity and excellent ionic conductivity, while maintaining the good

mechanical flexibility of the hydrogel. The incorporation of fumed silica effectively optimizes the micro-pore structure of the gel and increases ion transport channels, thereby enhancing ionic conductivity. More importantly, as an inorganic filler, it significantly improves the interfacial stability between the zinc anode and the electrolyte. By regulating the ion concentration distribution at the interface, it promotes rapid and uniform deposition of Zn^{2+} , effectively suppressing the formation and accumulation of irreversible by-products, thereby alleviating electrode passivation and corrosion. Experiments show that when the fumed silica content is 10 wt%, the assembled Zn||MnO₂ battery exhibits optimal performance. Even at high current densities, the battery maintains 67% of its capacity after 1000 cycles (higher than 33% with pure gelatin electrolyte). Moreover, the battery can still provide stable power when bent, demonstrating excellent potential for flexible applications.

4.4.3 | All-Solid Composite Electrolytes

In order to completely prevent hydrogen evolution and corrosion caused by water, electrolytes are developing towards “ultralow free water” or even “crystalline” states, which is the form closest to all-solid-state electrolytes. Typically, eutectic hydrate melts at room temperature are solidified in polymers. In polymers, almost all water molecules participate in the coordination of Zn^{2+} , with no free water, significantly widening the electrochemical window. Ion transport no longer relies on diffusion through a water molecule medium but occurs through the motion of polymer chain segments.

Zhao et al. adopted a strategy combining nanochannel engineering with solvated structure regulation to design a novel plastic crystal electrolyte (PCE) with fast ion transport channels [131]. This new electrolyte structure features a unique layered bilayer architecture, combining liquid-like high ionic conductivity with solid-state mechanical strength. The design aims to address the bottleneck in conventional all-solid-state electrolytes, where Zn^{2+} migration is slow due to strong electrostatic trapping, while eliminating the risk of leakage associated with liquid electrolytes. It utilizes ion-dipole interactions between trace water molecules and amphiphilic anions to induce the directional self-assembly of anions into layered structures. In the interlayer region, water molecules and anions together form a unique heterocoordination environment. This coordination structure effectively releases Zn^{2+} ions captured by anions and weakens the Coulombic interactions between ions, thereby activating fast Zn^{2+} migration channels within the robust solid-state framework. Experimental results show that this plastic crystal electrolyte enables the zinc anode to achieve up to 99.6% Coulombic efficiency for stripping/deposition. The assembled symmetric cells and full cells both exhibit excellent long-term cycling stability, demonstrating their potential as a next-generation electrolyte for high-safety miniature batteries.

Xu et al. achieved in situ generation of an ultrathin layered solid electrolyte interface (SEI) through a dual reaction strategy: the organic-rich upper layer buffers mechanical stress, while the inorganic-rich inner layer guides uniform zinc deposition [132]. The Zn||Zn symmetric battery demonstrates stable cycling for 400 h at a high current density of 100 mA cm^{-2} , with an accumulated zinc plating capacity reaching 67.5 Ah cm^{-2} .

4.5 | Catalytic Interfacial Regulation

In addition to regulating electrolyte chemistry and constructing composite electrolyte systems, recent studies have also begun to directly optimize Zn interfacial kinetics through catalytic regulation. This emerging strategy goes beyond conventional electrolyte design because it targets the interfacial transformation of hydrated Zn species, including desolvation, charge transfer, and subsequent surface diffusion. Compared with electrolyte systems that mainly regulate the upstream solvation environment, catalytic interfacial regulation acts more directly on the dynamic processes occurring at or near the electrodelectrolyte interface. Therefore, it provides a more focused route for lowering desolvation barriers, homogenizing Zn^{2+} flux, and guiding Zn nucleation and lateral deposition.

Beyond static solvation structure regulation, interfacial desolvation should be understood as a dynamic process during Zn plating. When solvated Zn^{2+} migrates from the bulk electrolyte to the Helmholtz layer, its coordination environment, ion pair state, and surrounding water orientation can continuously evolve under the local electric field. This dynamic evolution affects the desolvation barrier, charge transfer resistance, nucleation overpotential, and subsequent Zn atom diffusion. Recent studies have shown that dual region interfacial modulation, dual Helmholtz plane engineering, and facet-selective deposition strategies can accelerate Zn^{2+} desolvation and guide more ordered Zn growth [133–135]. Therefore, electrolyte design should not only regulate the initial solvation sheath in the bulk electrolyte, but also control the time dependent interfacial pathway from solvated Zn^{2+} to deposited Zn atoms.

