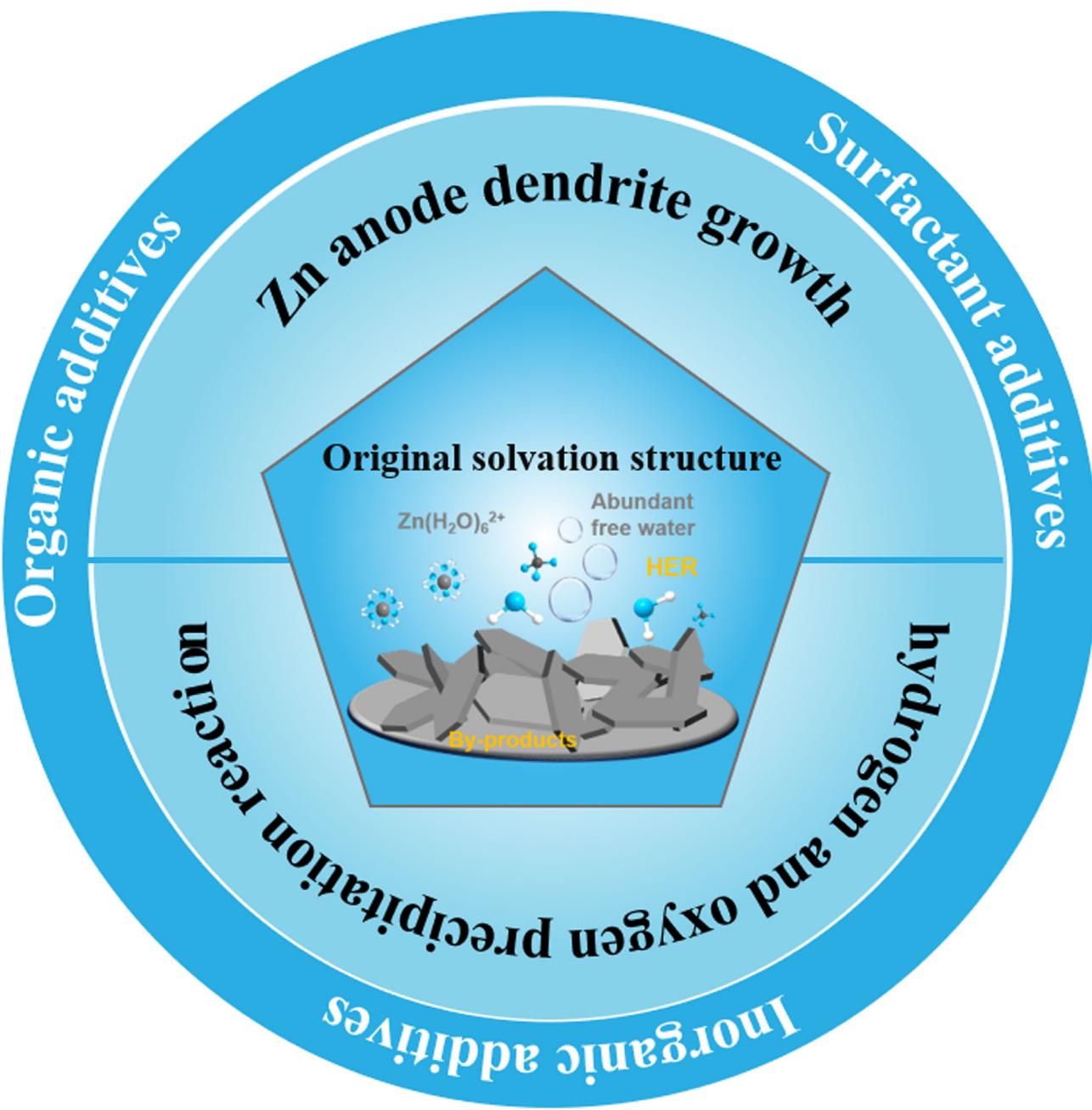


Electrolyte Additive Strategies for Stabilizing Zn Anodes in Zn²⁺ Energy Storage Devices

Kaifa Dong, Bingrong Shen, Xuan Xie,* Xin Wang, Yaping Jiang, Pengyun Xie, Hui Peng,* and Guofu Ma*



Zinc ion (Zn^{2+}) energy storage devices are considered promising candidates for next-generation energy storage technologies, offering advantages in safety, low cost, and environmental friendliness. However, their commercialization remains limited by numerous challenges, including precise regulation of the molecular conformational relationships of electrolyte additives, optimization of electrode–electrolyte interfacial stability, scalability of manufacturing processes, and comprehensive analysis of long-term degradation mechanisms. Pure Zn anode interfaces face numerous unavoidable challenges, including dendrite growth, corrosion, passivation, and hydrogen evolution reactions. This review summarizes recent advances in electrolyte additives for

Zn^{2+} energy storage devices, encompassing inorganic, organic, surfactant, and organic–inorganic composite additives, with a focus on the interaction mechanisms between additives, electrodes, and electrolytes. Furthermore, the optimal type and incorporation method of additives are discussed, emphasizing the positive impact of these factors on improving additive efficiency and performance. Finally, challenges and future directions for the development of electrolyte additives and advanced ZIHSs are proposed. This review aims to provide a comprehensive perspective to guide future research and development, advancing the efficiency, stability, and cost-effectiveness of aqueous Zn^{2+} energy storage devices.

1. Introduction

With the increasing global energy demand and environmental problems, the development of novel and efficient energy storage systems has become particularly important.^[1–4] Aqueous Zn^{2+} hybrid supercapacitors (ZIHSs) and Zn^{2+} batteries (ZIBs), as an emerging multivalent ion energy storage device, have become increasingly important in meeting the needs of modern societies for efficient, safe, and economical energy storage devices.^[5–8] Although lithium ions (Li^{+}) batteries dominate the energy storage system market due to their high energy density (250–300 Wh kg⁻¹ for commercial systems and up to 711 Wh kg⁻¹ for laboratory grade), voltage platform (3.0–4.3 V vs Li⁺/Li), and mature technology,^[9–11] However, the limited availability of lithium resources, cost issues, and safety risks associated with organic electrolytes have become a significant impediment to the development of stable energy storage devices.^[12–14] In contrast, Zn^{2+} energy storage devices offer significant low-cost advantages, primarily due to the greater abundant of Zn resources not only reduces reliance on limited resources but also mitigates supply chain risks.^[15,16] Despite significant progress in the development of Zn^{2+} energy storage devices, their performance remains strongly influenced by the electrolyte, which plays a crucial role in determining safety, energy conversion efficiency, and long-term stability.^[17,18] Therefore, optimizing the electrolyte composition is of paramount importance for enhancing both the energy efficiency and cycle life of Zn^{2+} energy storage devices.^[19,20]

In recent years, researchers have invested a great deal of effort in the design and optimization of electrolytes with the

aim of improving the energy conversion efficiency and cycling stability of ZIHSs.^[21] At present, many studies have been carried out for the modification of $ZnSO_4$ electrolyte, but its key bottleneck has not been fundamentally broken. For example, the narrow electrochemical stability window of $ZnSO_4$ electrolytes results in low energy density and compromised electrochemical stability of ZIHSs.^[22,23] Under extreme conditions, these electrolytes often suffer from substantial ionic conductivity loss, severely restricting their applicability at elevated or subzero temperatures. To overcome this limitation, it has been shown that the electrochemical stability window broadened and coulombic efficiency improved by introducing functional additives.^[24,25] One effective strategy is to introduce electrolyte salts or organic small molecules into the electrolyte to generate a stable interfacial protective layer *in situ* at the anode/electrolyte interface, thus inhibiting side reactions and structural degradation of the anode material and significantly enhancing the interfacial stability.^[26,27] For example, Xue et al. developed a dual electrolyte additives strategy to simultaneously achieve uniform nucleation and directional deposition of Zn^{2+} using citric acid (CA) and aspartame (APM).^[28] Experimental results and theoretical calculations reveal that the interaction between CA and Zn^{2+} enhances the hydrophobic character of Zn^{2+} , thereby suppressing the decomposition of H_2O . Meanwhile, CA also modulates the pH of the electrolyte, contributing to the stabilization of the acid/base environment.^[29,30] Furthermore, the adsorption of APM on the Zn anode surface facilitates the formation of a robust solid electrolyte interphase (SEI), which effectively inhibits dendrite formation and significantly improves the reversibility of Zn plating/stripping processes.^[31] Li et al. introduced the diethylenetriaminepenta (methyleneephosphonic acid) sodium salt (DS) into a conventional $ZnSO_4$ electrolyte thereby enabling the *in-situ* formation of an inorganic/organic hybrid bilayer interfacial architecture, which effectively suppresses Zn dendrite growth and mitigates the hydrogen evolution reaction (HER).^[32] Electrochemical measurements reveal that the Zn//Zn symmetric cell employing the DS-modified $ZnSO_4$ electrolyte exhibits an extended cycling stability exceeding 1700 h at 5 mA cm⁻², outperforming its counterpart using the unmodified $ZnSO_4$ electrolyte.

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Based on the above analysis, we first focus on the key challenges and issues that currently hinder the development of Zn^{2+} energy storage devices as a starting point, and highlight the key roles of electrolyte additives in the emerging strategies to improve energy conversion efficiency and cycling stability from inorganic additives, organic additives, and surfactant additives. By integrating molecular dynamics simulations with advanced interfacial characterization techniques, we provide in depth insights into how electrolyte additives influence the Zn^{2+} solvation structure, regulate interfacial ions transport and effectively passivate reactive sites. Finally, we outline the remaining challenges and offer perspectives on the future development of high-performance electrolyte additives.

