



Coanda-effect-inspired curved polyacrylonitrile/sodium lignosulfonate nanofiber interphase with a negatively charged surface for durable zinc anodes

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ABSTRACT

The interfacial instability of the Zn anode hinders practical aqueous Zn-ion batteries (ZIBs). Existing artificial layers focus on static field distribution and zincophilic sites but overlook dynamic ion transport near the electrode. Inspired by the Coanda effect, we construct a composite interfacial layer comprising polyacrylonitrile (PAN) and sodium lignosulfonate (LS) on Zn foil (PAN-LS@Zn) via in-situ electrospinning. The curved fibrous topologies of PAN, coupled with the negatively charged surface of LS, homogenize the Zn^{2+} flux and establish a local ion reservoir that substantially elevates the Sand's time, thereby mitigating concentration polarization. Concurrently, the preferential interaction between the functional groups and the (101)/(100) planes steers Zn nucleation along (002). Consequently, the PAN-LS@Zn anode enables a Zn//Zn symmetric cell to achieve a cycling life of 4345 h at $1 \text{ mA cm}^{-2}/1 \text{ mAh cm}^{-2}$, and sustains over 210 h even at a stringent depth of discharge of 68.5%. Paired with a $\text{Zn}_2\text{V}_2\text{O}_7$ cathode, the full battery retains 93.7% of its initial capacity after 1100 cycles at 10 A g^{-1} . Moreover, the practical viability is substantiated under low N/P ratios and in pouch cell configurations. This work opens a new avenue for durable ZIBs via Coanda-effect-inspired interface design.

1. Introduction

The extensive combustion of fossil fuels has dramatically accelerated global industrial productivity while concurrently exacerbating environmental pollution and global warming. In the pursuit of sustainable energy solutions, Li-ion batteries (LIBs) have emerged as promising alternatives to fossil fuels [1–3]. Despite substantial progress in enhancing the energy density and cycle life of LIBs, their widespread adoption remains constrained by inherent safety concerns and high manufacturing costs. In this context, aqueous Zn-ion batteries (ZIBs) have attracted significant interest as a next-generation platform for scalable energy storage, offering a unique combination of exceptional cost-effectiveness

and intrinsic safety [4,5]. Furthermore, the Zn metal anode features a mild redox potential (-0.76 V vs. SHE) along with high theoretical gravimetric and volumetric capacities (820 mAh g^{-1} and 5855 mAh cm^{-3} , respectively) [6–8]. Despite these advantages, the practical implementation of ZIBs is severely hindered by interfacial instability between the Zn metal anode and aqueous electrolytes. This incompatibility triggers a cascade of parasitic reactions, including the hydrogen evolution reaction (HER), electrochemical corrosion, and irregular dendrite formation, which collectively compromise the Coulombic efficiency (CE) and long-term cycling stability of ZIBs [9–14].

To date, various strategies for stabilizing the Zn anode interface have

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been developed, primarily encompassing interfacial engineering, electrolyte additive design, and hydrogel electrolyte application [4,15–20]. Among these, the construction of an artificial solid electrolyte interphase (SEI) has gained widespread recognition as an effective approach for achieving uniform Zn deposition, suppressing dendrite formation, and mitigating deleterious interfacial side reactions [21,22]. However, the majority of existing SEI studies are based on static electrode/electrolyte models, with a primary focus on electric field distribution and the design of zincophilic nucleation sites [23–25]. Critically, the effects of dynamic mass transfer and ion concentration evolution during electrolyte transport from the bulk to the electrode interface have rarely been considered, yet these factors critically govern Zn anode durability. In practical scenarios, Zn electrodeposition is strongly governed by interfacial ion transport dynamics. Zn dendrite formation is essentially a secondary nucleation and growth process of Zn metal crystals during electrodeposition, which follows classical crystal growth behavior and is inevitably influenced by ion fluid dynamics [26–28]. Uncontrolled interfacial ion flux induces rapid ion depletion at the interface, resulting in concentration polarization and destabilizing the interfacial electrochemical reaction. According to Sand's model, localized depletion of Zn^{2+} ions triggers stochastic nucleation and perturbs the local electric field [29,30]. Therefore, it is particularly critical to develop interfacial engineering approaches grounded in ion fluid regulation, with an emphasis on stabilizing electrolyte flow, maintaining a high interfacial Zn^{2+} concentration, and extending the Sand's time.

The three-dimensional (3D) nanofiber network constructed from polyacrylonitrile (PAN) not only possesses a unique curved morphology but also features abundant polar $-\text{CN}$ functional groups, which have been reported to effectively regulate ion transport [31,32]. The interconnected porous structure ensures uniform electrolyte infiltration and promotes homogeneous ion flux distribution [33]. Beyond these inherent advantages, the external curved structure of the cylindrical nanofibers is expected to influence electrolyte transport pathways based on fluid dynamic principles. Specifically, the curved fiber surfaces can induce a Coanda-effect-like behavior, wherein a fluid jet adheres to a convex curved surface and follows its curvature rather than its original trajectory [34–36]. This effect arises from the pressure gradient generated by the entrainment of ambient fluid, which forces the flow to remain adherent to the curved surface. By analogy, Zn^{2+} ions migrating through the electrolyte are anticipated to undergo a similar curvature-guided redirection along the contours of PAN fibers, thereby converting the rapid depletion characteristic of vertical ion flux into a sustained and replenished ionic supply within the curved interfacial layer. Such Coanda-effect-like regulation of ion flux offers considerable potential for homogenizing the interfacial distribution of Zn^{2+} ions, delaying the onset of Zn^{2+} depletion, and ultimately suppressing dendrite formation.

In this study, inspired by the potential of curved PAN nanofibers to induce Coanda-effect-like electrolyte flux regulation, we have successfully constructed a composite interfacial layer comprising PAN and sodium lignosulfonate (LS) on a Zn metal anode (denoted as PAN-LS@Zn) using an in-situ electrospinning technique. In this design, the cylindrical fiber topologies leverage their curved external surfaces along with electrostatic interactions to establish an interfacial Zn^{2+} buffering reservoir that mitigates concentration polarization caused by rapid ion depletion, thereby extending the Sand's time. Concurrently, the preferential interaction of PAN and LS with the (101) and (100) crystal planes steers Zn nucleation and subsequent growth toward the (002) plane. As a result, the PAN-LS@Zn anode enables a Zn//Zn symmetric cell to achieve a cycling life of 4345 h at 1 mA cm^{-2} and 1 mAh cm^{-2} , and sustain over 210 h at a high depth of discharge (DOD) of 68.5%. When integrated with a $\text{Zn}_2\text{V}_2\text{O}_7$ (ZVO) cathode, the full battery retains 93.7% of its initial capacity after 1100 cycles at 10 A g^{-1} . The practical relevance of this approach is further underscored by its successful operation under low negative/positive (N/P) ratios and in pouch cell configurations.

2. Results and discussion

The fabrication of an integrated interfacial layer for electrode stabilization was achieved through in-situ electrospinning onto a Zn substrate placed directly on the spinning collector, followed by solvent evaporation at 80°C (Fig. 1a). PAN is a well-established electrospinning precursor capable of forming continuous nanofibers with a well-defined curved, cylindrical topology essential for curvature-guided ion transport. It also exhibits excellent electrochemical and structural stability in aqueous electrolytes and is cost-effective and compatible with large-area electrospinning for practical scale-up [37]. These attributes render PAN superior to other polymer matrices lacking the requisite fiber-forming capability, polar functional groups, or electrochemical robustness. LS was introduced to complement the PAN framework through its unique structural and electronic properties. Its abundant negatively charged sulfonate groups ($-\text{SO}_3^-$) impart a strongly negative surface charge, enhancing electrostatic attraction toward Zn^{2+} . LS also exhibits a high electron-donating propensity that reinforces Zn^{2+} migration, along with a strong anchoring affinity that modulates desolvation by displacing coordinated water molecules, thereby showing promise in suppressing dendrite formation and promoting a flat, uniform morphology [38]. A control electrode, denoted as PAN@Zn, was fabricated by electrospinning from a precursor solution without the addition of LS. As shown in Fig. S1, electrospinning can produce a uniform interfacial layer on a large-area Zn metal substrate of $30 \text{ cm} \times 10 \text{ cm}$. Importantly, this method offers better coverage uniformity and higher production efficiency, which is particularly beneficial for the scale-up and practical adoption of this technology. A comprehensive characterization of the morphology and phase structure was performed on the prepared interfacial layers. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) observations reveal a well-defined PAN-LS architecture composed of straight, smooth nanofibers with an average diameter of approximately 100 nm, which together form a continuous and interconnected scaffold (Fig. 1b and c). The successful integration of LS into the PAN matrix was further corroborated by energy-dispersive X-ray spectroscopy (EDS) mapping, which demonstrated a uniform distribution of C, N, S, and O throughout the fiber network (Fig. 1d).

