



Amphiphilic self-assembly chemistry for dual-interface water regulation in long-life aqueous zinc-vanadium batteries

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ABSTRACT

Aqueous zinc-vanadium batteries (AZVBs) are severely limited by interfacial water-induced failure mechanisms, where uncontrolled water activity simultaneously triggers zinc dendrite growth at the anode and vanadium dissolution at the cathode. Existing strategies predominantly focus on stabilizing a single electrode interface, while the synchronized regulation of dual-electrode interfacial chemistry remains fundamentally challenging. Here, we report an amphiphilic molecular engineering strategy that enables interfacial water redistribution across both electrode interfaces using terpineol (TER) as a bifunctional electrolyte regulator. Owing to its hydrophilic-hydrophobic architecture, TER spontaneously constructs a self-assembled interfacial layer that simultaneously reconstructs Zn^{2+} solvation environments, disrupts hydrogen-bonded water networks, and repels reactive interfacial water from both zinc and vanadium surfaces. This dual-interface regulation homogenizes Zn^{2+} flux, suppresses hydrogen evolution and dendrite propagation, while concurrently inhibiting hydration-driven vanadium dissolution and cathode structural degradation. Consequently, the TER-regulated system achieves highly reversible Zn plating/stripping for over 4000 h at $1 \text{ mA} \cdot \text{cm}^{-2}$ and enables $\text{Zn}||\text{NH}_4\text{V}_4\text{O}_{10}$ (NVO) full cells with 99.65% capacity retention after 800 cycles at $5 \text{ A} \cdot \text{g}^{-1}$. More importantly, this work establishes an amphiphilic interfacial water regulation paradigm that synchronizes anode and cathode stabilization through one-molecule chemistry, offering a generalizable route toward durable aqueous multielectrode energy-storage systems.

KEYWORDS

zinc dendrites, vanadium dissolution, amphiphilic terpineol, electrolyte additive, interfacial water regulation

1 Introduction

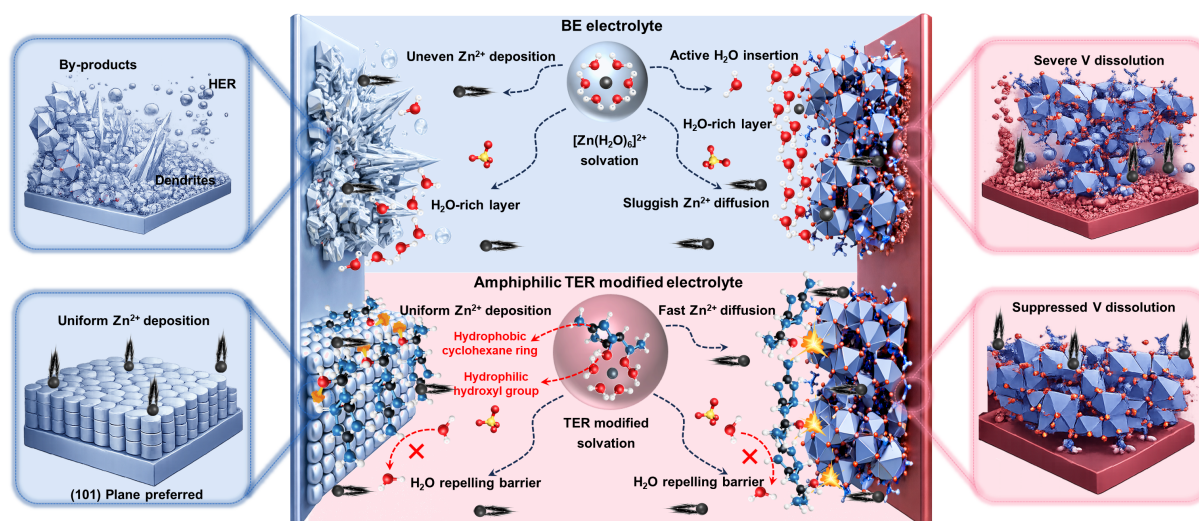
The growing global demand for sustainable, large-scale energy storage systems has accelerated the development of aqueous zinc-vanadium batteries (AZVBs), which are promising for grid-scale applications [1–3]. The zinc anode offers low cost, inherent safety, high theoretical capacity of $820 \text{ mAh} \cdot \text{g}^{-1}$ ($5855 \text{ mAh} \cdot \text{cm}^{-3}$), and a favorable redox potential of -0.762 V vs. the standard hydrogen electrode, enabling stable and efficient energy storage [4–6]. Vanadium-based cathodes provide high specific capacity ($> 300 \text{ mAh} \cdot \text{g}^{-1}$), rich structural diversity, and excellent redox activity, supporting rapid ion intercalation and high energy delivery in full-cell configurations [6–10]. Despite these advantages, their practical deployment is hindered by critical interfacial challenges (Scheme 1). At the zinc anode, water molecules induce parasitic hydrogen evolution reactions (HER)

and promote dendritic growth, leading to low zinc utilization, efficiency loss, and short cycle life [11–14]. Simultaneously, at vanadium-based cathodes, water molecules penetrate the electrode, accelerating vanadium dissolution and structural degradation, which result in active material loss and capacity fading [15–17]. While the aqueous environment contributes to high ionic conductivity and safety, its reactivity at both electrode interfaces fundamentally limits the performance and longevity of AZVBs.

To address these challenges, a variety of strategies have been developed to control water activity and stabilize the electrode interfaces. At the anode, protective coatings and solid electrolyte interphase (SEI) layers in polymeric form [18–22] or inorganic form [23–25] can isolate water molecules, suppress HER, and guide uniform Zn^{2+} deposition [26]. Electrolyte engineering

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Scheme 1 Schematic illustrations of electrode evolution of AZVBs in BE and TER-modified electrolytes.

approaches, including solvation structure optimization [27–29] and gel-based electrolytes [30–32] have been explored to reduce free water content and regulate ion transport. At the cathode, cathode-electrolyte interface (CEI)-like layers [33–35] and electrolyte modifications [36, 37] aim to block water penetration and prevent vanadium dissolution [38–41]. While these methods provide partial mitigation, they are often limited to one electrode and cannot simultaneously regulate both interfaces. Furthermore, some strategies compromise ionic conductivity, are prone to degradation during long-term cycling, or fail to fully suppress water-induced side reactions, leaving uneven ion flux, localized HER, and active material loss unresolved. Effective solutions therefore require a holistic approach capable of coordinating water activity and ion transport at both the anode and cathode, enabling synergistic stabilization and uniform electrochemical environments across the full cell.

In this work, we introduce amphiphilic terpineol (TER) as a multifunctional electrolyte additive capable of regulating water at both electrode interfaces. The hydrophilic hydroxyl group anchors onto the zinc anode and vanadium cathode surfaces, modulating Zn^{2+} solvation and displacing interfacial water, while the hydrophobic cyclic backbone forms a water-repelling barrier (Scheme 1). This amphiphilic self-assembly layer homogenizes local ion flux, suppresses HER, directs uniform zinc deposition, and preserves cathode integrity, resulting in enhanced electrochemical stability. TER-modified systems achieve ultra-stable Zn plating and stripping over 4000 hours at $1 \text{ mA}\cdot\text{cm}^{-2}$ and 900 hours at $5 \text{ mA}\cdot\text{cm}^{-2}$, along with high Coulombic efficiency, excellent rate capability, and 99.65% capacity retention over 800 cycles at $5 \text{ A}\cdot\text{g}^{-1}$ in full-cell configurations. These results demonstrate a mechanistically guided dual-electrode interfacial modulation strategy that overcomes water-induced limitations and enables long-lived, high-performance aqueous zinc-vanadium batteries.

