

High-flux horizontal ZHS layer and $\text{Zn}_{(101)}$ textured deposition enabled by alkyl glycoside molecules: A dual-effect interfacial regulation strategy for ultra-stable zinc anodes

Liang Tan¹✉, Min Zuo¹, Shijing Ou¹, Jiaqi Yang¹, Wenjin Ni^{1,2}, Ruiqing Long¹, Yijin Wu¹, Xijun Xu³✉, and Renzong Hu⁴✉

¹Hunan Provincial Key Laboratory of Functional Metal–Organic Compounds, School of Chemistry and Materials Science, Hengyang Normal University, Hengyang 421008, China

²School of Chemical Engineering, Xiangtan University, Xiangtan 411105, China

³School of Chemical Engineering and Light Industry, Guangdong University of Technology, Guangzhou 510006, China

⁴Guangdong Provincial Key Laboratory of Advanced Energy Storage Materials, School of Materials Science and Engineering, South China University of Technology, Guangzhou 510640, China



Cite this article: Nano Research, 2026, 19, 94908606. <https://doi.org/10.26599/NR.2026.94908606>

ABSTRACT: Rechargeable aqueous zinc-ion batteries (AZIBs) are considered the most promising electrochemical energy storage technologies due to their high safety and low cost. However, their practical application is hindered by persistent challenges at the Zn anode, including thermodynamic corrosion, hydrogen evolution, and dendrite growth in aqueous electrolytes. In this work, a nonionic molecule, n-octyl β -D-glucoside (8TG), is investigated as an additive for ZnSO_4 (ZS) electrolyte system. Experimental and theoretical calculations confirm that 8TG molecules tend to adsorb on the (001) plane of zinc hydroxide sulfate (ZHS), lowering its interfacial energy and thereby inducing lateral growth to form a dense protective layer. This layer exhibits an interlayer spacing of $> 8 \text{ \AA}$ and is rich in anions, which can lower the energy barrier for ion transport through an electrostatic repulsion–attraction effect. Furthermore, 8TG preferentially adsorb on the $\text{Zn}_{(002)}$ and $\text{Zn}_{(100)}$ facets during zinc electrodeposition. Through a spatial confinement effect, it guides the preferential deposition of Zn^{2+} along the $\text{Zn}_{(101)}$ plane, effectively suppressing dendrite growth. Benefiting from these multifunctional effects of 8TG, $\text{Zn}||\text{Zn}$ symmetric cells achieve a lifespan exceeding 2500 h at $2 \text{ mA} \cdot \text{cm}^{-2}$ and $1 \text{ mAh} \cdot \text{cm}^{-2}$, and maintain stable operation over 400 h even under $10 \text{ mA} \cdot \text{cm}^{-2}$ and $5 \text{ mAh} \cdot \text{cm}^{-2}$. Moreover, $\text{Zn}||\text{I}_2/\text{MnO}_2@\text{activated carbon (AC)}$ full cells assembled with 8TG additive exhibit nearly 100% capacity retention after 500 cycles. This work systematically elucidates the influence of 8TG additive on the growth orientation of ZHS and Zn dendrites, providing new insights into exploring electrolyte additives for high-performance AZIBs.

KEYWORDS: aqueous Zn-ion batteries, Zn anode, electrolyte additives, n-octyl β -D-glucoside, zinc hydroxide sulfate

1 Introduction

Aqueous zinc-ion batteries (AZIBs) are widely regarded as a promising candidate for next-generation grid-scale energy storage

systems, owing to their inherent advantages of operational safety, eco-friendliness, and abundant Zn resources [1–3]. However, their industrialization is hampered by continuous deteriorating interfacial issues of Zn anode, including the hydrogen evolution reaction (HER), uncontrollable parasitic reactions, and irreversible Zn dendrite growth during cycling [4–6]. Therefore, it is essential to explore a feasible strategy for constructing a long-term stability interface for Zn anode, which could not only avoid the contact of active H_2O molecules with the Zn anode to suppress the parasitic reactions but also facilitate highly efficient Zn^{2+} transport across the interface.

Received: December 15, 2025; Revised: February 14, 2026

Accepted: February 27, 2026

✉Address correspondence to Liang Tan, tanl1991@hynu.edu.cn; Xijun Xu, xuxijun2022@gdut.edu.cn; Renzong Hu, msrenzonghu@scut.edu.cn

Up to now, various approaches have been explored to settle interface problems of Zn anode, including artificial interface engineering [7–10] and electrolyte modification strategies [11–15]. For the former, which utilizes micro or mesoporous materials (e.g., metal–organic frameworks (MOFs) [16, 17], covalent organic frameworks (COFs) [18], or molecular sieves [19]), has demonstrated promising interface stabilization by simultaneously isolating the Zn anode from electrolyte while providing rapid ion transport pathways [20, 21]. Nevertheless, their practical application is severely hindered by the high synthesis costs and mechanical stress concentration at the coating interface, which inevitably leads to interlayer delamination during cycling [22–25]. This creates an urgent demand for low-cost and stable interface materials with a simple manufacturing process. Further, electrolyte modification, common approaches include highly concentrated “water-in-salt” systems [26], deep eutectic electrolytes [27], the incorporation of diluents [28], and the introduction of functional additives [29–31]. Among these, electrolyte additives stand out due to their cost-effectiveness and precise regulation, offering distinct advantages. They can preferentially adsorb onto the Zn anode surface through electrostatic interactions or undergo self-decomposition to form a dense solid electrolyte interphase (SEI)-like layer [32–34], effectively shielding the electrode from direct contact with H_2O molecules and thereby alleviating HER and corrosion reactions [35]. Additionally, these additives participate in reconstructing the solvation structure of Zn^{2+} , reducing the number of H_2O molecules in the primary solvation sheath and thus diminishing H_2O reactivity at the source. However, it is important to acknowledge that the inherent thermodynamic tendency of Zn toward spontaneous HER in aqueous media leads to persistent interfacial alkalization [36], making it challenging to entirely prevent the formation of byproducts, such as zinc hydroxide sulfate (ZHS, $\text{Zn}_4(\text{OH})_6\text{SO}_4 \cdot n\text{H}_2\text{O}$) during long-term cycling [37, 38]. Therefore, in the selection and design of additives, alongside their role in suppressing side reactions and promoting uniform Zn deposition, proactive consideration of their ability to modulate the crystallization morphology, distribution, and properties of ZHS is also a crucial aspect for enhancing long-term cycling stability.

Notably, ZHS [39], a naturally occurring byproduct on the Zn anode surface, exhibits unique potential as an artificial interfacial layer. Its intrinsic chemical inertness prevents additional parasitic reactions with the Zn substrate or electrolyte, and its bridge provides a strong contact with the Zn anode, circumventing localized stress [40]. Significantly, the brucite-like layered structure of ZHS with an interplanar spacing of $\sim 8 \text{ \AA}$ along the $\text{ZHS}_{(001)}$ plane [41], which creates nanochannels for domain-limited dehydration. These channels are size-matched to accommodate $[\text{Zn}(\text{H}_2\text{O})_6]^{2+}$ ions ($\sim 8.4 \text{ \AA}$ in diameter) [42, 43], thereby capably lowering the desolvation energy barrier. As previously reported, the intercalation of poly(dimethyldiallylamine) ions via sodium displacement enlarged the interlayer spacing of layered montmorillonite coatings, which subsequently facilitated the desolvation of Li^+ [44]. Meanwhile, a zinc benzene-1,3,5-tricarboxylate (Zn-BTC) MOF layer featuring $9.0 \text{ \AA}$ channels promoted partial dehydration of hydrated Zn^{2+} [45], whereas a zeolitic imidazolate framework-7 (ZIF-7) MOF coating with $2.9 \text{ \AA}$ channels facilitated the complete desolvation of Li^+ [46]. These findings suggest that a layered material with an optimized interlayer spacing, like ZHS, is well-suited for stabilizing the Zn anode and accelerating Zn^{2+} diffusion. However, ZHS coatings formed in conventional aqueous

electrolytes exhibit disordered vertical orientation growth, resulting in porous, non-compact architectures [47, 48]. Such microstructures consequently fail to establish continuous water-blocking barriers and even exacerbate dendrite formation due to inhomogeneous ion flux distribution. Therefore, achieving horizontally aligned ZHS nanosheets via crystal orientation control to form a compact layer, such as an SEI-like functional layer able to repel water while ensuring uniform ion conduction, represents a promising strategy to overcome the interfacial instability of Zn anodes.

Herein, a dual-effect interfacial modulation strategy was proposed to address these challenges. Leveraging the surface energy anisotropy of ZHS crystals, a nonionic molecular additive (n-octyl- β -D-glucopyranoside (8GT)) containing both hydrophilic and hydrophobic groups was selected to precisely regulate the nucleation and growth kinetics of ZHS through selective interfacial adsorption. It was found that 8GT induces the parallel-oriented growth of ZHS crystals along the $\text{ZHS}_{(001)}$ plane via an adsorption-stabilization effect, constructing a dense and horizontally aligned passivation layer under open-circuit or static conditions. Interestingly, during zinc electrodeposition, 8GT molecules preferentially adsorb onto the $\text{Zn}_{(002)}$ and $\text{Zn}_{(100)}$ crystal planes, utilizing a spatial confinement effect to induce the preferential deposition of Zn^{2+} along the $\text{Zn}_{(101)}$ plane. Benefiting from the unique dual-scene protection mechanism of 8GT, the $\text{Zn}||\text{Zn}$ symmetric cell achieves stable plating/stripping behaviors over 2500 h at $2 \text{ mA}\cdot\text{cm}^{-2}$ and $1 \text{ mAh}\cdot\text{cm}^{-2}$, and even maintains stable operation for $\geq 400 \text{ h}$ under $10 \text{ mA}\cdot\text{cm}^{-2}$ and $5 \text{ mAh}\cdot\text{cm}^{-2}$. Significantly, the $\text{Zn}||\text{Li}_2/\text{MnO}_2/\text{activated carbon (AC)}$ full cell assembled with this electrolyte retains a reversible capacity of $203 \text{ mAh}\cdot\text{g}^{-1}$ with $\sim 100\%$ capacity retention after 500 cycles. This work guides the selection of appropriate additives to modulate the formation of ZHS on the Zn surface, offering a new direction for the design of high-performance AZIBs.