The electrostatic interaction between the methylene groups ($-\text{CH}_2$) in PAN and the sulfonate sites in LS, corroborated by density functional theory (DFT) calculations in Fig. S2, is expected to improve the electric field distribution on the curved PAN matrix surface. Fourier-transform infrared (FTIR) spectra reveal the characteristic band of $-\text{SO}_3^-$ in LS at 1033.7 cm^{-1} and that of $-\text{CH}_2$ in PAN at 1454.1 cm^{-1} (Fig. S3). Compared with the spectra of pristine PAN and LS, the $-\text{SO}_3^-$ band in the PAN-LS composite shifts to 1039.5 cm^{-1} , while the $-\text{CH}_2$ band shifts to 1452.1 cm^{-1} . The opposite shifts in these two groups provide direct spectroscopic evidence, further confirming the strong electrostatic interaction between PAN and LS molecules [39]. Zeta potential measurements in Fig. S4 show that the value shifts to a more negative potential, from -16.8 mV (PAN) to -27.7 mV (PAN-LS), demonstrating that the incorporation of LS with the $-\text{SO}_3^-$ group increases the negative surface charge of the interfacial layer. This enhancement is expected to boost Zn^{2+} interfacial transport by the curved PAN-based fiber network. To clarify the spatial distribution of LS within the PAN-LS interphase, SEM coupled with EDS elemental mapping on both the surface and the cross-section of the PAN-LS@Zn electrode was performed (Figs. S5 and S6). The mapping results reveal a uniform distribution of C, N, O, and S elements throughout the entire PAN-LS fiber network with no detectable localized aggregation. Particularly, the homogeneous S signal across the mapped regions therefore provides direct evidence that LS is evenly dispersed within the PAN matrix. The wettability of different Zn-based anodes toward the aqueous electrolyte was evaluated by contact angle tests, as depicted in Fig. S7. The PAN-LS@Zn electrode exhibits a contact angle of 26.5° , in contrast to 50.2° for PAN@Zn and 77.2° for bare Zn, demonstrating that the synergistic effect of PAN and LS enhances the hydrophilicity of the Zn electrode. Furthermore, the wetting surface free

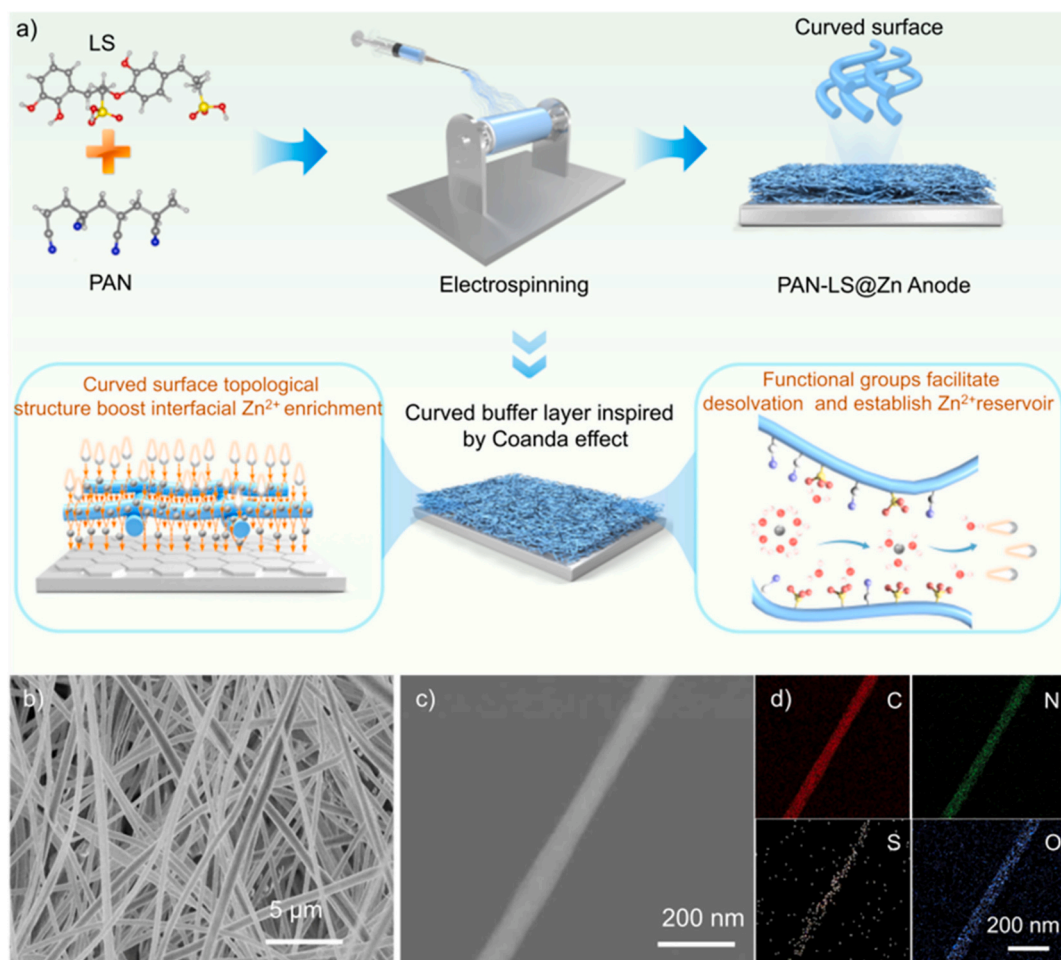


Fig. 1. a) Schematic illustration of the in-situ electrospinning preparation of the PAN-LS@Zn electrode and the Coanda-effect-inspired interfacial layer for stable mass transfer and deposition. b) SEM and c) TEM images of the PAN-LS composite nanofibers. d) Corresponding EDS elemental mapping of C, N, S, and O within a single fiber.

energy, calculated using Eq. S1, decreases from -899.3 to -3988.6 J mol $^{-1}$ upon incorporation of the PAN-LS layer (Fig. S8), indicating a stronger affinity between the modified electrode and the electrolyte [5]. The improved wettability ensures intimate contact between the aqueous electrolyte and the zincophilic functional groups within the interfacial layer, thereby maximizing the availability of these active sites for Zn $^{2+}$ coordination. This high hydrophilicity is expected to facilitate desolvation by promoting the interaction between Zn $^{2+}$ ions and the zincophilic groups. To evaluate the corrosion resistance of the PAN-LS coating, both bare Zn and PAN-LS@Zn electrodes were immersed in 2 M ZnSO $_4$ electrolyte for 3 days, after which SEM characterization was performed on the electrode surfaces (Fig. S9). For the bare Zn electrode, the post-corrosion SEM image reveals a rough and heterogeneous surface covered with unevenly distributed by-products, indicating that the bare Zn surface undergoes severe parasitic reactions and corrosion in the aqueous electrolyte. In contrast, after removing the PAN-LS coating from the PAN-LS@Zn electrode, the underlying Zn surface remains flat and uniform, with no observable by-product accumulation or corrosion pits. The preserved surface flatness of the protected Zn electrode further confirms the corrosion resistance of the PAN-LS interfacial layer and its ability to maintain the integrity of the underlying Zn substrate during electrolyte exposure.