As illustrated in Figure 11a,b, Dong et al. developed a suspension-electrolyte engineering approach by introducing electron-delocalized CeO_{2-x} nanoparticles into a conventional aqueous Zn electrolyte [136]. In the traditional electrolyte, as shown in Figure 11a, hydrated $[\text{Zn}(\text{H}_2\text{O})_6]^{2+}$ species undergo sluggish desolvation near the Zn surface. This slow interfacial process leads to uneven Zn^{2+} flux, dendrite formation, corrosion, and HER. In contrast, after introducing CeO_{2-x} nanoparticles, the interfacial behavior changes markedly, as shown in Figure 11b. The catalytic particles help regulate the Zn^{2+} solvation structure and dynamically modulate the Helmholtz layer near the Zn surface. Meanwhile, the defect-rich CeO_{2-x} redistributes the local electric-field, which helps homogenize Zn^{2+} flux and guide more uniform Zn deposition. Therefore, both dendrite growth and HER can be effectively suppressed. Benefiting from these effects, the Zn anode delivered a high Coulombic efficiency of 99.92% over 1200 cycles, and the symmetric cell achieved a long lifespan exceeding 6500 h with a low overpotential of 34 mV at 0°C. The corresponding full cell also maintained a capacity retention of 96.75% at 1 A g^{-1} under -20°C , demonstrating that catalytic regulation at the electrolyte level is an effective strategy for improving low-temperature Zn metal battery performance.

Further advancing this concept, Zhang et al. constructed a MXene-immobilized atomic catalyst layer by anchoring isolated Fe atoms on defect-rich MXene (SAFe@MXene) [137]. In the unmodified system, Figure 11c indicates that hydrated $[\text{Zn}(\text{H}_2\text{O})_x]^{2+}$ species approach the Zn surface and undergo

sluggish interfacial desolvation. Under such conditions, active water can participate in parasitic reactions, leading to pronounced HER and disordered Zn deposition. After introducing the SAFe@MXene layer, the interfacial process becomes markedly different. As presented in Figure 11d, the catalytic layer accelerates the dissociation of hydrated Zn species and reduces the participation of active water in side reactions. At the same time, the porous and defect-rich MXene framework improves interfacial wettability and ion transport, which helps homogenize Zn^{2+} flux and guide more uniform lateral Zn growth. A similar diffusion modulator concept has been used in Li metal anodes, where single metal atoms anchored in an interfacial layer guide metal-ion/atom transport and suppress dendritic growth [139]. This analogy highlights the importance of coupling desolvation acceleration with surface diffusion regulation. Therefore, both HER and dendritic deposition can be effectively restrained. Benefiting from these combined effects, the modified Zn anode maintained long-term stability over 1000 h at 2 mA cm^{-2} and still exhibited 800 h of stable cycling at 0°C. The full cell also delivered nearly 100% capacity retention after 1000 cycles at 1 A g^{-1} , demonstrating the effectiveness of atomic-catalyst interfacial regulation for low-temperature Zn metal batteries.