2 Introduction

As observed in the insert of Fig. 1(a), the polished Zn foil tarnished and emerged with visible byproducts after immersion in ZnSO_4 (ZS) electrolyte for 7 days. X-ray diffraction (XRD) patterns (Fig. S1 in the Electronic Supplementary Material (ESM)) confirm the formation of ZHS on this Zn foil. Consistently, scanning electronic microscopy (SEM) images (Figs. 1(a) and 1(b)) revealed randomly stacked ZHS nanosheets ranging from 2 to $10 \text{ }\mu\text{m}$. The cross-sectional SEM image (Fig. 1(c)) reveals a $\sim 9 \text{ }\mu\text{m}$ ZHS layer consisting of vertically aligned nanosheets with distinct gaps, indicating a non-dense microstructure. Conversely, the Zn foil surface exhibited a markedly different corrosion morphology in the electrolyte after introducing the n-octyl β -D-glucoside (8TG) additive (Fig. 1(d)). XRD patterns (Fig. S1 in the ESM) confirm the formation of ZHS, revealing significantly intensified (001) diffraction peaks. This observation suggests that the additive induces preferential crystal growth along these planes. Furthermore, SEM images (Figs. 1(e) and 1(f)) demonstrated that the ZHS nanosheets were not only considerably smaller and highly uniform in size ($1\text{--}2 \text{ }\mu\text{m}$), but also organized into a dense and horizontally aligned layer. Cross-sectional SEM image (Fig. 1(f)) indicated that this optimized ZHS layer had a thickness of $\sim 1 \text{ }\mu\text{m}$, which is substantially thinner than that of the control sample. Comparative analysis reveals that a thin passivation layer composed of

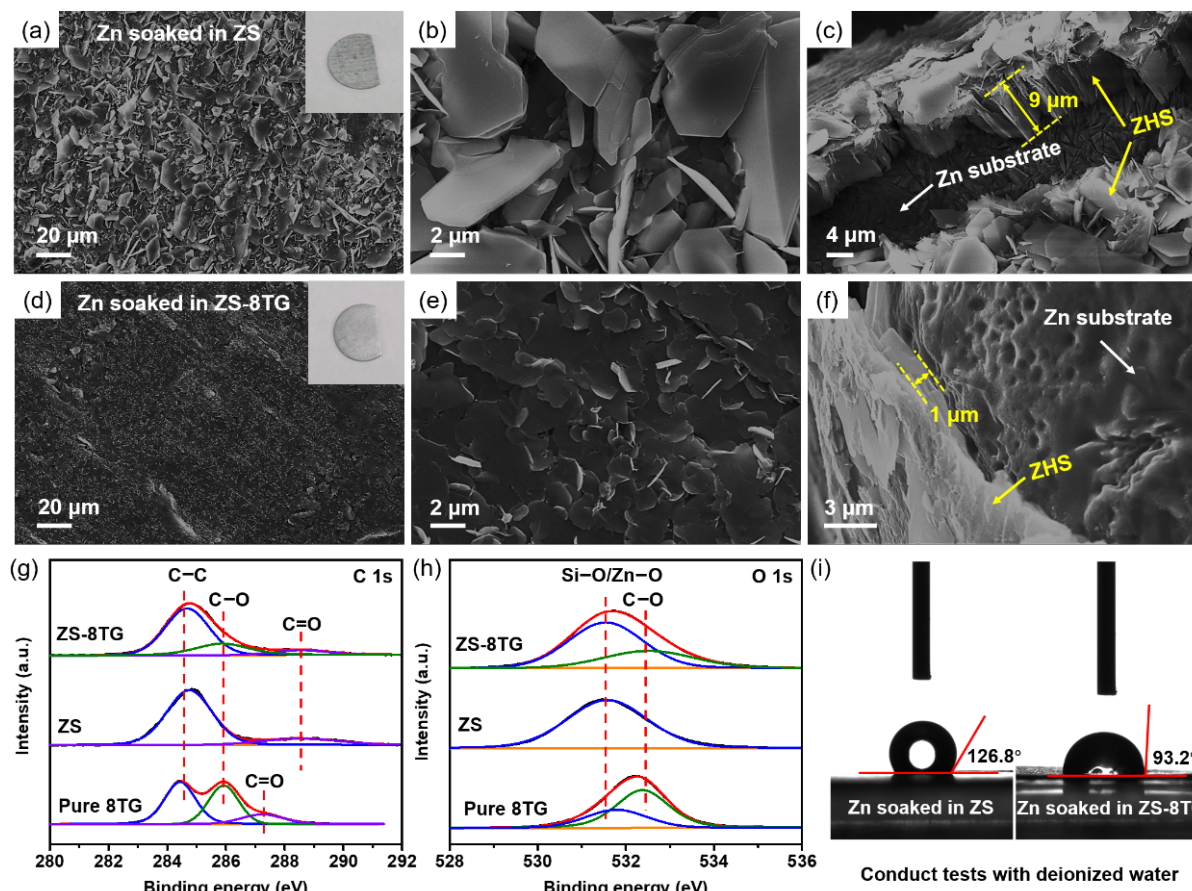


Figure 1 ((a) and (b)) SEM images and inserted optical diagram, and (c) lateral view of Zn foil surface after soaking in ZS electrolyte. ((d) and (e)) SEM images and inserted optical diagram, and (f) lateral view of Zn foil surface after soaking in ZS-8TG electrolyte. ((g) and (h)) C 1s and O 1s XPS spectra of the 8TG additive and Zn surface after soaking in both electrolytes. (i) Contact angle of deionized water on the Zn surface after soaking in both electrolytes.

horizontally stacked ZHS nanosheets forms on the Zn surface when immersed in an 8TG-containing ZS electrolyte. This compact layer effectively inhibits further growth of the passivation layer by blocking direct contact between Zn and the electrolyte. As observed in Fig. S2 in the ESM, Zn||Zn symmetric cells with ZS electrolyte exhibited visible swelling and electrolyte leakage after five days of standstill. In contrast, the cell utilizing ZS-8TG electrolyte maintained its original state without any deformation or leakage, thereby confirming its exceptional interfacial stability and effectiveness in inhibiting further hydrogen precipitation.

To further investigate the critical role of 8TG additives in promoting horizontal growth of ZHS, X-ray photoelectron spectroscopy (XPS) measurement was conducted on both pure additive samples and Zn surfaces following immersion in electrolytes with and without 8TG. The C 1s spectra (Fig. 1(g)) revealed characteristic peaks for C-C (~ 284.8 eV), C-O (~ 286 eV), and carbon dioxide species (~ 287.4 eV) from pristine 8TG samples. The Zn surface exposed to pure ZS electrolyte displayed only C-C (~ 284.8 eV) alongside carbonate signals at ~ 288.5 eV. However, the Zn surface from the ZS-8TG electrolyte exhibited a distinct C-O peak at ~ 286 eV, confirming the presence of adsorbed 8TG molecules. This finding was corroborated by the O 1s spectra (Fig. 1(h)), where the C-O signal from 8TG at 532.6 eV persisted even after rinsing, demonstrating a strong adsorption affinity between 8TG and the Zn foil. These results were further validated through theoretical calculations (Fig. S3 in the

ESM). The adsorption energy of 8TG molecules on the $\text{ZHS}_{(001)}$ facet was calculated to be -1.946 eV, which is significantly stronger than that of water molecules (-1.008 eV) and its adsorption on the $\text{Zn}_{(002)}$ facet (-0.935 eV). This preferential adsorption stabilizes the $\text{ZHS}_{(001)}$ surface, thereby influencing crystal growth orientation and facilitating horizontal alignment of ZHS nanosheets. Corresponding contact angle measurements were conducted to investigate the resulting wettability of the Zn surface after immersion in different electrolytes. As illustrated in Fig. 1(i), the Zn surface immersed in ZS electrolyte exhibited a water contact angle of 126.8° , indicating significant hydrophobicity attributed to the low surface energy characteristics of the vertically oriented ZHS passivation layer. In contrast, treatment with ZS-8TG electrolyte resulted in a markedly lower contact angle of 93.2° , reflecting enhanced hydrophilicity. Control tests performed on bare Zn foil with various electrolytes (Fig. S4 in the ESM) ruled out interference from residual surfactants, confirming that this reduced contact angle arises from the inherently higher surface energy associated with horizontally aligned $\text{ZHS}_{(001)}$ planes. This observation further elucidates that conventional ZS electrolyte leads to vertically or disordered stacked ZHS nanosheets owing to the maximized thermodynamic stability. In contrast, the horizontally oriented passivation layer induced by 8TG demonstrates improved compatibility within aqueous electrolytes. The dense and horizontal arrangement of this ZHS layer effectively obstructs water access to the Zn substrate, thereby inhibiting side reactions and significantly enhancing anode stability

and utilization. To verify whether similar behavior occurs with other surfactants, we further examined coconut diethanolamide (CDA), octylphenol ethoxylate (Triton X-100), polysorbate 20 (Tween 20), polyvinyl pyrrolidone (PVP), sodium dodecyl sulfonate (SDS), cetyl trimethylammonium bromide (CTAB), as well as alkyl poly glycoside (4TG), C6 alkyl glucoside (6TG), decyl glucoside (10TG), and lauryl glucoside (12TG). Notably, a similar horizontal stacking of ZHS nanosheets was also observed in other nonionic surfactant systems (Figs. S5–S7 in the ESM).

