



# The road ahead for hydrogel electrolytes in aqueous zinc metal batteries

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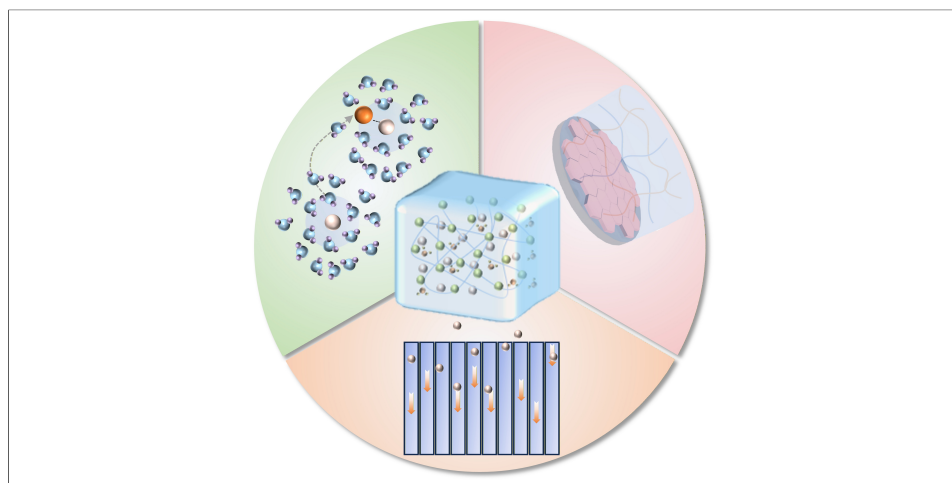
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## Abstract

Hydrogel electrolytes, enabled by tunable polymer networks, mechanical robustness, and water confinement, provide a promising platform for stabilizing Zn metal anodes in aqueous zinc metal batteries. Progress can be broadly viewed through three coupled modules: microenvironment control to regulate water activity and  $\text{Zn}^{2+}$  solvation, bulk transport engineering to sustain selective  $\text{Zn}^{2+}$  delivery, and interfacial design to tune desolvation, nucleation, and growth. Our perspective is that the central challenge is not the independent optimization of these modules, but the rate mismatch that can emerge among them under practical conditions: suppressing water activity alone may not sustain  $\text{Zn}^{2+}$  supply, improving bulk transport alone may not resolve interfacial kinetic barriers, and interfacial stabilization alone may remain ineffective if local ion delivery is insufficient. As a result, the practical operating window is often governed by how well these three processes are temporally and spatially coordinated, especially at high current densities and areal capacities. Additionally, scalable fabrication, full-cell architectures that accommodate electrode asymmetry, and sealed-cell management of gas evolution and pressure are needed.



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Aqueous zinc metal batteries (AZMBs) have attracted widespread attention in various energy storage applications due to their intrinsic high safety, low cost, and the high theoretical gravimetric capacity of zinc metal ( $820 \text{ mAh g}^{-1}$ )<sup>[1-3]</sup>. However, zinc anodes in aqueous environments undergo coupled failure pathways, including dendritic growth, hydrogen evolution, and corrosion. These processes continuously consume electrolyte and increase polarization, which sharply limits the stable operating range in terms of both current density and areal capacity<sup>[4]</sup>. For deformable and wearable devices, conventional separators and liquid electrolytes also suffer from interfacial contact degradation, insufficient mechanical stability, and leakage risk<sup>[5]</sup>. A central challenge is therefore to realize safe, stable, and controllable  $\text{Zn}^{2+}$  transport and deposition under aqueous conditions.

Hydrogel electrolytes (HEs) provide a platform to reshape electrochemical behavior in AZMBs through structural design and chemical tunability<sup>[6-7]</sup>. Their three-dimensional polymer networks enable quasi-solid operation while simultaneously serving as both separator and electrolyte, maintaining continuous ionic pathways and stable interfacial contact at the device scale<sup>[8]</sup>. Functional groups and additives in the network can regulate water coordination and the local  $\text{Zn}^{2+}$  solvation structure, making water chemistry a design variable rather than a passive outcome of the bulk electrolyte<sup>[9-11]</sup>.