2 Results and discussion

2.1 Zn^{2+} solvation and interfacial water network modulation by TER

To elucidate the impact of amphiphilic terpineol (TER) on the solvation structure of Zn^{2+} ions in aqueous electrolytes, quantum

chemical calculations were first performed to examine its electronic properties. Electrostatic potential (ESP) mapping (Fig. 1(a)) provides a direct visualization of the amphiphilic character of TER, the hydrophilic hydroxyl group exhibits pronounced electron density, while the hydrophobic cyclic backbone remains nonpolar. This asymmetric charge distribution allows the single hydroxyl group to act as a polar anchoring site, while the hydrophobic carbon framework can perturb the local organization of water molecules. As a result, the cooperative action of the hydrophilic hydroxyl group and hydrophobic backbone facilitates the modulation of the local Zn^{2+} solvation/interfacial environment [42–44]. Molecular orbital analysis (Fig. 1(b)) further reveals that the highest occupied molecular orbital (HOMO) of TER lies at a higher energy level than that of water, while its lowest unoccupied molecular orbital (LUMO) is lower than that of water, indicating a higher propensity for electron transfer and a stronger ability to influence interfacial electronic interactions and adsorption-induced charge redistribution [45]. Such electronic features underpin the stabilization of zinc deposition by forming a protective interfacial layer that suppresses hydrogen evolution while maintaining efficient Zn^{2+} transport. Molecular dynamics (MD) simulations were employed to further probe TER's influence on Zn^{2+} solvation within ZnSO_4 electrolytes (Fig. 1(c)). TER molecules preferentially occupy coordination sites adjacent to Zn^{2+} ions, partially displacing water molecules from the primary solvation shell. Radial distribution function (RDF) analysis (Fig. 1(d)) confirms the emergence of a $\text{Zn}-\text{O}(\text{TER})$ peak together with a decreased $\text{Zn}-\text{O}(\text{H}_2\text{O})$ intensity, demonstrating that, despite its relatively low concentration, TER can still participate in the local Zn^{2+} solvation/interfacial environment owing to its strong affinity toward Zn^{2+} and favorable interfacial adsorption capability, thereby perturbing the original Zn^{2+} solvation structure. Binding energy calculations (Fig. 1(e)) reveal that Zn^{2+} -TER interactions ($-221.354 \text{ kcal}\cdot\text{mol}^{-1}$) are significantly stronger than Zn^{2+} - H_2O interactions, highlighting the strong stabilizing effect of TER on the solvation shell, which reduces water reactivity and mitigates side reactions during zinc deposition.

Beyond direct Zn^{2+} coordination, TER also disrupts the hydrogen-bonding network of interfacial water. Raman spectroscopy (Fig. 1(f)) shows that increasing TER concentration shifts the O-H stretching vibration, reflecting weakening of tightly bound water clusters. Deconvolution analysis (Figs. 1(g) and 1(h)) indicates a reduction in strong hydrogen bonds from 46.61% in

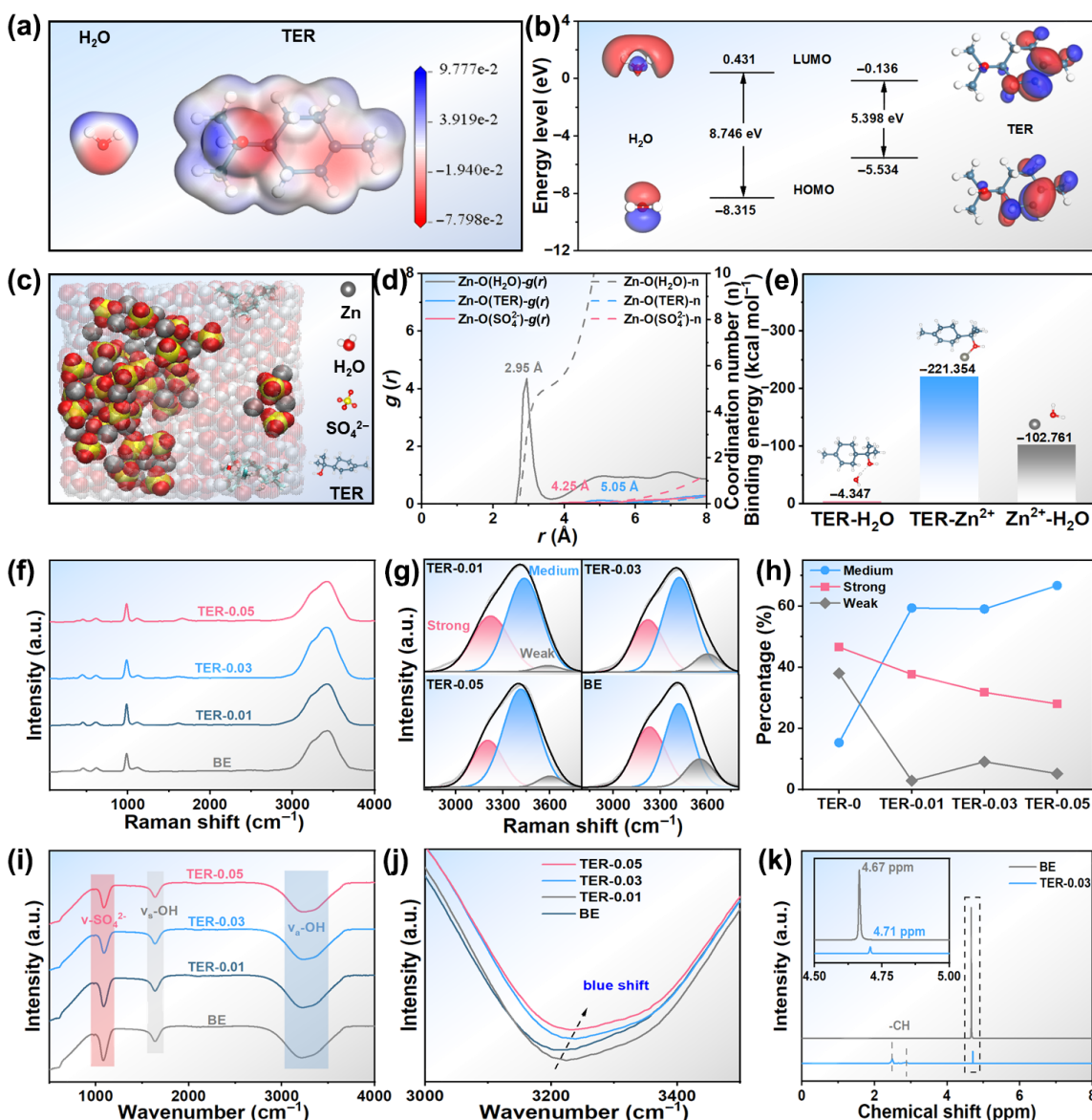


Figure 1 (a) ESP mapping of TER and H₂O. (b) Comparison of the LUMO/HOMO energy levels for TER and H₂O. (c) MD simulation snapshots of the TER-0.03 system within the ZnSO₄ electrolyte. (d) Radial distribution functions $g(r)$ and corresponding coordination numbers in the TER-0.03 system. (e) Calculated binding energies between Zn²⁺ and various components in the electrolyte system. (f) Raman spectra of electrolytes with different concentrations of TER. (g) Deconvoluted O-H stretching vibration peaks. (h) Distribution of the proportions of hydrogen bonds with different strengths. (i) FTIR spectra of electrolytes with different TER concentrations. (j) Magnified view of the FTIR peaks. (k) ¹H NMR spectra of the BE and TER-0.03 system.

the bare electrolyte to 28.02% in the TER-modified system, with corresponding increases in medium and weak hydrogen bonds [46]. Fourier-transform infrared (FTIR) spectra (Figs. 1(i) and 1(j)) further confirm this effect, showing a blue shift of the O-H vibration around 3200 cm⁻¹, indicative of weakened water network interactions [47]. Complementary ¹H NMR spectra (Fig. 1(k)) show a downfield shift from 4.67 to 4.71 ppm, consistent with decreased electron density around water protons, further evidencing TER-induced water network perturbation. Collectively, these computational and spectroscopic results demonstrate that TER simultaneously stabilizes Zn²⁺ solvation and modulates the interfacial water network. By partially replacing coordinated water and disrupting hydrogen-bonding clusters, TER establishes a dense, water-depleted interfacial layer that suppresses HER, promotes uniform Zn²⁺ diffusion, and lays the mechanistic foundation for enhanced anode stability in aqueous zinc-vanadium batteries.

