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Flexible Zinc Battery Electrolytes Based on PVA/PAA/PAM: From Ion Transport Regulation to Zinc Dendrite Suppression

Ke Lei, Ruining Wang *, Jing Deng

School of Textile Science and Engineering, Xi'an Polytechnic University, Xi'an, Shaanxi 710048, China

ABSTRACT

The rapid development of flexible wearable electronics has imposed comprehensive requirements on energy storage devices, including safety, thinness, stretchability, and high reliability. Traditional lithium batteries suffer from issues such as flammability, high cost, and insufficient flexibility. In contrast, flexible aqueous zinc batteries are regarded as an important alternative due to the abundance of zinc resources, moderate operating voltage, environmental friendliness, and high safety. However, their performance is limited by bottlenecks such as slow ion transport in the electrolyte, poor interfacial stability, and the tendency for zinc dendrite growth. PVA/PAA/PAM-based hydrogel electrolytes, with advantages such as film-forming ability, adhesiveness, water retention, and strong designability, provide a new approach for regulating Zn^{2+} migration and suppressing dendrite growth, and thus have become a current research hotspot. Flexible aqueous zinc-ion batteries are considered promising energy storage devices for wearable and portable electronics because of their intrinsic safety, low cost, and environmental compatibility. However, practical deployment remains hindered by sluggish ion transport in hydrogel electrolytes, unstable electrode/electrolyte interfaces, limited mechanical robustness, and severe zinc dendrite growth during repeated cycling. This study focuses on polyvinyl alcohol/polyacrylic acid/polyacrylamide (PVA/PAA/PAM)-based hydrogel electrolytes and systematically analyzes the structural basis and synergistic regulation mechanisms that enable improved ionic conduction and dendrite suppression. By integrating complementary molecular interactions, the composite hydrogel can construct continuous ion-conduction pathways, reduce the activation energy for ion migration, and regulate the Zn^{2+} solvation structure through reversible coordination with polar groups. In addition, crosslinking engineering, double-network or interpenetrating-network design, and nanofiller incorporation are effective strategies for balancing ionic conductivity, tensile durability, and interfacial stability. The introduction of a homogeneous polymer matrix can further homogenize the local electric field at the zinc anode surface, thereby mitigating tip-induced deposition and inhibiting dendrite propagation. From a device-level perspective, PVA/PAA/PAM composite electrolytes also improve dimensional stability, flexibility under bending, and operational reliability under repeated deformation. The review concludes that the cooperative design of polymer chemistry, network architecture, and ion-solvation regulation is the key route for overcoming the conductivity-mechanics-dendrite suppression trade-off in flexible zinc batteries. Future research should emphasize wide-temperature operation, self-healing interfaces, and scalable manufacturing to bridge the gap between laboratory demonstrations and practical applications.

KEYWORDS

Flexible zinc batteries; PVA/PAA/PAM hydrogel electrolyte; Ion transport regulation; Zinc dendrite suppression; Solvation structure engineering; Interfacial stability

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1. INTRODUCTION

1.1. Research Background and Significance

As global energy transition advances and the "dual-carbon" targets gain traction, the large-scale integration of intermittent renewables—wind and solar in particular—into power grids has created an urgent demand for energy storage systems that are intrinsically safe, low-cost, and readily recyclable. Concurrently, the rapid proliferation of flexible wearable electronics, electronic skins, soft robotics, and portable medical devices is redefining the role of an energy-storage unit: it is no longer a mere "energy warehouse" but must also offer bendability, stretchability, adhesiveness, and long-term reliability under dynamic conditions. Conventional lithium-ion batteries, despite their impressive energy density, rely on flammable organic electrolytes that are costly, raise interfacial security concerns, and, critically, when deployed in flexible formats, are prone to mechanical failure and thermal runaway—limitations that severely hinder their ability to meet the demands of next-generation distributed and flexible energy systems. Motivated by these constraints, aqueous zinc-ion batteries have gained prominence as a promising candidate for both large-scale stationary storage and flexible power sources, leveraging abundant zinc reserves, a relatively high theoretical capacity, and straightforward fabrication processes. Compared with lithium-based counterparts, the zinc anode offers a lower redox potential and superior intrinsic safety in aqueous media, drastically reducing fire and explosion risks—a non-negotiable feature for wearable applications. In 2023, Zhenglin et al. demonstrated that a dual-network gel electrolyte combined with a water-confinement strategy could yield a solid-state zinc metal battery with high energy density and suppressed dendrite growth, underscoring that the structural engineering of gel electrolytes directly and profoundly impacts zinc-battery performance [9]. However, translating aqueous zinc-ion batteries from laboratory prototypes to practical deployment remains obstructed by four deeply intertwined bottlenecks. During repeated zinc plating/stripping, uneven local current densities drive continuous dendrite nucleation and proliferation, which not only invite short-circuit risks but also progressively erode Coulombic efficiency and cycle life. Side reactions in aqueous environments—hydrogen evolution, corrosion, passivation-product formation, and interfacial instability—are particularly pronounced, continuously consuming active zinc and electrolyte and thereby accelerating capacity decay. Conventional liquid electrolytes suffer from non-uniform ion distribution, high leakage susceptibility, and weak mechanical support, making it difficult to sustain stable ion transport and interfacial contact under bending or twisting. Moreover, flexible devices impose far more stringent demands on mechanical endurance: the electrolyte must simultaneously conduct ions and withstand stretching, compression, torsion, and repeated flexing without functional collapse, which brings the notorious trade-off between high ionic conductivity and high mechanical strength to the forefront. Collectively, these issues are neither independent nor sequential; they interact and amplify each other, forming a systemic barrier that explains why isolated improvements in a single performance metric rarely lead to an overall breakthrough.

Against this backdrop, hydrogel electrolytes constitute a critical bridge that reconciles intrinsically safe aqueous batteries with high-performance flexible electronics. Unlike conventional liquid electrolytes, hydrogels are capable of stably immobilizing substantial amounts of water and salt ions within their three-dimensional crosslinked networks, thereby preserving the inherently high ionic mobility of aqueous systems while simultaneously endowing the electrolyte with desirable deformability and structural coherence. More critically, via judicious design of polymer functional groups, crosslinking chemistries, and network topologies, hydrogels enable multilevel regulation of the Zn^{2+} solvation structure, ionic transport pathways, and electrode–electrolyte interfacial behaviour—directly mitigating dendritic nucleation, suppressing parasitic side reactions, and improving interfacial homogeneity at their very origin. The representative polymer systems—PVA, PAA, and PAM—each offer distinct attributes: PVA excels in film-forming ability and mechanical backbone integrity; PAA provides abundant carboxyl coordination sites; and PAM stands out for

superior water retention and chain flexibility. Upon compounding these three polymers, a synergistic equilibrium emerges among ionic conduction, mechanical support, and interfacial stability, offering a more practically viable electrolyte solution for flexible zinc batteries. In essence, the hydrogel electrolyte is far more than a "solidified liquid"; it is a functional platform that, through structural engineering, integrally couples electrochemical processes, ion transport, and mechanical responsiveness within a single cohesive framework.

Given the above context, this review surveys recent advances in PVA/PAA/PAM-based hydrogel electrolytes for flexible zinc batteries, with the objective of systematically elucidating how these materials achieve optimised ionic conduction and effective dendrite suppression across three interconnected dimensions—material composition, structural engineering, and functional mechanisms—while also critically examining their application prospects and practical bottlenecks in flexible device configurations. The review is organised around three central themes: the fundamental material characteristics and mechanistic roles of PVA/PAA/PAM composite hydrogels; the synergistic regulation of ionic transport and zinc deposition behaviour; and the performance evaluation together with future development directions for their deployment in flexible zinc-battery systems.

1.2. Scope of the Review and Basis for Material Selection

In recent years, research on hydrogel electrolytes for flexible zinc-ion batteries has progressively transitioned from a narrow focus on water retention and film-forming ability toward the synergistic optimisation of ionic conduction, mechanical integrity, and interfacial stability—a shift that has clearly delineated the “electrolyte bulk” as the legitimate core subject of this review [1]. Within this conceptual framework, we select poly (vinyl alcohol) (PVA), poly (acrylic acid) (PAA), and polyacrylamide (PAM) as our primary analytical objects, grounded in the following rationale: these three polymers individually embody three pivotal functions—structural support, chemical coordination, and moisture regulation. PVA is renowned for its superior film-forming capacity and robust mechanical backbone; PAA, rich in carboxyl groups, proves amenable to tailoring the Zn^{2+} coordination environment and interfacial adhesion; and PAM, distinguished by its high water retention and flexible chain segments, facilitates the preservation of continuous ionic pathways. The three polymer systems are highly complementary in functionality, such that the deficiencies of any single system—whether in ionic conductivity, mechanical strength, or long-term stability—can be effectively remedied through blending, crosslinking, or interpenetrating-network designs. Moreover, the extensive body of existing studies on these three materials, along with their well-established modification routes and clear evolutionary trajectories, renders them particularly suitable for extracting general mechanistic principles and transferable design guidelines. Consequently, this paper presents a systematic review centred on ionic-conduction regulation and zinc-dendrite inhibition mechanisms in composite hydrogel electrolytes, taking PVA/PAA/PAM as the main thread of our investigation.

2. FUNDAMENTAL THEORIES AND CORE MECHANISMS OF FLEXIBLE AQUEOUS ZINC-ION BATTERIES

2.1. Working Mechanisms of Flexible Aqueous Zinc-Ion Batteries

Critically, the working mechanism of flexible aqueous zinc-ion batteries necessitates that the role of gel electrolytes transcends the mere function of an ionic conduit—they must concurrently withstand deformation-induced stress, preserve interfacial contact, and sustain electrochemical activity. From a device-configuration perspective, a typical cell comprises a cathode, a zinc anode, a gel electrolyte, and an encapsulation layer, wherein the gel electrolyte not only serves as the ion-transport pathway but also functions as a physical separator between electrodes and as an interfacial buffering layer. In

contrast to rigid counterparts, flexible devices demand that all functional strata maintain uninterrupted interfacial contact and stable charge transport under repeated bending and twisting. This integrated electrode–electrolyte–encapsulation design forms the foundational basis for flexible zinc-ion batteries to strike a viable balance between electrochemical performance and wearable compatibility.