Based on the performance comparison of 4TG to 12TG as electrolyte additives (Fig. S8 in the ESM), 8TG was selected as a prototype for a systematic investigation into the function and mechanism of nonionic surfactants in ZS electrolytes. Initially, Fourier transform infrared (FTIR) spectroscopy was employed to examine the influence of the 8TG additive on the solvation structure of the ZS electrolyte. As illustrated in Fig. 2(a), absorption peaks at approximately 1090, 1640, and 3300 cm^{-1} correspond to the telescopic vibrational signals of SO_4^{2-} , OH^- , and H_2O , respectively. It can be observed that the red shift in the SO_4^{2-} signal suggests that the 8TG additive slightly participates in modifying the solvation structure and subtly alters the chemical environment surrounding SO_4^{2-} within the electrolyte. Subsequently, nuclear magnetic resonance (NMR) spectroscopy was utilized to corroborate these FTIR findings. As depicted in Fig. 2(b), a signal at 1.2–1.3 ppm corresponds to alkyl chain hydrogens of the 8TG molecule. This represents an up-field shift towards higher fields, indicating an enhanced shielding effect within the aqueous electrolyte. In contrast, a slight downfield shift is observed for the C–OH group signal at approximately ~ 4.78 ppm. This aligns with weak interactions occurring with Zn^{2+} solvation structures. Meanwhile, there is no change noted for solvent water signals around ~ 4.7 ppm, confirming that 8TG does not disrupt bulk hydrogen-bonding networks. Theoretical calculations were conducted to analyze interaction forces between Zn^{2+} , SO_4^{2-} , 8TG molecules, and H_2O molecules. As shown in Fig. 2(c) and Fig. S9 in

the ESM, the binding energies of Zn^{2+} –8TG, Zn^{2+} – H_2O , 8TG– SO_4^{2-} , 8TG–8TG, 8TG– H_2O , and H_2O – H_2O are -0.89 , -0.83 , -1.10 , 0.84 , -0.69 , and -0.24 eV, respectively. These results indicate that 8TG preferentially interacts with Zn^{2+} and SO_4^{2-} , with the 8TG– SO_4^{2-} interaction being the strongest, consistent with the FTIR and NMR spectra. Such preferential association may partially sequester SO_4^{2-} , thereby weakening the strong Zn^{2+} – SO_4^{2-} interaction and accelerating the desolvation process. To provide further evidence supporting this phenomenon, molecular dynamics (MD) simulations were performed. As illustrated in Figs. 2(d) and 2(e) and Fig. S10 in the ESM, the average H_2O coordination number around Zn^{2+} was decreased from 5.31 in the pure ZS electrolyte to 5.21 in the ZS-8TG electrolyte. This study confirms that 8TG participates in the solvation structure of Zn^{2+} and partially displaces H_2O molecules. Collectively, these findings demonstrate the existence of weak interactions between 8TG and Zn^{2+} , which moderately attenuate the interaction between Zn^{2+} and H_2O , thereby facilitating the desolvation process.

To further investigate the impact of 8TG additives on the performance of the ZS electrolyte system, a series of electrochemical measurements was carried out. The electrochemical stabilization windows (ESW) for electrolytes with and without the 8TG additive were evaluated using linear scanning voltammetry (LSV). To eliminate interference from Zn^{2+} deposition behavior on HER, Na_2SO_4 was employed as an alternative electrolyte to replace ZS. As illustrated in Fig. 3(a), the Na_2SO_4 electrolyte exhibited HER and oxygen evolution reaction (OER) potentials of -0.976 and 1.438 V (vs. Ag/AgCl) at a current density of $10 \text{ mA}\cdot\text{cm}^{-2}$, respectively, resulting in an ESW of 2.414 V. Upon introducing 8TG, both HER potential shifted negatively to -1.117 V while OER potential shifted positively to 1.653 V, leading to an expanded ESW of 2.77 V. These results confirm that the addition of 8TG effectively suppresses both HER and OER activities, thus widening the ESW for aqueous electrolytes. Furthermore, Tafel curves were measured to evaluate the electrochemical corrosion behavior of Zn electrodes in both

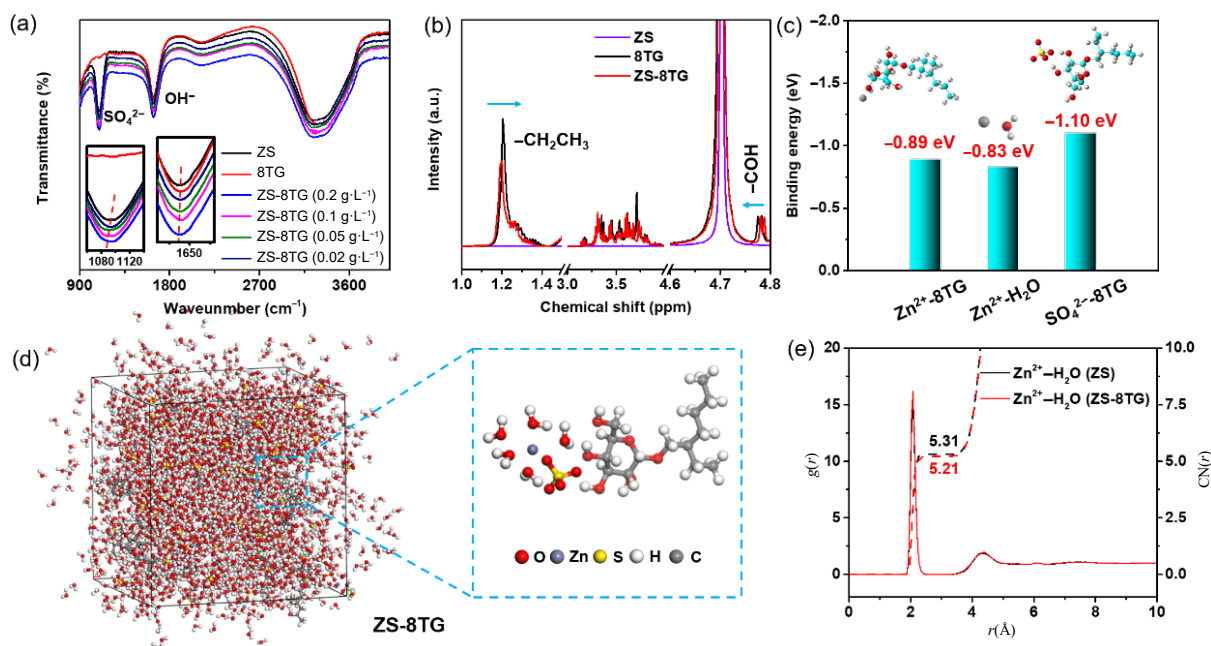


Figure 2 (a) IR and (b) NMR spectra of the different electrolytes. (c) Binding energy of the Zn^{2+} with 8TG additive or water molecules. (d) 3D snapshot of MD simulation cells in ZS-8TG electrolyte. (e) The radial distribution function of Zn^{2+} –O and the radius-dependent coordination numbers ($\text{CN}(r)$) in the ZS and ZS-8TG electrolytes.

electrolytes. As shown in Fig. 3(b), the Zn electrode with ZS electrolyte exhibited a corrosion potential of -20.3 mV along with a corrosion current density of 9.5×10^{-6} A·cm $^{-2}$. Conversely, when utilizing ZS-8TG electrolyte yielded a positively shifted corrosion potential at 9.1 mV alongside a reduced corrosion current density of 1.7×10^{-6} A·cm $^{-2}$, indicating significantly enhanced corrosion resistance. To investigate interfacial effects stemming from adsorption by 8TG, electrochemical impedance spectroscopy (EIS) of Zn||Zn symmetric cells was conducted. As depicted in Fig. 3(c), charge transfer resistance increased from 150 Ω within pure ZS electrolyte to 178 Ω upon addition of the additive. This modest increase can be attributed to the protective adsorption layer formed by 8TG, which serves to homogenize the electric field and mitigate the tip effect, thereby contributing to the inhibition of Zn dendritic growth. Further cyclic voltammetry (CV) tests (Fig. 3(d)) indicate that Zn nucleation necessitates a more negative potential in the ZS-8TG electrolyte compared to the pure ZS system, while maintaining a comparable peak current density. This observation suggests that finer Zn grains are formed without compromising interfacial Zn $^{2+}$ mass transport kinetics. Moreover, the deposition behavior of Zn $^{2+}$ was investigated using chronoamperometry (current–time (I – t)). A continuous increase in current can be observed with pure ZS electrolyte (Fig. 3(e)), indicating a two-dimensional (2D) diffusion-limited process for Zn $^{2+}$ on the surface of Zn. Conversely, a slow