The thickness of the PAN-LS layer was tailored by adjusting the electrospinning time (0.5, 1, and 1.5 h), yielding a series of PAN-LS@Zn electrodes for comparative study. Micrometer measurements show that the thicknesses of the three samples are 10, 15, and 20 μ m, respectively

(Fig. S10). As shown in Fig. S11, the cross-sectional SEM image of the as-prepared PAN-LS@Zn electrode reveals that the PAN-LS coating maintains intimate contact with the underlying Zn substrate, and the interface is free from any visible gaps, delamination, or cracks. This flawless interfacial adhesion confirms that the in-situ electrospinning method yields a highly reproducible and homogeneous protective layer devoid of macroscopic defects, providing a robust structural foundation for subsequent electrochemical cycling. The measured coating thickness is consistent with the micrometer measurements, further confirming that the in-situ electrospinning method produces a homogeneous interfacial layer whose thickness can be effectively tuned by varying the electrospinning time. Electrochemical impedance spectroscopy (EIS) measurements at different temperatures were performed on symmetric cells assembled with electrodes coated at different thicknesses to calculate the desolvation energy (E_a). The results corroborate that a 15 μ m PAN layer is optimal (15.6 kJ mol $^{-1}$) (Fig. S12). A thinner coating (10 μ m) provides insufficient regulation for the desolvation of hydrated Zn $^{2+}$ ions, resulting in a higher E_a value of 29.2 kJ mol $^{-1}$. Conversely, a thicker coating (20 μ m) increases the Zn $^{2+}$ transport resistance and concentration polarization, which elevates the E_a value to 27.4 kJ mol $^{-1}$. The diameter of the electrospun PAN-LS fibers was tailored by adjusting the diameter of the electrospinning needle. Using this approach, PAN-LS fibers with average diameters of approximately 60 nm, 100 nm, and 130 nm were successfully synthesized, as confirmed by SEM characterization (Fig. S13), which clearly resolves the distinct fiber diameters obtained under the different spinning conditions. This

confirms that the fiber diameter can be effectively and reproducibly tuned by controlling the needle diameter. The PAN-LS interphases with different fiber diameters were then applied to stabilize Zn anodes, and symmetric cells were assembled and evaluated at a high current density and areal capacity of $10 \text{ mA cm}^{-2}/10 \text{ mAh cm}^{-2}$. As shown in Fig. S14, the 100 nm fiber diameter delivers the longest cycling stability among the three samples, outperforming both the 60 nm and 130 nm counterparts. The observation that an intermediate diameter (100 nm) yields the best performance indicates that an optimal fiber diameter exists that maximizes the beneficial regulation of ion transport without introducing

excessive transport resistance. This structure-property relationship further underscores the importance of rational morphological design in engineering effective interfacial layers for stable Zn anodes.

Efficient and stable interfacial mass transfer dynamics are essential for optimizing Zn nucleation and growth behaviors. Galvanostatic intermittent titration technique (GITT) was performed on Zn//Zn symmetric cells to investigate interfacial polarization effects [40]. The cells were operated in an intermittent mode at a current density of 1 mA cm^{-2} , consisting of a 1-min deposition step followed by a 1-min rest. After a cumulative deposition time of 60 min, this cycle was followed by

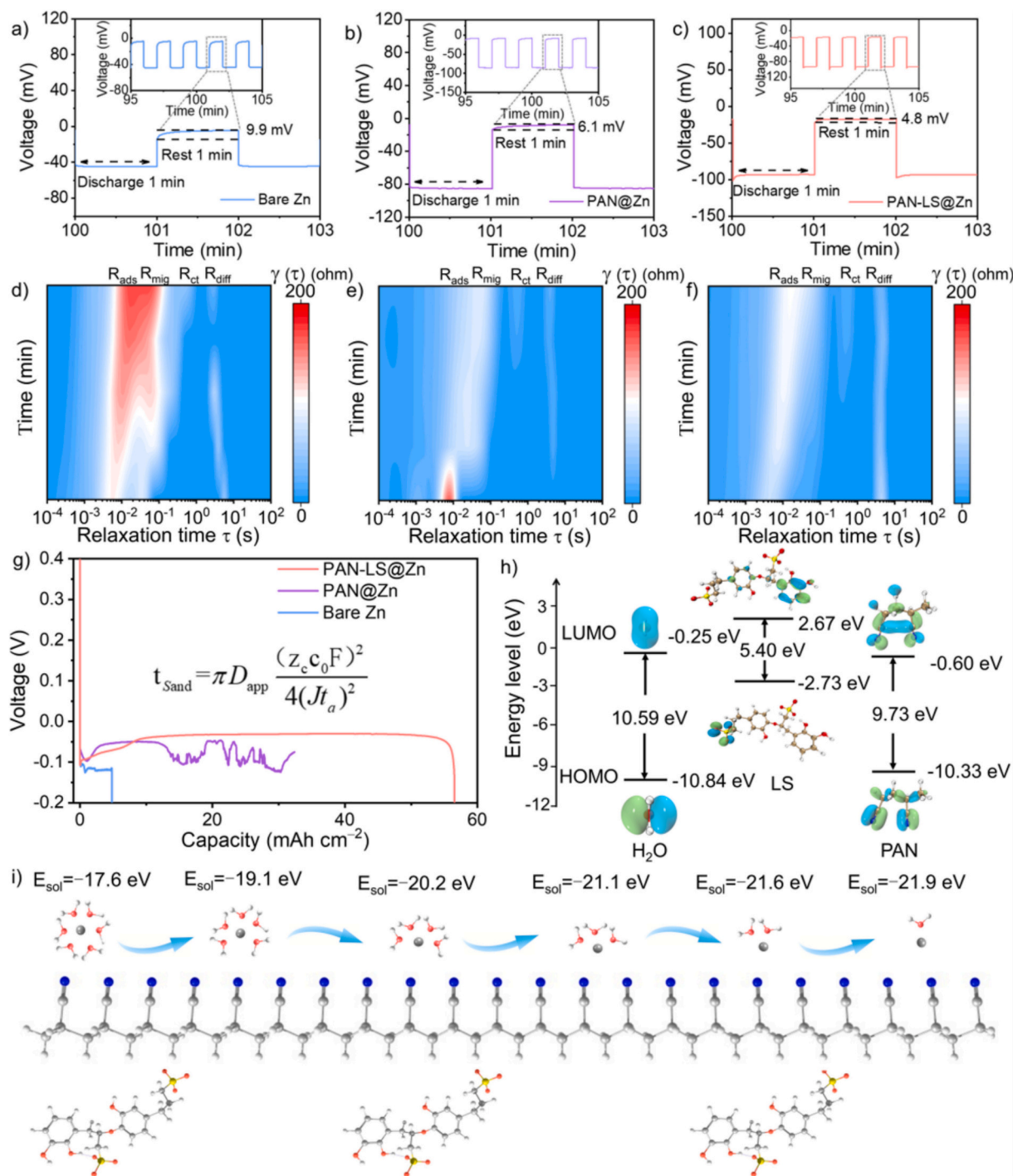


Fig. 2. GITT profiles of a) bare Zn, b) PAN@Zn, and c) PAN-LS@Zn at 1 mA cm^{-2} . DRT contour maps of d) bare Zn, e) PAN@Zn, and f) PAN-LS@Zn symmetric cells. g) Sand's capacity for Zn dendrite growth at 1 mA cm^{-2} . h) LUMO and HOMO energy levels of H_2O , LS, and PAN. i) Relative solvation energy of solvated Zn^{2+} migrating along the PAN-LS chain.

a 1-min dissolution phase and another 1-min rest. The resulting voltage relaxation profiles manifest stark differences among the three electrodes (Figs. 2a-c). PAN-LS@Zn displays a significantly reduced concentration polarization, with a relaxation voltage of only 4.8 mV, substantially lower than those of PAN@Zn (6.1 mV) and bare Zn (9.9 mV). This pronounced enhancement originates from the synergistic regulation of interfacial Zn^{2+} flux by the PAN and LS components. Their synergy effectively aligns ionic transport with deposition kinetics, thereby buffering the detrimental interfacial Zn^{2+} depletion that typically follows rapid Zn deposition. During the charging process, the external electric field drives Zn^{2+} ions toward the anode while directing SO_4^{2-} anions in the opposite direction. To quantitatively assess the Zn^{2+} concentration at the electrode/electrolyte interface, in situ Raman spectroscopy was performed by monitoring the SO_4^{2-} concentration at the counter electrode, specifically targeting the SO_4^{2-} vibrational band at 984 cm^{-1} (Fig. S15). In the case of the bare Zn electrode, a significant decrease in peak intensity is observed after approximately 10 min of electroplating, indicative of an unstable interfacial mass transport process and exacerbated concentration polarization [41]. Encouragingly, the Raman signal for the PAN-LS@Zn electrode remains consistently stable throughout the entire electroplating duration, once again attesting to the superior ion-regulating capability of the PAN-LS interfacial layer. Operando distribution of relaxation times (DRT) analysis was employed to comprehensively deconvolute the interfacial kinetic evolution during Zn plating. As plating proceeds, the impedance distributions of the bare Zn and PAN@Zn surfaces exhibit varying oscillations, reflecting inadequate interfacial mass transfer regulation (Fig. 2d and e). The PAN-LS@Zn electrode consistently displays more stable impedance values (Fig. 2f). More importantly, its adsorption resistance (R_{ads} , $\tau \approx 10^{-2}\text{ s}$), which corresponds to the adsorption process of $[\text{Zn}(\text{H}_2\text{O})_6]^{2+}$ ions at the electrode/electrolyte interface, is substantially smaller than those of the other two anodes [21]. This finding demonstrates that the PAN-LS interfacial layer can promote interfacial Zn^{2+} enrichment during deposition. Such superior dynamic stability establishes a physico-chemical foundation for regulating interfacial Zn^{2+} transport, which is a prerequisite for enhancing cycling durability.