In terms of the underlying energy-storage mechanism, the essential chemistry remains the reversible migration of Zn^{2+} coupled with electrode reactions: upon discharge, the zinc anode undergoes oxidative dissolution, releasing Zn^{2+} that migrates through the electrolyte and intercalates into the cathode host; this process is reversed during charging. Prior work has established that the high safety and superior rate capability of aqueous zinc batteries stem from the intrinsically low electrochemical activation barrier of zinc metal and the rapid ionic mobility in aqueous media—yet these very advantages are inevitably accompanied by hydrogen evolution, parasitic side reactions, and dendritic growth, all of which necessitate regulation through rational electrolyte engineering [10]. Indeed, single-ion-conducting or highly adhesive gel systems have provided partial validation of this design philosophy: they are capable of homogenising Zn^{2+} flux, enhancing the reversibility of zinc deposition/stripping, and offering a more stable operational window for quasi-solid-state flexible devices [11].

The introduction of flexible scenarios adds a further layer of complexity to the problem. In rigid batteries, the electrode–electrolyte interface remains relatively stable, whereas flexible counterparts are prone to interlayer microcracks, local delamination, and stress concentration under bending and stretching—consequences that result in disrupted ion-transport pathways and, in extreme cases, localized short circuits. Particularly during plating/stripping, should the interface fail to maintain conformal continuity upon deformation, the spatial heterogeneity of Zn^{2+} flux can provoke localized current crowding and preferential dendritic nucleation. In other words, flexibility tests not merely mechanical robustness, but critically, the dynamic stability of the interface: the material must not only bend, but also—while bent—sustain ion conduction, preserve conformal contact, and retain electrochemical integrity. Overall, the practical performance ceiling of flexible zinc batteries is dictated by the interface's ability to preserve structural and functional integrity under dynamic deformation, which in turn constitutes the central rationale for subsequent electrolyte design.

2.2. Ion Conduction Mechanism in Hydrogel Electrolytes

In hydrogel electrolytes, ion conduction proceeds via a carrier-mediated mechanism: unlike solid polymer electrolytes, where ion hopping relies on segmental motion of the polymer chains, Zn^{2+} ions and their solvation shells migrate predominantly through water molecules within the continuous aqueous-phase channels. In essence, the conduction process is better described as "aqueous-network-mediated ionic flow" rather than "polymer-skeleton-driven migration." As Weng et al. have demonstrated, the efficiency of ion transport in aqueous gel systems critically depends on the construction and sustained integrity of continuous water pathways; local dehydration or channel collapse markedly elevates the ionic diffusion resistance [16].

Three primary factors govern ionic conduction in hydrogel electrolytes. Water content determines whether continuous ion channels can be established in the first place: at an appropriate level, sufficient liquid-phase domains form within the gel to permit rapid ion diffusion; when too low, channels shrink or even disconnect; when excessively high, over-swelling dilutes the polymer network and compromises mechanical integrity. Zhu et al. have rightly emphasised that high ionic conductivity does not arise simply from "more water", but rather from a rational partition between confined water—which stabilises the solvation environment—and free water, which provides the fast migration pathways [25]. Crosslinking density, on the other hand, imposes a fundamental trade-off between structural constraint and channel architecture: low crosslinking leaves channels open but risks creep-induced instability, whereas high crosslinking densifies the framework but narrows the channels, elevating migration resistance. Through a double-network design, Li et al. have

demonstrated that judicious crosslinking can achieve concurrent optimisation of mechanical restraint and ion diffusion, preserving both microporous voids and hydration spaces while attaining appropriate structural densification [9]. Functional groups further modulate salt dissociation and ion-migration barriers via polar interactions, Zn^{2+} coordination, or hydrogen bonding. The hydroxyl moieties of PVA, carboxyl groups of PAA, and amide groups of PAM all influence the ionic solvation state; however, overly strong binding tends to trap ions near chain segments, thereby increasing the activation energy for migration. Li et al. have shown that tuning the ratio of oxygen- to nitrogen-containing functional groups can effectively tailor the coordination environment and migration kinetics of Zn^{2+}, enabling synergistic gains in both ionic conductivity and interfacial stability [10].

The three factors, although seemingly independent, are inherently coupled: even with ample water content, ion conduction remains impeded if crosslinking is excessively dense, as the conduction pathways become compressed; and regardless of how well functional groups are designed, the coordination environment cannot be established in the absence of water. Only when all three parameters fall within a reasonable "window" can hydrogel electrolytes achieve the desired combination of high ionic conductivity, excellent stability, and long-term durability. In essence, water content constructs the conduction pathways, crosslinking density preserves their structural integrity, and functional groups optimise migration efficiency—these three elements are mutually intertwined and collectively dictate the overall conduction performance. For the design of electrolytes intended for flexible zinc batteries, the synergistic interplay among these factors should be taken as the core guiding principle, and a unified regulatory framework that links aqueous pathways, network architecture, and chemical coordination must be established [1, 16].

2.3. Growth Mechanism and Adverse Effects of Zinc Dendrites

Zinc dendrites are not accidental surface imperfections but rather an inevitable consequence that undergoes progressive amplification, arising from the imbalance of charge and mass transport within aqueous zinc electrodes under the synergistic influence of high current density, restricted ion supply, and a non-uniform interfacial electric field. The driving force stems from local ion depletion and electric-field inhomogeneity: when the Zn^{2+} transport rate in a given region of the electrode surface falls short of the electron-supply rate, concentration polarisation intensifies, and the deposition front is preferentially directed toward sites of higher field intensity, thereby establishing a positive-feedback loop that favours tip growth. Tan et al. reported that natural honey can retard dendrite evolution by modulating the interfacial water environment [13], whereas Zhang et al. demonstrated that a carboxyl-containing bacterial cellulose gel electrolyte, when combined with an imidazole-type ionic liquid, markedly improves deposition uniformity [14]; both findings corroborate the critical role of uniform interfacial ion distribution in suppressing dendritic nucleation.

Dendrite evolution exhibits cross-scale characteristics. At the atomic scale, the nucleation stage already becomes uncontrollable: the interfacial adsorption layer and the basic salt generated by local alkalisation alter the surface diffusion pathways of zinc atoms, leading to concentrated nucleation at a limited number of high-energy sites. Based on the TLK (terrace-ledge-kink) model, terraces, ledges, and defect sites on the crystal surface exhibit distinct atomic attachment preferences, which promotes the formation of protrusions and their subsequent evolution into primary dendrite precursors—heterogeneous nucleation thus predetermines the inherent bias of the ensuing morphology. When the process extends to the mesoscopic scale, the coupling between the concentration field and the morphology field turns unstable: surface protrusions intensify the local electric field, attracting more Zn^{2+} ions and accelerating reduction, whereas the ions surrounding the protrusions are continuously depleted, causing the concentration gradient to rise steadily and establishing a positive-feedback loop—"the more protruded, the faster the growth; the faster the growth, the more protruded". Phase-field simulations have demonstrated that the propagation direction and branching frequency of dendrites are governed by interface instability under mass-transfer limitations [37].

To intuitively understand the aforementioned cross-scale evolution pathway, Figure 1 summarizes the typical process of zinc dendrites from atomic-scale nucleation to macroscopic failure.

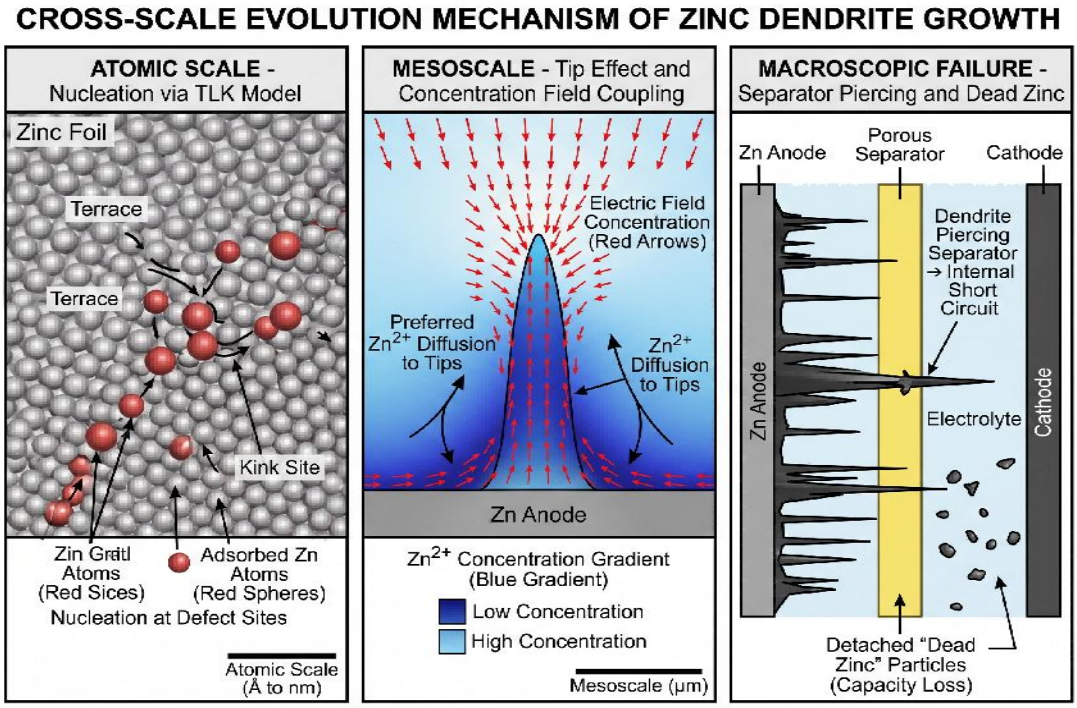


Figure 1. Schematic of the Cross-Scale Evolution Mechanism of Zinc Dendrites

The deleterious effects of dendrites propagate swiftly from microscale morphological irregularities to macroscale functional degradation: sharp dendrites pierce the separator, inducing short-circuiting and thermal runaway; the build-up of "dead zinc" results in reversible capacity loss and a drop in Coulombic efficiency; moreover, dendritic growth accelerates parasitic reactions—hydrogen evolution and corrosion in particular—thereby compromising the structural integrity of the electrodes. Consequently, dendrite suppression cannot be confined to mere surface "smoothing" or a simplistic enhancement of mechanical strength. Instead, targeted interventions must be directed separately at the two distinct stages—nucleation and growth: the former demands regulation of interfacial chemistry and uniformity of nucleation sites, whereas the latter calls for reconstruction of ion-transport pathways and electric-field distribution. Only by combining a "nip-in-the-bud" approach at the nucleation stage with a "guided homogenisation" strategy during growth can dendrite suppression evolve from passive "blocking" into active "regulation"—and this very principle serves as the logical cornerstone for the subsequent discussion of electrolyte structural design in Chapters 3 and 4.