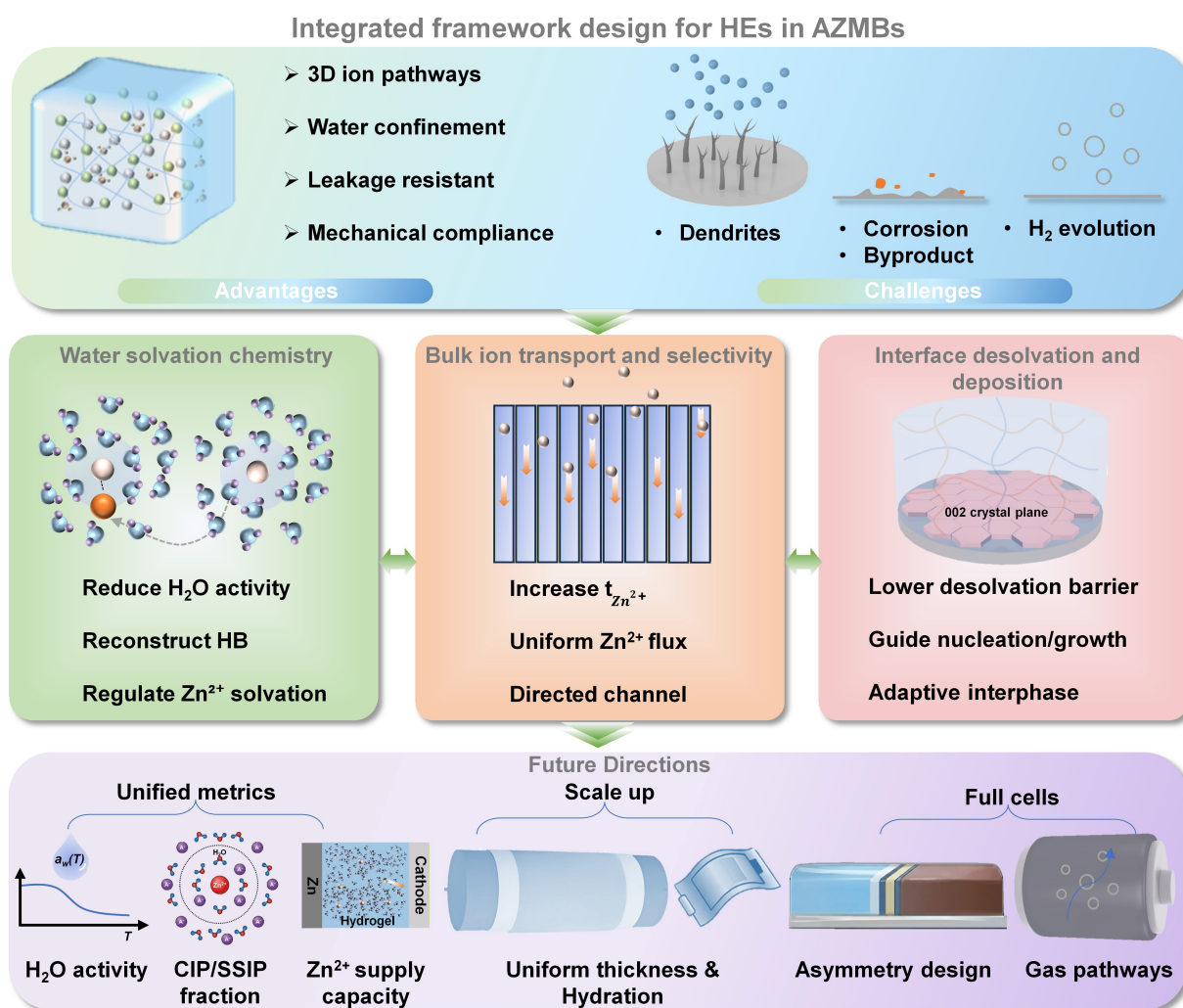
Under practical conditions of high current density and high areal capacity, however, intrinsic constraints remain. Compared with liquid electrolytes, effective  $\text{Zn}^{2+}$  transport in HEs is highly sensitive to pore connectivity, tortuosity, and the swelling state<sup>[12]</sup>. Sustained flux demands can therefore lead to stronger concentration polarization, interfacial  $\text{Zn}^{2+}$  depletion, and accelerated water-driven side reactions<sup>[10,13]</sup>. Microstructural and chemical heterogeneity in the network can further produce local flux variations and destabilize deposition morphology. Residual free water can still participate in hydrogen evolution, corrosion, and byproduct formation, thereby hindering the development of a stable and self-consistent interphase. These observations indicate that failure rarely reflects a single missing property, but instead arises from coupled constraints among water chemistry, transport, and interfacial reactions.