2.2 Adsorption and interfacial layer formation at the Zn anode by TER

The introduction of amphiphilic terpineol (TER) profoundly alters the zinc anode/electrolyte interface through strong adsorption and ordered molecular alignment. Contact angle measurements (Fig. 2(a)) show a pronounced decrease from 94.63° in the bare electrolyte to 59.27° upon TER addition, reflecting enhanced wettability and the formation of a densely packed interfacial layer. This ordered alignment of TER molecules disrupts the hydrogen-bonding network of interfacial water [48], reduces surface tension, and establishes a favorable environment for uniform zinc deposition. Density functional theory (DFT) calculations further elucidate the molecular-level interaction between TER and the Zn(002) surface (Fig. 2(b)). TER exhibits a stronger adsorption energy (-1.96 eV) than Zn (-0.29 eV), highlighting its preferential binding to the anode surface. Differential charge density analysis

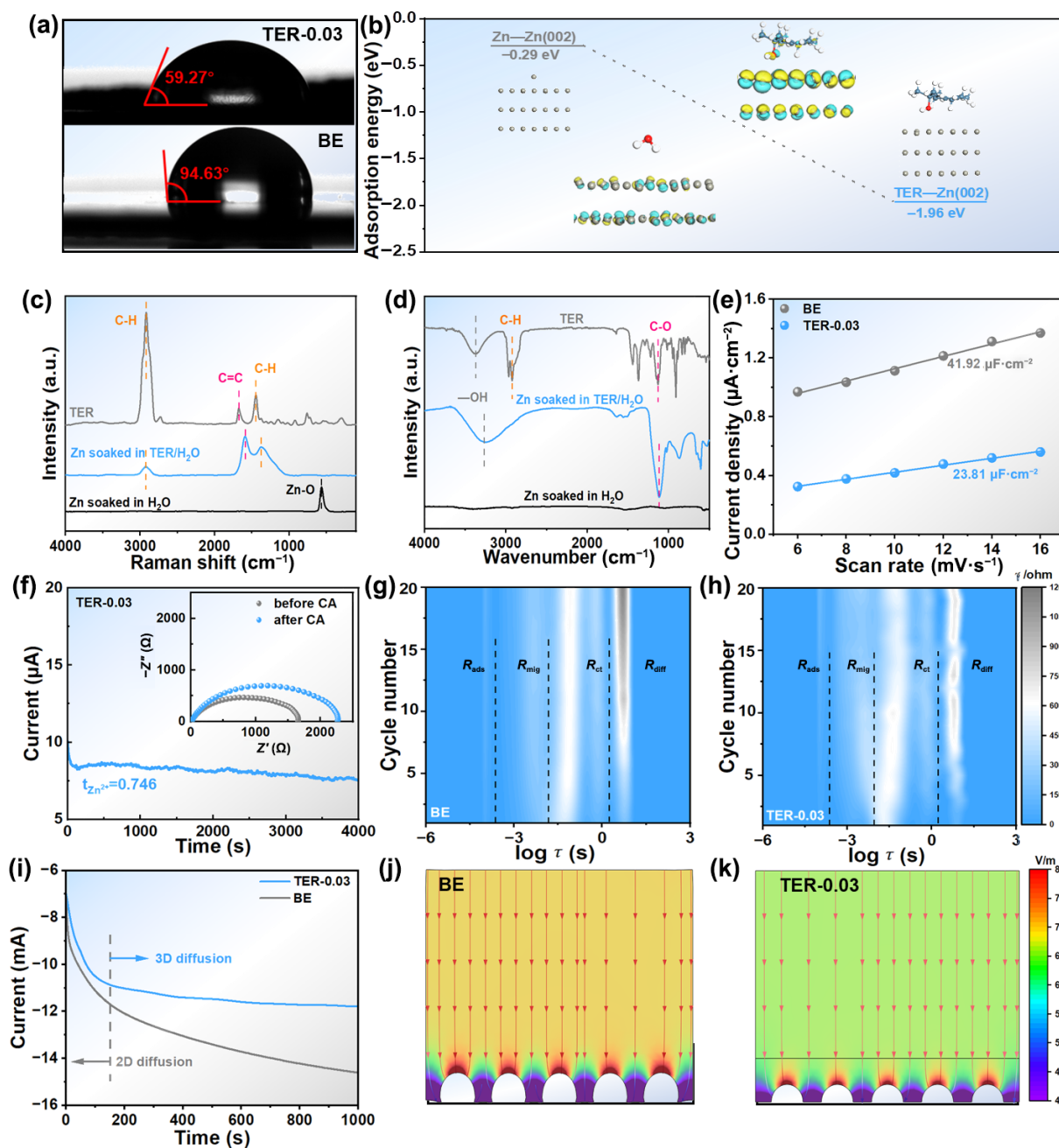


Figure 2 (a) Contact angle measurements of different electrolytes on the Zn anode surface. (b) Comparison of the adsorption energies of TER and water on the Zn(002) crystal plane. (c) Raman spectra and (d) FTIR spectra of Zn foils immersed in different electrolytes. (e) EDLC values derived from cyclic voltammetry (CV) curves. (f) Zn^{2+} transference number of the TER-0.03 electrolyte system. DRT plots of the (g) BE system and (h) TER-0.03 system during symmetric cell cycling. (i) CA curves on Zn anode surface in BE and TER system. Electric field distribution on the Zn anode surface in the (j) BE and (k) TER-0.03 electrolyte system.

confirms electron density transfer from Zn atoms to the hydroxyl oxygen of TER, indicating strong chemisorption. This adsorption not only stabilizes the interface but also facilitates the displacement of water molecules from the Zn^{2+} solvation shell, promoting efficient ion transport and suppressing side reactions. Spectroscopic analyses corroborate these findings. Raman spectra (Fig. 2(c)) display characteristic C–H and C=C skeletal vibrations from TER on the Zn surface, absent in the bare electrolyte. FTIR spectra (Fig. 2(d)) reveal distinct C–O and –OH stretching vibrations, further confirming the presence of a stable TER adsorption layer. Together, these results demonstrate that TER forms a robust interfacial layer capable of regulating water activity and electrostatic interactions at the anode surface.