2.4. Comparison of Intrinsic Properties of PVA/PAA/PAM Polymers

The reasons why PVA, PAA, and PAM have become the most frequently employed materials in gel-electrolyte research for flexible zinc-ion batteries do not lie in their individual perfection, but rather in their distinct and complementary strengths across mechanical support, water retention, and interfacial adaptation. PVA molecular chains are rich in hydroxyl groups, which readily form abundant hydrogen bonds and semi-crystalline domains—a structural feature that confers excellent film-forming ability, toughness, and mechanical strength, thereby enabling the electrolyte skeleton to preserve its structural integrity under repeated bending and deformation. However, high crystallinity also implies restricted segmental motion and insufficient free-volume distribution, resulting in relatively sparse ion-migration pathways and unremarkable intrinsic ionic conductivity. As Zhang Wei et al. have pointed out, without structural regulation, the internal crystalline regions of

PVA-based systems significantly impede the continuous migration of Zn^{2+}, making it difficult to meet the dual demands of high mechanical strength and high ionic conductivity simultaneously [1].

Compared with polyvinyl alcohol (PVA), polyacrylic acid (PAA) exhibits markedly enhanced hydrophilicity and higher ion-binding capacity, owing to the densely distributed carboxyl groups along its molecular chains. These carboxyl groups not only serve to immobilise and stabilise water molecules, thereby mitigating hydrogel dehydration, but also modulate local ion distribution through electrostatic interactions or coordination with Zn^{2+}, thus improving the uniformity of interfacial deposition to a certain degree. In a 2024 study, Ji Shanguo et al.introduced PAA into a composite hydrogel system and emphasised that the carboxyl-rich network of PAA contributes to enhanced water retention and interfacial wettability of the electrolyte, exerting a beneficial effect on the cycling stability of Zn–MnO_2 batteries [2]. These benefits, however, come with equally conspicuous drawbacks: the highly hydrophilic chain segments of PAA are prone to excessive swelling upon water uptake, which leads to relaxation of the network structure, a decline in mechanical strength, and even dimensional instability and interfacial delamination during prolonged cycling. In short, PAA excels in "water retention" and "zincophilicity," yet is susceptible to loss of structural integrity due to over-swelling.

In contrast, polyacrylamide (PAM) takes the amide group as its core functional unit and exhibits relatively strong polarity. It can not only form stable hydrogen bonds with water molecules, but also, under certain conditions, improve the wettability and adhesion between the electrolyte and electrode interfaces, thereby demonstrating considerable potential for interface adaptation in flexible devices. PAM possesses flexible chain segments and excellent processability, facilitating the construction of continuous and uniform gel networks, and is particularly suitable for fabricating flexible electrolyte layers with strong adhesion and rapid deformation responsiveness. In 2023, Li et al. reported that PAM-based gel systems exhibit favourable interfacial contact stability and deformation tolerance in flexible energy-storage devices, indicating that PAM holds certain advantages in buffering mechanical stress and reducing interfacial contact resistance [9]. However, the shortcomings of PAM are equally conspicuous: its water-retention capacity is generally weaker than that of PAA, rendering it more susceptible to water loss and cracking during prolonged operation; when used alone, its ability to directionally regulate Zn^{2+} migration is limited, and it cannot independently satisfy the dual requirements of dendrite suppression and long-term ionic conduction.

Accordingly, PVA, PAA, and PAM respectively exhibit the three typical characteristics of "robust skeleton, excellent water retention, and superior compatibility." However, no single polymer can simultaneously satisfy the comprehensive requirements of flexible zinc batteries—high ionic conductivity, excellent mechanical toughness, low interfacial impedance, and resistance to dendrite growth. The primary limitation of PVA lies in insufficient ion-transport channels; PAA suffers from excessive swelling; and PAM is deficient in both water retention and ion-regulation capability. This structural imbalance dictates that a single-component system can hardly break through the performance ceiling. Nevertheless, the above static comparison of intrinsic properties remains insufficient to unveil the failure-evolution path of each system under real cycling conditions: how crystallinity dynamically evolves upon repeated charge–discharge, under what conditions excessive swelling triggers electrode instability, and what constitutes the critical criterion for interfacial-homogenisation failure—these questions can only be addressed by incorporating performance-degradation behaviour under specific operating regimes. For this reason, building upon the intrinsic-property analysis, Chapter 3 will further examine the performance bottlenecks and failure mechanisms of single-polymer-based electrolytes in the actual operating environment of flexible zinc batteries, so as to provide a more comprehensive diagnosis of the root causes underlying their performance limitations.

Indeed, the intrinsic properties of PVA, PAA, and PAM outlined above do not exist in isolation. The material landscape for hydrogel electrolytes extends well beyond these three polymers—natural polymers, synthetic variants, and composite systems each possess distinct functional priorities and

performance boundaries. Appreciating this broader context helps clarify why PVA/PAA/PAM were chosen as the core subjects of this review through comparative analysis.

2.5. Overview of Other Hydrogel Electrolyte Systems

The research landscape for hydrogel electrolytes extends well beyond the PVA/PAA/PAM ternary system; in fact, diverse electrolyte formulations—ranging from natural polymers and synthetic variants to composite systems—have been actively explored for flexible zinc batteries, with each class displaying distinct performance characteristics and operational boundaries that are fundamentally rooted in differences in chemical structure.

Natural polymer hydrogels are typically represented by cellulose, chitosan, sodium alginate, gelatin, and starch, whose most prominent advantages lie in abundant reserves, biodegradability, and low cost. Zhang et al. constructed a gel electrolyte with excellent water-retention performance by combining carboxylated bacterial cellulose fibres with imidazole-type ionic liquids, which effectively improved the uniformity of zinc deposition [14]. However, natural polymers generally suffer from low mechanical strength, poor batch-to-batch consistency, and a narrow processing window—drawbacks that render them barely capable of independently meeting the stringent long-term cycling stability demanded by flexible devices; in most cases, they must be compounded with other components to realise practical application value.

Synthetic polymer hydrogels encompass a broad range of systems beyond PVA, PAA, and PAM. Among these, poly(N-isopropylacrylamide) (PNIPAm) has been exploited in intelligent responsive electrolyte design owing to its thermally responsive phase behaviour, whereas poly(ionic liquid)s have drawn considerable interest for their high ionic conductivity and wide electrochemical window. Qiu et al. achieved a marked improvement in the Zn^{2+} transference number by introducing sulfonic acid groups to anchor anions within a covalent organic framework (COF)/PAM composite system [11]. Synthetic polymers offer distinct advantages—precisely controllable molecular architecture, high batch-to-batch consistency, and ample scope for modification—yet their limitations are equally apparent: a single system typically accommodates only one or two performance metrics, and it remains challenging to concurrently fulfil the threefold requirements of ionic conductivity, mechanical strength, and interfacial stability.

Composite and hybrid hydrogels currently represent a frontier direction in the field, wherein researchers incorporate inorganic nanofillers—such as graphene oxide, MXene, and Laponite clay—into polymer networks to transcend the performance limitations of single materials through multiscale synergistic interactions among fillers, polymers, and salts. Chen et al. have pointed out that two-dimensional materials like MXene not only enhance internal interfacial continuity within the gel but also, via surface functional groups, exert a synergistic effect with Zn^{2+} to optimise the local electric-field distribution [7]. Tian et al., on the other hand, employed Laponite nanoclay to construct physical crosslinking points and a confined microenvironment, thereby markedly improving both water retention and deformation-recovery capacity [17]. Although such systems exhibit outstanding overall performance, their preparation procedures are relatively intricate, and the compatibility and long-term stability among disparate components still demand systematic validation.

Overall, the aforementioned systems each possess distinctive strengths across different dimensions: natural polymers excel in sustainability, synthetic polymers offer superior design flexibility, and composite systems stand out for their ability to deliver comprehensive performance breakthroughs. Nevertheless, none of these single systems can simultaneously accommodate the three core functions of structural support, ionic coordination, and water regulation. By contrast, the reason why the PVA/PAA/PAM ternary system is selected as the core research focus of this study lies precisely in the fact that each of the three components corresponds to one of the aforementioned functional orientations, and the three polymers can form a thermodynamically compatible synergistic structure via hydrogen-bonding associations and interpenetrating networks. This feature enables the

PVA/PAA/PAM system to strike a relatively sound balance between functional designability and engineering implementability, and also provides a specific material platform for the synergistic regulation strategy that will be elaborated in Chapter 4.