2. Issues and Challenges on Zn Metal Anode

Zn -based energy storage devices, particularly aqueous Zn^{2+} batteries (ZIBs) and ZIHSs have attracted extensive attention owing to their intrinsic advantages.^[33,34] Despite these appealing features, the practical application of Zn -based devices is severely

hindered by persistent challenges associated with the Zn anode, including dendritic Zn growth and parasitic side reactions.^[35] Although previous studies have demonstrated that employing mild electrolytes can partially mitigate these issues, Zn dendrite formation remains a pronounced problem even in assembled full cells.^[36,37] Concurrently, during repeated Zn plating/stripping, the Zn surface is prone to parasitic HER, which generates by-products that perturb the local microenvironment at the electrode-electrolyte interface.^[38] These intertwined issues significantly deteriorate the coulombic efficiency (CE) and compromise the long-term electrochemical stability of Zn^{2+} energy storage devices.^[39] The challenges and issues mentioned above are described in more detail below Figure 1.

2.1. Zn Dendrites

Early research on ZIBs and ZIHSs focused on optimizing high-performance anode materials. In this case, metallic Zn metals were often used directly as anode in aqueous electrolytes owing to high hydrogen evolution overpotential.^[40] However, in-depth studies have shown that in traditional aqueous electrolytes,



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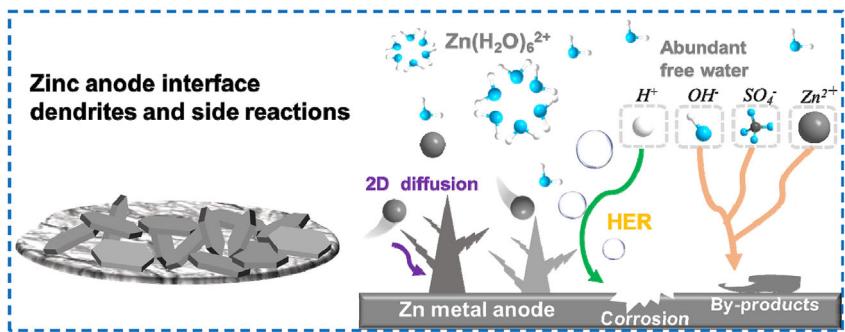


Figure 1. Schematic illustration of the challenges and problems faced by Zn metal anode.

Zn²⁺ migrates to the negative electrode surface in the form of a [Zn (H₂O)₆]²⁺ hexacoordinated solvated cluster. The coordinating water molecules in this cluster are strongly polarized by Zn²⁺, resulting in a significant weakening of the O-H bonds, which in turn increases the desolvation energy barrier and leads to localized charge density inhomogeneities. When desolvation is incomplete, "partially dehydrated" Zn²⁺ preferentially deposit at surface protrusions, forming microscopic tips; the concentrated electric field at these tips further attracts subsequent Zn²⁺, often leading to uneven deposition of Zn²⁺ on the anode surface and the formation of Zn dendrites.^[41] At the same time, loose by-products such as Zn₅(OH)₈(OAc)₂·xH₂O generated by side reactions exacerbate surface roughness, providing additional nucleation sites and forming a "tip-by-product" positive feedback loop that causes dendrites to continue to branch and penetrate the separator, ultimately leading to short circuits. These dendritic structures severely compromise battery performance and lifespan, and may even lead to safety risks.^[42] Upon charging, Zn²⁺ are electrochemically reduced on the anode surface by gaining electrons to form metallic Zn ($Zn^{2+} + 2e^- \rightarrow Zn$), but the uneven distribution of the electric field and ion concentration, which facilitate the dendrite growth.^[43,44] When multiple Zn dendrites form in proximity, they may cross-grow and form a more complex structure, which further aggravate the internal inhomogeneity of the cell and significantly elevate the risk of electrical short circuits.^[45,46] Moreover, the Zn²⁺ deposition behavior is highly sensitive to the local Zn²⁺ ion concentration and the spatial distribution of the electric field, both of which critically influence the uniformity and reversibility of Zn plating/stripping processes.^[44] During charge/discharge process, the nonuniform distribution of the electric field on the electrode surface leads to spatial variations in local field strength.^[45–48] According to Ohm's law, such electric field inhomogeneity directly leads to uneven current density distribution. In regions of higher current density, Zn²⁺ are reduced more rapidly, favoring the formation of protrusions that can subsequently evolve into dendritic structures. Simultaneously, gradients in local Zn²⁺ concentration exacerbate this effect by further promoting anisotropic growth, ultimately accelerating dendrite formation and compromising the stability of the Zn anode. According to Fick's law of diffusion, the presence of a concentration gradient triggers the migration of ions. However, due to the limited diffusion rate of Zn²⁺ in electrolytes, localized

accumulation of Zn²⁺ can lead to preferential and accelerated deposition in those regions, thereby inducing the Zn dendrites growth.^[49,50] In ZIHSs and ZIBs, when Zn foil is employed as the anode, such nonuniform deposition behavior becomes increasingly pronounced over repeated charge-discharge cycles.^[25] These Zn dendrites will continue to grow and may eventually penetrate the diaphragm, leading to direct contact between the positive and negative electrodes, and thus triggering an internal short circuit in the battery.^[51–53] Based on the above analysis, surface modification of Zn anode and regulation of microenvironment around Zn²⁺ can effectively inhibit the Zn dendrites growth, contributing to the development of Zn anode with excellent stability and reversibility. Among various strategies explored to address the challenges of Zn anodes, the incorporation of functional additives into conventional aqueous electrolytes has emerged as one of the most effective approaches. These additives can substantially enhance the Zn²⁺ ion transport kinetics and increase the nucleation overpotential, thereby promoting more uniform and compact Zn deposition.

2.2. Hydrogen Evolution Parasitic Reaction

In the aqueous electrolyte system, six water molecules in the solvated shell form a strong hydrogen bond network with Zn²⁺, and the bond energy of the OH bonds is significantly reduced due to metal ion polarization. At the Zn deposition potential, the water molecules within [Zn (H₂O)₆]²⁺ readily accept electrons and undergo proton reduction. Meanwhile, during charging, the relatively high reduction potential of Zn²⁺ at the Zn anode surface not only facilitates Zn²⁺ deposition but also promotes parasitic reduction of water molecules.^[41] This competitive HER leads to hydrogen gas production, with the reaction: $2H_2O + 2e^- \rightarrow H_2\uparrow + 2OH^-$. This process not only directly consumes electrons and reduces coulombic efficiency but also releases OH⁻, which increases the local pH at the interface, further promoting the formation of Zn(OH)₄²⁻ complexes. The HER at the Zn anode is mainly divided into two steps mechanism.^[53,54] The first step involves a charge-transfer process, wherein protons are reduced to adsorbed hydrogen atoms. This step is generally considered rate-determining and plays a crucial role in dictating the overall HER kinetics. First, it will significantly reduce the Coulomb efficiency of the battery, resulting in a large gap between the actual

battery output and the theoretical power.^[55,56] Second, it will increase the internal pressure of the battery, which may cause serious safety problems if the pressure continues to rise. The pH value of the electrolyte plays an important role in the rate of hydrogen precipitation. Generally, HER is more pronounced in an acidic electrolyte, where the abundance of protons provides favorable thermodynamic and kinetic conditions for the HER.^[56–58] However, the rate of hydrogen gas generation is not affected by pH alone. Instead, it is influenced by a combination of factors, including the nature of the electrode surface, the electrolyte composition, the overpotential for HER, and the interfacial charge-transfer kinetics. These parameters collectively determine the propensity and rate of HER, thereby impacting the electrochemical stability and efficiency of Zn-based aqueous energy storage systems.^[57–59] In addition, the ion transport capacity of the electrolyte also affects the rate of hydrogen precipitation reaction, when the ion transport capacity is strong, hydrogen ions can reach the electrode surface more quickly to participate in the reaction.^[60] At the same time, the state of the electrode surface should not be ignored. Surface inhomogeneities, including morphological irregularities of the Zn anode surface and the presence of impurities, will introduce additional active sites that providing proton reduction, thereby accelerating promote the HER.^[61] In some ZIBs and ZIHs, the HER can lead to the generation of large amounts of H₂ during the charging process.^[20] The generation of large amounts of hydrogen will cause the internal pressure of the batteries to rise rapidly, and if the cell exhibits poor sealing integrity, H₂ is very likely to leak out, which will bring great safety risks. In this context, many studies have suggested that reducing the H₂O activity in the electrolyte can inhibit HER. Compared to other modulation strategies, development of electrolyte additives is currently the most effective and simple method to inhibit HER.

3. Types of Electrolyte Additives

3.1. Inorganic Additives

In the field of ZIBs and ZIHs, the incorporation of inorganic electrolyte additives has emerged as a highly effective strategy for enhancing overall battery performance.^[62] These additives can effectively inhibit the formation of Zn dendrites, regulate the physicochemical properties of the electrolyte, and build a protective layer on the surface of the electrode, thus significantly enhancing the cycle stability and rate performance of the battery.^[47,63] At the same time, the inherent advantages of inorganic additives, including low cost, environmental friendliness, and structural tunability, which makes them ideal candidates for the development of high-performance, scalable aqueous ZIBs.^[64,65]

To alleviate the performance bottleneck caused by the irregular interfacial reaction between the solid-state electrode and the liquid electrolyte, Deng et al. successfully developed a high-density aqueous electrolyte with low water molecule activity and high cation transfer number by using metal oxide SrTiO₃ as an electrolyte additive (Figure 2a).^[66] The experimental results show

that the introduction of SrTiO₃ additives weakens the activity of water molecules, extends the onset potential of the oxygen evolution reaction (OER) in the electrolyte system from 1.8 V to above 2.2 V, and effectively inhibits the occurrence of side reactions at the Zn anode/electrolyte interface (Figure 2b,c). Density functional theory calculations revealed that the Zn (002) crystal plane exhibits a low affinity for hydrogen atom adsorption, which implies that simultaneous suppression of dendrites growth, HER and surface corrosion (Figure 2d). Based on the above advantages, the stability of the assembled Zn//Zn symmetric cell can be stabilized for more than 2000 h at 2 mA cm⁻², markedly outperforming cell assembled with conventional electrolytes (Figure 2e). In addition, when coupled with MnO₂ cathodes, the assembled Zn//MnO₂ full cell could reach a specific capacity of 328.2 mAh g⁻¹ after 500 cycles at 1 A g⁻¹, while maintaining superior rate performance and reaction kinetics (Figure 2f–h).