variation of the current density within the ZS-8TG electrolyte points towards a preference for three-dimensional (3D) diffusion modes, which aid in preventing dendrite formation on the Zn anode. The noted enhancements in deposition uniformity and stability prompted further investigation into both Zn $^{2+}$ transport and desolvation behaviors. As illustrated in Figs. 3(f) and 3(g) and Figs. S11 and S12 in the ESM, it was found that the migration number of Zn $^{2+}$ in ZS-8TG electrolyte (0.87) significantly exceeds that observed in pure ZS electrolyte (0.63). Meanwhile, there was a reduction in activation energy for Zn $^{2+}$ desolvation from 65.68 to 57.2 kJ·mol $^{-1}$, and it becomes even lower after introducing a horizontally oriented ZHS layer on the Zn surface (Fig. S13 in the ESM). In addition, the ionic conductivity of the ZS electrolyte increases from 25.13 to 37.82 mS·cm $^{-1}$ upon the addition of 8TG (Fig. S14 in the ESM). These findings demonstrate that the additive 8TG partially participates in forming a solvated sheath layer around Zn $^{2+}$; this not only facilitates desolvation by lowering energy barriers but also enhances ion transport efficiency. Consequently, symmetric cells composed of Zn||Zn with ZS-8TG electrolyte exhibit superior rate capabilities and reduced deposition overpotentials across a range of current densities from 0.5 to 10 mA·cm $^{-2}$ (Figs. 3(h) and 3(i)). This performance enhancement correlates with decreased charge-transfer resistance at the interface during Zn deposition (Fig. S15 in the ESM).

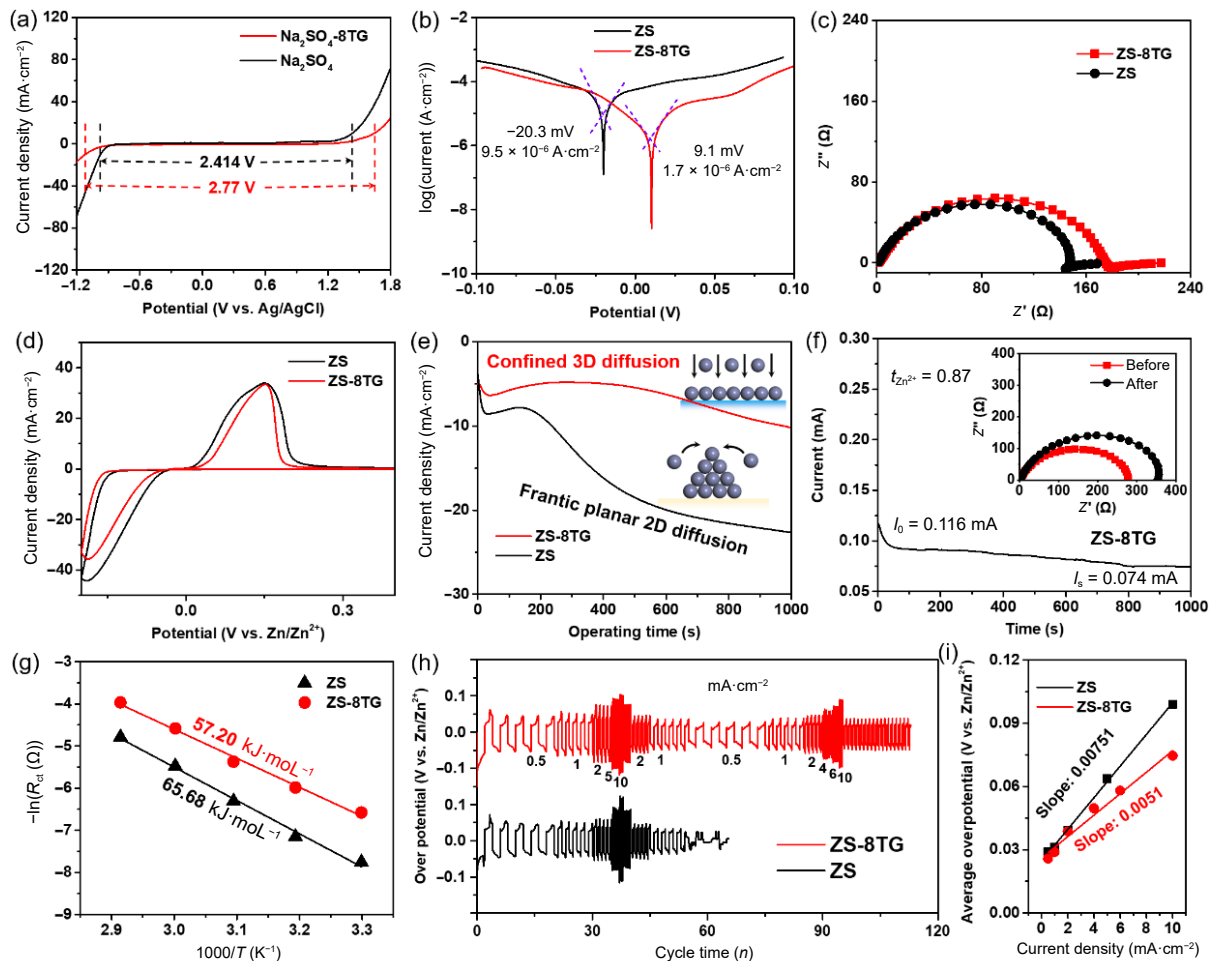


Figure 3 (a) LSV curves of the Zn electrode in Na $_2$ SO $_4$ and Na $_2$ SO $_4$ -8TG electrolytes. ((b)–(i)) Comparison of electrochemical performance in ZS and ZS-8TG electrolytes: (b) Tafel curves, (c) EIS curves, (d) CV curves, and (e) I – t curves of Zn electrode; (f) test curves for Zn $^{2+}$ migration number in ZS-8TG electrolyte; (g) activation energy test curves; and ((h) and (i)) rate curves and corresponding current/overpotential curves of the Zn||Zn cells.

To further evaluate the long-term cycling stability of the Zn anode in ZS-8TG electrolyte, the deposition/stripping behavior was monitored in Zn||Cu cells. As shown in Fig. 4(a), the initial Coulombic efficiency (ICE) in the ZS-8TG electrolyte reached 84.4% under the condition of $2 \text{ mA}\cdot\text{cm}^{-2}$ and $1 \text{ mAh}\cdot\text{cm}^{-2}$, significantly higher than that of the 68.8% observed in the pure ZS electrolyte. This observation indicates a lower degree of side reactions in the ZS-8TG electrolyte, which can be attributed to the inhibitory effect of the 8TG additive on side reactions such as HER. Furthermore, the Zn||Cu cells utilizing the ZS-8TG electrolyte demonstrate exceptional cycling stability over 2000 cycles, and exhibited a remarkably stable overpotential of 33, 31, 29, 25, and 25 mV at the 40th, 100th, 500th, 1000th, and 2000th cycles, respectively (Fig. 4(b)). In contrast, the cell with ZS electrolyte displayed an increasing overpotential of 47 and 52 mV at the 40th and 60th cycles, respectively. This increase is attributed to the accumulation of disordered byproducts that hinder Zn^{2+} diffusion. Ultimately, this degradation led to cell failure, characterized by a sharply fluctuating voltage curve. Moreover, there was a notable improvement in average Coulombic efficiency from 96.4% for ZS electrolyte to 99.77% with ZS-8TG electrolyte, further confirming the efficacy of 8TG in enhancing the reversibility and cycling stability of the Zn anode. Long-term cycling stability was additionally evaluated using Zn||Zn symmetric cells across various current densities (Figs. 4(c)–4(e)). The cells assembled with ZS-8TG electrolyte achieved

remarkable stability lasting over 2500 h at $2 \text{ mA}\cdot\text{cm}^{-2}$ and $1 \text{ mAh}\cdot\text{cm}^{-2}$, significantly outperforming those with ZS electrolyte, which failed after merely 50 h. Notably, the ZS-8TG-based cell sustained 800 h at $5 \text{ mA}\cdot\text{cm}^{-2}/2.5 \text{ mAh}\cdot\text{cm}^{-2}$ and 400 h at $10 \text{ mA}\cdot\text{cm}^{-2}/5 \text{ mAh}\cdot\text{cm}^{-2}$, whereas the ZS-based cells failed after only 49 and 80 h, respectively. Moreover, using Zn foils with thicknesses of $\sim 0.1 \text{ mm}$ ($\sim 60 \text{ mAh}$) and $\sim 0.009 \text{ mm}$ ($\sim 5 \text{ mAh}$), we systematically evaluated the long-term cycling performance under high depth-of-discharge (DOD) conditions of 33%, 40%, and 60% (shown in Fig. S16 in the ESM). These results strongly confirm that 8TG significantly enhances the cycling stability of the Zn anode, which can be attributed to its function in modulating the solvation structure and inducing the formation of a protective surface layer. To reveal the effect of the pre-formed horizontal ZHS interlayer, the Zn anode was pre-treated in ZS-8TG electrolyte. This Zn||Zn symmetric cell achieved stable cycling over 2350 h (Fig. S17 in the ESM), demonstrating that the ZHS interlayer provides robust and durable protection for highly reversible Zn deposition/stripping. Meanwhile, we also summarized representative electrolyte-additive studies reported in recent years (in Table S1 in the ESM), which demonstrate that the 8TG additive still offers notable advantages.