3. PERFORMANCE BOTTLENECKS OF SINGLE POLYMER-BASED HYDROGEL ELECTROLYTES

3.1. PVA-Based Electrolytes: The Inherent Trade-off Between Crystallinity and Ion Channels

The core challenge confronting PVA-based hydrogel electrolytes lies in the inherent difficulty of simultaneously achieving structural stability and ion accessibility. The abundant hydroxyl groups along PVA chains endow the material with excellent film-forming ability and crosslinkability, allowing it to construct a relatively well-defined three-dimensional network. However, the strong hydrogen-bonding interactions among PVA segments also promote the formation of highly crystalline domains, which in turn compress the free volume and restrict segmental mobility. These crystalline regions not only impede water uptake and retention but also create physical obstacles to Zn^{2+} migration pathways, causing the otherwise continuous ion channels typical of aqueous systems to become locally fractured, tortuous, or even blocked. In a single-component PVA system, this transport limitation imposed by high crystallinity is particularly severe, manifesting as low ionic conductivity, aggravated concentration polarisation, and inadequate rate capability—all of which render it difficult to meet the demands of flexible zinc-ion batteries for rapid charge–discharge and stable cycling. In essence, while PVA offers the necessary mechanical support and processing convenience, it achieves these at the cost of ion-transport efficiency, giving rise to a typical structure–performance paradox inherent to single-component systems. It is therefore imperative to introduce a second or even a third component to overcome this intrinsic bottleneck [1].

3.2. PAA-Based Electrolytes: The Contradiction Between Excessive Swelling and Mechanical Softening

The prominent advantage of PAA-based electrolytes lies in the densely distributed carboxyl groups along their molecular chains, which form strong interactions with water molecules and Zn^{2+} ions, thereby providing active sites for ion migration. Yet this very advantage also constitutes the root cause of their performance imbalance. Research by Jin et al. in 2023 demonstrated that although polycarboxyl-containing polymer hydrogels facilitate the construction of a reversible zinc-anode environment, their excessively strong hydrophilicity readily induces electrolyte swelling upon liquid uptake, which in turn weakens the compactness and structural stability of the network—indicating that PAA-based materials, while improving ion accessibility, are also plagued by the side effects of excessive swelling [15]. In line with this observation, Gao et al. further pointed out in 2023 that achieving high-performance output in flexible aqueous zinc-ion energy-storage systems necessitates a careful balance between mechanical integrity and interfacial stability of the hydrogel electrolyte; if the material continuously absorbs water and softens during cycling, the electrode contact state becomes unstable, local stress concentration emerges, and interfacial degradation ensues, ultimately constraining the long-term operational life of the device [16]. Thus, although the single PAA system possesses strong ion-binding capacity and interfacial affinity, the cascading problem of "high hydrophilicity—high swelling—low strength" remains unavoidable. Only through strategies such as compounding, crosslinking, or network reconstruction can mechanical softening and interfacial degradation be suppressed while preserving the advantage of ionic conduction.

3.3. PAM-Based Electrolytes: Shortcomings in Water Retention and Interfacial Adhesion

PAM-based hydrogel electrolytes have been widely adopted in flexible zinc-battery systems, owing to their flexible chain segments, mild gelation conditions, and favourable processing compatibility. However, their relatively loose network structure frequently gives rise to problems such as insufficient water retention, compromised structural integrity, and weak interfacial adhesion under prolonged cycling and complex deformation, making it difficult for PAM-based systems to stably maintain ion-transport pathways or effectively alleviate local electric-field concentration at the zinc-anode surface. A 2023 study by Siyu T. et al. indicated that constructing mechanically robust electrolyte membranes can significantly attenuate zinc dendrite growth and suppress side reactions, which indirectly suggests that, without adequate structural support and interfacial stability, PAM-based systems are prone to losing their regulatory effect on zinc deposition behaviour due to water loss and interfacial degradation during long-term operation [17]. The critical shortcoming of PAM-based electrolytes, however, is not merely the single issue of "decreased water content," but rather the failure to achieve a synergistic unification among water retention, mechanical properties, and interfacial adhesion. Only through crosslinking reinforcement, construction of composite networks, or introduction of strongly interacting components can these inherent deficiencies be remedied, thereby providing more reliable cycling support for flexible zinc batteries.

3.4. Inherent Limitations of Single-Component Systems: The "Impossible Triangle" of Conductivity, Mechanical Strength and Dendrite Inhibition

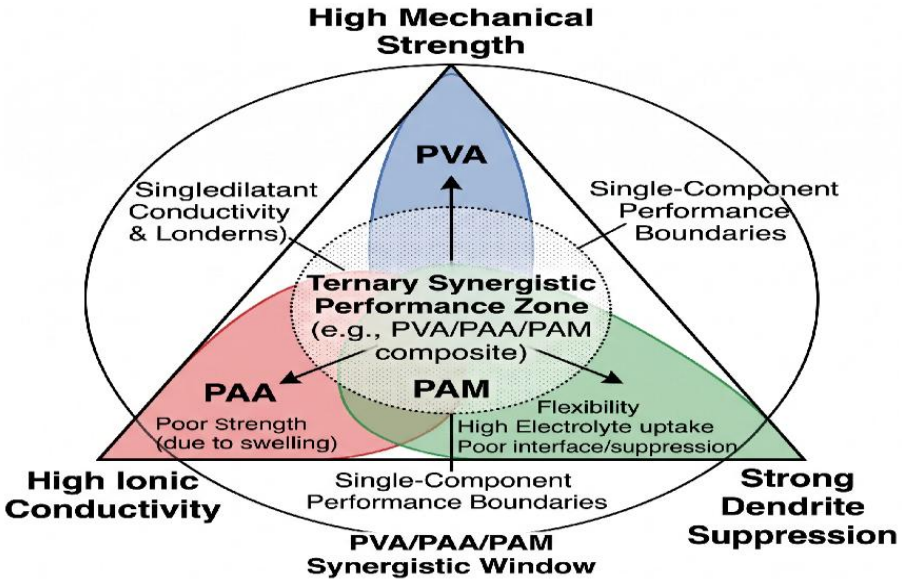


Figure 2. Schematic Diagram of the "Impossible Trinity" Bottleneck in Single Polymers

Single polymer hydrogel electrolytes have long been confronted with an unavoidable structural contradiction in flexible zinc batteries: ionic conductivity, mechanical strength, and dendrite-inhibition capacity generally cannot reach the desired levels simultaneously. PVA-based systems typically rely on relatively high crystallinity to maintain morphological stability, yet excessive crystalline domains reduce the free-volume fraction and weaken Zn^{2+} migration pathways. PAA-based systems, although rich in carboxyl groups that facilitate ion adsorption and interfacial bonding, suffer from a significant modulus decrease owing to severe water uptake and swelling. PAM-based systems possess flexible chain segments and strong water-retention capacity, but when used alone, they exert only a limited homogenising effect on the electric field at the zinc-anode surface and cannot fundamentally suppress dendritic nucleation. As Zuo et al. (2024) pointed out, PVA-based gel polymer electrolytes can achieve a certain trade-off between film-forming ability and

ion transport in zinc-ion devices, yet their performance enhancement still relies heavily on component synergy and structural optimisation—which implies that the single PVA system itself has an evident upper ceiling and cannot independently overcome the dual constraints of conductivity and mechanical properties [4]. This conclusion indirectly suggests that electrolyte design for flexible zinc batteries should no longer remain at the stage of "single-point optimisation," but rather shift toward multi-component synergistic regulation.

Table 1. Comparison of Key Properties of Single-Polymer Hydrogel Electrolytes

System	ionic conductivity	Mechanical modulus	Potential for controlling deposition morphology	Main inherent shortcomings:
PVA	secondary	higher	Limited	High crystallinity with restricted ion channels
PAA	higher	lower	secondary	Excessive swelling with a tendency for structural softening
PAM	Moderately high	secondary	Relatively limited	Insufficient homogenization capability of the interface

Note: 1) The performance comparison of various systems is based on the typical single-component hydrogel electrolytes reported in different literatures; 2) The actual ionic conductivity, mechanical modulus and deposition morphology are significantly affected by conditions including zinc salt concentration, crosslinking density, test temperature, current density/areal capacity and other factors. The qualitative trends presented in the table only reflect the intrinsic material properties of each system, and do not constitute an absolute performance ranking.

From the comparison, PVA primarily serves as a structural backbone, PAA is mainly responsible for ion adsorption, and PAM tends to provide water retention and chain flexibility. However, each of the three materials inevitably compromises one critical performance dimension—either ionic conductivity, mechanical stability, or dendrite-suppression capability. For this reason, Chapter 4 moves beyond the conventional "weakness-compensation" approach of single-material systems and instead adopts a synergistic PVA/PAA/PAM composite strategy, which, through network-structure reconstruction, cooperative functional-group coordination, and interfacial electric-field regulation, achieves concurrent enhancement of all three performance aspects.

4. SYNERGISTIC REGULATION STRATEGIES OF PVA/PAA/PAM-BASED GEL ELECTROLYTES

To achieve the aforementioned synergistic objectives, this chapter systematically sorts out the regulation strategies of PVA/PAA/PAM-based gel electrolytes from three dimensions: physical construction of ion transport channels, regulation of zinc ion solvation chemistry, and synergistic stabilization of interfacial electric field mechano-electrochemistry. Figure 3 first presents the overall schematic of the tripartite synergy at the molecular level, which serves as the structural reference for the elaboration in subsequent sections.