Zhang et al. demonstrated that the incorporation of TiOSO₄ as an inorganic salt additive into ZnSO₄ solution significantly improved the performance enhancement of ZIBs.^[67] The experimental results indicate that the incorporation of TiOSO₄ enhances the desolvation kinetics of Zn²⁺ and promotes stable interfacial reactions at both the Zn anode and the NH₄V₄O₁₀ cathode. Specifically, the strong coordination between TiOSO₄ molecules and Zn²⁺ accelerated the desolvation process, while concurrently forming an electrostatic shielding layer on the surface of the Zn anode. This effect not only ensures the reversibility of Zn deposition behavior but also improves the cyclic stability of ZIBs. Similarly, Yoo et al. employed vanadium oxide sulfate (VOSO₄) as an electrolyte additive to self-assemble a protective layer at the Zn anode, mitigating anodic corrosion, hydrogen evolution, and by-product generation.^[68] VOSO₄ also prevents vanadium dissolution at the V₂O₅ cathode, maintains the stability of the cathode structure, and promotes the interfacial Zn²⁺ diffusion.

Furthermore, the introduction of inorganic colloidal electrolytes not only regulates the dissolution environment of hydrated Zn²⁺, but also acts as a lubricant to improve the transfer of Zn²⁺. Inspired by the role of hemoglobin in promoting oxygen transport during human respiration, Rong et al. designed an inorganic colloidal electrolyte (CaSiO₃-ZnSO₄, CS-ZSO) that effectively suppresses Zn dendrite formation and mitigates the accumulation of interfacial by-products (Figure 3a).^[69] Combined structural characterization and DFT calculations reveal that the incorporation of CaSiO₃ as an electrolyte additive effectively modulates the pH of the electrolyte and tailors the solvation environment of hydrated Zn²⁺. This modulation leads to an optimized interfacial electric field between the Zn anode and the electrolyte and achieved uniform Zn²⁺ deposition (Figure 3b). As a result, the formation of Zn dendrites and the accumulation of interfacial by-products are significantly suppressed.^[70] Based on this, Zn//Zn symmetric cells, NH₄V₄O₁₀//Zn full cell and AC//Zn capacitors assembled with CS-ZSO electrolyte exhibit very excellent electrochemical performance compared with ZnSO₄ electrolyte (Figure 3c). Similarly, Cao et al. developed an inorganic colloidal electrolyte (LMS + ZSO) by incorporating a lithium magnesium silicate (LMS) into a ZnSO₄-based electrolyte, which simultaneously facilitates Zn plating/stripping behaviors and effectively suppresses

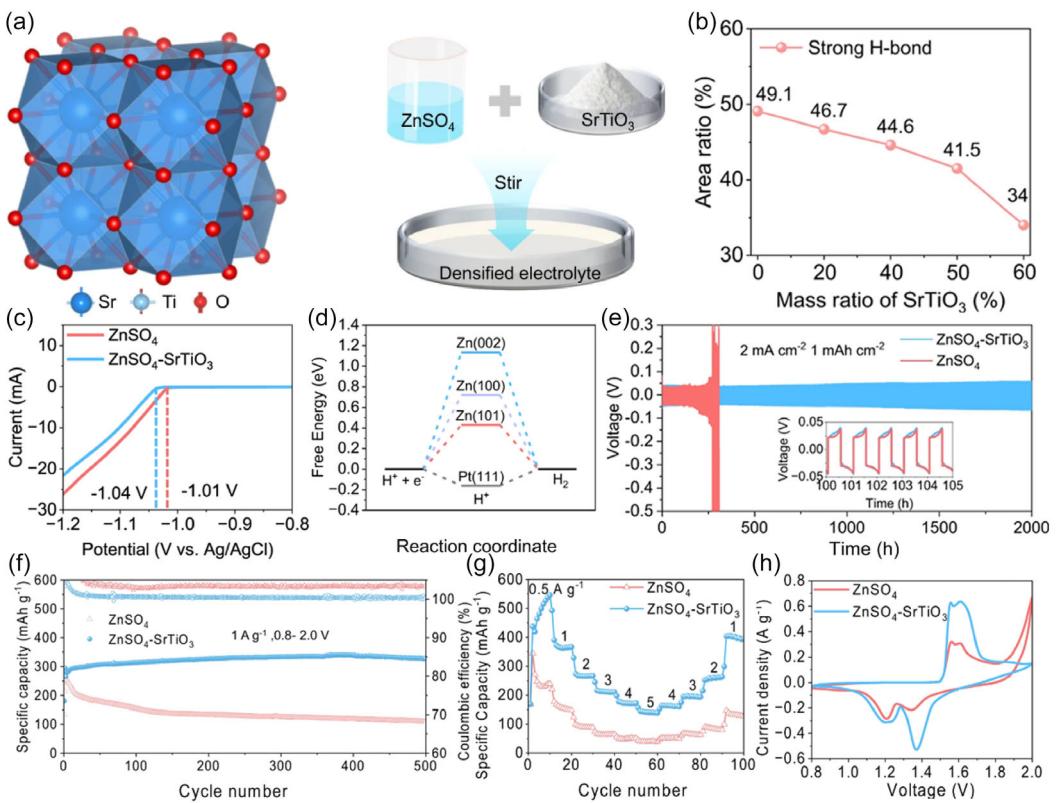


Figure 2. a) The crystal structure of SrTiO₃ and schematic diagram of densified aqueous electrolytes, b) the ratio of fitting strong H-bond area of electrolytes with various SrTiO₃ contents, c) the linear sweep voltammetry (LSV) for test of HER, d) the free-energy of HER on Zn (101), Zn (100), Zn (002), and Pt (111), e) Zn/Zn symmetric cells operated at 2 mA cm⁻², 1 mAh cm⁻², f) long-term galvanostatic cycling performance in various electrolyte at a current density of 1 A g⁻¹, g) the rate performance of the full cells, h) the cyclic voltammetry (CV) profiles at a scan rate of 1 mV s⁻¹.^[66]

side reactions at the Zn-LMS + ZSO electrolyte interface (Figure 3d–e).^[71] Due to its unique three-dimensional chain-layered architecture, LMS exhibits a pronounced affinity for H₂O. Both experimental observations and DFT calculations reveal that LMS-H₂O has a higher adsorption energy (0.12 eV). In contrast, the desolvation energy barrier is relatively low (56.62 kJ mol⁻¹), which suggests that the desolvation of Zn²⁺ occurs more readily in the LMS + ZSO electrolyte than in the ZSO electrolyte (Figure 3f). These results suggest that the ability of LMS to significantly enhance the interfacial Zn²⁺ transport kinetics by modulating the anode–electrolyte interaction. Based on the advantages of the hybrid electrolyte (1%LMS + ZSO), the assembled Zn//Zn symmetric cell can operate continuously for 1000 h at 0.5 mA cm⁻², which is 6.7 times that of the ZSO electrolyte (Figure 5g). In addition, the Zn//Ti cell assembled with 1% LMS + ZSO electrolyte showed an average CE value of 98.5% after 200 cycles at 2.5 mA cm⁻², significantly better than the Zn//Ti cell assembled with ZSO (Figure 3h).

Moreover, inorganic carbon nanomaterials can also be used as multifunctional electrolyte additives to stabilize Zn anode. For example, Zhang et al. introduced inorganic carbon nanomaterials (ICN) with rich surface polarity and low lattice mismatch as electrolyte additive for ZnSO₄ solution, which significantly improved the electrochemical performance of the Zn anode.^[72] Experiments and density functional theory (DFT) calculations

demonstrate that the abundant polar groups (-COOH, -OH) on the surface of ICN regulated the Zn²⁺ dissolution structure and reduced the H₂O activity.^[73] Secondly, ICN exhibits preferential adsorption on the Zn anode surface, forming a passivation protection interface that inhibit Zn corrosion and dendrite growth.^[74] Based on these synergistic effects, the ICN electrolyte additive enables a remarkably extended cycling stability of 900 h at 1 mA cm⁻². Furthermore, the Zn//VOx full cell assembled with ICN-modified ZnSO₄ electrolyte delivers a discharge capacity of 186 mAh g⁻¹ after 800 cycles, retaining 82.2% of its initial capacity at 1 A g⁻¹.