To investigate the influence of the 8TG additive on Zn^{2+} deposition, the resulting interfacial microstructure of Zn anodes after cycles was characterized. The XRD patterns (Fig. 5(a)) reveal

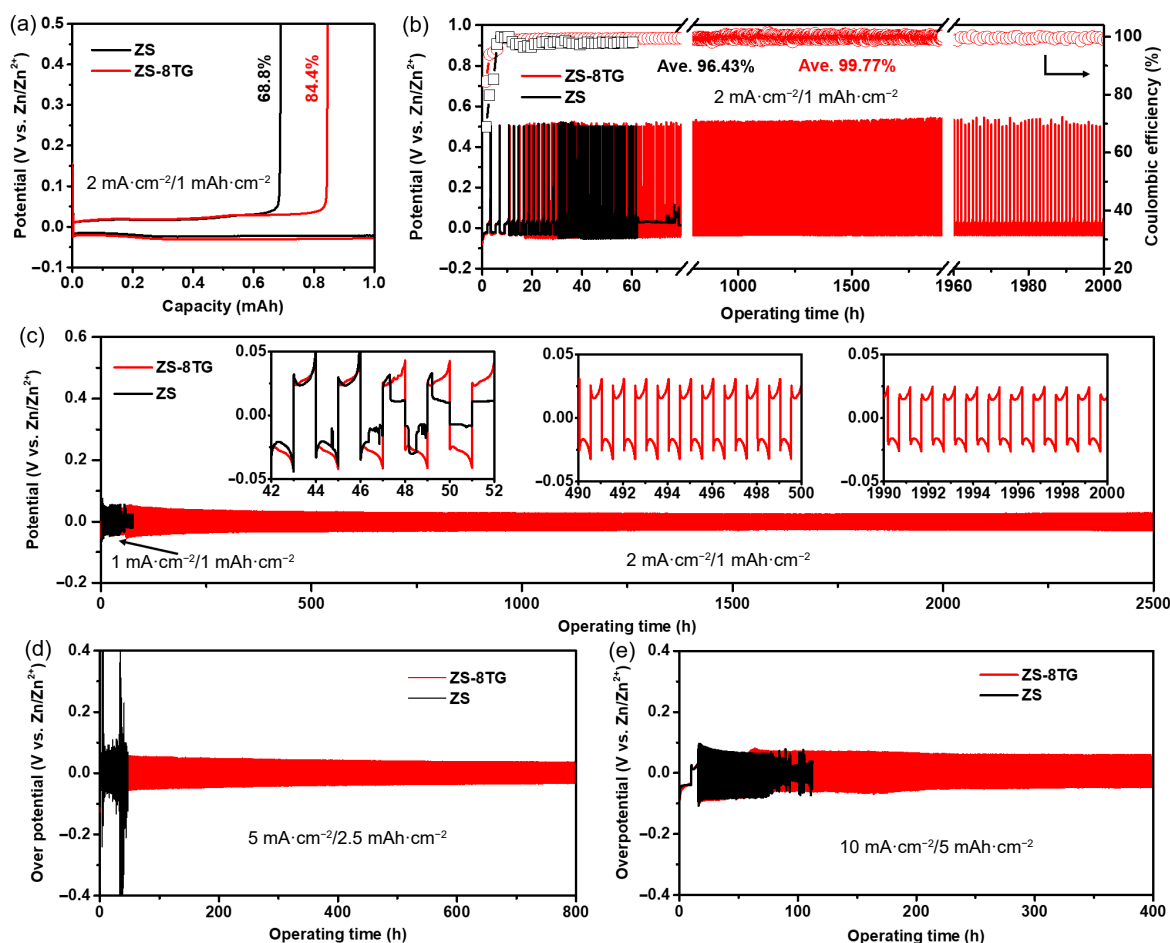


Figure 4 (a) Initial Zn plating/stripping curves. (b) Long-term cycling curves and corresponding Coulombic efficiency of the Zn||Cu half cells with different electrolytes under $2 \text{ mA}\cdot\text{cm}^{-2}$ and $1 \text{ mAh}\cdot\text{cm}^{-2}$. (c) Long-term cycling and section cycling curves of Zn||Zn half cells with different electrolytes under $2 \text{ mA}\cdot\text{cm}^{-2}$ and $1 \text{ mAh}\cdot\text{cm}^{-2}$. ((d) and (e)) Long-term cycling curves of Zn||Zn half cells with different electrolytes under different current densities.

that Zn and ZHS both exist after 100 cycles in ZS and ZS-8TG electrolytes. However, a more intense ZHS diffraction peak of the Zn anode cycled in the ZS electrolyte indicates persistent and severe side reactions during cycling. While for cycling in the ZS-8TG, a weaker ZHS peaks of Zn anode confirm effective suppression of these reactions, which is attributed to the protective horizontal ZHS layer mitigating HER. These results are fully consistent with the superior initial and average Coulombic efficiency achieved with the ZS-8TG electrolyte. Furthermore, the intensity ratio of the $Zn_{(002)}/Zn_{(101)}$ crystal planes decreased slightly from 0.513 to 0.445 after cycling in the ZS electrolyte. In contrast, this ratio dropped significantly to 0.283 in the ZS-8TG electrolyte, indicating a pronounced preferential deposition of Zn along the $Zn_{(101)}$ crystal plane, which promotes a denser packing morphology. To eliminate the interference from the intrinsic $Zn_{(101)}$ diffraction peak of the pristine Zn foil, we re-deposited a Zn layer on a Cu substrate and performed XRD measurement; the results still support our conclusion (shown in Fig. S18 in the ESM). SEM images corroborate this structural evolution (Figs. 5(b) and 5(c)). The Zn anode from the ZS electrolyte was covered with abundant flocculent material intertwined with glass fibers, leading to significant accumulation of hexagonal lamellar ZHS byproducts. Conversely, the Zn anode cycled in ZS-8TG electrolyte exhibited a smooth and

densely packed surface consistent with oriented deposition along $Zn_{(101)}$ planes and was covered by a thin continuous layer of ZHS (Figs. S19(a)–S19(c) in the ESM). XPS measurement and depth profiling analysis conducted on cycled Zn anodes from the ZS-8TG electrolyte revealed a distinct C–O signal (Fig. 5(d)), indicating the existence of adsorbed 8TG molecules or their derivatives, with an interfacial layer thickness of approximately 20 nm (Fig. S20 in the ESM). The Si 2p and S 2p spectra (Figs. S19(d) and S19(e) in the ESM) obtained from the ZS-electrolyte sample concurrently confirmed the significant presence of glass fiber residues and ZHS byproducts, which is consistent with the observed via SEM. To elucidate the mechanism underlying the increased exposure of the $Zn_{(101)}$ plane, theoretical calculations were conducted. The adsorption energies of 8TG on the $Zn_{(002)}$, $Zn_{(100)}$, and $Zn_{(101)}$ facets were calculated to be -0.935 , -0.922 , and 0.248 eV, respectively (Fig. 5(e)). This indicates that 8TG preferentially adsorbs on the $Zn_{(002)}$ and $Zn_{(100)}$ facets, creating a steric hindrance that regulates Zn^{2+} deposition toward the less-adsorbed $Zn_{(101)}$ plane. Additionally, the Zn^{2+} adsorption energies on different Zn facets after 8TG modification further support this conclusion (Fig. S21 in the ESM). Furthermore, molecular orbital analysis revealed a higher highest occupied molecular orbital (HOMO) energy of 8TG (-7.436 eV) than of H_2O (-8.263 eV) (Fig. 5(f)), suggesting 8TG is more