More recently, Cheng et al. proposed a self-cascade catalytic strategy based on atomically dispersed Bi within deficient $\text{LaMnO}_{3.15}$ (SABi/U-LMO) [138]. This design regulates not only the initial desolvation step. Instead, it targets the continuous kinetic process from $[\text{Zn}(\text{H}_2\text{O})_6]^{2+}$ dissociation to subsequent Zn atom diffusion and growth. This emphasis on post-desolvation diffusion is consistent with studies on Li metal anodes, where atomic metal catalysts promote surface diffusion and delocalized deposition to reduce localized metal accumulation [140]. For Zn metal anodes, similar diffusion regulation is also important for achieving compact and planar deposition. As illustrated in Figure 11e, the incorporation of atomic Bi induces appropriate electronic-structure regulation in the perovskite framework. This change lowers the d-band center and generates a favorable nonbonding state near the Fermi level, which helps accelerate catalytic desolvation, suppress HER, and redistribute the interfacial electric-field. Consequently, Zn deposition becomes more ordered and tends to proceed in a planar manner rather than through localized dendritic growth. The catalytic effect on interfacial water is further supported by the in situ sum frequency generation spectroscopy (SFG) spectra in Figure 11f [141]. Compared with bare Zn, the SABi/U-LMO@Zn interface shows a clear reconstruction of the O–H spectral features, indicating that the hydrated Zn species undergo desolvation more readily under applied bias. This trend is also reflected in Figure 11g, where the area ratio continuously increases at the SABi/U-LMO@Zn interface, suggesting a more favorable interfacial water environment for Zn^{2+} dissociation and transport. Therefore, desolvation and post-desolvation migration are regulated in a cascade manner rather than as separate processes. Similar self-tandem catalytic concepts have also been reported in multivalent Mg–S batteries, where serially assembled atomic reactors promote Mg^{2+} desolvation and sulfur conversion [142]. This comparison suggests that catalytic acceleration of multivalent-ion desolvation may be a general strategy for improving sluggish interfacial kinetics. Benefiting from this integrated kinetic modulation, the Zn anode achieved a high Coulombic efficiency of 99.59% over 2000 cycles and a long lifespan of 5000 h