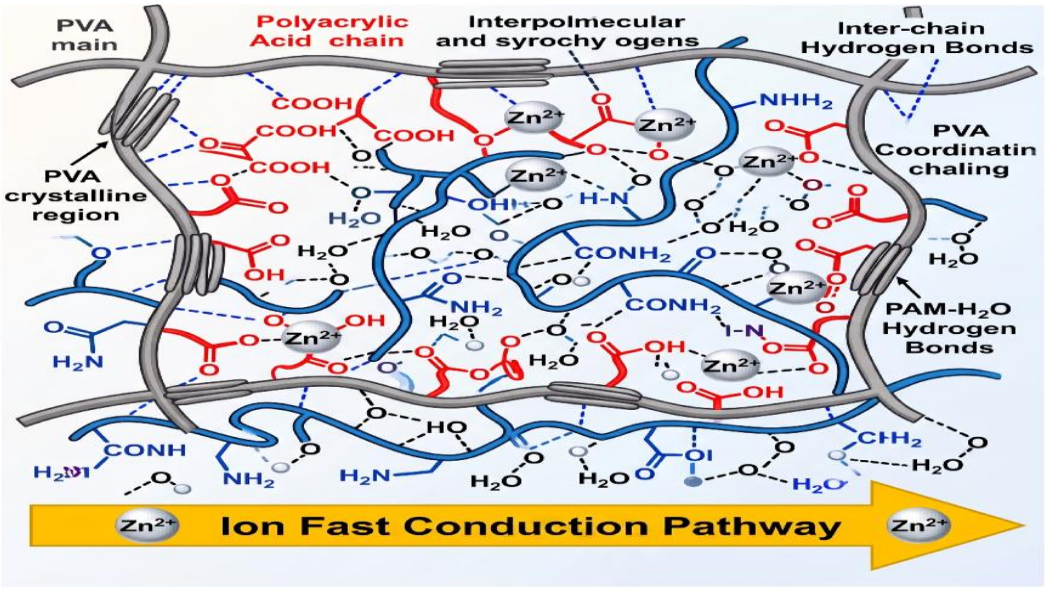


Figure 3. Schematic Diagram of the Ternary Synergistic Molecular Interaction Mechanism of PVA/PAA/PAM

4.1. Physical Construction of Ion Transport Channels and Strategies for Activation Energy Reduction

4.1.1. Crosslinking Density Regulation and Porous/Oriented Structure Design

The core of crosslinking-density regulation and porous/oriented structure design lies in reconstructing the internal spatial topology of the gel so as to minimise Zn^{2+} migration pathways, reduce transport tortuosity, and furnish continuous, low-resistance channels for ions while preserving mechanical support. In 2024, Kim et al. constructed a nanofibre network via electrospinning and demonstrated that such an architecture can form a highly interconnected skeleton, enabling more efficient ion migration along inter-fibre gaps, which effectively lowers interfacial polarisation and enhances the rate capability of batteries—this work suggests that building oriented continuous channels fundamentally improves ion-transport efficiency more effectively than merely increasing salt concentration [38]. In a 2024 study, Li et al. further proposed a bilayer gel-electrolyte design, in which an inner layer with high fluidity ensures rapid ion transport while an outer layer with high strength maintains structural integrity and resists deformation-induced instability; the synergistic interplay between the two layers achieves a balance between conduction rate and mechanical durability, illustrating that spatially partitioned regulation of crosslinked networks can overcome the conventional "high conductivity–low strength" trade-off [34]. In 2023, Duan et al. reported a construction route for three-dimensional porous electrolyte structures, showing that the introduction of interconnected pores markedly improves internal wettability and ion accessibility, thereby promoting a more uniform Zn^{2+} distribution across the entire electrode interface, reducing local deposition risks, and delaying dendritic nucleation—this finding further corroborates that pore formation is not merely a conductivity-enhancement strategy but also a critical route to optimising deposition uniformity [29]. Collectively, crosslinking-density regulation and porous/oriented structure design are not isolated morphological modifications; rather, they constitute a systematic engineering approach centred on reducing migration resistance while simultaneously reconciling mechanical constraints with interfacial uniformity, thereby offering a valuable paradigm for constructing efficient and stable ion-conduction networks in PVA/PAA/PAM-based flexible electrolytes.

4.1.2. Double Network (DN) and Interpenetrating Network (IPN) Structures

Double network (DN) and interpenetrating polymer network (IPN) structures offer a viable route for flexible gel electrolytes to simultaneously achieve mechanical support and ionic conduction. The first network typically functions as a rigid skeleton that resists deformation and prevents structural collapse of the electrolyte during cycling; the second network, composed predominantly of flexible chain segments, is responsible for constructing continuous ion-migration pathways, thereby establishing a synergistic balance between mechanical strength and ionic conduction. Notably, in the PVA/PAA/PAM ternary composite system, abundant intermolecular hydrogen bonds can form among hydroxyl, carboxyl, and amide groups—this dynamic physical crosslinking endows the interpenetrating network with thermodynamic compatibility, enabling uniform interpenetration of the three polymer chains at the molecular scale, which fundamentally distinguishes the system from simple physical blends and provides a structurally homogeneous continuous framework for subsequent uniform ion coordination and stress transmission. The PAM/LCNF double-network system proposed by Li et al. in 2023 demonstrates that coupling a rigid fibre network with flexible polymer chains simultaneously achieves physical confinement and water confinement: the former improves the overall modulus of the gel through the spatial constraint of nanofibres, while the latter retards water loss and stabilises the Zn^{2+} migration environment, enabling the electrolyte to maintain high structural integrity under bending and prolonged cycling [9]. In a consistent vein, the work of Hang et al. in 2023 further illustrates the application potential of double-network hydrogels in zinc–air batteries, indicating that multi-network support can significantly enhance both water-retention capacity and interfacial stability, thereby alleviating the problems of water loss and deactivation under ambient conditions and improving the continuous output capability of flexible devices [33]. At a more refined network-design level, the ionic–covalent double network of BC-IL/PAM constructed by Zhang et al. embodies the structural merits of interpenetrating networks: ionic bonds impart a degree of dynamic reversibility, while the covalent network provides a stable skeleton; the synergy of the two not only improves toughness and fatigue resistance but also affords a more uniform transport pathway for Zn^{2+}, reducing the risk of local ion accumulation. Overall, through a "rigidity–flexibility coupling" structural design, double and interpenetrating networks enhance mechanical reliability while sustaining efficient ion transport, laying a solid foundation for transitioning flexible zinc-battery electrolytes from laboratory performance optimisation to practical device applications.

4.1.3. Reinforcement by Inorganic/Organic Nanofillers

The incorporation of inorganic or organic nanofillers represents a pivotal strategy for enhancing the overall performance of PVA/PAA/PAM-based gel electrolytes, with the core mechanism lying in the construction of a more efficient and stable ion-transport environment through multiscale coupling interactions among fillers, polymers, and salts. In a 2023 study, Sun et al. demonstrated that ZnO nanofillers can promote zinc-salt dissociation by amplifying the interfacial space-charge effect, while simultaneously creating additional activation sites for ion migration within the gel network, thereby achieving concurrent improvements in ionic conductivity and electrochemical stability—this finding indicates that nanofillers are not merely mechanical reinforcing agents but also functional regulatory units for ionic conduction [19]. Furthermore, Bao et al. proposed that zeolite molecular sieves, by virtue of their regular microporous channels and high specific surface area, can provide ions with more directional migration pathways and attenuate the disordered diffusion of solvent molecules through pore-confinement effects, enabling more focused and controllable Zn^{2+} transport, which underscores the unique value of structurally ordered fillers in constructing oriented conduction networks [24]. In line with this perspective, studies by Chen et al. on MXene and related nanomaterials reveal that such two-dimensional highly conductive fillers not only improve internal interfacial continuity within the gel but also engage in synergistic interactions with metal ions via surface functional groups, thereby optimising the local electric-field distribution and mitigating uneven zinc deposition [7]. In the Laponite-based quasi-solid electrolyte system, Tian et al. found that lamellar nanoclay can establish physical crosslinking points and confined microenvironments

that enhance water retention and deformation-recovery capability, while simultaneously providing relatively continuous low-resistance pathways for ion migration—a combination particularly suited to the bending tolerance and long-term stability demanded by flexible devices [17]. Broadly speaking, ZnO focuses on promoting salt dissociation and ion activation, zeolite emphasises ordered pore channels and directional transport, MXene and its analogues prioritise interfacial coupling and field homogenisation, and Laponite highlights structural support, water retention, and quasi-solid stability; although these fillers diverge in their specific functional orientations, they converge on the same overarching objective—namely, synergistically improving ionic conduction efficiency and zinc-deposition stability through nanoscale structural regulation.

4.2. Regulation of Zinc Ion Solvation Structure and Optimization of Coordination Chemistry

4.2.1. Regulation of Solvation Structure via Functional Group Coordination

The core premise of functional-group coordination in regulating Zn^{2+} solvation structures is to reshape the ionic microenvironment at the molecular level, redirecting zinc-ion transport from a regime of free diffusion toward one of controlled migration, thereby simultaneously enhancing ionic-conduction uniformity and deposition stability. A 2023 study by Li et al. demonstrated that polar groups—carboxyl and hydroxyl in particular—can reconstruct the solvation sheath of Zn^{2+} through weak coordination and hydrogen-bonding interactions, weakening the strong binding of water molecules to zinc ions, reducing the desolvation energy barrier, and thus improving interfacial charge-transfer kinetics; this work confirms that the electrolyte is not a passive conduit but a functional medium capable of actively modulating the Zn^{2+} coordination equilibrium [10]. In the same year, Jin et al. further revealed that the negative charges carried by carboxyl groups within the polyanionic backbone can spatially homogenise Zn^{2+} flux, mitigating local current-density concentrations and suppressing dendritic nucleation—the essence of this effect lies not merely in zinc-ion adsorption, but in the redistribution of ionic flow through the fixed charge field to reconstruct a smoother deposition interface [15]. Tian et al. (2023) emphasised the value of chain–liquid synergy, showing that flexible segmental motion couples with liquid-phase ion diffusion to guide directional Zn^{2+} migration, thereby circumventing interfacial instability arising from disordered transport—a finding that underscores the ideal electrolyte should possess both the constraining force of a solid framework and the transport continuity of a liquid phase [28]. A 2024 report by Wang et al. focused on activating the intrinsic zincophilicity of polyacrylamide (PAM) systems, pointing out that enhancing amide–Zn^{2+} interactions markedly improves ion adsorption and interfacial wetting, consequently increasing deposition density on the zinc-anode surface [18]. Collectively, the regulation of Zn^{2+} solvation via functional groups can be categorised into three primary modes: the coordination mode, which directly reconstructs the solvation sheath through polar sites such as carboxyl, hydroxyl, and amide groups; the charge mode, which homogenises Zn^{2+} flux via fixed charges from polyanions; and the synergistic mode, which achieves directional ion migration through the coupling of segmental motion and liquid-phase diffusion. Together, these mechanisms demonstrate that the essence of this chemical strategy is not a simple increase in affinity sites, but rather a precise molecular-level programming of Zn^{2+} behaviour.