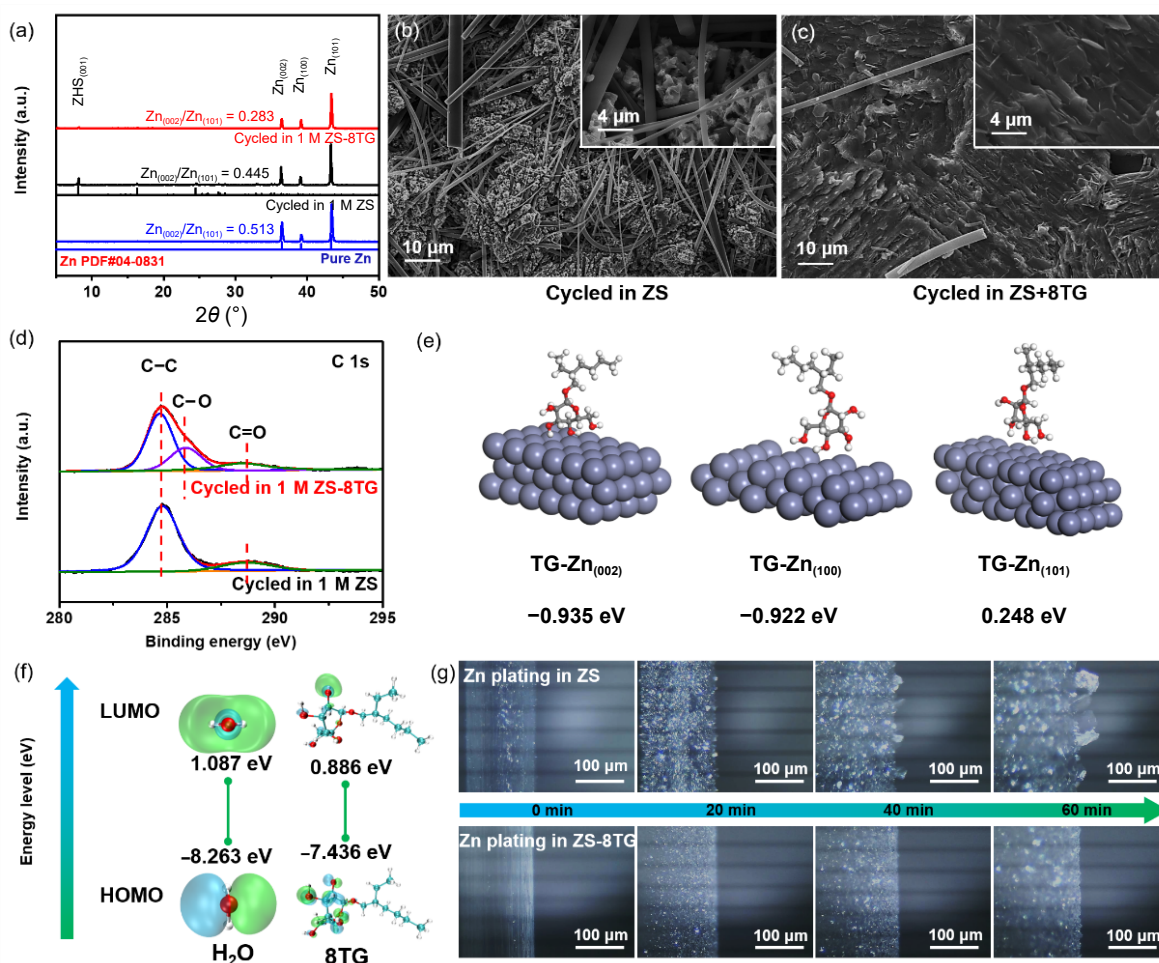


Figure 5 (a) XRD patterns of pure Zn foil and Zn electrodes cycled in different electrolytes. (b) and (c) SEM images of Zn electrode surface cycled in ZS and ZS-8TG electrolytes. (d) C 1s XPS spectra of the Zn surface after cycling in ZS-8TG electrolyte. (e) Adsorption energy of 8TG molecule on the $Zn_{(002)}$, $Zn_{(100)}$, and $Zn_{(101)}$ crystal planes. (f) calculated HOMO and lowest unoccupied molecular orbital (LUMO) energy level of H_2O and 8TG. (g) Cross-sectional Zn plating morphology in the symmetrical Zn cell with 5 mA·cm $^{-2}$ with different electrolytes using an *in-situ* optical microscope.

susceptible to reduction, which corroborates the origin of the C–O peak observed in XPS spectra. To visually monitor the Zn^{2+} deposition dynamics, *in-situ* optical microscopy was employed to observe the surface of the Zn electrode. As shown in Fig. 5(g), after 40 min of deposition at a current density of $5 \text{ mA}\cdot\text{cm}^{-2}$, distinct dendrites appeared on and gradually enlarged across the surface of Zn electrodes cycled in ZS electrolyte. Conversely, when using ZS-8TG electrolyte, uniform thickening occurred without observable dendritic protrusions on the Zn electrode's surface. This confirms that incorporating 8TG effectively suppresses Zn dendrite formation while enhancing cycling lifespan for zinc anodes. Such observation was further verified by *in-situ* EIS measurements and the corresponding distribution of relaxation times (DRT) analysis [49] (Fig. S22 in the ESM).

To evaluate the possibility of the 8TG additive for the practical application, $\text{Zn}||\text{I}_2/\text{MnO}_2@\text{AC}$ full cells ($\text{I}_2:\text{MnO}_2$ mass ratio was about 7:3) were fabricated and measured with ZS and ZS-8TG electrolytes (Fig. S23 in the ESM). As depicted in Fig. 6(a), CV curves of the cell with ZS-8TG electrolyte showed two well-defined redox pairs at 1.15 and 1.31 V, corresponding to the I_2/I^- and $\text{Mn}^{4+}/\text{Mn}^{3+}$ redox couples, respectively. In contrast, the corresponding CV peaks with the ZS electrolyte emerged at lower potentials of 1.12 and 1.25 V, indicating a slight voltage hysteresis. Furthermore, EIS results (Fig. S24(a) in the ESM) showed a marginally lower interfacial impedance for the cell with ZS-8TG electrolyte ($\sim 18 \Omega$) than that of the ZS electrolyte ($\sim 20 \Omega$), indicating enhanced ionic conductivity at the cathode surface with the 8TG additive. Rate performance tests (Fig. 6(b) and Figs. S24(b) and S24(c) in the ESM) revealed that the ZS-8TG cell delivered higher reversible capacities across various rates, maintaining $76 \text{ mAh}\cdot\text{g}^{-1}$ even at a high rate of 30 C. In addition, in the aging test (Fig. 6(c)), after 24 h, the cell with ZS-8TG electrolyte achieved a Coulombic efficiency of 82.55%, significantly surpassing that of the cell with ZS (61.54%, Fig. S25(a) in the ESM), which suffered from severe iodine loss (Fig. S26 in the ESM). Moreover, the charge–discharge plateaus (Fig. 6(d) and Fig. S25(b) in the ESM)

aligned with the CV results. In terms of long-term cycling performance (Fig. 6(e)), the cell with the ZS-8TG electrolyte retained a specific capacity of $203 \text{ mAh}\cdot\text{g}^{-1}$ after 500 cycles with minimal decay, whereas the cell with the ZS electrolyte showed a notable decay from 180 to $108 \text{ mAh}\cdot\text{g}^{-1}$ after 300 cycles. Furthermore, the pouch cell with ZS-8TG electrolyte successfully powered 17 light-emitting diodes (LEDs) (Fig. 6(e), inset). In summary, the 8TG additive significantly enhances the reversibility, rate capability, and long-term cycling stability of $\text{Zn}||\text{I}_2/\text{MnO}_2@\text{AC}$.

3 Conclusions

In summary, to address the cycling stability issue of zinc anodes, this work proposes a dual-effect interfacial modulation strategy based on the 8TG additive. The 8TG additive induces the parallel growth of ZHS along its (001) crystal plane through an adsorption–stabilization effect, forming a dense and horizontally aligned protective layer under static conditions. This layer effectively isolates the electrolyte from the zinc substrate, thereby significantly suppressing corrosion reactions. The ZHS interfacial layer exhibits an interlayer spacing of $> 8 \text{ \AA}$ and is rich in interlayer anions, which lowers the ion-transport energy barrier via an electrostatic repulsion–attraction effect, achieving a high zinc ion migration number ($t_{\text{Zn}^{2+}}$) of 0.87. Furthermore, during electrodeposition, 8TG preferentially adsorbs on the $\text{Zn}_{(002)}$ and $\text{Zn}_{(100)}$ crystal planes and, through a spatial confinement effect, guides the preferential deposition of Zn^{2+} along the $\text{Zn}_{(101)}$ plane, effectively inhibiting dendrite growth. Benefiting from its multifunctional protection, the $\text{Zn}||\text{Zn}$ symmetric cells with ZS-8TG electrolyte achieved exceptional cycling stability over 2500 h at $2 \text{ mA}\cdot\text{cm}^{-2}$ and $1 \text{ mAh}\cdot\text{cm}^{-2}$, and maintained stability for more than 400 h even at $10 \text{ mA}\cdot\text{cm}^{-2}$ and $5 \text{ mAh}\cdot\text{cm}^{-2}$. The assembled Zn–Cu cells maintained an average Coulombic efficiency of 99.77% for 2000 cycles, and $\text{Zn}||\text{I}_2/\text{MnO}_2@\text{AC}$ full cells retained nearly 100% capacity after 500 cycles. The 8TG electrolyte additive not only enhanced the interfacial stability of Zn anodes but also induced the

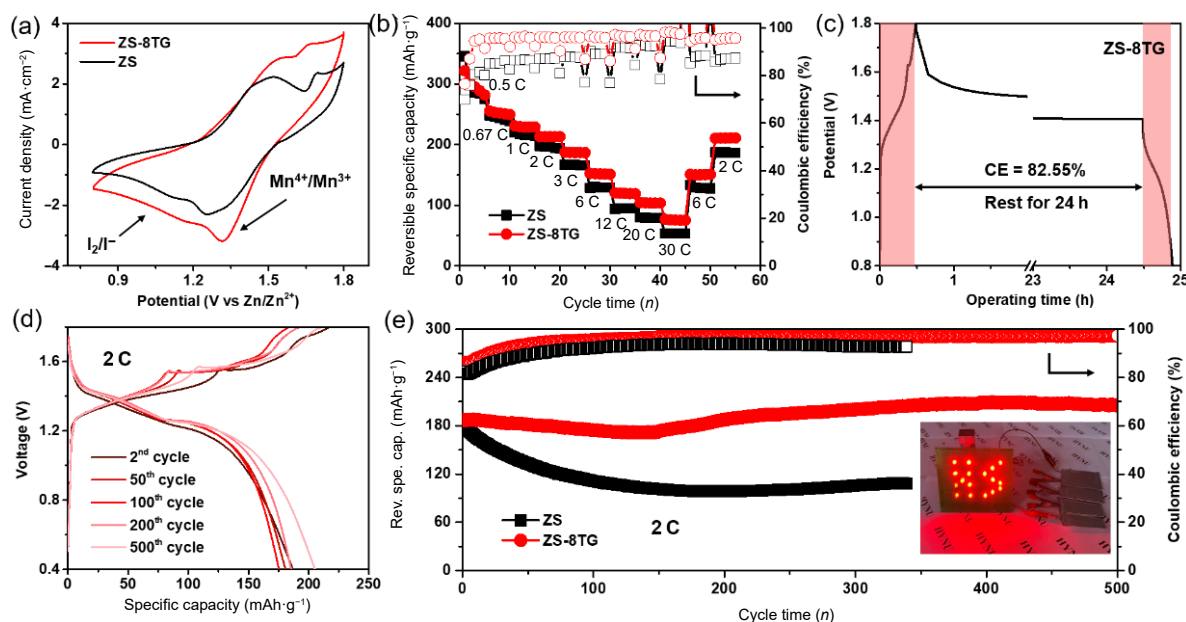


Figure 6 (a) The CV curves of the full cells with different electrolytes. (b) Rate curves of the cycled full cells. (c) The charge and then discharge curves after standing for 24 h. (d) The charge/discharge curves in different cycles with ZS-8TG electrolyte. (e) Long-term cycle capability of both full cells and illustration of lit LEDs (rev. spe. cap. refers to reversible specific capacity).

packing orientation of ZHS, suppressing the side reactions. This work opens an avenue for the development of high-performance AZIBs.