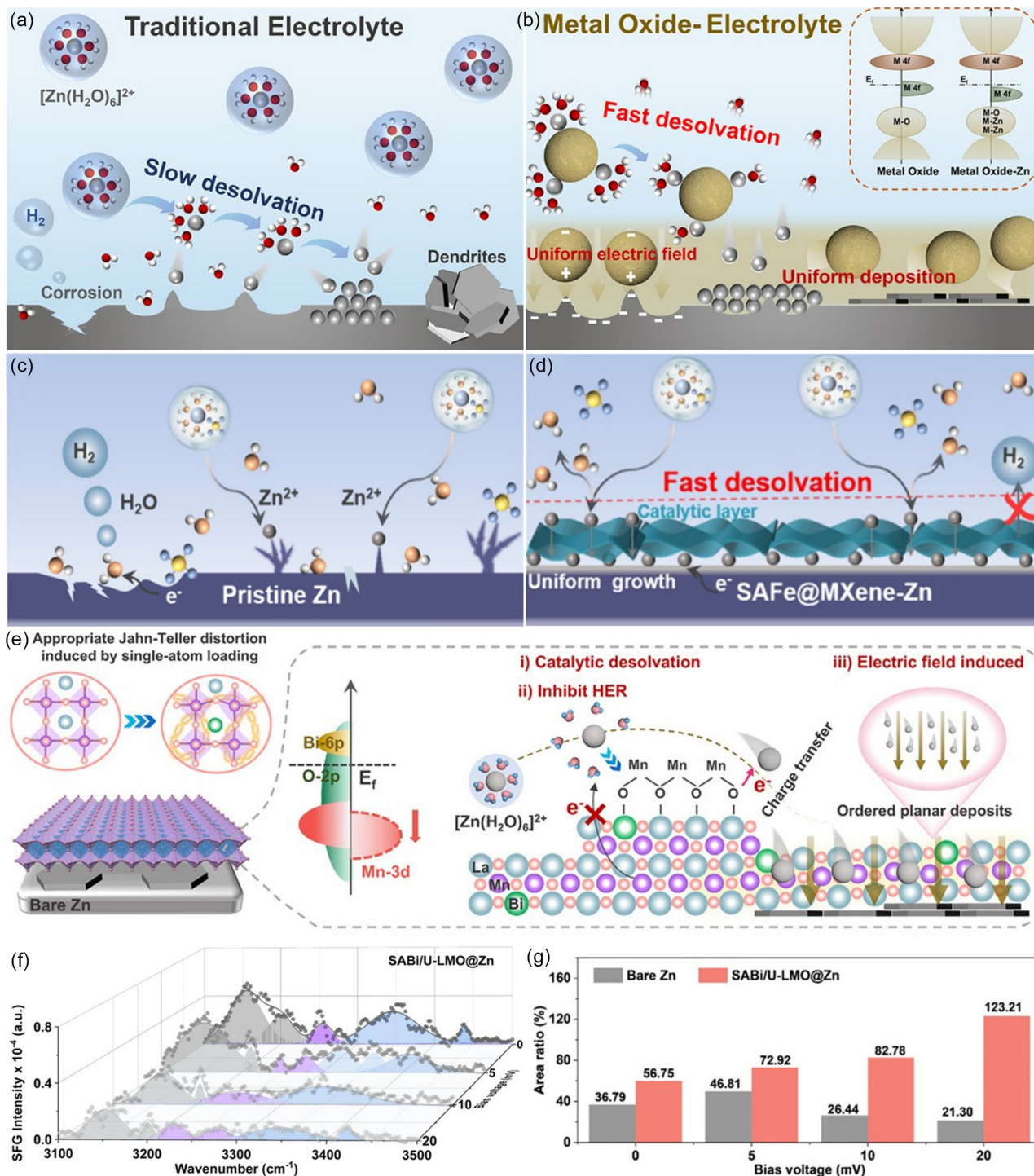


FIGURE 11 | Schematic illustration and calculation of EDCO effect on electrolyte. Schematic illustration of Zn^{2+} solvation structure and Zn deposition behavior in (a) traditional electrolyte and (b) metal oxide-traditional electrolyte [136]. Reproduced with permission. Copyright 2025, Wiley-VCH. Schematic illustrations of (c) dendrite growth and HER on pristine Zn and (d) synergistic desolvation mechanism of catalysis with porous sieving from the SAFe@MXene nanosheets avoiding side reactions and dendrite growth [137]. Reproduced with permission. Copyright 2025, American Chemical Society. (e) Schematic diagram of self-cascade catalytic strategy with tunable electronic structure for zinc ion deposition [138]. (f) The SFG spectra of different states of Zn^{2+} solvation structure under the OH region within the SABi/U-LMO@Zn/electrolyte interface. All spectra have been normalized to their maximum for better visualization [138]. (g) The ratio of strongly bound water and loosely bound water [138]. Reproduced with permission. Copyright 2026, Wiley-VCH.

at 1 mA cm^{-2} under 0°C . The full cell maintained nearly 100% capacity retention after 900 cycles at 1 A g^{-1} under -20°C , demonstrating the effectiveness of self-cascade catalysis for durable low-temperature AZIBs.

These catalytic and interfacial engineering studies further confirm that Zn deposition is governed by a dynamic sequence of desolvation, charge transfer, surface diffusion, and nucleation, rather than by static solvation structure alone.