Classical predictions from Sand's model were further employed to elucidate the regulatory effect of the interfacial layer on ionic transport (Fig. 2g). According to this framework, Sand's time (t_{Sand}) defines the critical threshold where the Zn^{2+} concentration at the electrode surface drops to zero under an applied current density. Operation beyond Sand's time inevitably generates an ion-depleted zone at the interface, compelling further deposition to occur within the bulk electrolyte, a process that fundamentally marks the onset of dendritic growth [26,29]. At a current density of 1 mA cm^{-2} , symmetric cells using PAN@Zn and bare Zn anodes exhibit dendritic failure after merely 12.8 h and 4.8 h of deposition, respectively. Remarkably, the PAN-LS@Zn anode supports a continuously stable deposition process (53.1 h), lasting until the Zn source in the counter electrode is nearly exhausted (up to 53.1 mAh cm^{-2}). The 53.1 h of stable Zn deposition observed for the PAN-LS@Zn symmetric cell at 1 mA cm^{-2} should be interpreted as the maximum experimentally achievable deposition duration under the current testing conditions. The theoretical areal capacity of the $100\text{ }\mu\text{m}$ Zn foil is 58.5 mAh cm^{-2} , which corresponds to a maximum deposition duration of approximately 58.5 h at a current density of 1 mA cm^{-2} before the Zn source in the counter electrode is fully consumed. The value of 53.1 h approaches this theoretical limit imposed by the Zn foil thickness, indicating that the deposition process remains stable throughout the entire consumption of the available Zn reservoir. This significantly prolonged Sand's time confirms that the guided enhancement of Zn^{2+} migration, conferred by the curved PAN architecture and the electrostatic interaction of LS, effectively delays Sand's time and establishes a more robust interfacial environment for Zn^{2+} replenishment. The electrochemical behavior of the Zn surface can be captured by the current evolution during chronoamperometry measurements under an applied overpotential of 200 mV over 200 s (Fig. S16). Following the brief initial

nucleation and two-dimensional (2D) diffusion stages, PAN-LS@Zn exhibits a markedly suppressed current relative to bare Zn and PAN@Zn, indicating a transition to stable 3D diffusion of Zn^{2+} ions. This observation suggests that PAN-LS can homogenize the interfacial electric field distribution, suppress dendrite growth, and enhance the cycling durability of the Zn anode [42].

Moreover, the curved geometry and electrostatic synergy of the interfacial layer also increase the Zn^{2+} transference number and facilitate the desolvation process. Chronoamperometry and EIS were combined to measure the Zn^{2+} transference number (Fig. S17). The Zn^{2+} transference number for PAN-LS@Zn (0.69) is higher than that for PAN@Zn (0.51) and bare Zn (0.35), indicating that the interfacial layer promotes Zn^{2+} migration and alleviates the hindrance caused by solvated water molecules and anions [43]. The temperature-dependent charge transfer resistance (R_{ct}) of the symmetric cell was further determined by EIS, with comparative analyses performed on different electrode configurations (Fig. S18). According to the Arrhenius equation, the E_a value of PAN-LS@Zn was calculated to be 15.6 kJ mol^{-1} , which was significantly lower than that of PAN@Zn (21.4 kJ mol^{-1}) and bare Zn (22.3 kJ mol^{-1}). This reduced energy barrier indicates that the PAN-LS interfacial layer facilitates the desolvation of hydrated Zn^{2+} ions [44]. The PAN-LS interphase facilitates efficient desolvation and establishes a favorable interfacial environment that promotes Zn^{2+} transport while partially restricting anion mobility. This is consistent with the observation that the PAN-LS@Zn system achieves a higher Zn^{2+} transference number compared with PAN@Zn and bare Zn.

To gain insight into the Zn^{2+} transport kinetics through the PAN-LS interfacial layer, DFT calculations were carried out to uncover the underlying mechanism by which this interfacial layer modulates the coordination and transport of solvated Zn^{2+} ions. As presented in Fig. S19, the results demonstrate that the binding energies between Zn^{2+} ions and the polar functional groups ($-\text{SO}_3^-$ and $-\text{CN}$) in PAN and LS are considerably higher than those between Zn^{2+} ions and H_2O . This stronger affinity suggests that both PAN and LS can reconstruct the primary solvation shell of Zn^{2+} ions by displacing coordinated H_2O molecules. Further analysis from molecular orbital energy level analysis discloses that the highest occupied molecular orbital (HOMO) energy level of LS (-2.73 eV) is considerably higher than that of H_2O (-10.84 eV), implying a greater electron-donating propensity of LS. This property, in turn, reinforces the PAN network's ability to guide Zn^{2+} migration (Fig. 2h). Moreover, the lowest unoccupied molecular orbital (LUMO) energy level of PAN (-0.60 eV) is lower than that of H_2O (-0.25 eV), which is beneficial for suppressing $\text{H}-\text{O}$ bond dissociation in solvated water molecules during the desolvation process and mitigating the HER [45]. These findings are also supported by linear sweep voltammetry (LSV) measurements (Fig. S20), in which the PAN-LS@Zn electrode demonstrates the most negative HER overpotential. The migration energy barrier of hydrated Zn^{2+} ions along the PAN-LS chain was determined by calculating the variation in solvation energy (Fig. 2i). The solvation energy of Zn^{2+} coordinated with six water molecules is -17.6 eV . At the early stage of interaction with the interfacial layer, adjacent $-\text{CN}$ groups engage in electrostatic attraction with Zn^{2+} , forming $[\text{Zn}(\text{H}_2\text{O})_5(\text{C}_3\text{H}_3\text{N})]^{2+}$, which elevates the solvation energy to -19.1 eV . Under the synergistic action of PAN and LS, following progressive desolvation to the point where only a single water molecule remains in the hydrated Zn^{2+} solvation structure, the solvation energy ultimately reaches -21.9 eV . This finding reveals that PAN-LS exhibits a strong anchoring affinity for Zn^{2+} ions, thereby inducing substantial interfacial ion enrichment and effectively modulating both desolvation and the ensuing deposition kinetics [46]. The PAN-LS layer contains abundant polar functional groups ($-\text{CN}$ from PAN and $-\text{SO}_3^-$ from LS) that exhibit strong binding affinity toward Zn^{2+} ions. This zincophilic character enables the layer to reconstruct the Zn^{2+} solvation shell, facilitate desolvation, and promote interfacial Zn^{2+} enrichment, thereby creating a local ion reservoir that buffers concentration polarization and homogenizes the interfacial ion flux.

To verify the protective role of the PAN-LS interfacial layer for Zn anodes during stripping/plating, the cycling performance of Zn//Zn symmetric cells with different anodes was compared. As illustrated in Fig. 3a, at 1 mA cm^{-2} and 1 mAh cm^{-2} , the PAN-LS@Zn symmetric cell delivers stable voltage hysteresis and exceptional cycle stability over 4345 h, dramatically outperforming PAN@Zn (667 h) and bare Zn (93 h). To isolate the contributions of the curved fiber topology and the negatively charged LS component, a control electrode was prepared by direct spin-coating a PAN/LS mixture onto Zn foil without electrospinning (denoted as PS@Zn). SEM characterization (Fig. S21) reveals that PS@Zn exhibits a dense, featureless film morphology, entirely

lacking the curved, cylindrical nanofiber architecture of PAN-LS@Zn. This confirms that electrospinning is essential for constructing the curved PAN topology, whereas PS@Zn retains the chemical composition but lacks the structural motif. At 1 mA cm^{-2} with a capacity of 1 mAh cm^{-2} , the PS@Zn cell, which lacks the nanofiber structure, is stable for only 180 h. These results allow the individual contributions to be deconvoluted as follows: the curved PAN architecture primarily contributes to the structural regulation of ion flux and to electric field homogenization, as evidenced by the comparison between PAN@Zn (667 h) and bare Zn (93 h). The curved fiber network alone extends cycle life by homogenizing the interfacial electric field and providing