Electronic Supplementary Material: Supplementary material (details of experimental section; Figs. S1–S26: XRD patterns, optical images, adsorption energy, contact angle, SEM images, CV curves, 3D snapshot of MD simulation cells, current–time curve, AC impedance test curves, the activation energy test curves, ionic conductivities, AC impedance test curves, XPS depth profiling spectra, adsorption energies, DRT plots, charge and then discharge curves of the full cells, ultraviolet–visible (UV–vis) spectra, long-term cycling performance; and Table S1) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908606>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

Acknowledgements

This work was financially supported by the Hengyang Normal University Science Foundation Project (No. 2024HSKFJJ023) and the Natural Science Foundation of Hunan Province (No. 2024JJ7064).

Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

L. T.: Experimental design, data curation, writing manuscript, investigation, funding acquisition. M. Z.: Formal analysis, methodology, data curation, investigation, writing manuscript. S. J. O.: data curation, methodology. J. Q. Y.: data curation. W. J. N.: Software, supervision. R. Q. L.: Project administration, supervision, funding acquisition. Y. J. W.: Methodology, validation. X. J. X.: Conceptualization, writing – review & editing, supervision. R. Z. H.: Conceptualization, writing – review & editing, supervision. All the authors have approved the final manuscript.

Use of AI statement

None.

References

- [1] Shang, Y.; Kundu, D. A path forward for the translational development of aqueous zinc-ion batteries. *Joule* **2023**, *7*, 244–250.
- [2] Zhu, C. Y.; Li, P. Z.; Xu, G. Y.; Cheng, H.; Gao, G. Recent progress and challenges of Zn anode modification materials in aqueous Zn-ion batteries. *Coord. Chem. Rev.* **2023**, *485*, 215142.
- [3] Wang, T. T.; Zhang, Y.; You, J. H.; Hu, F. Recent progress in aqueous zinc-ion batteries: From fundamental science to structure design. *Chem. Rec.* **2023**, *23*, e202200309.
- [4] Chen, J. P.; Zhao, W. Y.; Jiang, J. M.; Zhao, X. L.; Zheng, S. H.; Pan, Z. H.; Yang, X. W. Challenges and perspectives of hydrogen evolution-free aqueous Zn-ion batteries. *Energy Storage Mater.* **2023**, *59*, 102767.
- [5] Loh, J. R.; Xue, J. M.; Lee, W. S. V. Challenges and strategies in the development of zinc-ion batteries. *Small Methods* **2023**, *7*, 2300101.
- [6] Nie, C. H.; Wang, G. L.; Wang, D. D.; Wang, M. Y.; Gao, X. R.; Bai, Z. C.; Wang, N. N.; Yang, J.; Xing, Z.; Dou, S. X. Recent progress on Zn anodes for advanced aqueous zinc-ion batteries. *Adv. Energy Mater.* **2023**, *13*, 2300606.
- [7] Wang, D. D.; Liu, H. X.; Lv, D.; Wang, C.; Yang, J.; Qian, Y. T. Rational screening of artificial solid electrolyte interphases on Zn for ultrahigh-rate and long-life aqueous batteries. *Adv. Mater.* **2023**, *35*, 2207908.
- [8] Weng, J. Q.; Zhu, W. Q.; Yu, K.; Luo, J.; Chen, M. X.; Li, L. Y.; Zhuang, Y. H.; Xia, K. L.; Lu, Z. X.; Hu, Y. J. et al. Enhancing Zn-metal anode stability: Key effects of electrolyte additives on ion-shield-like electrical double layer and stable solid electrolyte interphase. *Adv. Funct. Mater.* **2024**, *34*, 2314347.
- [9] Wen, Q.; Fu, H.; Cui, R. D.; Chen, H. Z.; Ji, R. H.; Tang, L. B.; Yan, C.; Mao, J.; Dai, K. H.; Zhang, X. H. et al. Recent advances in interfacial modification of zinc anode for aqueous rechargeable zinc ion batteries. *J. Energy Chem.* **2023**, *83*, 287–303.
- [10] Huang, Z. X.; Liu, H.; Liu, Y. X.; Tan, L.; Wu, Y. J. Surfactant-assisted hierarchical silver coating enhances the anode surface stability toward zinc stripping/plating. *Mater. Today Chem.* **2023**, *30*, 101545.
- [11] Huang, K. X.; Zeng, X. G.; Zhang, D.; Wang, Y. J.; Lan, M.; Wen, C. Y.; Guo, Y. Tailoring crystallization zinc hydroxide sulfates growth towards stable zinc deposition chemistry. *Nano Res.* **2024**, *17*, 5243–5250.
- [12] Chen, J. T.; Bai, S.; Zhang, X. X.; Qiu, J. K.; Liu, Z. P.; Zhang, Y. N. Self-adsorbing electrolyte additive promoting Zn(002) deposition on Zn anode for aqueous zinc-ion battery. *J. Colloid Interf. Sci.* **2025**, *696*, 137870.
- [13] Yang, Q. H.; Wang, L. F.; Wang, X. Y.; Zhen, M. M. Progress and challenges of electrolyte modulation in aqueous zinc-ion batteries. *Rare Met.* **2024**, *43*, 2940–2967.
- [14] Yu, J.; Zhao, F. X.; He, J. L.; Li, A.; Que, C. N.; Chen, Z.; Tan, L.; Li, Z. L.; Wu, Y. J. Regulating zinc hydroxide sulfate (001) preferential growth and Zn (002) deposition by trace dibenzene-sulfonimide additive toward long cycle lifespan for aqueous Zn-ion batteries. *Chem. Eng. J.* **2024**, *497*, 154795.
- [15] Yu, H. M.; Chen, D. P.; Zhang, L. J.; Huang, S. Z.; Zhou, L. J.; Kuang, G. C.; Wei, W. F.; Chen, L. B.; Chen, Y. J. Electrolyte engineering for optimizing anode/electrolyte interface towards superior aqueous zinc-ion batteries: A review. *Trans. Nonferrous Met. Soc. China* **2024**, *34*, 3118–3150.
- [16] Zhang, W. W.; Qi, W. T.; Yang, K.; Hu, Y. Y.; Jiang, F. Y.; Liu, W. B.; Du, L. Y.; Yan, Z. H.; Sun, J. C. Boosting tough metal Zn anode by MOF layer for high-performance zinc-ion batteries. *Energy Storage Mater.* **2024**, *71*, 103616.
- [17] Feng, K. Y.; Zhao, Y. Y.; Liu, Z.; Yu, Y. J. Long cycle life aqueous zinc-ion battery enabled by a ZIF-N protective layer with electron-withdrawing group and zincophilicity on the Zn anode. *J. Colloid Interf. Sci.* **2025**, *678*, 76–87.
- [18] Liang, L. P.; Su, L.; Zhang, X.; Wang, Y. Q.; Ren, W. R.; Gao, X. P.; Zheng, L. Q.; Lu, F. Synergistically regulating Zn-ion flux and accelerating ion transport kinetics via zincophilic covalent organic framework interlayer for stable Zn metal anode. *Chem. Eng. J.* **2024**, *485*, 149813.
- [19] Xu, J.; Yang, Y. T.; Dai, Q. Y.; Zheng, Z. Y.; Cao, Y. J.; Cheng, Y. W.; Peng, B.; Ma, L. B.; Wang, Y. G. Towards ultra-stable wide-temperature zinc-ion batteries by using ion-sieving organic framework membrane. *Angew. Chem., Int. Ed.* **2025**, *64*, e202423118.
- [20] Li, S. W.; Li, M.; Guo, A. H.; Feng, Z. M.; Zhang, Z. K. Interfacial engineering of Zn anodes with zincophilic polyethylene terephthalate coating for high-performance and ultra-stable aqueous zinc-ion batteries. *J. Colloid Interf. Sci.* **2026**, *704*, 139397.
- [21] Duan, J. W.; Dong, J. M.; Cao, R. R.; Yang, H.; Fang, K. K.; Liu, Y.; Shen, Z. T.; Li, F. M.; Liu, R.; Li, H. L. et al. Regulated Zn plating