3.2. Organic Additives

Besides inorganic additives, organic additives (small molecules, polymers, etc.) have also emerged as promising additives for enhancing the electrochemical performance of ZIBs.^[75] Their mechanism of action is like inorganic additives, which is mainly reflected in the modulating the interfacial structure and deposition behavior of Zn anode, thus effectively inhibiting the dendritic growth and corrosion reactions.^[76,77] In addition, organic additives can self-assemble at the interface between the Zn electrode and the electrolyte to form a stable interfacial layer, thereby regulating the solvation structure, deposition kinetics, and the interfacial charge transfer process of Zn²⁺, which helps to reduce

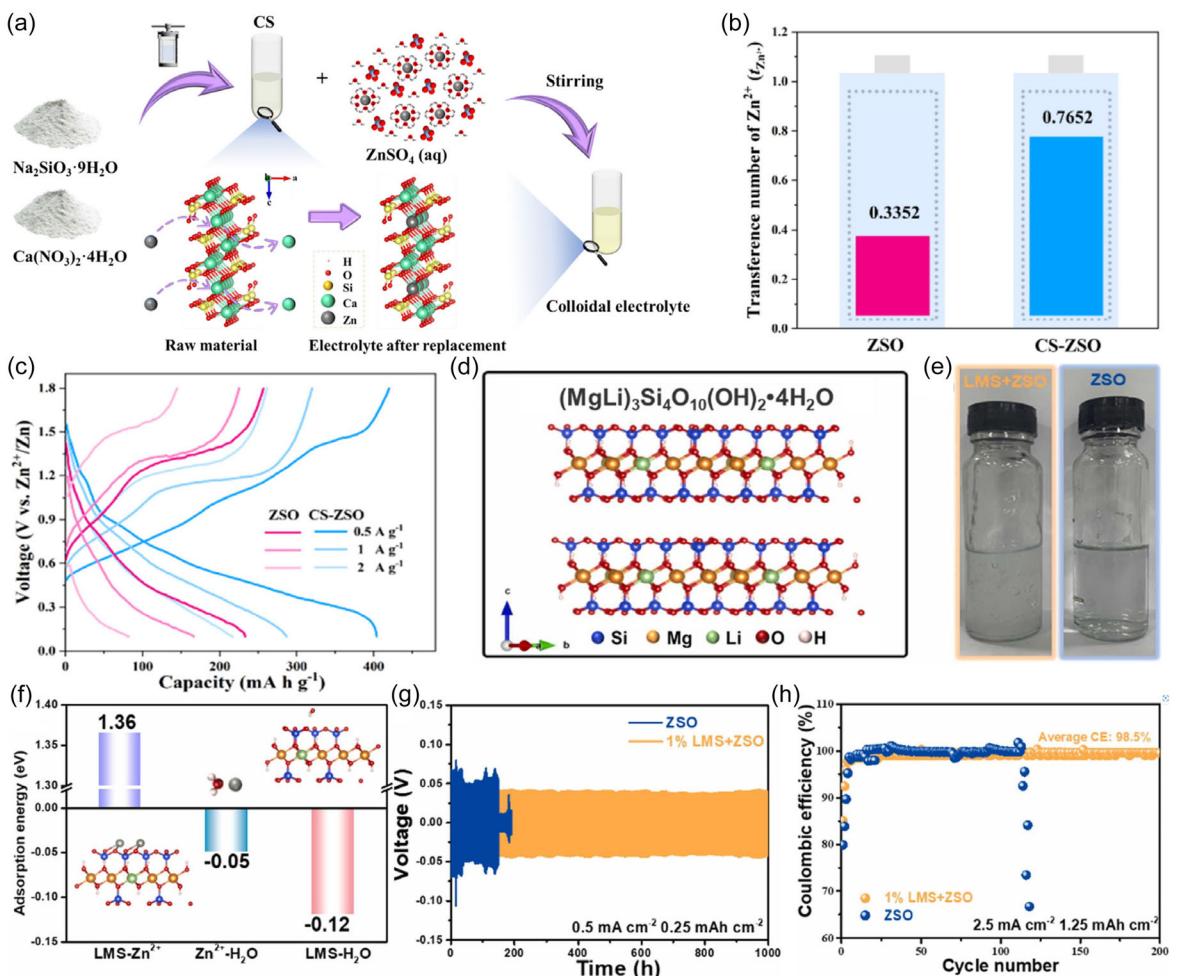


Figure 3. a) Schematic diagram of the synthesis process of the CS-ZSO electrolyte, b) comparison of the Zn^{2+} transfer numbers in ZSO and CS-ZSO, c) charge/discharge profiles,^[69] d) Structure of LMS. e) Optical images of LMS + ZSO and ZSO electrolytes, f) Calculated adsorption energy via DFT calculations, g) Cycling stability of Zn//Zn symmetric batteries in 1%LMS + ZSO and ZSO electrolytes at 0.5 mA cm^{-2} , h) CE of Zn//Ti batteries.^[71]

the H_2O induced side-reaction activity on Zn anode, ultimately improving the reversibility and stability of the Zn anode.^[78,79]

Wang et al. used melamine (M) as a multifunctional electrolyte additive to significantly modulate the electrolyte environment and optimize electrode–electrolyte interface.^[80] Theoretical calculations showed that the M molecule with two coordination environments of N atoms can interact with H_2O by strong hydrogen bonding, thus disrupting the original solvation sheath and reconstructing the hydrogen bonding network.^[81,82] Meanwhile, M molecule coordination with Zn^{2+} to form a dynamic electrostatic shielding layer, which effectively inhibits the dendritic growth and the occurrence of side reactions.^[83] Thanks to these advantages, Zn//Zn symmetric cells assembled with the $\text{ZnSO}_4 + \text{M}$ electrolyte have excellent cycling stability of over 1100 h at 2 mA cm^{-2} and 1 mAh cm^{-2} , and the Zn//Cu asymmetric cell also exhibits 1900 cycles with almost 100% coulomb efficiency.

To overcome the inherent limitations of purely anionic/cat-ionic additives in stabilizing the Zn anode, Wang et al.^[80] introduced a zwitterion, sulfonated amphoteric betaine (DMAPS)

into the ZnSO_4 electrolyte, which effectively addressed the uni-directional modulation associated with conventional anionic/cat-ionic additives (Figure 4a). Experimental and DFT calculations demonstrate that the $-\text{NR}_4^+$ and $-\text{SO}_3^-$ moieties within the DMAPS molecular chain readily dissociate under an applied electric field, thereby promoting enhanced ion transport in the electrolyte. Specifically, the $-\text{NR}_4^+$ group adsorbs on the Zn surface leads to the formation of a water-deficient Helmholtz layer, which effectively suppresses both Zn corrosion and dendritic growth.^[84,85] Concurrently, $-\text{SO}_3^-$ as a “gripper” to anchor Zn^{2+} , thereby fundamentally destroying the solvated sheath structure of Zn^{2+} (Figure 4b,c).^[86–88] Thanks to the unique multifunctionality of DMAPS, the Zn anode in the Zn//Zn symmetric cell assembled with DMAPS + ZnSO_4 has a significantly suppressed corrosion current and corrosion potential compared to those using pristine ZnSO_4 electrolyte (Figure 4d). Meanwhile, and Zn//Cu cell employing a $\text{ZnSO}_4 + \text{DMAPS}$ as the electrolyte can be cycled stably for 3000 h with an average CE of over 99% (Figure 4e). Furthermore, Zn//NPHC ZHS using DMAPS + ZnSO_4 electrolyte can achieve capacity retention of more than 90% at

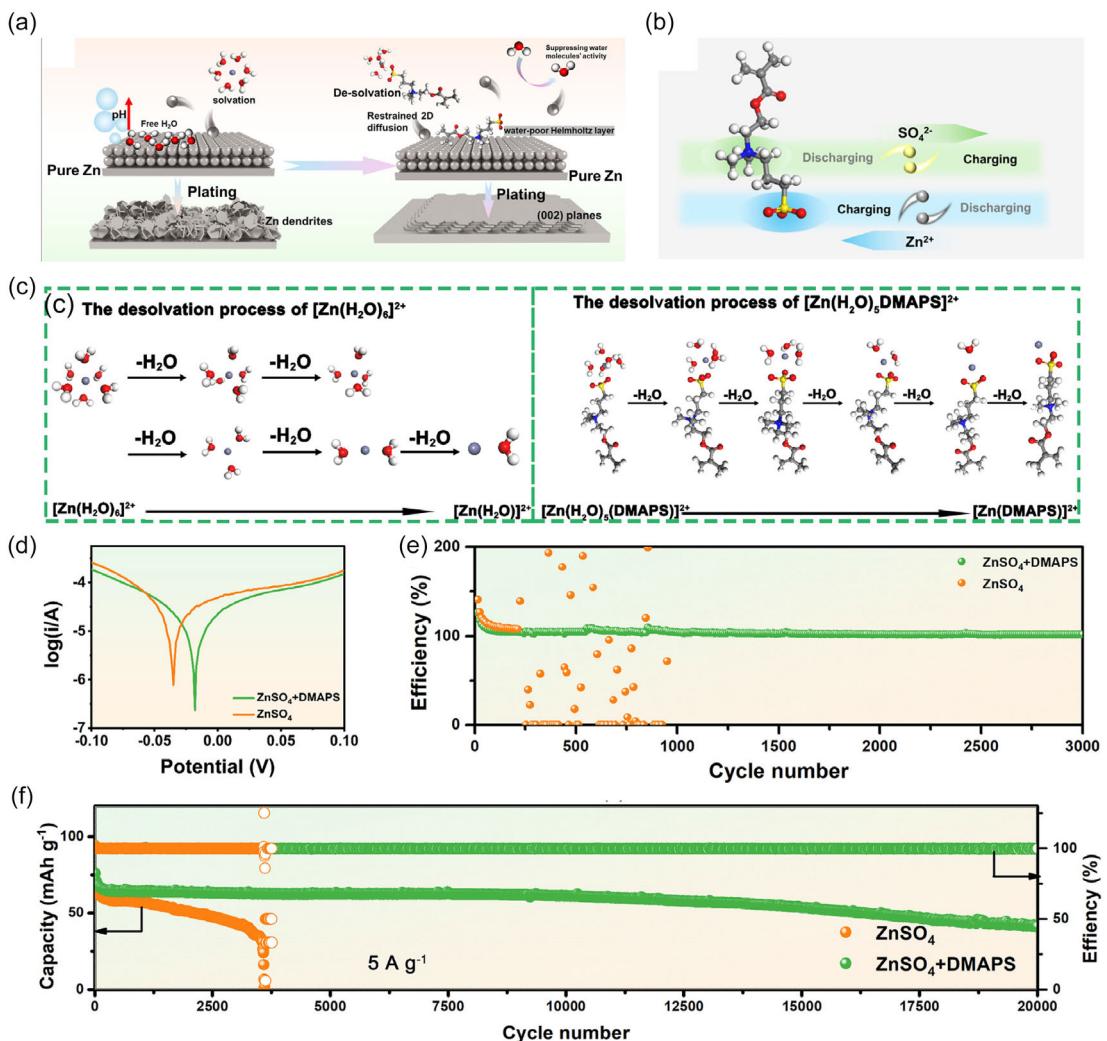


Figure 4. a) Schematic diagram of DMAPS as an electrolyte additive that is used to regulate the electrolyte environment and interface engineering, b) Schematic representation of the interaction between DMAPS molecules and other molecules, c) Simulation routes for desolvation of two solvated sheath structures, d) Tafel test of a Zn//Zn symmetry cell in two different electrolytes, e) Coulombic efficiency of Zn//Cu cell in different electrolytes, f) Long-term plots illustrating the performance of Zn//NPHC ZIHGs in various electrolytes.^[80]