The TER layer also profoundly modulates the electrochemical environment at the zinc interface. Electric double-layer capacitance (EDLC) measurements reveal a significant decrease from $41.92 \mu\text{F}\cdot\text{cm}^{-2}$ in the bare electrolyte to $23.81 \mu\text{F}\cdot\text{cm}^{-2}$ in the TER-modified system (Fig. 2(e), Fig. S1 in the Electronic Supplementary Material (ESM)), indicating dense TER coverage and displacement of high-dielectric water molecules. According to the Helmholtz model [49], this increase in effective interfacial thickness optimizes the local electric field distribution, reduces water-mediated parasitic reactions, and promotes uniform Zn^{2+} nucleation and deposition. Complementarily, the desolvation energy barrier of Zn^{2+} decreases from $61.29 \text{ kJ}\cdot\text{mol}^{-1}$ to $59.69 \text{ kJ}\cdot\text{mol}^{-1}$ in the TER system (Fig. S2 in the ESM), and the

Zn^{2+} transference number ($t_{\text{Zn}^{2+}}$) rises from 0.533 to 0.746 (Fig. 2(f), Fig. S3 in the ESM), reflecting enhanced ionic mobility and accelerated interfacial charge transfer. These combined effects ensure more homogeneous ion flux and mitigate localized overpotential spikes that typically lead to dendritic growth.

Distribution of relaxation times (DRT) analysis (Figs. 2(g)–2(h), Fig. S4 in the ESM) confirms that the TER layer maintains stable charge transfer and ion diffusion pathways throughout prolonged cycling. Chronoamperometry (CA) tests (Fig. 2(i) in the ESM) demonstrate a smoother and more stable current response for the TER-modified system, with nucleation overpotential (Fig. S5 in the ESM) increasing from 40.50 mV in the bare electrolyte to 54.05 mV, this moderate increase suggests suppressed random nucleation at highly active sites, which favors homogeneous 3D Zn deposition and dendrite-free morphology. Moreover, the impact of TER on Zn^{2+} deposition kinetics and long-term stability is further validated through Multiphysics simulations and electrochemical tests. COMSOL simulations (Figs. 2(j) and 2(k) in the ESM) indicate that TER homogenizes the local electric field, effectively suppressing tip effects at protrusions that drive preferential deposition. Collectively, these results indicate that the amphiphilic TER adsorption layer not only suppresses interfacial side reactions but also guides uniform Zn^{2+} deposition, providing a robust mechanistic basis for long-term dendrite-free operation and enhanced anode durability.

2.3 Interfacial regulation and dendrite suppression by TER

The incorporation of amphiphilic TER into ZnSO_4 electrolyte substantially alters the Zn/electrolyte interface, promoting uniform zinc deposition while simultaneously suppressing parasitic reactions. Tafel polarization measurements (Fig. 3(a)) reveal that the TER-0.03 system exhibits a positive shift in corrosion potential (E_{corr}) and a pronounced reduction in corrosion current density (I_{corr}) relative to the BE, indicating that the chemisorbed TER layer forms a dense and continuous barrier. This barrier effectively isolates the Zn surface from water molecules, mitigating self-corrosion and enhancing interfacial stability. Linear sweep voltammetry (LSV, Fig. 3(b)) further demonstrates that the HER overpotential increases significantly in the TER-modified system. By occupying reactive sites and disrupting the hydrogen-bonding network at the interface, TER raises the kinetic barrier for water reduction, effectively suppressing hydrogen evolution during both plating and stripping cycles, thereby preserving the integrity of the anode surface. X-ray diffraction (XRD) analysis of cycled electrodes provides complementary evidence for TER's stabilizing effect (Fig. 3(c)). In the BE system, peaks at 9.8° , 21.2° , and 31.3° corresponding to basic zinc sulfate (BZS, JCPDS PDF#38-0383) appear, reflecting the accumulation of insulating by-products from water-induced side reactions. In contrast, TER-0.03 electrodes display only the characteristic metallic Zn peaks (JCPDS PDF#04-0831), demonstrating that TER preserves interfacial pH, inhibits by-product formation, and maintains electrochemical activity. *In situ* optical microscopy visually confirms these findings, showing extensive gas bubble formation and protruding dendrites in the BE system (Fig. 3(d)), while TER-modified surfaces remain smooth, planar, and free from dendritic protrusions (Fig. 3(e)), underscoring TER's dual role in mitigating both HER and dendrite growth.

To elucidate the impact of TER on Zn crystallographic evolution, the relative texture coefficient (RTC) of primary Zn

planes was analyzed over deposition times of 1, 5, 10, and 20 h (Fig. S6 in the ESM): $\text{RTC}_{(hkl)} = \frac{I_{(hkl)}/I_{0(hkl)}}{\sum (I_{(hkl)}/I_{0(hkl)})} \times 100$ [46, 50], where $I_{(hkl)}$ denotes the measured intensity of a given plane and $I_{0(hkl)}$ is the corresponding standard intensity from the Zn reference pattern (JCPDS PDF#04-0831). In the TER-0.03 system, the (101) plane progressively dominates (Fig. 3(f)), promoting dense and uniform Zn deposition. Conversely, the BE system exhibits competitive growth among multiple planes, correlating with irregular dendrite formation and increased corrosion susceptibility. Scanning electron microscopy (SEM) further corroborates these findings. Zn surfaces cycled in BE display a porous, mossy structure with severe corrosion pits (Fig. 3(g)), whereas TER-0.03 deposits form a dense, smooth, and layered morphology (Fig. 3(h)), consistent with RTC analysis. 3D laser scanning confocal microscopy (Figs. 3(i) and 3(j)) and corresponding surface roughness analysis reveal a significant reduction for TER-0.03, demonstrating its effective leveling effect. By homogenizing the local current density, guiding uniform Zn^{2+} nucleation, and stabilizing the interfacial environment, TER enables dendrite-free deposition while maintaining efficient ion transport. Collectively, these results demonstrate that TER not only suppresses hydrogen evolution and side reactions but also directs preferential crystallographic growth. The synergistic interplay of electric double-layer modification, surface chemisorption, and crystallographic control provides a robust mechanistic foundation for enhanced cycling stability and dendrite-free behavior of Zn anodes in both symmetric and full-cell configurations.

2.4 Cycling stability and electrochemical performance enhancement by TER

To determine the optimal concentration of TER, Zn||Zn symmetric cells were tested in electrolytes containing 0.01, 0.03, and 0.05 M TER under $5 \text{ mA}\cdot\text{cm}^{-2}$ and $5 \text{ mAh}\cdot\text{cm}^{-2}$ (Fig. S7 in the ESM). The 0.03 M system exhibits the most stable voltage profile with minimal polarization and negligible fluctuations during extended cycling. In contrast, cells with 0.01 M TER show pronounced voltage drift due to insufficient interfacial coverage, while 0.05 M TER results in increased polarization, likely caused by excessive surface adsorption impeding Zn^{2+} transport. These results indicate that 0.03 M TER provides an optimal balance between effective interfacial coverage and unhindered ion transport, and this concentration was therefore adopted for all subsequent experiments. The TER-0.03 system demonstrates remarkable long-term cycling stability compared with the BE. At $1 \text{ mA}\cdot\text{cm}^{-2}$ and $0.5 \text{ mAh}\cdot\text{cm}^{-2}$, the TER-0.03 symmetric cell operates stably for over 4000 h, whereas the BE system exhibits rapid voltage fluctuations and fails after 272 h (Fig. 4(a)). At $5 \text{ mA}\cdot\text{cm}^{-2}$ and $5 \text{ mAh}\cdot\text{cm}^{-2}$ (Fig. 4(b)), TER-0.03 sustains dendrite-free cycling beyond 900 h, while the BE system fails within 200 h. Even under high current densities of $10 \text{ mA}\cdot\text{cm}^{-2}$ (Fig. S8 in the ESM), TER-0.03 maintains stable cycling for 300 h, whereas BE shows early short-circuiting. Under high areal capacity conditions of $5 \text{ mA}\cdot\text{cm}^{-2}$ and $20 \text{ mAh}\cdot\text{cm}^{-2}$ (Fig. 4(c)), TER-0.03 achieves stable operation for over 300 h with a Zn utilization of 47.87%, while the BE system deteriorates rapidly after 80 h. These results clearly demonstrate that the TER adsorption layer dramatically enhances cycling lifespan and interfacial stability, even under harsh operating conditions.