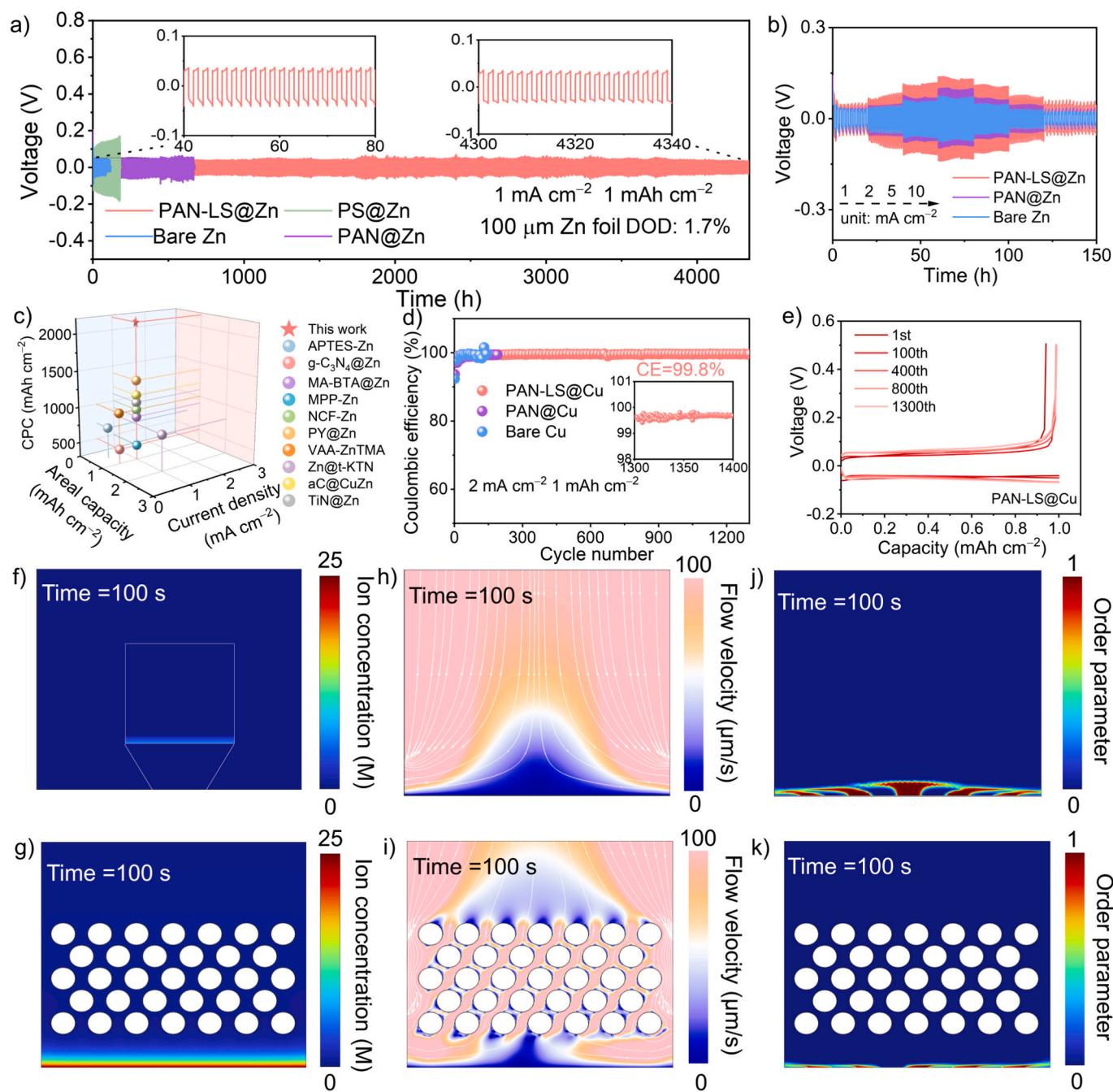


Fig. 3. a) Long-term cycling stability of symmetric cells at 1 mA cm^{-2} and 1 mAh cm^{-2} . b) Rate performance of symmetric cells with different anodes. c) Key parameter comparison with reported literature. d) CE measurements of asymmetric cells. e) Voltage profiles of the PAN-LS@Cu-based asymmetric cell. Simulated Zn^{2+} concentration distribution at the f) bare Zn and g) PAN-LS@Zn electrode interfaces. Simulated electrolyte flow velocity at the h) bare Zn and i) PAN-LS@Zn electrode interfaces. Simulated phase-field morphology at the j) bare Zn and k) PAN-LS@Zn electrode interfaces.

interconnected pathways for uniform ion-flux distribution, but it remains insufficient to regulate rapid ion transport without the LS component. The negatively charged LS component primarily enhances interfacial Zn^{2+} enrichment and desolvation, as demonstrated by the comparison between PS@Zn (180 h) and bare Zn (93 h), in which the electrostatic interaction with Zn^{2+} ions provides only moderate improvement. In addition, the dense coating structure of PS@Zn leads to a significantly increased overpotential and elevated interfacial ion transport resistance. To evaluate the mechanical stability of the interphase under prolonged electrochemical cycling, cross-sectional SEM characterization was further performed on the PAN-LS@Zn electrode after 50 cycles in a symmetric cell (Fig. S22). The interface between the PAN-LS coating and the Zn foil remains clean and intimately bonded, with no observable signs of cracking, delamination, compaction, or structural collapse. This observation confirms that the PAN-LS interphase maintains its mechanical integrity and protective function throughout extended cycling, accommodating the continuous morphological and stress variations associated with repeated Zn plating/stripping. To assess whether LS leaches from the PAN-LS layer during prolonged electrochemical cycling, the chemical state of the PAN-LS composite layer was characterized after the PAN-LS@Zn symmetric cell was cycled at $1 \text{ mA cm}^{-2}/1 \text{ mAh cm}^{-2}$ for 50 cycles. As shown in Fig. S23, the FTIR spectrum of the PAN-LS layer after 50 cycles retains the characteristic $-\text{SO}_3^-$ band of LS, confirming that the sulfonate groups remain within the composite layer after extended cycling and demonstrating that LS is stably retained during repeated Zn plating/stripping. Upon increasing the current density from 1 to 10 mA cm^{-2} , the PAN-LS@Zn symmetric cell maintains stable operation across different current densities and exhibits superior rate performance (Fig. 3b). The exchange current density of the PAN-LS@Zn cell, derived from the voltage hysteresis, is determined to be 10.8 mA cm^{-2} , a lower value compared to those of PAN@Zn (13.4 mA cm^{-2}) and bare Zn (19.2 mA cm^{-2}) cells (Fig. S24). This observation indicates that the PAN-LS layer favorably modulates the kinetics of initial Zn nucleation, thereby suppressing the rapid dendrite proliferation typically induced by locally elevated current densities [41]. To further assess the practical applicability of the PAN-LS interfacial layer, shelving-recovery tests were conducted on Zn//Zn symmetric cells at a current density of 1 mA cm^{-2} and a fixed capacity of 1 mAh cm^{-2} , incorporating a 20 h rest period after every 10 cycles (Fig. S25). The symmetric cell employing the PAN-LS@Zn anode delivers stable operation exceeding 820 h, with a cumulative resting time of 400 h, surpassing the performance of the PAN@Zn and bare Zn cells. More importantly, to test the deep cycling capability, we conducted measurements at a high DOD of 68.5% (Fig. S26), a stringent condition that demands efficient utilization of the Zn anode. Remarkably, PAN-LS@Zn delivers superior stability exceeding 210 h under this high DOD, underscoring its robustness for practical applications requiring deep charge/discharge cycles. Benchmarking against some state-of-the-art artificial interfacial layers reported in recent literature shows that PAN-LS@Zn ranks at the top in critical aspects such as resistance to elevated current densities, extraordinarily high cumulative Zn plating capacity, and extended cycle life (Fig. 3c and Table S1). Collectively, these performance gains highlight PAN-LS as a paradigm-shifting interfacial design for enabling practical rechargeable Zn metal batteries.