5 A g⁻¹ and maintain high CE of about 100% after 20,000 cycles, significantly exceeding that of ZHS using pure ZnSO₄ electrolyte (Figure 4f). Similarly, Tao et al.^[89] investigated a bifunctional interface was constructed with the N-decyl-N, N-dimethyl-3-ammonia-1-propane sulfonic acid zwitterion. On the Zn electrode, the interface forms a self-assembled hydrophobic layer and a polarized layer. Among them, the electric field generated by the polarized layer can effectively weaken the “tip effect”, which is a key issue in electrochemical processes.^[90] Specifically, the weakening of the “tip effect” can accelerate the transfer of Zn²⁺ and enhance electroplating/stripping process of Zn, thus improving the reaction efficiency. In addition, the hydrophobic layer inhibits the formation of Zn hydroxyl sulfate, which often affects cell performance and stability. The implementation of these two layers significantly improved the cycling stability of the battery.^[91]

In addition, polymer additives also contribute significantly to the stabilization of Zn anode. By constructing cross-linked network structure at the electrode surface, polymers serve as a

barrier that effectively prevents the direct contact between the electrolyte and the electrode, thus suppressing the occurrence of side reactions and enhancing interfacial integrity.^[92] At the same time, the polymer can also regulate the ionic conductivity and viscosity of the electrolyte, thereby optimizing the overall performance of the battery. For example, Huang et al. proposed an “amphoteric polymer strategy” with buffer-adsorption mechanism utilizing the amphiphilic polymer poly (L-glutamic acid) (PGA), effectively addresses key challenges in Zn²⁺ batteries, including poor reversibility and the parasitic HER (Figure 5a).^[93] The experimental results show that the abundant amphoteric functional groups (-COOH, -NH₂, -CONH₂) in PGA can buffer the pH value in the electrolyte to stabilize the electrolyte and further reduce the by-product generation/accumulation. At the same time, PGA can be adsorbed on the Zn anode surface to improve the deposition behavior of Zn.^[94] When using PGA + ZnSO₄ electrolyte, the assembled Zn//Zn symmetric cells can work stably for more than 1600 h at 2 mA cm⁻² and

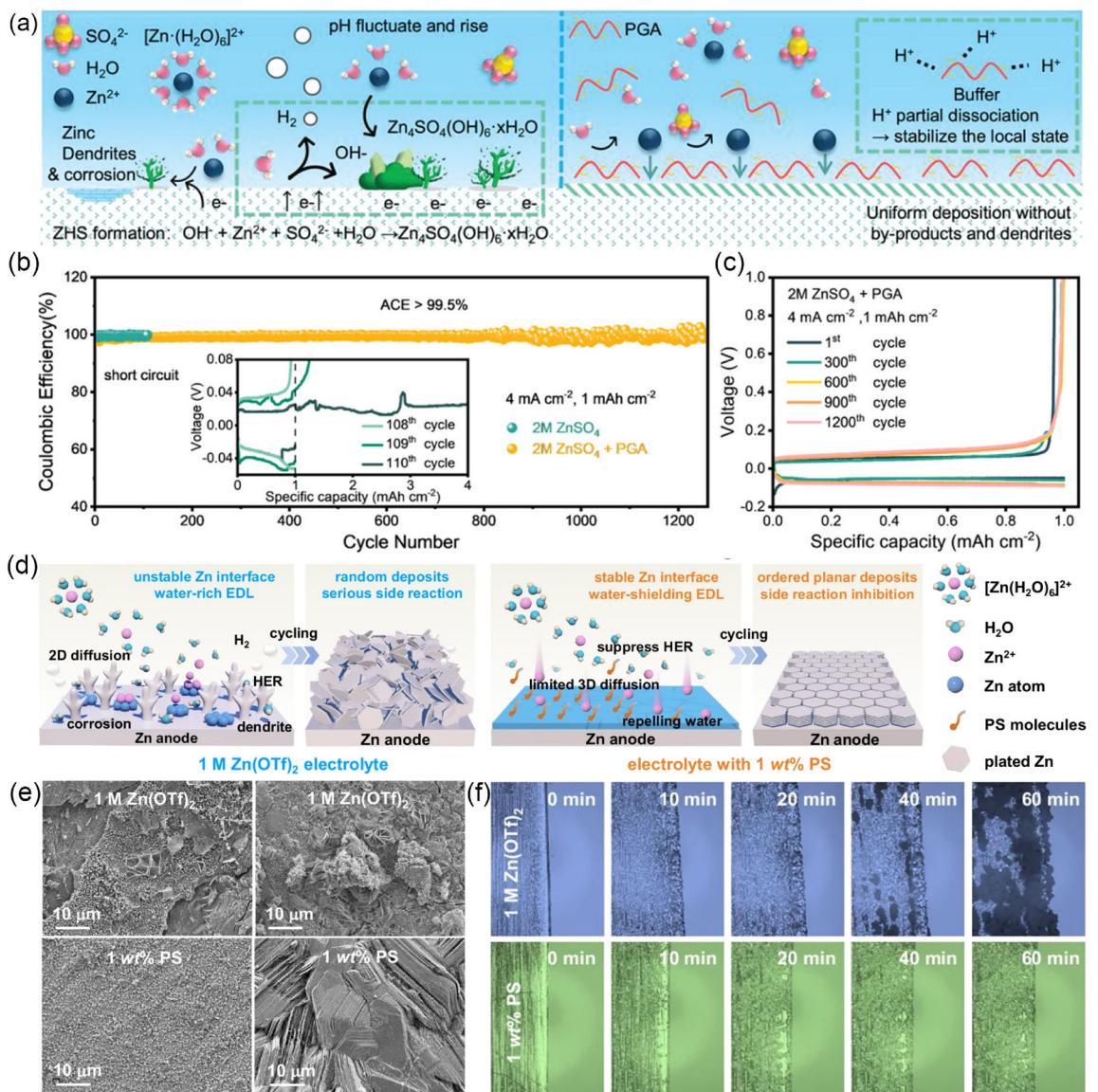


Figure 5. a) Schematic diagram demonstrating the undesired parasitic reactions (left) and the buffer-assisted stabilization(right) of the internal environment, b) CE of Zn//Cu cells at 4 mA cm^{-2} with a capacity of 1 mAh cm^{-2} and corresponding voltage profiles at various cycles in ZnSO_4 electrolyte with PGA at c) 4 mA cm^{-2} , 1 mAh cm^{-2} , [95] d) 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, e) SEM images of Cu electrodes and Zn anodes in 1 M Zn(OTf)_2 electrolyte and electrolyte with 1 wt% PS after 50 cycles at 2 mA cm^{-2} and 2 mAh cm^{-2} , f) three-dimensional in situ Raman spectra of v-O-H during Zn plating process.^[97]

1 mAh cm^{-2} , which is 10 times longer than that of the bare cells. Under the 4 mA cm^{-2} and 1 mAh cm^{-2} , the Zn//Cu asymmetric cell employing a PGA + ZnSO_4 as the electrolyte can be stably cycles for 1250 times with the average CE maintained at 99.5%, and show stable high reversibility and low voltage polarization. In contrast, the bare cell without PGA failed after only 100 cycles (Figure 5b,c).

The in-situ formation of Znophilic nanoprotective interlayers on the Zn anode surface has emerged as an effective strategy to suppress dendritic growth and mitigate parasitic side reactions. Zhang et al. incorporated perfluoropolymer (Nafion) as an additive into an aqueous electrolyte to construct a Zn^{2+} directed transport channel, offering a valuable design strategy for

engineering an ultra-stable Zn/electrolyte interface.^[95] Due to the low adsorption energy of Nafion molecules on Zn metal (-2.09 eV), Nafion nanofilms were spontaneously formed on the Zn anode surface during the initial charging cycle.^[96,97] This ultrathin interfacial layer with Znophilic $-\text{SO}_3$ functional groups still guides the directional electrodeposition of Zn^{2+} along the (002) crystal surface under high current density, thus inhibiting the formation of Zn dendrites.^[96,98] Electrochemical evaluations revealed that Zn//Cu cell employing the Zn(OTf)_2 -NAF electrolyte delivered an average CE of 99.71% over 4500 cycles. Moreover, symmetric Zn//Zn cell with Zn(OTf)_2 -NAF electrolyte operated stably for over 7000 h at 5 mA cm^{-2} . These performances markedly surpass those achieved with the

Zn (OTF)₂ electrolyte alone, highlighting the significant advantages conferred by the incorporation of Nafion. In addition, Peng et al. proposed a rational strategy for engineering water-rich electrical double layer (EDL) and SEIs in ZIBs by employing large-sized nonionic amphiphilic polysorbate (PS) as functional electrolyte additive (Figure 5d). Combined theoretical calculations and experimental results demonstrate that PS molecule exhibits preferential chemisorption and directional alignment on the Zn anode surface, leading to the formation of an EDL that effectively suppresses parasitic reactions.^[99] Meanwhile, the PS molecule facilitate the construction of an SEI, which promote a more uniform electric field distribution and guide the orientation Zn deposition, thereby enabling highly reversible plating/stripping behavior and significantly enhancing Zn utilization (Figure 5e,f).^[24] Based on the above advantages, the Zn//Zn symmetric cell incorporating the PS-containing electrolyte delivers an extended cycling stability of up to 8060 h at 1 mA cm⁻² and 1 mAh cm⁻². Furthermore, Zn//Cu cell with PS-containing electrolyte exhibit a high CE of 99.2% and outstanding cycling stability exceeding 3900 cycles. When assembled into full cells with V₂O₅/rGO cathode, the Zn/V₂O₅/rGO maintains excellent electrochemical activity and stability.