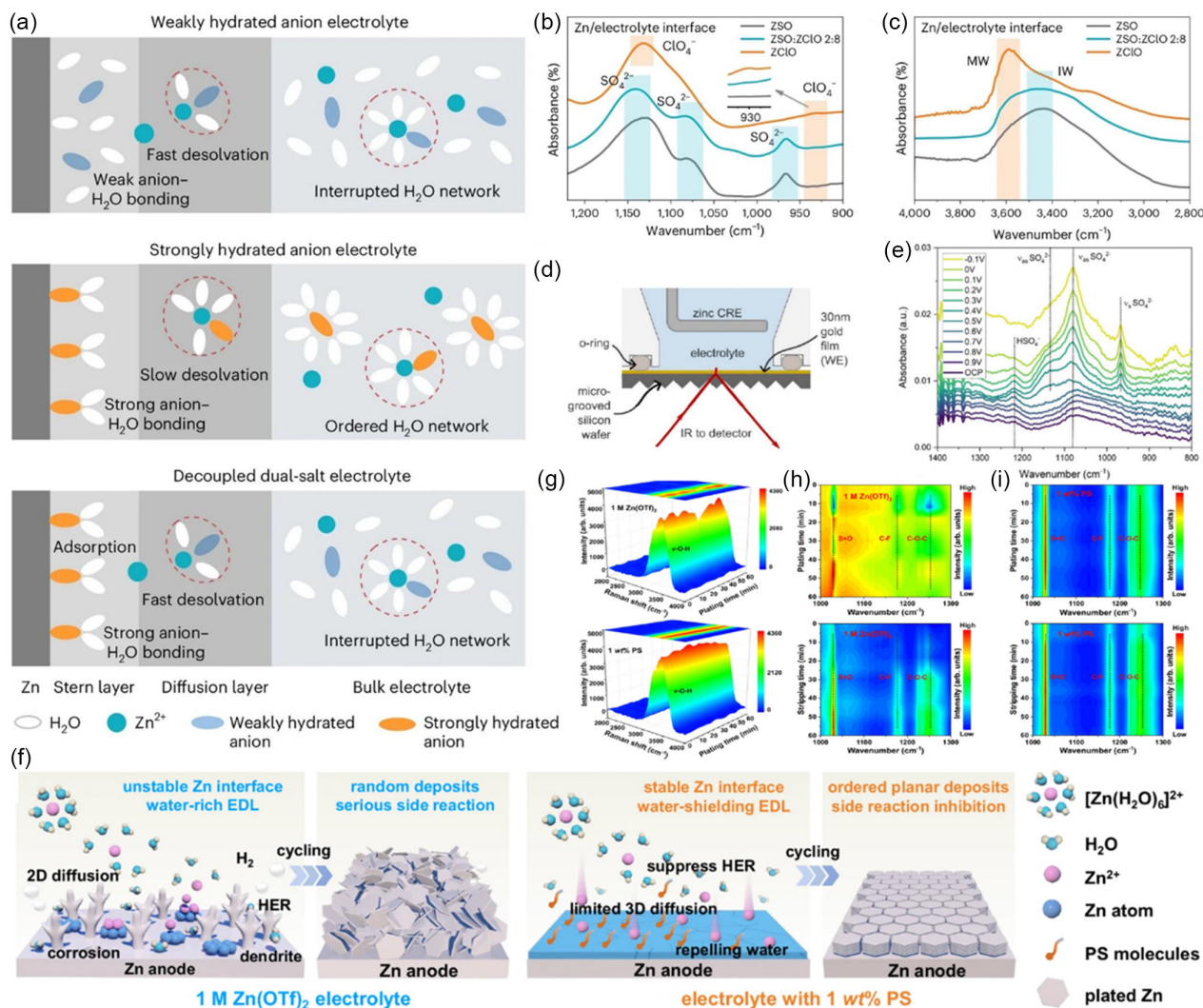


FIGURE 12 | (a) Schematic illustrating the trade-off between HER mitigation and antifreezing capability in single-salt electrolytes with weakly hydrated anions (top) and strongly hydrated anions (middle), and the specific anion distribution in a DDSE, engineered to overcome the trade-off (bottom) [143]. (b, c) ATR-SEIRAS spectra of equilibrium adsorption species at the Zn electrode/electrolyte interface in the 900–1250 cm⁻¹ range (b) and in the 2800–4000 cm⁻¹ range (c) for ZSO, ZSO:ZClO 2:8 and ZClO electrolytes. The blue bars denote peaks associated with SO₄²⁻ and IW, and orange bars highlight peaks associated with ClO₄⁻ and MW [143]. Reproduced with permission. Copyright 2025, Springer Nature. (d) Instrumental schematic for SEIRAS spectroelectrochemical cell [144]. (e) Potential dependent ATR-SEIRAS spectra for 1 m aqueous ZnSO₄ at negative potentials [144]. Reproduced with permission. Copyright 2024, American Chemical Society. (f) Schematic illustration of the effects of the PS molecule on electrolyte and interfacial chemistry for 1 m Zn(OTf)₂ electrolyte and electrolyte with 1 wt% PS [145]. (g) Three-dimensional in situ Raman spectra of ν-O–H during Zn plating process, (h, i) In situ FTIR spectra of Zn plating/stripping process [145]. Reproduced with permission. Copyright 2025, Springer Nature.

4.6 | Characterization of Bulk and Interfacial Electrolyte Behaviors

The catalytic regulation strategies discussed above highlight the decisive role of local interfacial processes in Zn²⁺ desolvation, HER suppression, and Zn deposition. However, these interfacial processes cannot be fully understood by bulk electrolyte analysis alone. Conventional electrolyte characterizations, such as Raman spectroscopy, Fourier transform infrared spectroscopy (FTIR), nuclear magnetic resonance spectroscopy (NMR), viscosity measurements, and ionic conductivity tests, provide useful information on Zn²⁺ solvation structures, hydrogen bond networks, and ion transport. Nevertheless, they mainly reflect the average properties of the bulk electrolyte. In practical AZIBs, Zn²⁺ desolvation, Zn nucleation, hydrogen evolution, corrosion,

SEI formation, and by-product evolution occur mainly at electrode/electrolyte interfaces. Therefore, the local ion distribution, water orientation, electric-double-layer structure, and interfacial reaction pathway may differ markedly from the bulk electrolyte behavior. As summarized in Figure 12a–i, advanced operando characterization techniques are essential for distinguishing these two environments.