CE measurements were conducted to assess the plating/stripping reversibility. In these tests, an interfacial-layer-modified Cu electrode serves as the working electrode, while a bare Zn foil functions as the counter electrode [47]. This configuration eliminates interference from the underlying Zn source, enabling a more accurate assessment of the CE throughout the cycling process. As illustrated in Fig. 3d, the PAN-LS@Cu electrode sustains stable operation while delivering a high average CE of 99.8% over 1428 cycles at $2 \text{ mA cm}^{-2}/1 \text{ mAh cm}^{-2}$. In contrast, both the bare Cu and PAN@Cu electrodes undergo rapid failure, primarily due to severe side reactions and non-uniform Zn deposition. Moreover, the corresponding voltage profiles demonstrate that the PAN-LS@Cu

electrode also exhibits remarkably stable voltage hysteresis with highly reproducible plateaus over extended cycling (Figs. 3e and S27), suggesting well-regulated ionic flux and a highly reversible electrochemical interface. To further elucidate the regulatory role of the PAN-LS interfacial layer in Zn deposition, finite element simulations were performed to analyze the interfacial concentration field, ion flux, and phase field during the deposition process. The simulation results reveal that the bare Zn electrode exhibits a dramatic decline in surface Zn^{2+} concentration approaching near-depletion (Fig. 3f), indicating that Sand's time has been reached and the system has entered a diffusion-limited regime [48]. Remarkably, after the same duration, the PAN-LS@Zn interface preserves a robust and homogeneous Zn^{2+} concentration (Fig. 3g). This persistent and sufficient Zn^{2+} supply effectively overcomes interfacial ion transport limitations, thereby mitigating concentration polarization. These advantageous features are ascribed to the combination of the exposed curved structure of PAN fibers and the LS-enhanced negative potential.

The outstanding performance of the PAN-LS interfacial layer was further corroborated by simulations of interfacial ion flux. Under practical operating conditions, the electrolyte is inevitably subject to fluid dynamic fluctuations. The uncontrolled ion flows severely disrupt local ion concentration in the vicinity of the electrodes and exacerbate dendritic growth [24]. Upon imposing a simulated flow velocity of $100 \mu\text{m s}^{-1}$ (Fig. 3h), the bare electrode surface becomes engulfed by chaotic turbulent vortices, accompanied by significant local attenuation within 100 s. In stark contrast, the introduction of the PAN-LS architecture fundamentally alters the flow dynamics. Within the fibrous network, the flow velocity is redistributed in a more uniform and controlled manner (Fig. 3i). This regulation is analogous to the Coanda effect in fluid mechanics, where the negatively charged convex curvature of the fibers induces the electrolyte stream to adhere to the fiber surfaces. This surface adherence effectively buffers local Zn^{2+} depletion immediately after rapid reduction, thereby functioning as an interfacial ion storage reservoir. Additionally, finite-element simulations were also conducted at electrolyte flow velocities of 50 and $200 \mu\text{m s}^{-1}$ to cover a broad range of flow conditions. The simulation results demonstrate that, within the PAN-LS interfacial layer system, stable regulation of interfacial ion transport can be achieved under all the examined flow conditions (Fig. S28). Specifically, the electrolyte flow velocities through the interfacial layer reach 14, 16, and $20 \mu\text{m s}^{-1}$ for applied flow velocities of 50, 100, and $200 \mu\text{m s}^{-1}$, respectively (Fig. S29). These results indicate that the PAN-LS architecture effectively redistributes and moderates electrolyte flow regardless of the applied flow velocity, thereby maintaining a uniform and controlled ion flux at the electrode/electrolyte interface. The altered ion transport kinetics and interfacial concentration distribution described above also exert a profound impact on the ultimate morphology of Zn deposition products. Phase-field simulations were carried out to gain deeper insight into these divergent deposition outcomes. As illustrated in Fig. 3j, within 100 s, the bare Zn electrode develops well-defined dendritic structures, a clear indication that uncontrolled ion migration triggers morphological instability. Benefiting from the interfacial layer composed of negatively charged convex curved fibers, the PAN-LS@Zn electrode maintains a relatively flat and uniform deposition pattern after ion flux stabilization (Fig. 3k), underscoring the critical role of the PAN-LS architecture in suppressing dendrite formation [49]. This morphology regulation is directly responsible for the substantially improved cycling stability and CE observed experimentally. Over extended cycling, this well-regulated deposition behavior translates into suppressed parasitic side reactions, minimized electrolyte consumption, and prolonged anode lifespan. Thus, the combination of favorable ion transport kinetics, homogeneous concentration field, and flat deposition morphology underpins the exceptional performance of the PAN-LS@Zn anode, highlighting the importance of Coanda-effect-inspired integrated interfacial design in advanced ZIBs.

The prolonged effectiveness of the PAN-LS interfacial layer in

modulating Zn deposition was further validated through deposition process monitoring and phase characterization of the resulting deposits. The cross-sectional mapping shows that the newly deposited Zn partially overlaps with the PAN-LS elemental signals (Fig. S30), indicating that the deposited Zn is localized at the interface between the PAN-LS layer and the underlying Zn substrate. The Zn deposition behavior of bare Zn and PAN-LS@Zn was investigated using in situ optical microscopy under 10 mA cm^{-2} . As shown in Fig. 4a, Zn protrusions become visible on the bare Zn electrode within 10 min of plating initiation. These protrusions continue to grow as current flows, eventually evolving into bulky protuberances after 40 min, which may induce short-circuiting and severely

compromise battery operation. Nonetheless, the PAN-LS@Zn electrode demonstrates substantially more uniform deposition morphology (Fig. 4b). This marked improvement arises from the interfacial layer's ability to generate a homogeneous Zn^{2+} flux at the electrode/electrolyte interface, thereby regularizing the electroplating pattern. Moreover, confocal laser scanning microscopy (CLSM) was further employed to characterize the 3D morphological evolution of different anodes. The bare Zn anode exhibits a rough deposition surface featuring distinct voids and protuberances (Fig. 4c). In contrast, the PAN-LS@Zn anode displays a uniform plating morphology with flat height profiles (Fig. 4d), which underscores the critical role of the interfacial layer in regulating