- and stripping by a multifunctional polymer-alloy interphase layer for stable Zn metal anode. *Adv. Sci.* **2023**, *10*, 2303343.
- [22] Zhang, Q. H.; Su, Y. W.; Shi, Z. X.; Yang, X. Z.; Sun, J. Y. Artificial interphase layer for stabilized Zn anodes: Progress and prospects. *Small* **2022**, *18*, 2203583.
- [23] Li, L. Y.; Zheng, Y. S.; Xu, J.; Peng, B.; Zhu, G. Y.; Wu, J. X.; Ma, L. B.; Jin, Z. Structural and interfacial engineering strategies for constructing dendrite-free zinc metal anodes. *ACS Energy Lett.* **2024**, *9*, 3269–3289.
- [24] Yang, J. J.; Zhao, R.; Wang, Y. S.; Hu, Z. F.; Wang, Y. H.; Zhang, A. Q.; Wu, C.; Bai, Y. Insights on artificial interphases of Zn and electrolyte: Protection mechanisms, constructing techniques, applicability, and prospective. *Adv. Funct. Mater.* **2023**, *33*, 2213510.
- [25] Yu, H. M.; Zhang, X. F.; Wang, Y. X.; Li, M. L.; Chen, W.; Hu, Z.; Zhu, M. S.; Huang, Y. Activation and stabilization strategies of aluminum metal anode toward high performance aqueous Al metal batteries. *Adv. Mater.* **2025**, *37*, 2507164.
- [26] Zhang, L.; Rodríguez-Pérez, I. A.; Jiang, H.; Zhang, C.; Leonard, D. P.; Guo, Q. B.; Wang, W. F.; Han, S. M.; Wang, L. M.; Ji, X. L. ZnCl₂ “water-in-salt” electrolyte transforms the performance of vanadium oxide as a Zn battery cathode. *Adv. Funct. Mater.* **2019**, *29*, 1902653.
- [27] Cao, L. S.; Li, D.; Hu, E. Y.; Xu, J. J.; Deng, T.; Ma, L.; Wang, Y.; Yang, X. Q.; Wang, C. S. Solvation structure design for aqueous Zn metal batteries. *J. Am. Chem. Soc.* **2020**, *142*, 21404–21409.
- [28] Cao, X. S.; Xu, W.; Zheng, D. Z.; Wang, F. X.; Wang, Y.; Shi, X.; Lu, X. H. Weak solvation effect induced optimal interfacial chemistry enables highly durable Zn anodes for aqueous Zn-ion batteries. *Angew. Chem., Int. Ed.* **2024**, *63*, e202317302.
- [29] Wei, J.; Zhang, P. B.; Sun, J. J.; Liu, Y. Z.; Li, F. J.; Xu, H. F.; Ye, R. Q.; Tie, Z. X.; Sun, L.; Jin, Z. Advanced electrolytes for high-performance aqueous zinc-ion batteries. *Chem. Soc. Rev.* **2024**, *53*, 10335–10369.
- [30] Ding, W. H.; Zhou, G. L.; Guo, X. J.; Liu, C.; Wang, T. S.; Fu, Y. S.; Yin, J. Z.; Zhang, L. L.; Ang, E. H. Revolutionizing aqueous Zn-ion batteries: Precision control of H₂O activity and Zn deposition through ammonium oxalate additive. *Chem. Eng. J.* **2024**, *481*, 148544.
- [31] He, Z. Q.; Yu, H. M.; Fu, M.; Li, Q. Y.; Li, C. L.; Li, T. B.; Zhang, C. X.; Chen, Y. J.; Chen, L. B. Competitive solvation with regulated ion-coordination chemistry toward dendrite-free and long-life Zn metal anodes. *Energy Storage Mater.* **2024**, *70*, 103469.
- [32] Zhu, Y. X.; Ge, M. Y.; Ma, F. C.; Wang, Q. H.; Huang, P.; Lai, C. Multifunctional electrolyte additives for better metal batteries. *Adv. Funct. Mater.* **2023**, *34*, 2301964.
- [33] Zhu, K. Y.; Xie, W. L.; Yang, W. S. Aqueous zinc-ion pouch cells: From failure mechanisms to practical solutions. *Angew. Chem., Int. Ed.* **2025**, *64*, e202516233.
- [34] Li, C. L.; Liao, T.; Chen, D. P.; You, T. C.; Jiang, X. Z.; Xu, M. H.; Yu, H. M.; Zhou, G.; Li, G. H.; Chen, Y. J. Fabrication of carbon-coated V₂O_{5-x} nanoparticles by plasma-enhanced chemical vapor deposition for high-performance aqueous zinc-ion battery composite cathodes. *Chin. Chem. Lett.* **2025**, *36*, 110557.
- [35] Lee, Y. H.; Jeoun, Y.; Lee, S. H.; Kim, J. H.; Kim, S. Y.; Yu, S. H.; Ahn, K. S.; Sung, Y. E. Byproduct reverse engineering to construct unusually enhanced protection layers for dendrite-free Zn anode. *Chem. Eng. J.* **2023**, *464*, 142580.
- [36] Zhao, Q. G.; Yu, X. Y.; Xue, J. Y.; Zhang, M. H.; Li, Z. G.; Wang, J. H.; Yang, Y.; Zou, Y. G.; Qiao, Y.; Sun, S. G. Competitive tradeoff between Zn deposition and hydrogen evolution reaction on Zn-metal anode. *ACS Energy Lett.* **2024**, *9*, 4102–4110.
- [37] Yu, X. Y.; Chen, M.; Li, Z. G.; Tan, X.; Zhang, H. T.; Wang, J. H.; Tang, Y. L.; Xu, J. P.; Yin, W.; Yang, Y. et al. Unlocking dynamic solvation chemistry and hydrogen evolution mechanism in aqueous zinc batteries. *J. Am. Chem. Soc.* **2024**, *146*, 17103–17113.
- [38] Rana, A.; Paul, S.; Bhadouria, A.; Nguyen, J. H.; Koons, J. F.; Li, C. G.; Das, A.; Roy, K.; Tackett, B. M.; Dick, J. E. Interfacial pH gradients suppress HER at high currents in zinc metal batteries. *Joule* **2025**, *9*, 102167.
- [39] Verma, V.; Kumar, S.; Manalastas, W. Jr.; Srinivasan, M. Undesired reactions in aqueous rechargeable zinc ion batteries. *ACS Energy Lett.* **2021**, *6*, 1773–1785.
- [40] Lim, W. G.; Li, X. L.; Reed, D. Understanding the role of zinc hydroxide sulfate and its analogues in mildly acidic aqueous zinc batteries: A review. *Small Methods* **2024**, *8*, 2300965.
- [41] Xiang, Z. C.; Qiu, Y. B.; Guo, X. P.; Qi, K.; Xu, Z. L.; Xia, B. Y. Inherited construction of porous zinc hydroxide sulfate layer for stable dendrite-free Zn anode. *Energy Environ. Sci.* **2024**, *17*, 3409–3418.
- [42] Cai, Z. J.; Wang, J. P.; Lian, S. T.; Chen, J. W.; Lang, F.; Li, Z.; Li, Q. Regulating the Zn electrode/electrolyte interface toward high stability-insights from the resting time impact on Zn electrode performance. *Adv. Funct. Mater.* **2025**, *35*, 2401367.
- [43] He, J. F.; Tang, Y. C.; Liu, G. G.; Li, H. Q.; Ye, M. H.; Zhang, Y. F.; Yang, Q.; Liu, X. Q.; Li, C. C. Intrinsic hydrogen-bond donors-lined organophosphate superionic nanochannels leveraging high-rate-endurable aqueous Zn batteries. *Adv. Energy Mater.* **2022**, *12*, 2202661.
- [44] Shen, Y. X.; Gu, J. N.; Zhu, Z. Q.; Zhang, L. M.; Lv, X. J.; Li, Y. M. Fast Li⁺ de-solvation kinetics with PDDA intercalated-montmorillonite hybrid artificial interface layer on Cu substrate for lithium metal batteries in a wide climate temperature. *Adv. Funct. Mater.* **2025**, *35*, 2414835.
- [45] Wang, L. P.; Zhang, B.; Zhou, W. H.; Zhao, Z. W.; Liu, X.; Zhao, R. Z.; Sun, Z. H.; Li, H. P.; Wang, X.; Zhang, T. S. et al. Tandem chemistry with Janus mesopores accelerator for efficient aqueous batteries. *J. Am. Chem. Soc.* **2024**, *146*, 6199–6208.
- [46] Jiang, Y. Z.; Wan, Z.; He, X.; Yang, J. R. Fine-tuning electrolyte concentration and metal-organic framework surface toward actuating fast Zn²⁺ dehydration for aqueous Zn-ion batteries. *Angew. Chem., Int. Ed.* **2023**, *62*, e202307274.
- [47] Yu, J. X.; Chen, M. F.; Ma, H.; Liu, W. H.; Tian, Q. H.; Chen, J. Z. Integrating inorganic Zn-ion conductor with nanocellulose towards separator-free and long-life aqueous zinc ion batteries. *J. Colloid Interf. Sci.* **2026**, *701*, 138635.
- [48] Chen, R. W.; Zhang, W.; Huang, Q. B.; Guan, C. H.; Zong, W.; Dai, Y. H.; Du, Z. J.; Zhang, Z. Y.; Li, J. W.; Guo, F. et al. Trace amounts of triple-functional additives enable reversible aqueous zinc-ion batteries from a comprehensive perspective. *Nano-Micro Lett.* **2023**, *15*, 81.
- [49] Wan, T. H.; Saccoccio, M.; Chen, C.; Ciucci, F. Influence of the discretization methods on the distribution of relaxation times deconvolution: Implementing radial basis functions with DRT tools. *Electrochim. Acta* **2015**, *184*, 483–499.



This is an open access article under the terms of the Creative Commons Attribution 4.0 International License (CC BY 4.0, <https://creativecommons.org/licenses/by/4.0/>).

© The Author(s) 2026. Published by Tsinghua University Press.