Figure 12a illustrates the trade-off between HER mitigation and antifreezing capability in single salt electrolytes with weakly hydrated anions (top) and strongly hydrated anions (middle), and shows the specific anion distribution in a decoupled dual-salt electrolyte (DDSE) designed to overcome this trade-off [143]. ATR-SEIRAS spectra in the 900–1250 cm⁻¹ range (Figure 12b) and 2800–4000 cm⁻¹ range (Figure 12c) reveal equilibrium

adsorption species at the Zn electrode/electrolyte interface for ZSO, ZSO:ZClO 2:8, and ZClO electrolytes. The blue bars denote peaks associated with SO_4^{2-} and interfacial water (IW), while the orange bars indicate ClO_4^- and bulk water (MW) [138]. These results show that the interfacial electrolyte composition differs from the bulk, and that SO_4^{2-} preferentially stabilizes interfacial water and suppresses HER.

The SEIRAS spectroelectrochemical cell schematic is shown in Figure 12d, while Figure 12e displays potential dependent ATR-SEIRAS spectra for 1 M aqueous ZnSO_4 at negative potentials [144]. These techniques monitor interfacial adsorption and mass evolution dynamically, providing more reliable information on how electrolyte components accumulate or reorganize at the Zn surface during plating/stripping.

Interfacial water structure and activity are also critical, as they govern HER and Zn^{2+} desolvation. Figure 12f illustrates how polysorbate (PS) molecules modify the electrolyte and interfacial chemistry for 1 M $\text{Zn}(\text{OTf})_2$ electrolyte and 1 wt% PS electrolyte [145]. The additive forms a water-shielding electric double layer (EDL) at the Zn surface, reducing HER and promoting uniform Zn^{2+} flux. Figure 12g shows three-dimensional in situ Raman spectra of O–H vibrations during Zn plating, while Figure 12h,i display in situ FTIR spectra of Zn plating/stripping [141]. These methods confirm that interfacial water can be effectively regulated by additives, and not just by bulk electrolyte composition.

Beyond the representative Zn/electrolyte interfacial characterizations summarized in Figure 12, interphase resolved analysis is also necessary for understanding electrolyte-derived surface chemistry. X-ray photoelectron spectroscopy (XPS), cryogenic XPS (cryo-XPS), time-of-flight secondary ion mass spectrometry (ToF-SIMS), transmission electron microscopy (TEM), and cryogenic TEM (cryo-TEM) can reveal the chemical composition, depth distribution, and morphology of SEI or interfacial layers formed on Zn anodes. For instance, depth sensitive characterization has been used to identify robust bilayer SEI structures on Zn electrodes, where different inorganic and organic components jointly regulate Zn^{2+} transport, mechanical stability, and side reaction suppression [146]. Related interphase designs in Li metal batteries have shown that ordered organic/inorganic dual-layered structures can improve interfacial stability by separating mechanical protection from ion-transport regulation [147]. Although the interphase chemistry differs between Li and Zn systems, this comparison emphasizes the need to resolve both composition and spatial distribution when evaluating electrolyte derived SEI layers. Such information is difficult to obtain from bulk electrolyte spectra. Therefore, interphase resolved characterization is important for clarifying how additives, anions, or solvent molecules decompose, adsorb, or reorganize at the Zn surface during cycling.

The cathode/electrolyte interface also requires operando characterization. In many AZIB systems, especially Mn- and V-based cathodes, proton participation, local pH fluctuations, active material dissolution, and by-product formation can strongly affect cycling stability. Operando pH monitoring has been used to reveal $\text{H}^+/\text{Zn}^{2+}$ coinsertion behavior in aqueous $\text{Zn}/\text{V}_2\text{O}_5$ batteries, indicating that local proton activity cannot be simply inferred from the initial bulk electrolyte composition [148]. Similarly, operando X-ray diffraction (operando XRD), in situ Raman

spectroscopy, and inductively coupled plasma mass spectrometry (ICP-MS) can help track cathode phase evolution, by-product formation, and active material dissolution. These methods are important for clarifying how cathode side interfacial reactions couple with Zn anode stability through the electrolyte.