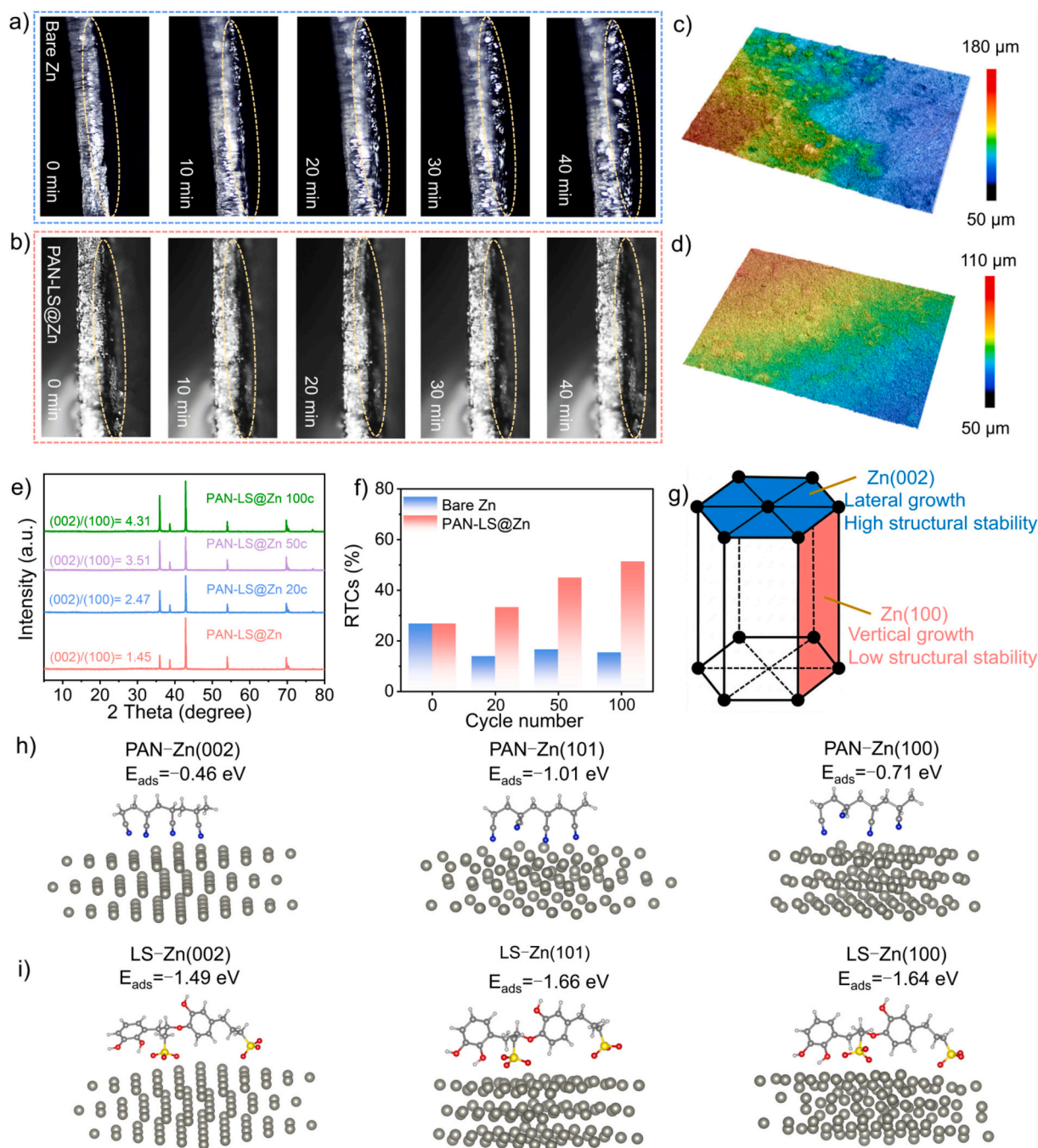


Fig. 4. In situ optical microscopy images of a) bare Zn and b) PAN-LS@Zn during the deposition processes at 10 mA cm^{-2} . 3D topography of CLSM for c) bare Zn and d) PAN-LS@Zn after cycling. e) XRD patterns of the PAN-LS@Zn electrode at different cycle numbers. f) RTCs of the deposited Zn on different electrodes at various cycle numbers. g) Schematic illustration of Zn deposition on different crystal planes. Adsorption binding energies of h) PAN and i) LS on different crystallographic planes of Zn.

Zn^{2+} flux to achieve a planar deposition morphology. SEM imaging further demonstrates the deposition morphologies of different anodes (Fig. S31). On the cycled bare Zn anode, porous dendritic deposits are observed, with glass fibers tightly “welded” into the structure. This signifies uncontrolled Zn growth, where metallic dendrites have even pierced the separator, raising safety concerns and leading to premature cell failure. The observation that the PAN-LS@Zn anode retains a uniform and compact electroplating morphology after 100 cycles is consistent with the underlying function of the PAN-LS interfacial layer in directing homogeneous Zn^{2+} migration at the Zn/electrolyte interface. To complement the morphological evidence described above, Raman mapping was further employed to characterize the spatial distribution of basic by-products on the electrode surface. As depicted in Fig. S32, intense and heterogeneous Raman signals attributed to basic Zn sulfate compounds are observed on the cycled bare Zn electrode, indicating severe accumulation of by-products that exacerbate surface inhomogeneity. The PAN-LS-protected electrode exhibits nearly undetectable Raman signals, demonstrating that the interfacial layer optimizes ion flux while mitigating parasitic side reactions, which collectively facilitate the formation of a flat and uniform electrode surface.

To elucidate the crystal structure evolution of the deposited Zn, X-ray diffraction (XRD) analysis was performed at different stages of cycling (Fig. 4e). For the PAN-LS-modified electrode, the diffraction peak intensity corresponding to the Zn (002) plane increases progressively with cycle number, and the (002)/(100) peak intensity ratio increases from 1.45 to 4.31 (Fig. S33), indicating a gradually enhanced preferential orientation of the (002) crystallographic plane. The bare Zn electrode shows non-oriented deposition (Fig. S34). The relative texture coefficients (RTCs) were also calculated to quantitatively evaluate the evolution of the (002) texture for both the PAN-LS@Zn and bare Zn electrodes before and after cycling (Fig. 4f). The RTC of the (002) plane for the PAN-LS@Zn electrode increases markedly from an initial value of 26.7% to 51.3% after 100 cycles, while the bare Zn electrode decreases to 15.3% over the same cycling period, demonstrating that the PAN-LS interfacial layer pronouncedly enhances the preferential (002) orientation during repeated Zn plating/stripping. This preferred (002)-oriented growth facilitates the formation of a flat and uniform deposition morphology, thereby effectively suppressing dendrite formation (Fig. 4g). DFT calculations were performed to understand the mechanism behind the preferential crystal plane exposure by evaluating the adsorption energies (E_{ads}) of PAN and LS on various Zn facets (Fig. 4h and i). The calculated binding energies reveal that, for both molecules, the E_{ads} values on (100) and (101) facets are considerably higher than those on the (002) facet. These results indicate that the constituent molecules of the interfacial layer possess a stronger thermodynamic affinity for the (100) and (101) planes, while exhibiting relatively weaker interaction with the (002) plane [50]. This selective adsorption behavior consequently directs Zn^{2+} ions to preferentially deposit along the (002) direction, which presents a lower energy barrier for growth, thereby effectively promoting the exposure and development of the (002) crystal plane.

To better assess the practical applicability of PAN-LS@Zn, full batteries were constructed using ZVO as the cathode material, and their electrochemical performance was evaluated. The characteristic diffraction peaks in the XRD pattern are in good agreement with the standard reference, confirming the successful synthesis of phase-pure ZVO (Fig. S35). The corresponding SEM image further reveals that the as-prepared ZVO exhibits a well-defined, uniform rod-like morphology, which is favorable for efficient ion transport (Fig. S36). Cyclic voltammetry (CV) measurements confirm that no extraneous redox peaks are detected at 1 mV s⁻¹ for both electrode configurations (Fig. S37), underscoring the negligible influence of the PAN-LS coating on the fundamental redox reactions. Notably, the PAN-LS@Zn//ZVO full battery delivers a higher current density than its bare Zn//ZVO counterpart, clearly demonstrating accelerated reaction kinetics and enhanced

charge storage capability [51]. Moreover, galvanostatic charge-discharge (GCD) curves were recorded across a range of current densities to further probe the energy storage behavior and rate capability. At 0.2 A g⁻¹, the PAN-LS@Zn//ZVO battery achieves a small polarization voltage of 467.5 mV, which is significantly superior to the 573.2 mV observed for the bare Zn//ZVO battery (Fig. 5a). As the current density increases from 0.2 to 10 A g⁻¹, the PAN-LS@Zn//ZVO battery delivers reversible capacities of approximately 327.8, 305.4, 274.1, 241.5, and 219.5 mAh g⁻¹, respectively (Figs. 5b and S38). When the current density is returned to 0.2 A g⁻¹, a reversible capacity of approximately 326.7 mAh g⁻¹ is still retained, confirming the remarkable reversibility and rate capability of the PAN-LS@Zn-based battery. These results confirm that the PAN-LS interfacial layer mitigates interfacial side reactions by regulating mass transport, promoting the desolvation of hydrated Zn^{2+} ions, and alleviating concentration polarization, thereby extending the cycling lifespan of the Zn anode.

The EIS results in Fig. 5c show that the R_{ct} value of the PAN-LS@Zn//ZVO full battery is 230 Ω , which is lower than that of the Zn//ZVO full battery (412 Ω). This notable reduction in R_{ct} can be attributed to the suppression of byproduct formation and the faster Zn^{2+} transport kinetics enabled by the PAN-LS@Zn anode [52]. The cycling stability of the PAN-LS@Zn//ZVO full battery was systematically benchmarked against that of the bare Zn//ZVO counterpart. As presented in Fig. 5d, the PAN-LS@Zn-based battery delivers exceptional long-term durability over 1100 cycles at 10 A g⁻¹, achieving a near-unity CE ($\approx 100\%$) and an outstanding capacity retention of 93.7%. By stark contrast, the bare Zn//ZVO full battery suffers from severe capacity degradation, only maintaining approximately 51.9% of its initial capacity over the same cycling period. The origin of the initial capacity difference can be attributed to the improved electrochemical reaction kinetics at the Zn anode interface, which enhances the accessible capacity of the full cell [53]. Specifically, at the bare Zn anode, uncontrolled interfacial ion flux leads to rapid local Zn^{2+} depletion, concentration polarization, and sluggish desolvation kinetics. These kinetic limitations impose an additional overpotential on the full cell, prematurely terminating the discharge process before the ZVO cathode reaches its intrinsic capacity limit. In contrast, the PAN-LS interfacial layer facilitates desolvation, homogenizes the interfacial Zn^{2+} flux, and lowers the charge-transfer barrier, thereby enabling the ZVO cathode to discharge more fully and achieve a higher capacity. Closer examination of the GCD profiles reveals a pronounced capacity decay within the initial 200 cycles for the bare Zn//ZVO cell, which can be reasonably ascribed to the progressively diminished reversibility of Zn plating and stripping processes at the bare Zn anode interface. The PAN-LS@Zn//ZVO cell exhibits minimal capacity loss while maintaining sustained electrochemical reversibility throughout cycling (Fig. 5e), unequivocally demonstrating its robust long-term cycling stability. Self-discharge behavior was further evaluated by monitoring the voltage decay over 48 h at full charge. Under this condition, the bare Zn anode configuration suffers from substantial capacity loss, retaining only 73.9% of its capacity when discharged to 0.2 V. The PAN-LS@Zn system preserves 81.9% of its capacity under identical conditions (Fig. S39).