In this perspective, we analyze the interdependence among  $\text{Zn}^{2+}$  solvation chemistry, selective bulk transport, and interfacial desolvation and deposition. We discuss how these coupled processes define a sustainable operating window for AZMBs. We then summarize recent progress in three corresponding design dimensions. First, microenvironment regulation aims to reduce free-water activity and reshape the solvation structure. Second, transport regulation seeks to increase the  $\text{Zn}^{2+}$  transference number ( $t_{\text{Zn}^{2+}}$ ), weaken ion pairing, and optimize transport pathway geometry. These modifications are expected to enhance effective  $\text{Zn}^{2+}$  flux and reduce polarization. Third, interfacial design aims to promote uniform nucleation and improve long-term contact stability. Finally, we outline priorities for scale-up, including achieving spatially uniform fabrication in terms of thickness and hydration, developing structural solutions for anode-cathode asymmetry in full cells, and implementing engineering controls for gas management and pressure evolution in sealed devices. The overall framework is illustrated in [Figure 1](#).

## PROGRESS IN HYDROGEL ELECTROLYTES

### Regulating the water network and solvation structure

In AZMB systems, free water activity, hydrogen-bond (HB) network connectivity, and the number of water molecules in the primary  $\text{Zn}^{2+}$  solvation sheath jointly govern the kinetics of hydrogen evolution, corrosion, and byproduct formation. HEs leverage polymer networks and functional components to tune water speciation and the local coordination environment of  $\text{Zn}^{2+}$ , thereby mitigating water-driven side reactions<sup>[14]</sup>.



**Figure 1.** Integrated framework design for HEs in AZMBs. HB: Hydrogen-bond; CIP: contact ion pair; SSIP: solvent-separated ion pair; HE: hydrogel electrolyte; AZMB: aqueous zinc metal battery.

A widely adopted strategy is to reconstruct the HB network through multipoint interactions, decreasing the fraction of free water and lowering bulk water activity. Polyhydroxy molecules and other multidentate hydrogen-bond donors can simultaneously engage water molecules and polymer chains. For instance, the introduction of sucrose into polyacrylamide-based hydrogels simultaneously regulates the HB network and the Zn<sup>2+</sup> solvation structure, enabling Zn||Zn symmetric cells to operate stably for over 3,970 h at 0.5 mA cm<sup>-2</sup> and -10 °C. The corresponding Zn||VO<sub>2</sub> full cell also exhibits excellent durability, sustaining 10,000 cycles at 1 A g<sup>-1</sup> even at 0 °C<sup>[15]</sup>. However, it is important to note that excessive immobilization of water or an overly compact polymer network can reduce the concentration of mobile charge carriers, increase viscous resistance, and generate locally starved Zn<sup>2+</sup> microenvironments<sup>[16]</sup>. Moreover, spatially nonuniform microenvironments can induce local flux heterogeneity, exacerbating polarization under high-rate conditions<sup>[17]</sup>.

A second approach targets the Zn<sup>2+</sup> solvation sheath directly by introducing functional groups or molecules that compete with water in a controlled manner, partially replacing water molecules within the first solvation shell<sup>[18]</sup>. Cosolvents and urea-based molecules can coordinate with Zn<sup>2+</sup> while simultaneously tuning gel swelling through hydrogen bonding. For instance, tetramethylurea incorporated into polyvinyl alcohol or

polyacrylamide gels can lower water activity and adjust solvation composition, contributing to improved corrosion resistance and cycling stability at elevated temperatures<sup>[9,19]</sup>. In zwitterionic polyelectrolyte gels, increasing salt concentration can reshape solvation through changes in ionic strength and preferential ion–polymer interactions<sup>[20]</sup>.