4.2.2. Strategies for Single-Ion Conduction and Transference Number Enhancement

A high Zn^{2+} transference number is one of the critical parameters governing the operational stability of flexible zinc batteries, as its essential role lies in minimising the parasitic migration of anions under the applied electric field, thereby suppressing concentration polarisation, homogenising the interfacial ion distribution, and promoting more continuous and uniform zinc deposition on the anode surface. Qiu et al. have proposed that introducing sulfonic acid groups to anchor anions within a COF/PAM composite system can markedly curtail the free mobility of anions, thereby concentrating the charge-carrier contribution more heavily on Zn^{2+} itself; this in turn elevates the transference number

and improves the interfacial electrochemical environment, not only enhancing the directionality of ionic flux but also, to some extent, restraining dendrite formation induced by local current-density concentrations [11]. In a complementary approach focused on conductivity enhancement, Wang Haoran et al. (2024) reported a high-conductivity hydrogel polymer electrolyte constructed via a PVA/chitosan composite that establishes a more continuous ion-philic network, which optimises internal ion-transport pathways and improves Zn^{2+}migration efficiency while maintaining mechanical flexibility, thereby demonstrating that multi-polymer synergy can simultaneously boost overall ionic conductivity and provide more stable channels for Zn^{2+}migration [5]. Taken together, these findings indicate that the optimisation of single-ion conductivity and transference number is not an isolated materials-parameter adjustment but rather a systematic strategy—encompassing anion immobilisation, channel reconstruction, and polymer-network synergy—that ultimately achieves concentration-polarisation mitigation, uniform zinc deposition, and overall electrolyte-performance enhancement [1,5].

4.2.3. Synergy of Water-Salt Concentration and Regulation of "Active Water"

In PVA/PAA/PAM-based gel electrolytes for flexible zinc batteries, water is far more than an inert solvent background—it is a core variable that governs ionic migration, interfacial reactions, and the intensity of parasitic side processes. The concept of "active water" emphasises that not all water molecules play identical electrochemical roles: those with higher mobility are more prone to participate in hydrogen evolution, side reactions, and interfacial instability. Reducing the proportion of free water and reconstructing the water existing state essentially compresses the occurrence space of side reactions at the source. A 2024 study by Zhu et al. revealed a significant synergistic interplay between water content and salt concentration: when water molecules are co-constrained by the polymer network and solutes, the solvation environment of Zn^{2+} is remodelled, enabling simultaneous optimisation of ionic transport and electrochemical stability—indicating that a system with low free water and strongly bound water is more favourable for achieving efficient and stable zinc deposition [25]. This conceptual framework is further corroborated by studies on water-environment regulation via small-molecule additives; the work of Tan et al. and Wu et al. demonstrates that introducing components capable of interacting with water molecules or Zn^{2+} can attenuate water activity, suppress side reactions, and improve interfacial homogeneity, revealing that "water regulation" essentially reconstructs the electrolyte microenvironment through both chemical and structural pathways. Consequently, in the PVA/PAA/PAM composite gel system, the synergistic regulation of water–salt concentration is not a mere adjustment of component ratios, but rather a systematic remodelling—centred on the core concept of "active water"—of solvation structure, ionic migration pathways, and interfacial reaction kinetics, which provides critical support for high-stability flexible zinc batteries.

4.3. Synergistic Enhancement of Interfacial Electric Field Homogenization and Mechano-Electrochemical Stability

The core concept of interface engineering is currently shifting from the traditional "passive inhibition" paradigm to "active regulation": rather than merely relying on increasing electrolyte mechanical strength to delay battery failure, it reconstructs the electrode–electrolyte interfacial microenvironment to synergistically orchestrate ion transport, desolvation kinetics, and deposition morphology, thereby curtailing dendrite nucleation at its origin. Zhou et al. proposed an oriented dipole-induced electric-field strategy that establishes a directional polarisation barrier at the interface, which on one hand facilitates rapid Zn^{2+} desolvation and on the other induces preferential zinc growth along the low-surface-energy (002) crystal plane, yielding a dense and uniform deposition layer. In a complementary approach, Yang et al. utilised a thermally repairable interface to achieve dynamic closure of microvoids between the gel and the zinc surface, mitigating local current-density concentrations and improving both interfacial continuity and mechanical integrity over prolonged cycling. The PAA dually-modified gel reported by the Shangnuo team exhibits superior interfacial stability in zinc–air batteries, where its carboxyl-coordination and hydrophilic network jointly buffer

parasitic side reactions while maintaining the accessibility of the three-phase interface. Furthermore, polymer protective coatings, by constructing artificial interface layers, can homogenise electric-field distribution, isolate detrimental side reactions, and enhance interfacial adhesion. The core effect of interface regulation is intuitively illustrated in Figure 4. Table 2 summarises the core logic, representative works, and key performance improvements of these three strategic categories. Overall, physical strategies address the "transport" issue, chemical strategies target the "deposition" problem, and interface strategies determine the "stability" of the system; only the synergy of all three can propel flexible zinc batteries from mere functional prototypes toward long-term durable operation.

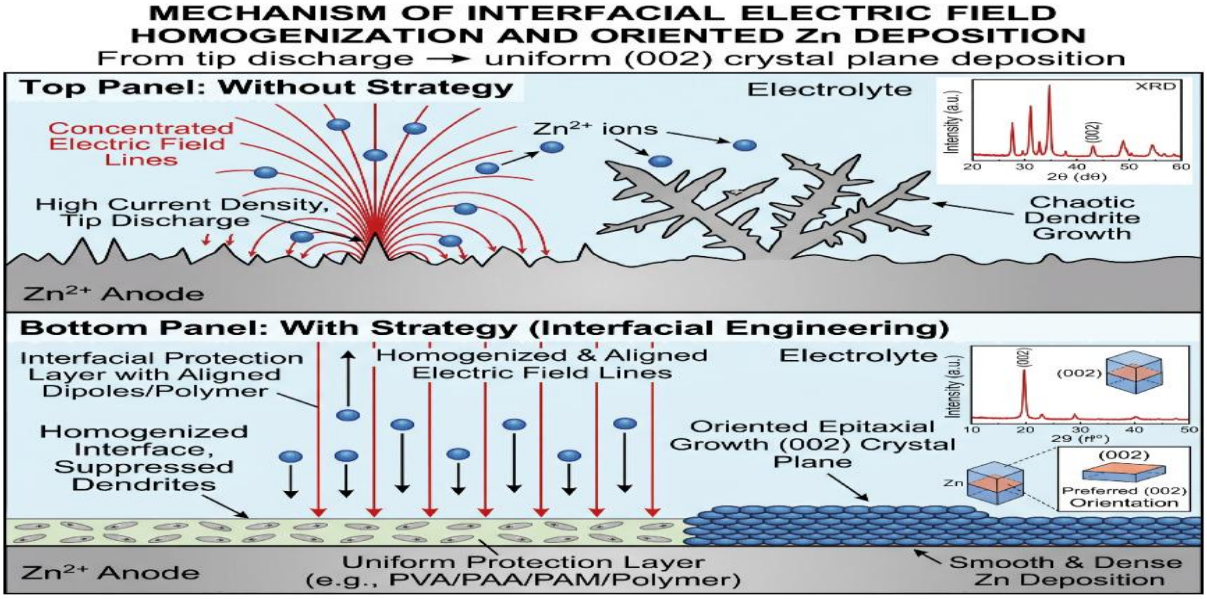


Figure 4. Schematic Diagram of Interfacial Electric Field Homogenization and Oriented Deposition Mechanism

Table 2. Comprehensive Comparison of Three Major Regulation Strategies for PVA/PAA/PAM-Based Flexible Zinc Battery Electrolytes

Strategy Dimension	Physical Strategy	Chemical Strategy	Interface Strategy
Core Objective	Reduce ion migration resistance and establish a continuous low-resistance pathway	Regulate the solvation structure of Zn^{2+} to optimize deposition behavior	Homogenize the interfacial electric field to enhance mechanical-electrochemical synergistic stability
Main Methods	Cross-linking density regulation, porous/oriented structures, dual networks/porous-interpenetrating networks, nanofiller composites	Coordination regulation of functional groups, single-ion conductivity, water-salt concentration synergy, and additive regulation	Oriented dipole-induced electric field, thermal repair interface, polymer protective coating
Representative Literature	Kim et al. [38], Li et al. [34], Duan et al. [29], Li et al. [9], Hang et al. [33], Zhang et al. [15], Sun et al. [19], Bao et al. [24], Tian et al. [17], Chen Ze et al. [7]	Li et al. [10], Jin et al. [15], Tian et al. [28], Wang et al. [18], Qiu et al. [11], Zhu et al. [25], Tan et al. [13], Wu et al. [31]	Zhou et al. [20], Yang et al. [26], Shang Nuo et al. [3], Cai et al. [22]
Key Performance Enhancements	Electrical conductivity: 235.7 mS/cm; 9.75 mS/cm at -50 °C; 43.76 mS/cm	Migration number: 0.89; Coordination number: 4.49; Energy density: 236.8 Wh/kg; Cycle life: 2250 h	Strong (002) texture deposition; 76.4 mW/cm² under 150° bending; stable cycling for 500 hours
Typical Implementation Method	Continuous channel of PVA/PAA nanofibers; Proton-rich network of PVA/chitosan; Three-dimensional porous structure of PAM/gelatin; Dual-network confinement of PAM/LCNF	The carboxyl group of PAA coordinates and reconstructs the solvation shell; the amide group of PAM activates zinc affinity; the sulfonic group of COF/PAM anchors anions.	The PAA dual-modified gel alleviates adverse reactions; the gelatin thermally induced sol-gel transition fills interfacial voids; the P2VP coating homogenizes the electric field distribution.