To extend the above electrochemical evaluation, a PAN-LS@Zn//ZVO full battery with a deliberately low N/P ratio of 3.6 was fabricated, aiming to assess the influence of a high Zn utilization rate on overall cell performance. The long-term cycling stability of this configuration was subsequently examined. In coin-cell format, the device delivers an appreciable capacity of 186.5 mAh g⁻¹ at 2 A g⁻¹ after 200 cycles, retaining 73.5% of its initial capacity (Fig. 5f). This demonstration of reasonably stable cycling under a low N/P ratio, which imposes a more realistic and demanding condition where the Zn anode is subjected to deeper stripping/plating cycles, indicates that the PAN-LS interfacial layer effectively suppresses dendrite growth and parasitic side reactions even under high Zn utilization scenarios, thereby preserving anode integrity. As a more rigorous validation of the interfacial layer's practical utility, a pouch cell was assembled (Fig. 5g). The as-assembled PAN-

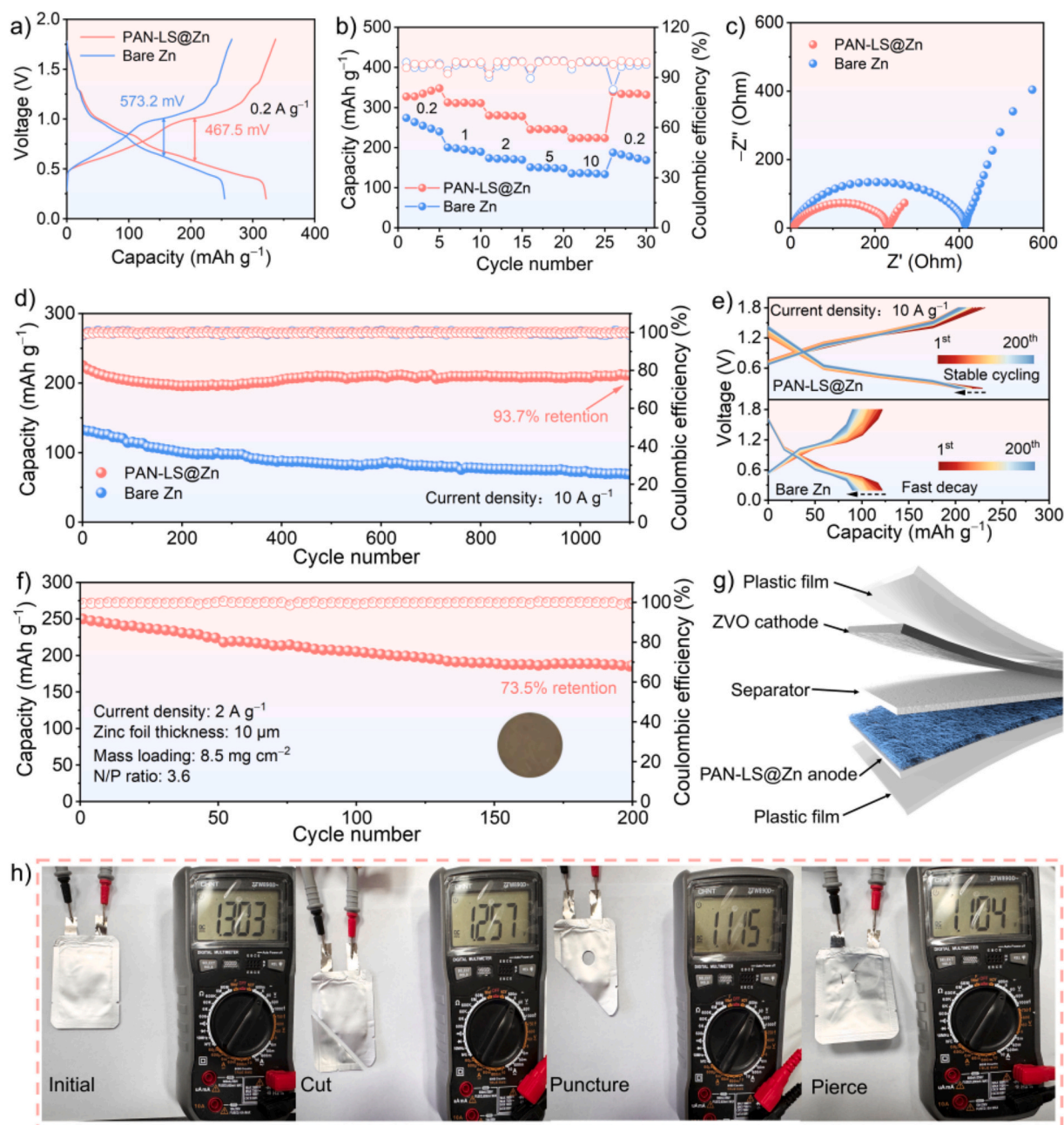


Fig. 5. Comparative electrochemical performance of PAN-LS@Zn//ZVO and Zn//ZVO full batteries. a) GCD curves recorded at a current density of 0.2 A g^{-1} . b) Rate capability at various current densities. c) Nyquist plots of the EIS data. d) Long-term cycling stability at 10 A g^{-1} . e) Selected GCD curves from first to 200th cycle. f) Cycling performance of the PAN-LS@Zn//ZVO coin cell under a low N/P ratio of 3.6. g) Schematic diagram illustrating the assembly process of the pouch cell. h) Open-circuit voltage measurements and safety evaluations of a pouch cell under destructive mechanical testing, including cutting, puncturing, and piercing.

LS@Zn//ZVO pouch cells readily power a light bulb, highlighting their considerable promise for practical energy-supplying devices (Fig. S40). This real-world demonstration confirms that the PAN-LS@Zn anode not only delivers sufficient output voltage and current to drive functional electronic loads but also maintains stable power output without abrupt failure. Furthermore, the battery exhibits exceptional safety and structural integrity even under mechanical abuse conditions, including cutting, puncturing, and piercing (Fig. 5h). These extreme tests simulate catastrophic damage scenarios such as sharp object penetration or impact during daily use. Collectively, these results demonstrate that the PAN-LS interfacial layer significantly enhances both the performance and safety of ZIBs, paving the way for their practical deployment in next-generation aqueous energy storage devices.

3. Conclusion

In summary, a PAN-LS interfacial layer was fabricated in situ on a Zn metal anode via a facile electrospinning method. Through the synergy of curved fiber topologies and electrostatic interactions, this rationally designed layer induces Coanda-effect-like ion flux regulation, creating an interfacial Zn^{2+} storage reservoir that buffers concentration polarization and extends the Sand's time toward the theoretical Zn capacity limit. In addition, the adsorption behavior of the PAN and LS components at the interface facilitates oriented Zn deposition. Electrochemical evaluations demonstrate that the Zn//Zn symmetric cell with the PAN-LS@Zn anode achieves a cycling life of 4345 h at 1 mA cm^{-2} and 1 mAh cm^{-2} , and remains stable for over 210 h even under a high DOD of 68.5%. Moreover, the asymmetric cell featuring the PAN-LS interfacial

layer delivers a high average CE of 99.8% over 1428 cycles. When paired with a ZVO cathode, the full battery retains 93.7% of its initial capacity after 1100 cycles at 10 A g⁻¹. The practical viability of this strategy is further confirmed under low N/P ratio and in pouch cell configurations. By offering precise control over interfacial ion mass transfer at the molecular scale, this interface engineering strategy paves the way for designing high-performance ZIBs.

CRediT authorship contribution statement

Tianyu Zhang: Writing – original draft, Formal analysis, Data curation, Conceptualization. **Haolan Tao:** Validation, Software, Methodology. **Feng Yang:** Validation, Data curation, Conceptualization. **Le Zhou:** Formal analysis, Data curation. **Wenzhuo Gao:** Formal analysis. **Bin-Bin Xie:** Visualization, Validation, Software, Methodology. **Hongfei Wang:** Writing – original draft, Validation, Funding acquisition, Conceptualization. **Yong Hu:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cej.2026.182687>.

Data availability

The authors do not have permission to share data.

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