Overall, microenvironment engineering aims to lower water activity while optimizing  $\text{Zn}^{2+}$  solvation. In our view, its effectiveness should be evaluated not only by its ability to suppress water-driven side reactions, but also by whether it preserves sufficient and spatially uniform  $\text{Zn}^{2+}$  supply. Otherwise, reducing water activity alone may simply shift the bottleneck from parasitic chemistry to transport limitations under high-rate or high-loading conditions<sup>[21]</sup>. We argue that microenvironment engineering remains limited in practice by the lack of standardized protocols for quantifying water activity and solvation structure under operating, rather than equilibrium, conditions.

### Selective bulk ion transport and decoupling

In HEs, bulk ion transport is not fully captured by ionic conductivity alone. It is also governed by  $t_{\text{Zn}^{2+}}$ , the ion association state, and the connectivity and tortuosity of transport pathways<sup>[22–27]</sup>. Low  $t_{\text{Zn}^{2+}}$  (or dominant anion transport) can result in the rapid formation of concentration gradients under an applied current, leading to severe concentration polarization<sup>[28,29]</sup>. This can cause interfacial  $\text{Zn}^{2+}$  depletion, higher local overpotential, and nonuniform nucleation, which may in turn promote dendritic growth and accelerated side reactions<sup>[22,30]</sup>. Transport regulation, therefore, targets a higher effective  $\text{Zn}^{2+}$  flux, reduced anion contribution, and lower tortuosity, so that ion supply remains more uniform.

Hydrogels with polyanionic backbones or anion binding sites immobilize anions through electrostatic interactions, increasing  $t_{\text{Zn}^{2+}}$  and alleviating concentration polarization. A sulfonate-based polyanionic hydrogel ( $t_{\text{Zn}^{2+}} = 0.79$ ) reshapes the  $\text{Zn}^{2+}$  solvation shell and regulates the interfacial potential distribution, enabling stable  $\text{Zn}||\text{Zn}$  cycling for over 1,100 h at 5 mA cm<sup>-2</sup><sup>[22]</sup>. An anion-philic design combining weak cation-dipole coordination with strong anion adsorption ( $t_{\text{Zn}^{2+}} = 0.71$ ) further supports pouch cell operation at 4.9 mAh cm<sup>-2</sup> over 270 cycles<sup>[31]</sup>. Regulating the microenvironment to weaken  $\text{Zn}^{2+}$ -counterion pairing can thus enhance the mobile  $\text{Zn}^{2+}$  fraction and flux responsiveness. Meanwhile, the incorporation of ordered porous fillers or oriented channels can lower tortuosity and promote uniformity, collectively contributing to a more stable ion supply at high areal capacities and current densities<sup>[23,32,33]</sup>.

Enhanced bulk transport, however, does not necessarily translate into interfacial stability<sup>[34]</sup>. Interfacial reactions and morphology evolution are highly localized and can be amplified by electrode roughness and reaction heterogeneity<sup>[35]</sup>. Even with an enhanced ion supply, desolvation, nucleation, and growth can still become the dominant lifetime limiting steps. This motivates direct regulation of interfacial processes. In our view, meaningful progress requires a standardized framework that reports  $t_{\text{Zn}^{2+}}$ , ionic conductivity, and concentration polarization together under identical operating conditions, enabling identification of the true rate-limiting transport parameter.

### Interfacial desolvation and deposition regulation

After  $\text{Zn}^{2+}$  reaches the electrode surface, it undergoes desolvation, nucleation, and growth. Even when bulk transport is improved, a high desolvation barrier, elevated local water activity, or a nonuniform interfacial ion supply can still lead to heterogeneous nucleation and unstable deposition. This is often accompanied by the thickening of byproduct layers and rising polarization<sup>[36]</sup>.