Recently, Ren et al. introduced biocompatible betaine into aqueous electrolytes, restructuring the solvation shell through "molecular crowding" to maintain high ionic conductivity at -30 °C and induce Zn (002)-oriented deposition.^[100] The same team further developed a Water-in-DMSO deep eutectic system, which generates ZnF₂-rich SEI *in situ*, achieving more than 10,000 h of dendrite-free cycling and 60% DOD deep discharge. These two approaches respectively demonstrate the new paradigms of "green small molecule crowding" and "deep eutectic solvation," suggesting that the coupling of organic additives with deep eutectic strategies may become a promising direction for next-generation aqueous Zn electrolyte design that balances low-temperature adaptability and high depth of discharge.^[101]

Organic salt additives, owing to their unique molecular structure and unable functional groups have demonstrated notable efficacy in modulating the interfacial chemistry of Zn anode. Cui et al. used green and low-cost carboxymethyl cellulose sodium (CMC-Na) as an additive to the ZnSO₄ electrolyte to effectively enhance the cycling stability of ZHS.^[102] Compared to Zn²⁺, Na⁺ in CMC-Na exhibit a lower redox potential, which promotes the formation of an electrostatic protection layer on the Zn anode surface, thus inducing the (002) crystal plane-oriented deposition of the Zn²⁺ during electrochemical cycling.^[103] In addition, the abundant hydrophilic functional groups (-OH and -COO-) in CMC-Na modulate the structure of Zn²⁺ solvation sheath by disrupt the hydrogen-bonding network of the original electrolyte, which effectively suppresses dendrite formation and mitigates parasitic side reactions.^[81,104] Electrochemical test results show that the Zn//Zn cells assembled with ZnSO₄+CMC-Na (0.13) electrolyte could be stably cycled for more than 1600 h at 4 mA cm⁻² and 1 mAh cm⁻², which is much better than the cells without additive.

To address the limitations of poor electrolyte compatibility, intrinsic flammability and limited regulatory functionality of

conventional organic additives, Peng and co-workers introduced a sodium salt of POPSO (POPSO-Na) as an additive into an aqueous ZnSO₄ electrolyte,^[105] which effectively suppressed Zn dendrite formation, HER, thereby significantly enhancing the electrochemical stability and reversibility of the Zn anode (Figure 6a). Specifically, the POPSO-Na molecule with high hydrophilic and adsorption capacity can partially displace the structural water coordinated to Zn²⁺ and reduce the activity of H₂O molecules, thereby mitigating HER (Figure 6b).^[106,107] Moreover, the Na⁺ in POPSO-Na contribute to form an interfacial protective layer on the cathode surface, which replaces the structural water around the Zn²⁺ and promotes a more homogeneous diffusion of Zn²⁺, thus effectively inhibiting the dendritic growth (Figure 6c, d).^[102] Electrochemical measurements reveal that Zn//Zn and Zn//Cu cells employing ZnSO₄+POPSO-Na electrolyte exhibit markedly enhanced cycling stability, sustaining stable operation for over 1600 and 2000 h, respectively. In contrast, cells utilizing ZnSO₄ alone display significantly inferior longevity (Figure 6e). The ZHS with ZnSO₄+POPSO-Na electrolyte demonstrates exceptional cycling stability, retaining 99.6% of its initial capacity after 9000 cycles at a current density of 4 A g⁻¹, which exceeds the performance of the ZIHS assembled with ZnSO₄ electrolyte only under the same conditions.^[108]

3.3. Surfactant Additives

Surfactants play a significant role in aqueous ZIHSs by forming an adsorbent layer on the electrode surface and improving electrode wettability and ion transport properties.^[109,110] The surfactants can form oriented molecular layers to increase the hydrophilicity of the electrode and enhance the rate of ion adsorption and desorption.^[111,112] Furthermore, surfactants can also regulate the surface tension and viscosity of the electrolyte, contributing to enhance the cycling stability of aqueous ZIHSs.^[113,114]

In 1998, Kan et al. have reported the key role of surfactants in enhancing the stability of Zn anode. Experimental results showed that the incorporation of Triton X-100 at concentrations ranging from 0.02 to 500 ppm into an electrolyte composed of 2.5 M ZnCl₂ and 2.0 M NH₄Cl (pH = 4.0) markedly suppresses Zn dendrite growth during charge/discharge process,^[115] and ultimately enhance the cycling stability of the Zn-polyaniline secondary battery.^[116] Later, quaternary ammonium surfactants (cetyltrimethyl ammonium bromide, dodecyltrimethylammonium bromide, etc.) as representative cationic surfactants, were also identified as effective electrolyte additives capable of suppressing dendrite formation and mitigating parasitic side reactions, thereby significantly enhancing the stability of Zn anode.^[117]

Zhang et al. introduced a cost-effective cationic surfactant, dodecyltrimethylammonium chloride (DTAC), into a conventional ZnSO₄ electrolyte to improve the interfacial stability and cycling performance of the Zn anode (Figure 7a).^[116] Experimental results reveal a progressive blue shift in the O-H stretching vibration peaks with increasing DTAC concentration, implying that the DTAC additive can disrupt the hydrogen-bonding network and modulates the local solvation structure of H₂O (Figure 7b).

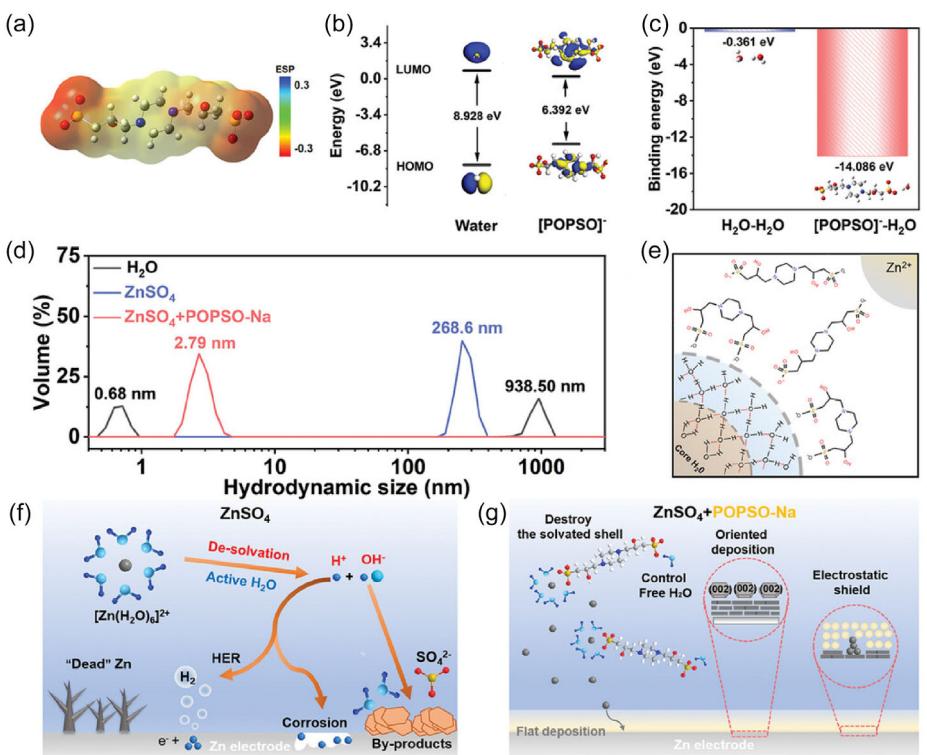


Figure 6. a) Electrostatic potential diagram of $[POPSO]^-$. b) schematic diagram of reverse micelle structure in electrolyte. Diagram of $ZnSO_4$ and $ZnSO_4+POPSO-Na$ electrolyte, c) in situ optical microscope images of galvanizing processes at current density of 10 mAh cm^{-2} in different electrolytes, d) the binding energy of $Zn\text{-H}_2\text{O}$ and $Zn\text{-}[POPSO]$, e) long-term cycling performance of $Zn//Zn$ batteries using two different electrolytes.^[105]

Furthermore, DFT calculations demonstrate that DTA^+ preferentially adsorb in a vertical orientation on the Zn anode surface, leading to the formation of a hydrophobic layer composed of dodecyl chains, which effectively inhibits interfacial side reactions (Figure 7c). Benefiting from the protection of the hydrophobic layer, the Zn^{2+} nucleation overpotential is elevated, while 2D diffusion at the anode surface is limited, thereby promoting the uniform deposition of Zn and effectively suppressing dendritic growth (Figure 7d). Electrochemical test results show that the $Zn//Zn$ symmetric cells with DTAC additive can operate for 2000 h at 1 mA cm^{-2} and 1 mAh cm^{-2} . Moreover, the $Zn//MnO_2$ full cell assembled with DTAC/ $ZnSO_4$ as the electrolyte showed an initial capacity of $149.44 \text{ mAh g}^{-1}$ at 5 A g^{-1} , and a capacity retention of 83.02% after 2000 cycles, significantly better than that of the cell assembled with a pure $ZnSO_4$ electrolyte, which fully demonstrated the key role of DTAC in stabilizing the Zn negative electrode (Figure 7e,f).

In addition, Zheng and co-workers incorporated various cationic ammonium-based additives into a conventional aqueous electrolyte,^[118] effectively modulating the interfacial properties of Zn anodes, and thus markedly enhanced the reversibility of Zn metal plating and stripping processes. By integrating theoretical calculations, molecular dynamics simulations, and experimental investigations, it was demonstrated that tetramethylammonium cations with well-defined molecular geometry and charge distribution can effectively regulate the solvation shell structure of $Zn(H_2O)_6^{2+}$. Thus, the $Zn//Zn$

symmetric cell assembled with BE + TMA can be stably cycled for over 4000 h at 0.5 mA cm^{-2} and 0.5 mAh cm^{-2} . Similarly, the $Zn//Cu$ cell can be stably operated for 2100 cycles under the same operating conditions with an average CE of 99.8%, which is far superior to that of the most recent additives reported to date.