5. APPLICATION EVALUATION OF MULTIVARIATE COMPOSITE SYSTEMS IN FLEXIBLE ZINC-BASED DEVICES

5.1. Flexible Zinc-Ion Batteries: Transition From Coin Cells to Flexible Devices

The remarkable specific capacity and cycling stability demonstrated by laboratory-scale coin cells over short test periods are largely attributable to their closed configuration, constant applied pressure, and idealised electrode–electrolyte contact—conditions that effectively mask the inherent deficiencies of electrolytes under real-world service environments. When transferred to flexible device scenarios, however, repeated bending, twisting, stretching, and confined packaging space rapidly exacerbate non-uniform ion transport, interfacial debonding, and local current crowding,

giving rise to a substantial gap between laboratory feasibility and device applicability. To bridge this gap, Li et al. (2024) proposed a bilayer gel electrolyte for fibrous batteries, in which an outer layer provides mechanical protection while an inner layer ensures ionic conduction; this functional division significantly enhances both structural integrity and electrochemical stability under bending, illustrating that flexible devices demand not a mere miniaturisation of cells but a structural re-engineering of electrolytes to accommodate deformation [34]. Zuo et al. introduced PVA-based gel polymer electrolytes into zinc-ion hybrid capacitors, capitalising on the electrolyte's excellent film-forming ability and flexible network to improve electrode–electrolyte contact, while simultaneously balancing ion migration and mechanical support—a result that underscores the strong engineering-translation potential of PVA-based systems in flexible energy storage. Ji et al. further validated the practical value of PAM/MMT composite hydrogels in Zn–MnO_2 batteries, where the synergistic reinforcement of montmorillonite nanosheets and the water-retaining PAM network affords a more stable interfacial environment and durable cycling output, thereby confirming the advantages of composite hydrogels in coping with the complex operational demands of flexible devices. In essence, the transition from coin cells to flexible devices is a shift from static performance demonstration to dynamic service verification, and the electrolyte stands as the pivotal hub determining the success of this transition—it must not only conduct ions but also sustain structural integrity, stabilise the electric field, and suppress side reactions under deformation, water loss, and interfacial disturbance. Only through concerted design across materials, architectures, and devices can flexible zinc-ion batteries evolve from functional prototypes into truly available, durable, and practical power sources.

5.2. Extensional Thinking on Expansion into Zinc-Air Systems: Shift of Focus on Water Retention and Interfacial Stability

The working environment of flexible zinc-air batteries differs fundamentally from that of closed zinc-ion systems: the cathode side demands continuous gas diffusion to sustain the oxygen reduction and evolution reactions, which fundamentally reshapes the core challenge confronting gel electrolytes. While the central dilemma in closed ZIBs centres on reconciling dendrite suppression with ionic conduction, the obstacles encountered in open ZABs pivot instead around water-retention durability, resistance to CO_2 poisoning, and the dynamic stability of the solid–liquid–gas three-phase interface—three dimensions that are largely independent of Zn^{2+} coordination chemistry, thereby compelling us to reassess the relevance of the PVA/PAA/PAM system through the lenses of water management and environmental tolerance.

We first turn to water-retention performance. The abundant carboxyl groups in poly (acrylic acid) (PAA) exhibit strong binding affinity toward water molecules—a distinct advantage in open-air configurations, as they can retard hydrogel dehydration and thus sustain both the continuity of ion-transport channels and the hydrated microenvironment essential for the oxygen reduction reaction. Shang et al., assembling a neutral zinc–air battery with PAA-dually-modified hydrogels, found that the hydrophilicity of the carboxyl network indeed lowers the water-loss rate over prolonged cycling, thereby affording the battery an extended lifespan under non-sealed conditions [3]. From a structural-design perspective, Hang et al. demonstrated that the value of double-network hydrogels in flexible zinc–air batteries (ZABs) lies primarily in mechanical support: the rigid-flexible integrated network not only resists bending deformations but also retards water evaporation through physical confinement, enabling the device to maintain a more stable output during continuous discharge [33].

Nevertheless, for the PVA/PAA/PAM system to gain practical traction in zinc–air batteries (ZABs), the most pressing challenge at present lies in its CO_2 resistance. CO_2 from ambient air diffuses through the gel electrolyte to the air cathode and reacts with OH^- in the near-neutral or alkaline environment to form carbonates, which directly passivate the catalyst's active sites and block gas-diffusion channels, resulting in a precipitous decline in ORR/OER activity. This issue, moreover, is largely independent of Zn^{2+} coordination chemistry and is instead governed by the CO_2 permeability of the

gel, interfacial pH stability, and gas-mass-transfer efficiency. To be candid, systematic investigations into the CO_2 tolerance of PVA/PAA/PAM systems remain rather scarce; existing reports are mostly confined to dual modification of PAA or double-network construction aimed at water retention [3, 33], leaving a substantial gap before a truly integrated design—one that reconciles water retention, CO_2 blocking, and oxygen conduction—can be realised.

Moreover, flexible deformation adds another layer of complexity to the stability of the three-phase interface. Repeated bending and stretching generate microcracks and even local debonding between the gel and the air cathode, which not only disrupts gas-diffusion pathways but also impairs ionic-conduction channels; once local current density becomes non-uniform, the reaction homogeneity can no longer be sustained. This issue is particularly acute in fibre-shaped and thin-film devices, where the demands on interfacial adhesion toughness, deformation recovery rate, and long-term fatigue resistance are substantially more stringent than those for static batteries [16, 36].

Ultimately, whether the PVA/PAA/PAM system can secure a practical niche in flexible zinc–air batteries hinges on achieving an engineering-feasible trade-off among three interconnected aspects: water retention, CO_2 resistance, and interfacial mechanical matching. Yet current research along this line remains at a fairly preliminary stage, and future efforts should prioritise the introduction of functional fillers capable of adsorbing or converting CO_2 to mitigate gas-permeation side effects, the development of gradient-structured or triple-layered gels to decouple the functions of the water-retention layer and the CO_2-blocking layer, and the clarification of the progressive failure mechanism of the three-phase interface under deformation through in-situ characterisation. Prior to these breakthroughs, the discussion on ZABs in this section serves only as a preliminary extension of the application boundary of the PVA/PAA/PAM system, while the central theme of this review remains the regulation of ionic conduction and dendrite suppression in closed zinc-ion batteries.

5.3. Electrolyte Configuration Adaptation for Fiber-Type/Film-Type Devices

Substantial distinctions exist between fibrous and thin-film flexible devices in their electrolyte requirements: the former prioritises weavability, bendability, and continuous interface-coating capability, whereas the latter places greater emphasis on large-area uniform film formation, controllable thickness, and the stability of maintaining overall ion transport under repeated deformation. For fibrous configurations, the electrolyte must not only form a continuous, dense, and non-detachable ion-conducting layer on fine-diameter curved surfaces but also preserve interface integrity during stretching, torsion, and weaving processes; thin-film devices, in contrast, rely more heavily on the flatness, mechanical consistency, and in-plane ion-distribution uniformity of the electrolyte—otherwise, local stress concentrations readily trigger performance degradation. Wang et al., in their review, pointed out that the key reason gel electrolytes have gained broad adoption in flexible wearable electronics lies in their ability to establish a dynamic balance among softness, adhesion, and ionic conductivity, while adapting to the processing requirements and service environments of different device morphologies through network-structure design [8]. Furthermore, the carboxylated hydrogel electrolyte proposed by Cong et al. (2023) leverages the synergistic interplay between chain segments and the liquid phase to markedly improve interfacial ion migration and zinc-deposition behaviour, demonstrating that in either fibrous or thin-film devices, rational chemical-bond construction and the establishment of continuous transport pathways constitute the core routes to enhanced configuration compatibility [28].

6. ANALYSIS OF CORE BOTTLENECKS AND CONSTRAINING FACTORS

As is evident from the systematic overview above, although PVA/PAA/PAM-based hydrogel electrolytes have shown considerable promise in overcoming the performance bottlenecks of flexible

zinc-ion batteries—the three-dimensional "physics–chemistry–interface" synergistic framework offering a systematic route to reconciling the inherent trade-offs among ionic conductivity, mechanical strength, and dendrite suppression—this framework nonetheless remains at a transitional stage from proof-of-concept to practical deployment, with four core bottlenecks constraining its further advancement.

Insufficient adaptability across a wide temperature range stands as the primary bottleneck. Wearable electronics, outdoor sensors, and vehicle-mounted storage systems are often simultaneously subjected to high-temperature solar radiation, sub-zero freezing, and frequent thermal cycling; should the electrolyte function stably only within a narrow window, its ionic transport, interfacial contact, and mechanical integrity will rapidly deteriorate. Duan et al. investigated all-climate hydrogel electrolytes operational from −50 °C to 100 °C and proposed that "passive tolerance" under extreme temperatures can be realised through inhibition of ice crystallisation, stabilisation of water states, and retention of network flexibility [29]. Yet whether this passive-tolerance strategy has reached its performance ceiling, and whether its operating temperature range can be further extended via material design, remain open questions. The concept of temperature-adaptive electrolytes introduced by Qu et al.—which actively modulates ion-migration pathways through the synergistic interplay of dynamic bonds, tunable solvation structures, and phase transitions [27]—offers a promising alternative, but its compatibility with the PVA/PAA/PAM system has yet to be systematically validated.

Dynamic interfacial instability represents a particularly acute challenge in flexible scenarios; under continuous mechanical stresses—bending, stretching, and compression—during device operation, microcracks, interfacial debonding, and local contact instability readily arise between electrodes and electrolytes, which in turn disrupts ion transport and engenders non-uniform current distribution. The thermally repairable interface technology developed by Yang et al. achieves active recovery of interfacial structure through reconfigurable interactions, shifting the paradigm from post-failure remediation to adaptive maintenance during operation [26]. However, most existing self-healing systems rely on external stimuli to trigger the repair process, whereas flexible batteries can hardly provide such triggering conditions as heating or illumination in practical applications; how to achieve intrinsic self-healing—capable of autonomously repairing dynamic damage at room temperature without any external trigger—remains a bottleneck that urgently needs to be overcome.