Interfacial regulation generally follows two complementary routes. One is to build structure-guided interfaces that promote uniform nucleation and controlled growth. For example, introducing silk fibroin into

hydrogels can yield an ordered,  $\beta$ -sheet-rich network via solvent-induced assembly. Zincophilic polar groups in this network can tune  $\text{Zn}^{2+}$  solvation and promote preferential growth along the (002) plane, thereby reducing dendrite formation and side reactions<sup>[6]</sup>.

The second route is to construct dynamic, adaptive interfaces that preserve interfacial contact stability during cycling. For instance, a zinc alginate hydrogel interlayer uses zincophilic carboxylate groups and a negatively charged network to guide  $\text{Zn}^{2+}$  transport, repel  $\text{SO}_4^{2-}$ , and facilitate interfacial desolvation. Meanwhile, its dynamic flexibility accommodates volume changes and repairs microcracks during cycling, sustaining stable Zn deposition for over 2,200 h without dendrite or byproduct formation<sup>[37]</sup>. These examples highlight that bulk ion supply and interfacial desolvation kinetics must be co-designed, yet we note that desolvation barriers and their dependence on local water activity remain poorly quantified relative to the field's focus on dendrite morphology.

## OUTLOOK

Based on the above discussion, we suggest that the next stage of HE design should move beyond the isolated optimization of individual properties. A key challenge is the emergence of rate mismatches among water regulation, bulk  $\text{Zn}^{2+}$  delivery, and interfacial reaction kinetics, particularly under practically relevant current densities and areal capacities. The resulting concentration polarization can elevate local overpotential and may locally increase water activity, potentially accelerating hydrogen evolution, corrosion, and byproduct accumulation. Approaches that mainly immobilize water or boost apparent conductivity may therefore be counterproductive if they reduce the density of mobile charge carriers, lower  $t_{\text{Zn}^{2+}}$ , or increase spatial heterogeneity.

Progress will benefit from a unified, quantitative evaluation framework that links chemistry, transport, and interfaces. At a minimum, studies should report water activity, descriptors of  $\text{Zn}^{2+}$  solvation and ion pairing, and the effective  $\text{Zn}^{2+}$  flux under practical loads. Ion association can be quantified as the fraction of contact ion pairs (CIPs) versus solvent-separated ion pairs (SSIPs), extracted using a defined protocol [e.g., Raman or infrared (IR) spectral deconvolution, or molecular dynamics (MD) statistics]. Flux should be benchmarked together with concentration polarization and interfacial  $\text{Zn}^{2+}$  depletion, rather than inferred from conductivity alone. Interfacial assessment should capture desolvation kinetics, nucleation uniformity, and the evolution of impedance and byproduct layers during cycling. Such reporting can identify the rate-limiting process for a given design and guide rate matching across the three coupled domains.

Translation to practical applications also introduces constraints that remain underdiscussed. Scalable manufacturing will likely require spatially uniform thickness, hydration, and crosslink density across large areas. Full cells impose anode-cathode asymmetry, so hydrogel designs should be validated under realistic electrode loadings and capacity ratios, where cathode compatibility, interfacial contact, and water balance may dominate failure. Sealed devices further convert water and gas dynamics into engineering boundary conditions. Long-term stability is expected to depend on providing controlled pathways and buffering volume for gas, and on monitoring or limiting pressure buildup. If these engineering factors are overlooked, it may lead to significant spatial performance variations across devices, premature battery failure under actual electrode loadings, and unsafe pressure buildup caused by uncontrolled gas evolution. All these issues will hinder the reliable translation of laboratory achievements into practical AZMBs. Addressing these coupled materials and engineering challenges within a shared quantitative framework will be essential for advancing HEs from laboratory demonstrations toward robust, deployable AZMBs.

## DECLARATIONS

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Supervision: Zeng, X.; Liu, S.

Critical revision of the manuscript: Zhang, Y.; Yang, B.; Li, W.; Zhu, H.

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Not applicable.

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### Conflicts of interest

All authors declare that there are no conflicts of interest.

### Ethical approval and consent to participate

Not applicable.

### Consent for publication

Not applicable.

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