Taken together, advanced interfacial characterization provides a bridge between bulk electrolyte structure and electrochemical performance. As illustrated by the representative examples in Figure 12, ATR-SEIRAS and potential dependent SEIRAS can reveal interfacial anion adsorption, water structure, and potential driven electrolyte reorganization at the Zn surface. In situ Raman and FTIR further track the evolution of interfacial water during Zn plating/stripping. Beyond these techniques, interphase resolved methods such as XPS, cryo-XPS, ToF-SIMS, TEM, and cryo-TEM can identify the chemical composition and spatial distribution of SEI layers, while cathode side operando methods can reveal local pH evolution, proton participation, phase transformation, and active material dissolution. These complementary techniques help clarify how electrolyte components redistribute from the bulk to the interface and how this redistribution regulates Zn^{2+} desolvation, HER, interphase formation, cathode/electrolyte degradation, and Zn deposition behavior. Therefore, future electrolyte studies should combine bulk solvation analysis with operando and interface sensitive characterization to establish a more reliable relationship among electrolyte composition, interfacial chemistry, and battery performance in AZIBs.

5 | Conclusions and Prospects

In this review, recent advances in electrolyte engineering for AZIBs have been systematically summarized from the perspective of solvation chemistry and interfacial stabilization. The working principles of AZIBs were first introduced, with emphasis on the role of electrolytes in governing Zn^{2+} transport, electrode/electrolyte interfacial reactions, and charge storage behavior. The major electrolyte-related challenges were then discussed, including uncontrolled Zn dendrite growth, HER, cathode dissolution, anode corrosion, and poor adaptability under extreme operating conditions. These issues are closely associated with the strong hydration of Zn^{2+} , the high activity of water molecules, and the instability of electrode/electrolyte interfaces in aqueous environments.

To address these challenges, various electrolyte regulation strategies have been developed. Solvent engineering can tune the Zn^{2+} solvation structure and reduce water activity, thereby mitigating water induced side reactions. Electrolyte salt optimization, especially through highly concentrated or anion regulated systems, can reconstruct the primary solvation sheath of Zn^{2+} , broaden the electrochemical stability window, and promote more reversible Zn plating/stripping. Functional additives provide a flexible and cost-effective approach to regulate interfacial electric-field distribution, Zn nucleation behavior, and surface side reactions. Composite electrolytes further integrate ion-transport regulation, mechanical reinforcement, and interfacial stabilization. More recently, catalytic interfacial regulation has offered a targeted route to accelerate Zn^{2+} desolvation and guide uniform Zn deposition. Together, these strategies demonstrate that electrolyte design has evolved from simple component optimization toward

integrated regulation of solvation structure, interfacial kinetics, and deposition behavior.

Despite this progress, several issues still limit the practical implementation of advanced AZIB electrolytes. In particular, the practical limitations of different electrolyte systems should be carefully considered. Highly concentrated electrolytes can effectively suppress water activity and broaden the electrochemical stability window, but their high salt consumption, increased viscosity, relatively high cost, and possible transport limitations may hinder large scale application. DES-based and hydrated eutectic electrolytes provide strong coordination environments and improved interfacial stability, yet their sluggish ion transport and high viscosity may restrict rate capability and low-temperature operation. Organic-aqueous hybrid electrolytes can improve temperature adaptability, but solvent volatility, toxicity, flammability, and long-term compatibility should be evaluated. Additive-based strategies are economical and efficient, but additive depletion, concentration sensitivity, and possible side reactions during long-term cycling remain concerns. Composite electrolytes improve safety and mechanical stability, while their ionic conductivity, electrode contact, water retention, and scalable fabrication still require further optimization. One important challenge is maintaining interfacial stability under practical operating conditions. Many electrolyte systems show excellent performance under moderate current density, low areal capacity, or excess electrolyte conditions. However, under high rate operation, high cathode loading, high Zn utilization, lean electrolyte, or thick electrode configurations, rapid Zn^{2+} consumption and intensified concentration polarization may disturb interfacial ion flux and local electric-field distribution. These effects can accelerate dendrite growth, HER, corrosion, and by-product accumulation. Therefore, future electrolyte evaluation should place greater emphasis on practically relevant conditions, rather than relying only on mild laboratory tests.