The engineering gap in large-scale preparation is by no means negligible. Admittedly, the ideal microstructure achieved in the laboratory through precise temperature control, slow film formation, and stepwise cross-linking proves exceedingly difficult to replicate in roll-to-roll continuous production; the variability of parameters such as stirring uniformity, solvent evaporation rate, and cross-linking reaction synchronicity is substantially amplified, giving rise to batch-to-batch inconsistencies in ion-channel distribution, mechanical properties, and interfacial adhesion. Zhang Wei et al. have pointed out that overly complex multi-step processes are inherently hard to scale, whereas excessive simplification tends to compromise conductivity, water retention, and mechanical integrity [1]. Zhu et al. further emphasised that the coupled interplay among water content, salt concentration, and polymer network architecture directly governs the electrolyte's stability across varying temperature and humidity regimes; if one pursues only high conductivity under ideal laboratory conditions while disregarding the practical operational window, the all-too-familiar scenario emerges—excellent performance in the cell but rapid failure in the field [25].

The insufficient depth of mechanistic understanding severely constrains the improvement of rational design capability. For the transport behaviour of Zn^{2+} within hydrogels, the macroscopic "aqueous-channel-facilitated ion migration" model is broadly accepted, yet the precise regulatory role of the polymer network at the molecular level remains elusive—whether Zn^{2+} migrates via "hopping" along chain surfaces through coordination bond breakage and reformation, or preferentially "drifts" through water-rich microdomains mediated by water carriers, remains unresolved. These two scenarios imply fundamentally distinct optimisation strategies: the former calls for enhancing the coordination activity of functional groups, whereas the latter demands preserving the continuity and

low tortuosity of aqueous pathways. Likewise, the nucleation process of zinc dendrites persists as a "black box"; ex-situ observations can only capture the final post-growth morphology, leaving the physicochemical events at the instant of nucleation largely unknown—what dictates nucleation-site selectivity, what is the critical local current-density threshold, and how does the competition between the interfacial adsorption layer and Zn^{2+} reduction evolve? Zhang et al. have unveiled the coupled instability of concentration and morphology fields during dendrite propagation via phase-field simulations [37], yet these results urgently require cross-validation through in-situ or operando characterisation. Techniques such as operando TEM offer an unprecedented window for directly observing nucleation and propagation, but their current applicability is more suited to verifying local mechanisms than serving as a universal characterisation tool [12], owing to the cumbersome sample preparation, the vacuum environment that deviates from realistic aqueous conditions, and the potential electron-beam perturbation of the genuine reaction pathway.

Overall, the four categories of bottlenecks discussed above are not mutually independent; the gaps in mechanistic understanding constrain the precision of materials design, while the functional limitations of the materials in turn magnify the uncertainties inherent in engineering scale-up. Collectively, these issues stem from an insufficient appreciation of the electrolyte's actual behaviour under realistic operating conditions. Consequently, the crux of achieving a genuine breakthrough lies not in tackling each bottleneck in isolation, but in establishing an integrated research paradigm that spans from mechanistic elucidation through materials design to manufacturing engineering—and this will be the central focus of the next chapter.

7. BREAKTHROUGH PATHWAYS AND DEVELOPMENT DIRECTIONS

Building upon the analysis of the four core bottlenecks delineated in Chapter 6, this chapter outlines the prospective breakthrough pathways for PVA/PAA/PAM-based hydrogel electrolytes across three interconnected dimensions—fundamental understanding, materials innovation, and engineering manufacturing—which collectively form a progressive closed-loop paradigm in which mechanistic insight guides design, design informs manufacturing, and manufacturing, in turn, provides feedback to validate and refine the underlying cognition.

Deepened mechanistic understanding serves as the prerequisite for any rational design, and the most urgent task at present is to develop multi-scale in-situ characterisation methods adapted to aqueous operating conditions—enabling researchers to directly trace Zn^{2+} transport pathways, solvation-structure evolution, and the initial events of dendritic nucleation within realistic electrolyte environments. The combination of first-principles calculations, molecular dynamics, and phase-field simulations holds promise for elucidating, at the atomic scale, the regulatory effects of different functional groups on Zn^{2+} adsorption energy, migration barriers, and interfacial affinity [10], while also reconstructing the spatiotemporal evolution of the deposition front at the mesoscopic level [37]. Yet simulation results must be tightly coupled with in-situ experimental data for closed-loop validation: computation alone cannot capture the full complexity of real systems, whereas experiments alone struggle to access atomic-scale details. Although techniques such as operando TEM currently suffer from deviations from realistic working conditions, the development trajectory is clear—chip-based sample holders and ultrathin liquid-phase windows are being refined to enable observations that progressively approximate true aqueous environments [12]. Once direct visualisation is achieved, the field can move beyond the current paradigm that relies on ex-situ characterisation and inference, providing an empirical foundation—rather than conjectural conclusions—for the structural design of electrolytes.

Transitioning from the current "static optimisation" paradigm to a "dynamic adaptation" framework represents the next core challenge in material design. At present, the design logic for PVA/PAA/PAM-based electrolytes remains largely static: researchers optimise a fixed formulation and expect it to deliver sustained performance throughout the entire battery lifecycle. Real operating

conditions, however, are inherently dynamic—temperature fluctuations, mechanical deformation, and local ionic concentration gradients continually shift the electrolyte's "optimal state". The design philosophy for next-generation electrolytes should therefore embrace dynamic adaptation, whereby the electrolyte senses environmental changes and actively adjusts its internal state to preserve stable performance. Temperature-adaptive electrolytes, for instance, have already demonstrated the viability of this concept by lowering transport resistance at low temperatures while mitigating excessive water loss at elevated temperatures through dynamic bonds and tunable solvation structures [27]. Self-healing functionality represents another crucial facet of dynamic adaptation; the thermal healing technology developed by Yang et al. has proven that interfacial damage can be actively repaired [26], yet the next horizon is to achieve intrinsic self-healing that operates autonomously at room temperature without any external trigger. In terms of interfacial affinity, the coordination capability of PAA's carboxyl groups has been confirmed [15], but a quantitative relationship linking their density, spatial distribution, and coordination strength has yet to be established. Future efforts should focus on the precise molecular-level orchestration of functional groups—such that the structure facilitates rapid Zn^{2+} desolvation while avoiding overly strong binding that impedes ion migration, approaching the ideal state of moderate coordination strength, uniform distribution, and dynamic reversibility. Furthermore, compounding the PVA/PAA/PAM system with fluoropolymers, ionic liquids, or nanofillers such as MOFs and COFs holds promise for achieving a better balance among water retention, electrochemical stability, and Zn^{2+} transference number [11].

The transition from laboratory validation to large-scale production confronts formidable manufacturing-science challenges, even when mechanistic understanding and material design have advanced considerably. The crux of this transition lies in establishing a quantitative relationship model linking formulation, process, and performance. Zhang Wei et al. have pointed out that a new balance must be struck between formulation design and process manufacturability: overly complex multi-step procedures are difficult to scale, whereas excessive simplification tends to compromise ionic conductivity and mechanical integrity [1]. Potential pathways to resolve this contradiction include developing one-step crosslinking systems suitable for continuous production, introducing self-assembly strategies to simplify film formation, and leveraging machine learning to optimise process parameters. Environmental adaptability represents another rigid threshold; Zhu et al. have emphasised that pursuing high conductivity under ideal laboratory conditions while neglecting the practical operational window frequently leads to rapid failure in real-world applications [25]. Duan et al. achieved passive tolerance under extreme temperatures through ice-crystallisation inhibition [29], yet the progression from passive tolerance to active response still demands material-level breakthroughs—for instance, incorporating low-freezing-point solvents or deep-eutectic systems to broaden the liquid-phase operating temperature range. Moreover, although PVA, PAA, and PAM are bulk chemical raw materials, subsequent modification and crosslinking steps can substantially elevate manufacturing costs. Future engineering research should therefore seek a judicious balance between performance enhancement and process simplification, while striving for compatibility with existing industrial production lines wherever possible.

Overall, the development of PVA/PAA/PAM-based hydrogel electrolytes calls for the establishment of a closed-loop along the causal chain of cognition, design, and manufacturing. Critically, the breakthroughs in these three dimensions are temporally sequential: only after in-situ characterisation clarifies the underlying mechanism can it guide the precise design of materials; and only once material design reaches maturity can engineering development obtain feasible input conditions. Accordingly, in the short term, priority should be given to developing operando characterisation methods; in the medium term, efforts should concentrate on intelligent and self-healing materials; and in the long term, a manufacturing-oriented integrated formulation-process optimisation system must be put in place. Once this closed loop is realised, it will not only accelerate the maturation of the PVA/PAA/PAM system but also offer a transferable methodological framework for designing other flexible energy-storage electrolytes, ultimately delivering safe and sustainable energy solutions for wearable electronics, smart textiles, and flexible sensing.

8. CONCLUSIONS

This review has systematically delineated the critical pathway by which PVA/PAA/PAM-based hydrogel electrolytes govern ionic conduction and zinc-dendrite suppression in flexible zinc batteries, revealing that the underlying performance imbalance of single-component systems stems from an inherent structural conflict among ionic conductivity, mechanical robustness, and dendrite-inhibition capability. Critically, the performance bottleneck of flexible aqueous zinc batteries is not solely attributable to the ionic conductivity of any single material, but rather to the long-term misalignment among internal ion migration, interfacial reactions, and mechanical stability within the electrolyte. PVA contributes a robust framework with excellent film-forming ability and mechanical toughness, providing deformation support for the gel system; PAA, leveraging its abundant polar groups, enhances Zn^{2+} affinity and solvation-regulation capability, thereby homogenising ionic distribution; and PAM, through its superior water retention and segmental flexibility, improves electrolyte–electrode interfacial compatibility and mitigates local concentration polarisation. The synergy of the three polymers is by no means a mere superposition—instead, through network complementarity, functional-group cooperation, and interfacial coupling, they collectively construct an integrated system that simultaneously delivers high conductivity, sound mechanical strength, and outstanding dendrite resistance. It can therefore be concluded that future electrolyte design for flexible zinc batteries should adhere to the tripartite principle of "physical channel confinement, chemical coordination homogenisation, and interfacial electric-field regulation": enhancing migration efficiency through structural constraints, stabilising zinc deposition via coordination equilibrium, and inhibiting uncontrolled growth through interfacial modulation—thereby advancing flexible zinc batteries from material-level optimisation toward systematic maturation.

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