In addition, anionic surfactants have demonstrated the ability to effectively suppress the HER and corrosion, while simultaneously modulating Zn^{2+} deposition behavior, owing to their strong hydrophilicity and favorable interfacial adsorption properties. For example, Wei et al. investigated the role of highest occupied molecular orbital (HOMO) energy levels in modulating coordination and interfacial adsorption by selecting three representative non-sacrificial anionic surfactants as additives, including sodium dodecylbenzene sulfonate (SDBS), sodium dodecyl sulfate (SDS), and sodium p-ethylbenzene sulfonate (SEBS).^[110,119] Combined experimental and DFT calculations revealed that SDBS exhibits the strongest coordination and adsorption effects than SDS and SEBS, which is attributed to its highest HOMO energy level (Figure 8a). The incorporation of SDBS into the $ZnSO_4$ electrolyte effectively modulates the Zn^{2+} solvation structure and decreases water activity, thereby significantly expanding the electrochemical stability window and suppressing the dendrite formation and HER (Figure 8b-d). Electrochemical measurements reveal that the $Zn//Zn$ symmetric cell employing a SDBS + $ZnSO_4$ electrolyte exhibits an exceptional cycling stability exceeding 3200 h at 2 mA cm^{-2} and 2 mAh cm^{-2} , this lifetime is

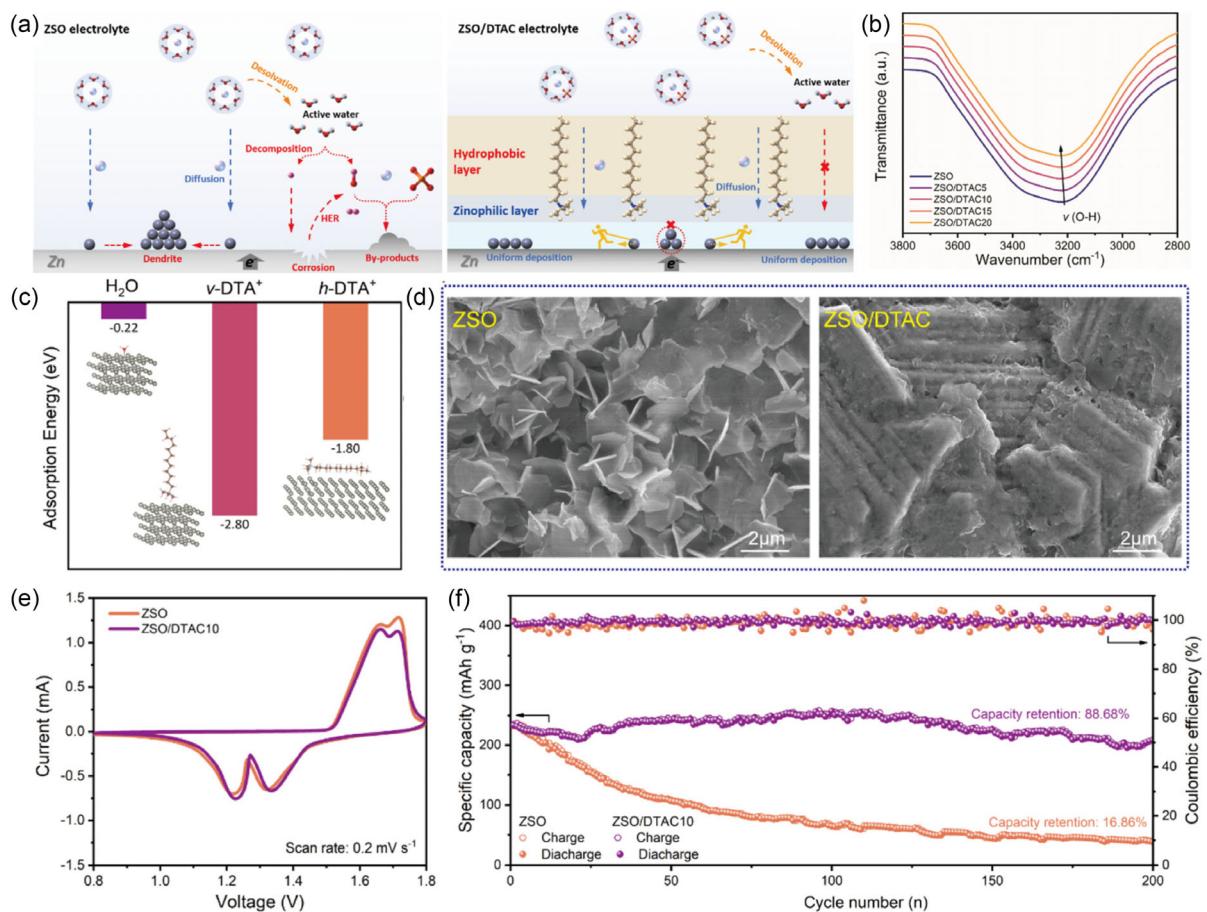


Figure 7. a) Schematic illustration of different reaction processes of Zn²⁺ solvation structure and the corresponding interaction between Zn anode and electrolyte in ZSO and ZSO/DTAC electrolytes, b) FTIR spectra of ZSO electrolytes with different DTAC concentration, c) the adsorption configuration and adsorption energy of H₂O and DTA⁺ cations on Zn (002) facet, d) SEM images of the Zn foil surface after cycling in the two electrolytes, e) CV curves of Zn//MnO₂ full cells in ZSO and ZSO/DTAC10 electrolytes at a scan rate of 0.2 mV s⁻¹, f) cycling performance at the current density of 0.1 A g⁻¹.^[116]

approximately 30 times longer than that of the cell with ZnSO₄ electrolyte alone (Figure 8e). Similarly, the assembled Zn//Cu cell exhibits a high CE of 98.15% after 800 cycles at 0.2 mA cm⁻² and 0.2 mAh cm⁻². In Figure 8f, the Zn//NH₄V₄O₁₀ pouch cell using SDBS + ZnSO₄ showed a high discharge capacity of 214 mAh g⁻¹ over 500 cycles, which far exceeded the discharge capacity of pure ZnSO₄.

To elucidate the influence of surfactant charge on Zn anode behavior, Xie et al. employed cetyltrimethylammonium bromide (CTAB) and SDBS as representative cationic and anionic additives, respectively, to construct positively and negatively charged self-assembled interfacial layers on the Zn surface.^[113] Experimental results reveal that introduction of CTAB induces electrostatic repulsive forces, which control the three-dimensional diffusion process of Zn²⁺ and produces loose Zn deposits. In contrast, the addition of SDBS enhances the electrostatic attraction with Zn²⁺ and facilitates a two-dimensional diffusion process, which favors the formation of a dense dendrite-free on the Zn anode surface, and ultimately improved the reversibility of the Zn anode in the ZnSO₄+SDBS electrolyte containing. As a result, the Zn//Zn cell with SDBS exhibits a cycle life of over 1800 h at 4 mA cm⁻², and a capacity retention of 85% after 1000 cycles at 1 A g⁻¹ for

the Zn//MnO₂ battery. Furthermore, it was successfully predicted that SDS and disodium lauriminodipropionate (DSL) could also produce similar dendrite-free morphology and highly reversible Zn anode by modeling both instantaneous nucleation and continuous nucleation models.

3.4. Organic-Inorganic Composite Additives

In aqueous Zn²⁺ batteries, single organic or inorganic additives often only unilaterally modulate the electrolyte solvent sheath or metal interface, making it difficult to simultaneously suppress dendrite formation, hydrogen evolution, and side reactions. In recent years, researchers have proposed an “organic-inorganic composite additive” strategy: organic functional groups are used to restructure the Zn²⁺ solvation structure, reducing the desolvation barrier; inorganic ions or nanocomposite components are then used to establish an electrostatic shielding layer and a physical barrier at the interface, thereby synergistically improving cycling stability and Coulombic efficiency. This strategy is considered one of the most promising electrolyte design approaches due to its simple process and scalable preparation. Using natural sodium humate (NaHA) as a model system,^[120] Wang et al.