The rate capability of TER-0.03 is also superior (Fig. 4(d)), maintaining well-defined voltage plateaus as the current density

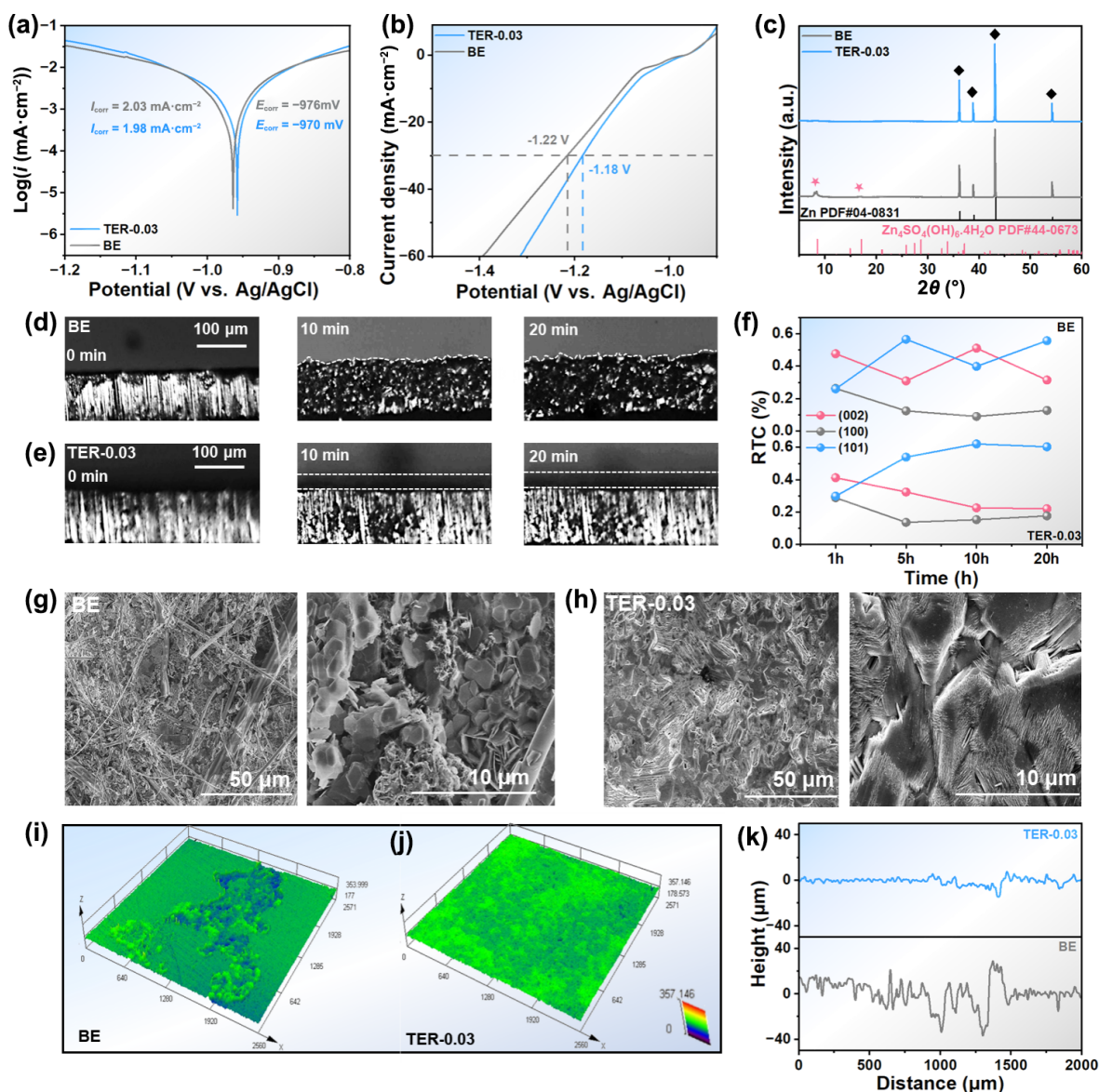


Figure 3 (a) Tafel polarization curves and (b) LSV curves of the Zn anode in BE and TER-0.03 electrolytes. (c) XRD patterns of cycled Zn anodes. *In situ* optical microscopy images of the Zn deposition process in the (d) BE system and (e) the TER-0.03 system. (f) Evolution of the RTC of different crystal planes during Zn deposition. SEM images of the cycled Zn anode surface in the (g) BE system and (h) TER-0.03 system. 3D confocal microscopy morphologies of cycled Zn anodes in the (i) BE system and (j) the TER-0.03 system. (k) Statistical surface roughness values extracted from the corresponding 3D morphology images.

increases from 1 to 10 mA·cm⁻², the TER-0.03 system consistently exhibits stable voltage plateaus as the current density increases from 1 to 10 mA·cm⁻², suggesting that interfacial polarization does not deteriorate significantly. By fitting the relationship between current density and overpotential, the exchange current density (i_0) was calculated using the equation $i = i_0 \frac{F}{2RT} \eta$ [51], where i is current density, i_0 is the exchange current density, F and R are Faradic constant and gas constant, T is temperature constant and η is overpotential. The calculated i_0 values (Fig. 4(e)) confirm that although the TER molecules form an adsorption layer at the interface, their unique amphiphilic structure does not impede ion transport. Instead, it promotes interfacial charge transfer by accelerating the desolvation process. Coulombic efficiency (CE) measurements in Zn||Cu half-cells (Fig. 4(f)) further demonstrate highly reversible Zn plating/stripping in TER-0.03, with an average CE of 98.74% (Fig. 4(g)), while the BE system suffers rapid decay to 92.4% (Fig. S9 in the ESM) due to irreversible Zn loss and dendrites formation.

Comparison with state-of-the-art aqueous Zn-ion electrolytes (Fig. 4(h)) positions TER-0.03 at the forefront in both cumulative capacity and cycling lifespan [52–67]. Mechanistically, this enhanced performance arises from the dense TER layer at the Zn/electrolyte interface, which (i) homogenizes local current density to suppress dendrite nucleation, (ii) stabilizes the electric double layer and mitigates the interfacial side reactions, and (iii) promotes efficient Zn²⁺ desolvation and transport. Collectively, these synergistic effects enable dendrite-free, highly reversible Zn deposition under extended cycling and high-current conditions, providing a robust platform for long-lived aqueous zinc metal batteries.