The trade-off between electrolyte functionality and practical applicability also requires further consideration. Highly concentrated electrolytes can effectively suppress water activity and broaden the electrochemical stability window, but their high salt consumption, increased viscosity, and relatively high cost may hinder large scale application. DES-based and hydrated eutectic electrolytes provide strong coordination environments and improved interfacial stability, yet their sluggish ion transport may restrict rate capability and low-temperature performance. Organic-aqueous hybrid electrolytes can extend the operating temperature range, but solvent volatility, toxicity, flammability, and long-term compatibility should be carefully assessed. Additive-based strategies are attractive because of their low dosage and high tunability, but additive depletion, concentration sensitivity, and possible side reactions during prolonged cycling remain unresolved. Composite electrolytes improve safety and mechanical stability, whereas their ionic conductivity, electrode contact, water retention, and scalable fabrication still need further optimization. These limitations indicate that future electrolyte design should balance electrochemical performance with cost, safety, manufacturability, and environmental compatibility.

Wide temperature adaptability is another key requirement for practical AZIBs. At low temperatures, electrolyte freezing, increased viscosity, and sluggish Zn^{2+} desolvation can severely

limit ion transport and interfacial charge transfer. At elevated temperatures, water evaporation, accelerated Zn corrosion, additive decomposition, and interphase instability may become more pronounced. Therefore, electrolyte systems should be designed to maintain sufficient ionic conductivity, regulated water activity, fast desolvation kinetics, and stable electrode/electrolyte interfaces over a broad temperature range. This requires a more comprehensive understanding of how solvation structures, hydrogen bond networks, and interfacial reactions evolve under different thermal conditions.

In addition, future mechanistic studies should move beyond static bulk electrolyte analysis toward operando, interface-sensitive, and multiscale investigations. Techniques such as attenuated total reflection surface-enhanced infrared absorption spectroscopy, SFG, electrochemical quartz crystal microbalance, differential electrochemical mass spectrometry, cryo-XPS, ToF-SIMS, and operando XRD can provide direct information on interfacial ion adsorption, water orientation, Zn^{2+} desolvation, HER, SEI evolution, and cathode/electrolyte degradation. Meanwhile, molecular dynamics and ab initio molecular dynamics simulations can reveal Zn^{2+} solvation structures, hydrogen bond networks, ion pair states, and interfacial desolvation pathways at the molecular level. Combining operando characterization with MD/AIMD simulations and multiscale modeling will help establish a clearer relationship among electrolyte composition, dynamic interfacial behavior, Zn nucleation, and long-term cycling stability. Such integrated understanding is essential for developing practical, durable, and cost-effective AZIBs electrolytes.

Acknowledgments

Dr. Jian Wang thanks the fellowship support provided by the Alexander von Humboldt Foundation. The authors thank financial support from the National Natural Science Foundation of China (grant nos. 22572217, 22279161, 22309144, 52202244, and 52436005), Opening funding from Key Laboratory of Engineering Dielectrics and Its Application (Harbin University of Science and Technology) (grant no. KFM202507, Ministry of Education), Open Project of Inner Mongolia Engineering Research Center of Lithium-Sulfur Battery Energy Storage (grant no. MDK2025071), China Postdoctoral Science Foundation (grant no. 2024M751176), and the Research Foundation for Advanced Talents of Jiangsu University, China (grant no. 22JDG010).

Open Access funding enabled and organized by Projekt DEAL.

Funding

National Natural Science Foundation of China (22572217, 22279161, 22309144, 52202244, 52436005); Opening funding from Key Laboratory of Engineering Dielectrics and Its Application (KFM202507); Open Project of Inner Mongolia Engineering Research Center of Lithium-Sulfur Battery Energy Storage (MDK2025071); China Postdoctoral Science Foundation (2024M751176); Research Foundation for Advanced Talents of Jiangsu University (22JDG010).

Conflicts of Interest

The authors declare no conflicts of interest.

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