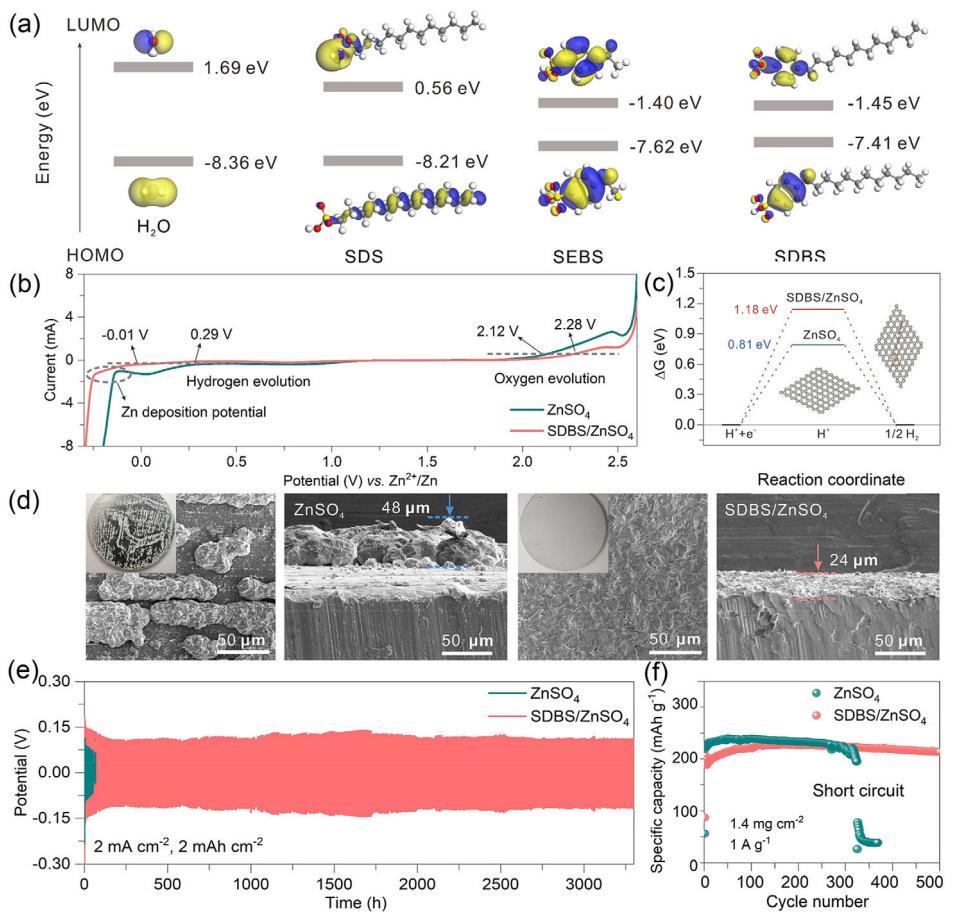


Figure 8. a) The LUMO and HOMO isosurfaces of different molecules, b) the LSV profiles of ZnSO_4 and SDBS/ZnSO_4 electrolytes, c) the hydrogen barrier of the ZnSO_4 and SDBS/ZnSO_4 electrolytes, d) the surface and cross-sectional SEM images of deposited Zn in the ZnSO_4 and SDBS/ZnSO_4 electrolytes at 10 mA cm^{-2} for 1 h, e) voltage profiles of Zn/Zn symmetric batteries at different current densities 2 mA cm^{-2} , f) the cycling performance of $\text{Zn}/\text{NH}_4\text{V}_4\text{O}_{10}$ pouch batteries at 1 A g^{-1} .^[119]

demonstrated a typical organic–inorganic synergistic effect (Figure 9a): its hydrophilic carboxyl groups strongly coordinate with Zn^{2+} , breaking the $[\text{Zn}(\text{H}_2\text{O})_6]^{2+}$ solvent sheath and reducing active water molecules; the hydrophobic aromatic-alkane skeleton adsorbs on the zinc surface, forming a water-poor organic protective film; and the Na^+ forms an electrostatic shielding layer at high electric fields, homogenizing interfacial ion flow (Figure 9b,c). DFT and MD calculations confirm that this additive increases the Zn^{2+} diffusion coefficient by approximately 10% and significantly reduces the desolvation barrier (Figure 9d,e). Experimentally, a $1 \text{ mA cm}^{-2}/1 \text{ mAh cm}^{-2}$ symmetric Zn/Zn battery exhibits a lifespan exceeding 600 h, a Zn/Cu battery achieves an average Coulombic efficiency of $\approx 99\%$ over 1600 cycles, and a Zn/AC hybrid capacitor retains 90% of its capacity after 10,000 cycles at 3 A g^{-1} (Figure 9f). Furthermore, the deposited layer exhibits a dense (002) orientation and is dendrite-free, demonstrating excellent scalability.

NaHA, with its “single molecule, dual functionality,” achieves all the elements of organic–inorganic synergistic regulation, balancing cost, sustainability, and scalability. It establishes a new paradigm for the integrated “molecular design-interface engineering-device validation” approach for aqueous Zn^{2+}

electrolyte additives and provides a replicable research framework for subsequent composite additive systems.

4. Summary and Outlook

Zn^{2+} energy storage devices are considered promising candidates for next-generation energy storage owing to their intrinsic safety, cost-effectiveness, and environmental benignity. Nevertheless, the practical deployment of their remains hindered by persistent challenges associated with Zn metal anodes, including uncontrolled dendrite formation, parasitic side reactions among the various strategies explored to mitigate these issues, particularly through the rational incorporation of functional additives, has emerged as a highly effective and adaptable approach, offering opportunities to modulate interfacial chemistry and stabilize Zn deposition behavior. In this review, we comprehensively summarize recent progress in electrolyte additive engineering from inorganic additives, organic additives, and surfactant-type additives for Zn^{2+} energy storage devices. We highlight the distinct physicochemical functions of each class in regulating interfacial processes and promoting the electrochemical stability of

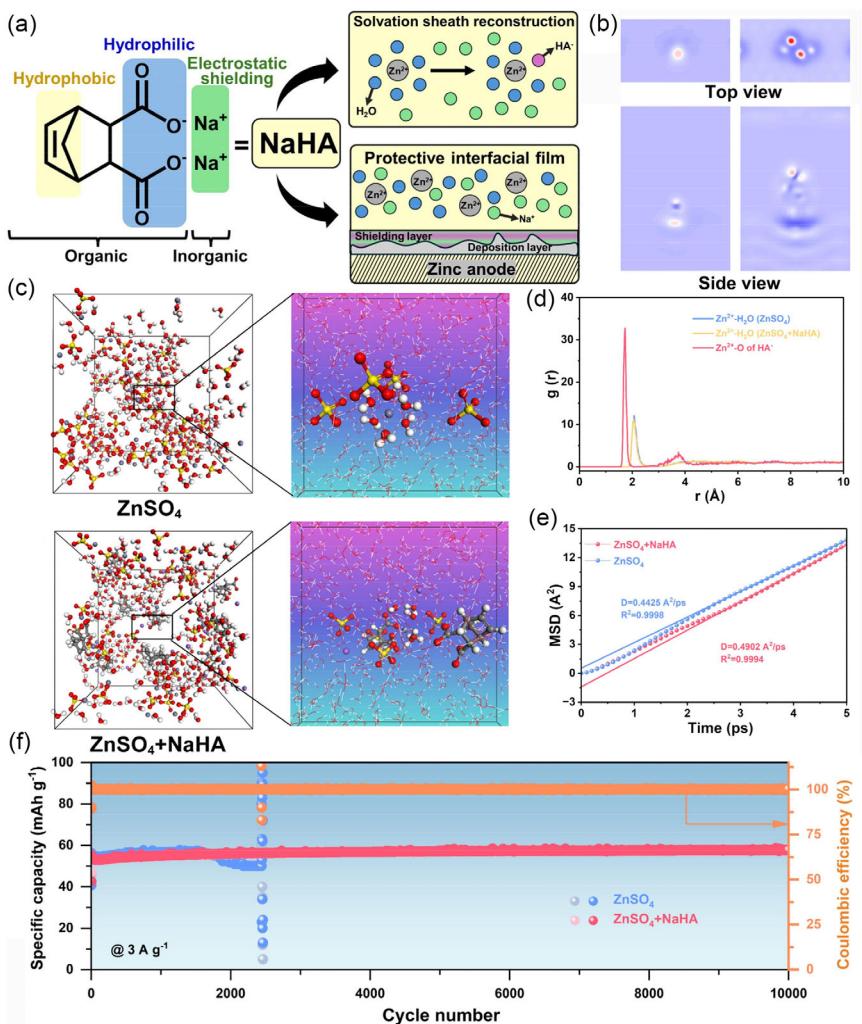


Figure 9. a) Mechanism of NaHA as an electrolyte additive to change the electrolyte environment and interface environment, b) charge density difference of H₂O and HA⁻ upon adsorption on Zn anode surface, c) ZnSO₄ + NaHA and ZnSO₄ electrolyte model, d) RDFs of Zn²⁺-O (H₂O) in different electrolyte models, e) MSD of Zn in two electrolyte models, and f) cycle stability of ZICs with different electrolytes.^[120]

Zn anode. Despite these encouraging advances, several critical challenges remain unresolved, and we further discuss future opportunities and design principles that warrant consideration for the practical implementation of Zn²⁺ energy storage devices. 1. With the progressive elucidation of the failure mechanisms associated with Zn anode, it is evident that current additives exhibit limited effectiveness in suppressing Zn dendrite formation and fail to substantially enhance the cycling stability and reversibility. Therefore, it is necessary to explore more organic compounds or nanomaterials with multifunctionality as additives. In particular, the development of advanced additives that concurrently suppress dendrites formation, enhance Zn deposition reversibility, and maintain robust performance across a wide temperature range is essential to meet diverse operational of Zn²⁺ energy storage devices. 2. Currently, traditional additive research mostly relies on the experimental trial-and-error method, which has bottlenecks such as low efficiency, extended development cycles and high cost. In contrast, the rational design strategies guided by DFT provides a new paradigm for the discovery and

optimization of functional additives. DFT enables quantitative evaluation of key molecular interactions, including adsorption energy, coordination strength, and molecular orbital level between additive molecules and Zn anode. These calculations can effectively predict the interfacial modulation tendency and electrochemical stability, thereby facilitating rapid screening and performance evaluation of additives while significantly narrowing experiments scope. Combined with high-throughput experimental platforms and in-situ characterization techniques, the additives development process can be markedly accelerated. 3. Research on Zn²⁺ energy storage devices involves several disciplinary fields, including materials science, electrochemistry, computational science. Given the complexity and multifaceted nature of the challenges involved, it is increasingly clear that no single disciplinary method can comprehensively and deeply solve all the problems. By leveraging the complementary strengths and methodologies of diverse fields, it becomes possible to achieve systematic optimization spanning from the rational design of material microstructures to the enhancement of

macroscopic battery performance. 4. In the development of new additives, full consideration should be given to their cost, environmental friendliness and scalability to ensure their feasibility in practical applications. For example, the use of abundant and low-cost additive raw materials can effectively reduce the cost of ZIHSS and ZIBs. At the same time, focusing on the environmental impact of additives and selecting environmentally friendly additives to reduce environmental pollution is crucial to promoting the sustainable development of ZIHSS and ZIBs.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords: electrolyte additives · hydrogen evolution reaction · Zn dendrites · Zn^{2+} energy storage devices

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