2.5 Device-level performance and full-cell interface stabilization by TER

To evaluate the practical applicability of the TER additive, Zn||NH₄V₄O₁₀ (NVO) coin cells were assembled and systematically investigated (Fig. 5(a)). Rate capability

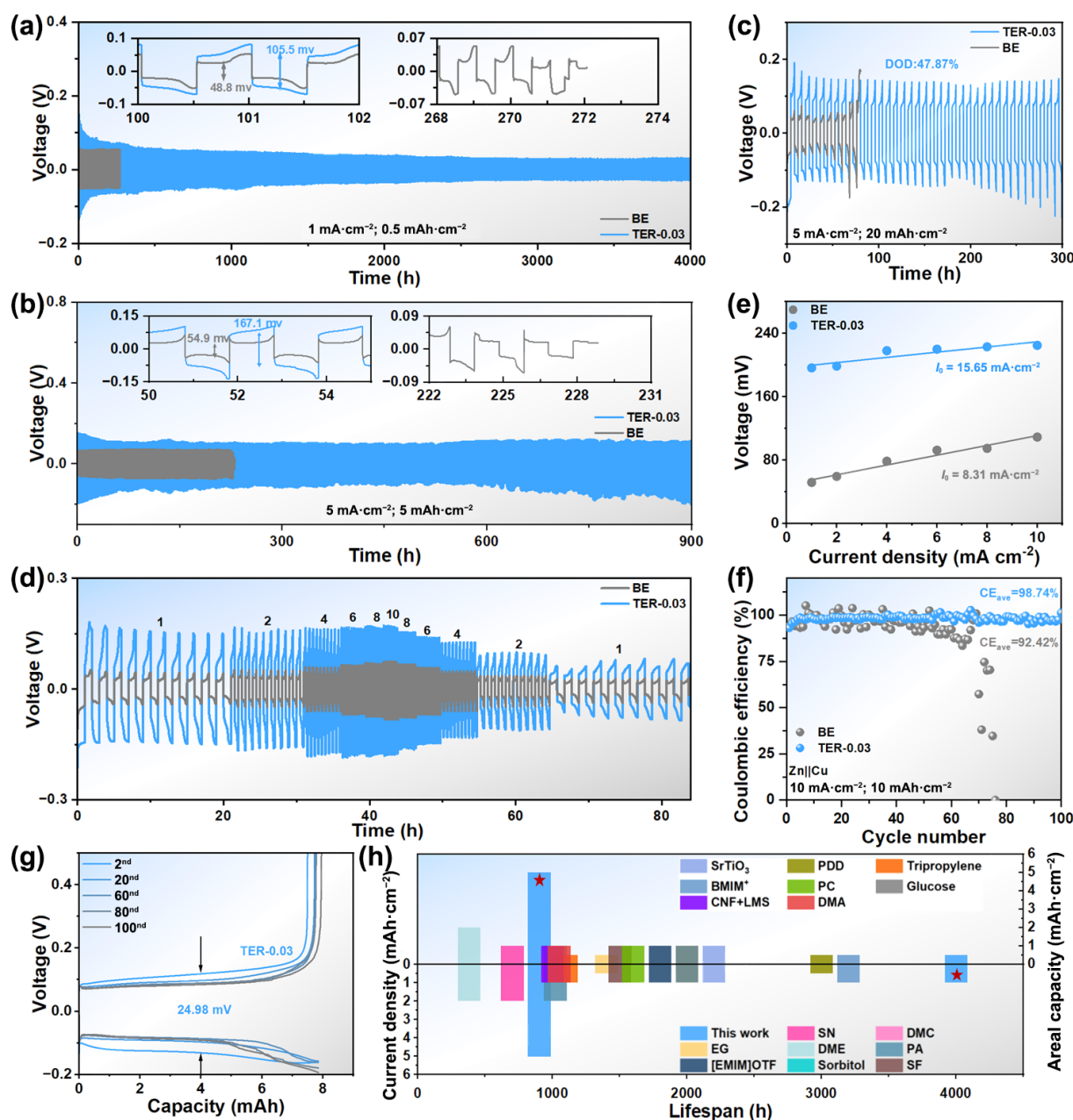


Figure 4 (a)–(c) Long-term cycling voltage-time profiles of Zn||Zn symmetric cells at different current densities and areal capacities. (d) Rate performance of the symmetric cells. (e) Linear fitting of the overpotential data at different current densities and the corresponding calculation of I_0 . (f) CE of Zn||Cu half-cells in BE and TER-0.03 system. (g) Voltage distribution characteristics of the TER-0.03 system during the initial cycling stage. (h) Performance comparison of this work with other reported works.

measurements (Fig. 5(b)) reveal that the TER-0.03 system delivers discharge capacities of 250.35, 236.54, 220.12, 206.45, and 163.30 $\text{mAh}\cdot\text{g}^{-1}$ at current densities of 0.4, 0.8, 1.0, 2.0, and 5.0 $\text{A}\cdot\text{g}^{-1}$, respectively. In comparison, the bare electrolyte (BE) system exhibits lower capacities of 230.92, 221.88, 215.10, 184.97, and 114.12 $\text{mAh}\cdot\text{g}^{-1}$ at the same rates. Upon returning to 0.4 $\text{A}\cdot\text{g}^{-1}$, TER-0.03 fully recovers to 234.12 $\text{mAh}\cdot\text{g}^{-1}$, whereas BE recovers only to 216.34 $\text{mAh}\cdot\text{g}^{-1}$, demonstrating superior rate performance, high reversibility, and rapid Zn^{2+} intercalation/de-intercalation kinetics in the TER-modified system. These results indicate that the amphiphilic TER layer not only accelerates ion transport but also effectively mitigates voltage polarization under high-rate operation, providing a stable interfacial environment for both the anode and cathode. Galvanostatic charge-discharge (GCD) curves further support these findings. As the current density increases, BE

cells display escalating voltage hysteresis and progressive capacity loss (Fig. 5(c)), while TER-0.03 cells maintain nearly overlapping charge-discharge plateaus (Fig. 5(d)), indicative of a stabilized interface and enhanced Zn^{2+} kinetics. Cyclic voltammetry (CV), shows identical redox peak positions for both systems (Fig. 5(e), Fig. S10 in the ESM), confirming that TER does not alter the fundamental electrochemical reaction mechanism [68]. However, the integrated CV area for TER-0.03 is larger, consistent with higher capacity. The b -values derived from CV scans at multiple rates (Fig. 5(f), Fig. S11 in the ESM) approach 0.92, compared to 0.90 in BE (Fig. S12 in the ESM), highlighting that charge storage in TER-0.03 is predominantly surface-controlled pseudocapacitive, enabling fast interfacial Zn^{2+} transport and efficient utilization of the electrode surface [69].

The interfacial stability of full cells was further assessed through

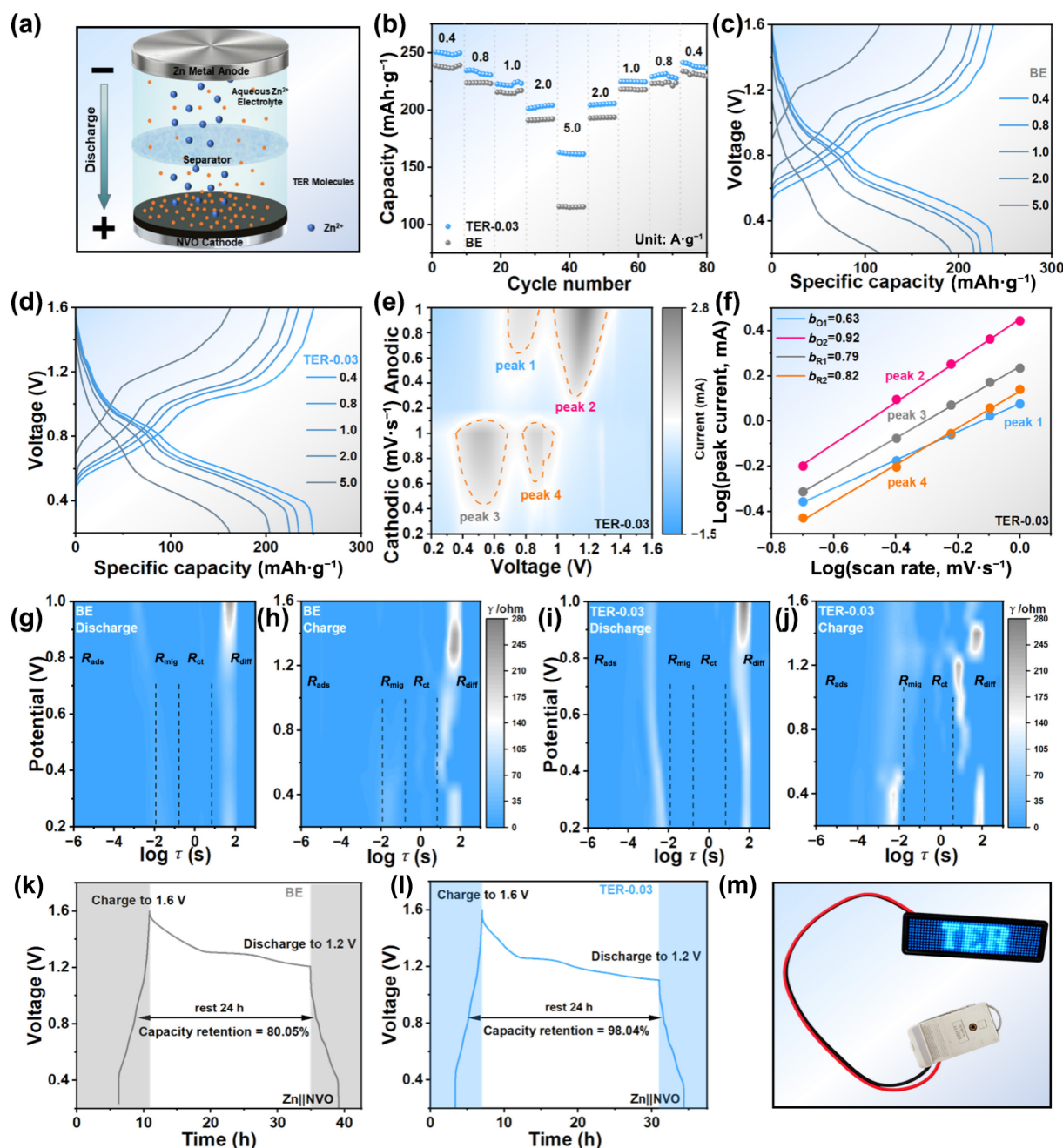


Figure 5 (a) Schematic illustration of the Zn||NVO full-cell configuration. (b) Rate performance comparison of BE and TER-0.03 cells at different current densities. GCD curves of the (c) BE system and (d) TER-0.03 system at various current densities. (e) CV contour map of TER-0.03 system. (f) Distribution of the b -value corresponding to the capacitive contribution. DRT contour maps of the BE system full cell during (g) discharging and (h) charging process. DRT contour maps of the TER-0.03 system full cell during (i) discharging and (j) charging process. Capacity retention after 24 h rest for (k) BE and (l) TER-0.03 cells. (m) Practical demonstration of a coin cell array using TER-0.03 electrolyte powering a custom LED panel.

DRT analysis. In BE cells, DRT heatmaps during discharge (Fig. 5(g)) and charge (Fig. 5(h), Fig. S13 in the ESM) reveal broadening and shifting of peaks corresponding to charge-transfer (R_{ct}) and ion-diffusion (R_{diff}) resistances, indicative of by-product accumulation and structural degradation over cycling. In contrast, TER-0.03 cells exhibit highly symmetric and stable DRT signals during both discharge (Fig. 5(i)) and charge (Fig. 5(j), Fig. S14 in the ESM), with minimal peak drift and overall lower impedance. These results confirm that the chemisorbed TER layer maintains a highly conductive and robust solid/liquid interface, suppressing interfacial side reactions, enhancing charge transfer, and ensuring uniform ion diffusion throughout cycling. Practical self-discharge

behavior was examined by testing capacity retention after resting the cells for 24 h. The BE system retains only 80.08% of its initial capacity (Fig. 5(k)), whereas TER-0.03 retains 98.04% (Fig. 5(l)), indicating that the TER-derived amphiphilic interfacial adsorption layer effectively reduces the direct exposure of active water molecules at the electrode/electrolyte interface. This water-poor interfacial microenvironment suppresses spontaneous parasitic reactions and alleviates vanadium dissolution under open-circuit conditions. This enhanced interfacial stability directly contributes to long-term capacity retention and consistent electrochemical performance, even during extended idle periods. Finally, the real-world feasibility of TER-modified electrolytes was demonstrated

using a coin cell array to power a custom light emitting diode (LED) display (Fig. 5(m)). The array maintained stable and continuous illumination over extended periods, highlighting the combined benefits of interfacial water regulation, dendrite suppression, and rapid Zn^{2+}/V -ion kinetics in practical devices. Together, these results confirm that TER simultaneously enhances rate capability, cycling stability, and device-level performance, offering a versatile strategy for achieving high-performance, long-lived aqueous zinc-vanadium batteries.

2.6 Vanadium dissolution suppression and cathode stabilization by TER

Continuous dissolution of vanadium-based cathode materials in

aqueous electrolytes is a major contributor to capacity decay and performance deterioration in $\text{Zn}||\text{NVO}$ full cells [62]. The amphiphilic TER molecule effectively constructs a water-poor interfacial microenvironment on the NVO cathode surface, providing robust structural stabilization during cycling. Low-current cycling at $0.4 \text{ A}\cdot\text{g}^{-1}$ (Fig. 6(a)) demonstrates that the TER-0.03 full cell exhibits an initial discharge capacity of approximately $252.62 \text{ mAh}\cdot\text{g}^{-1}$ and retains 82.65% of this capacity after 60 cycles, with smooth charge-discharge plateaus and minimal voltage polarization (Fig. 6(b)). In contrast, the BE system starts at $219.25 \text{ mAh}\cdot\text{g}^{-1}$ and drops to 31.58% of its initial capacity over the same period, accompanied by pronounced voltage hysteresis. At a high current density of $5 \text{ A}\cdot\text{g}^{-1}$, TER-0.03 maintains 99.65% capacity retention over 800 cycles (Fig. 6(c)), whereas BE retains

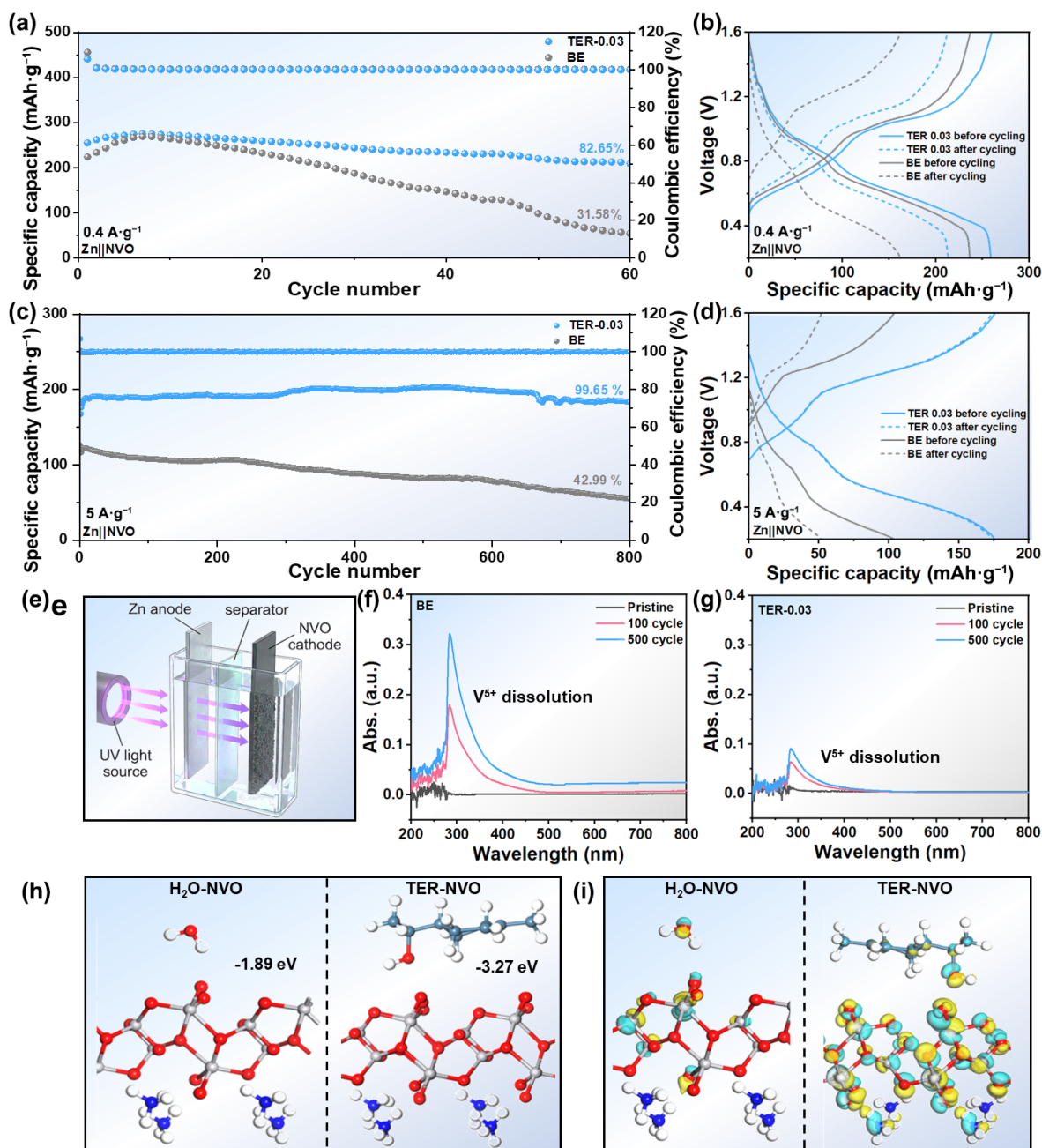


Figure 6 (a) Long-term cycling performance of $\text{Zn}||\text{NVO}$ full cells at a low current density of $0.4 \text{ A}\cdot\text{g}^{-1}$. (b) Corresponding GCD curves at $0.4 \text{ A}\cdot\text{g}^{-1}$. (c) Long-term cycling performance at a high current density of $5 \text{ A}\cdot\text{g}^{-1}$. (d) Corresponding GCD curves at $5 \text{ A}\cdot\text{g}^{-1}$. (e) Schematic of the UV-vis dissolution experiment for evaluating vanadium leaching. UV-vis absorption spectra of electrolytes after cycling in (f) BE and (g) TER-0.03 system. (h) DFT-calculated adsorption energies of H_2O and TER on the NVO(001) surface. (i) Differential charge density maps of H_2O and TER on the NVO(001) surface.

only 42.99% (Fig. 6(d)), highlighting TER's remarkable ability to stabilize the cathode under fast kinetics and prolonged cycling. The TER-0.03 system also demonstrates excellent rate performance, sustaining over 165 mAh·g⁻¹ at 5 A·g⁻¹, indicative of rapid Zn²⁺ intercalation/de-intercalation and efficient ion transport even under high-current operation. Overall, although deeper discharge at relatively low current densities may lead to more pronounced vanadium dissolution and structural relaxation on the cathode side, TER addition effectively mitigates capacity decay and cathode structural degradation over a wide range of current densities.

The suppression of vanadium dissolution by TER is directly evidenced by UV-vis spectroscopy (Fig. 6(e)). Electrolytes from cycled BE cells display a strong absorption peak at 310 nm, characteristic of V⁵⁺ species (Fig. 6(f)), confirming substantial vanadium dissolution [70, 71]. In contrast, TER-0.03 electrolytes show only minimal absorbance at this wavelength (Fig. 6(g)), indicating nearly complete inhibition of vanadium dissolution [37]. This effect arises from TER's amphiphilic structure, hydrophilic hydroxyl groups anchor to the NVO surface via strong chemisorption, while the hydrophobic cyclic backbone forms a barrier that repels water molecules, effectively suppressing hydration-driven vanadium extraction. DFT calculations further support this mechanism (Fig. 6(h)). TER binds to the NVO(001) surface with an adsorption energy of -3.27 eV, significantly more negative than water (-1.89 eV), confirming preferential adsorption and spontaneous displacement of interfacial water. Differential charge density analysis (Fig. 6(i)) shows pronounced electron accumulation between TER hydroxyl oxygen and vanadium atoms, highlighting strong chemisorption and local electronic stabilization of the cathode surface. This amphiphilic interfacial layer not only inhibits vanadium dissolution but also preserves the lattice integrity of NVO, maintains structural order, and minimizes side reactions, thereby sustaining high capacity and excellent cycling stability over prolonged operation.

Together, these results demonstrate that TER establishes a robust, water-repelling, and chemisorbed interfacial layer that simultaneously suppresses vanadium dissolution, stabilizes cathode structure, and enables efficient ion transport. The initial higher capacity, coupled with sustained retention over hundreds of cycles, confirms that dual-interface regulation by amphiphilic TER is an effective strategy to enhance both the electrochemical performance and long-term durability of aqueous Zn||NVO full cells, particularly under high-rate and extended cycling conditions.

3 Conclusion

In summary, amphiphilic terpineol (TER) serves as an effective dual-interface regulator for aqueous Zn||NH₄V₄O₁₀ (NVO) batteries. The molecular structure of TER, featuring a polar hydroxyl group and a hydrophobic cyclic backbone, enables its coordinated interfacial function at both electrodes. At the Zn anode, the hydroxyl group promotes interfacial anchoring, while the hydrophobic moiety helps reduce the local exposure of active water molecules, thereby suppressing hydrogen evolution, corrosion, and uneven Zn deposition. At the NVO cathode, TER adsorption constructs a localized water-poor interfacial microenvironment, alleviating water-induced side reactions and vanadium dissolution while preserving cathode structural stability. Benefiting from this amphiphilic dual-interface regulation, the TER-0.03 electrolyte delivers stable Zn plating/stripping for over 4000 h at 1 mA·cm⁻² and 900 h at 5 mA·cm⁻², with an average

Coulombic efficiency of 98.74%. Moreover, Zn||NVO full cells exhibit 99.65% capacity retention after 800 cycles at 5 A·g⁻¹ and 98.04% capacity retention after 24 h resting. This work highlights amphiphilic molecular engineering as a feasible strategy for stabilizing both anode and cathode interfaces in aqueous zinc-based batteries.

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Declaration of conflicting interests

The authors declare no conflicting interests regarding the content of this article.

Data availability

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

Electronic Supplementary Material: Supplementary material (detailed experimental procedures for material synthesis, characterization techniques and measurement methods, and computational methods and procedures. Some preliminary characterization results and control experiments) is available in the online version of this article at <https://doi.org/10.26599/NRE.2026.9120252>.

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