

Ionic Polymers in Aqueous Zinc–Iodine Batteries

Qian Chen^a, Jonas Wilhelm^b, Jean-François Gohy^c, Jiajia Huang^{a*}, and Feili Lai^{d*}^a School of Chemical Engineering, Zhengzhou University, Zhengzhou 450001, China^b Faculty of Mechanical Engineering, RWTH Aachen University, Aachen 52062, Germany^c Institute for Condensed Matter and Nanosciences (IMCN), Bio- and Soft Matter (BSMA), Université Catholique de Louvain (UCL), Louvain-la-Neuve 1348, Belgium^d Shanghai Key Laboratory of Atomic-Level Intelligent Manufacturing of Materials and Devices, State Key Laboratory of Metal Matrix Composites, School of Materials Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, China

Abstract Aqueous zinc–iodine batteries (AZIBs) have attracted considerable interest for large-scale energy storage owing to their high safety, low cost, and abundant resources. However, their further development is still limited by several key issues, including polyiodide shuttling, sluggish iodine-conversion kinetics, and unstable Zn anode interfaces. Ionic polymers, which combine charged functional sites, continuous macromolecular frameworks, and tunable transport channels, have emerged as promising materials for regulating these coupled processes. In this feature article, recent advances in ionic polymers for AZIBs are systematically summarized from three perspectives: iodine-cathode hosts, electrolyte and separator ion-transport regulation, and zinc-anode protection. Particular attention is given to the underlying mechanisms and structural design principles related to polyiodide confinement, Zn²⁺ transport regulation, interfacial desolvation, and reaction environment optimization. Based on this, the evolution from physical confinement to charge engineering is highlighted, and the remaining challenges and future opportunities are briefly discussed. This feature article aims to provide useful guidance for the rational design of ionic polymers for high-performance AZIBs.

Keywords Ionic polymers; Aqueous zinc–iodine batteries; Polyiodide shuttling; Ion-transport regulation; Zinc-anode protection

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1 INTRODUCTION

Energy has long been the cornerstone of the advancement of human civilization. In the context of carbon neutrality, the rapid

development of renewable energy technologies has accelerated the transition of energy systems to cleaner and more sustainable forms.^[1–3] Renewable energy sources such as solar,^[4–6] tidal,^[7–9] and wind energy^[10–12] have attracted increasing attention because of their abundance, cleanliness, and sustainability. However, their intermittent and stochastic nature creates substantial challenges in grid integration.^[13,14] Therefore, large-scale renewable energy utilization requires safe, flexible, cost-effective, and efficient energy-storage systems. Battery energy storage systems (BESS) can effectively enhance grid flexibility through rapid charge and discharge processes, particularly in systems that integrate diverse renewable energy sources.^[15–18] Lithium-ion batteries (LIBs) have dominated the current BESS market because of their high energy density and long cycle life,^[19,20] while sodium-ion batteries (SIBs) have emerged as low-cost alternatives.^[21] Nevertheless, both LIBs and SIBs still face challenges related to safety risks,^[22–24] environmental contamination,^[25–27] and the complex manufacturing processes associated with organic electrolytes.^[23,28] Therefore, developing safer, more economical, and more environmentally friendly aqueous battery systems remains highly desirable. Aqueous zinc-iodine batteries (AZIBs) have emerged as one of the most promising energy storage systems owing to the high theoretical capacity of the Zn anode, including a gravimetric capacity of 820 mAh·g^{−1} and a volumetric capacity of 5855 mAh·cm^{−3}, as well as the intrinsic safety of aqueous electrolytes.^[29,30] Among the vari-

* Corresponding authors, E-mail: huangjiajia@zzu.edu.cn (J.H.)
E-mail: feililai@sjtu.edu.cn (F.L.)

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ous cathode materials, iodine has attracted extensive research interest owing to its high theoretical specific capacity of 211 mAh·g⁻¹ and favorable iodine redox chemistry in aqueous electrolytes.^[31,32] However, the commercialization of AZIBs is hindered by three coupled bottlenecks: active material shuttling, sluggish solid–liquid conversion kinetics, and dendritic growth on the anode side.^[33]

To address these bottlenecks, early research efforts primarily focused on porous hosts and ion-sieving membranes, such as porous carbon^[34] and metal-organic framework (MOF)-based ionic-sieve membranes,^[35] to restrict polyiodide migration. However, purely physical confinement strategies have intrinsic limitations. On the one hand, polyiodide species are strongly solvated in aqueous media, enabling their continuous migration under concentration gradients. Physical adsorption dominated by van der Waals interactions is thermodynamically insufficient to fully counteract the solvation-driven diffusion. On the other hand, a structural mismatch exists between the rigid inorganic frameworks and the dynamic volume evolution of iodine species during the charge-discharge processes. Such mechanical incompatibility often results in interfacial degradation and progressive capacity fading during prolonged cycling. Although “passive blocking” strategies can provide short-term improvements, they rarely ensure long-term stability. These limitations have stimulated a paradigm shift from geometric confinement to interfacial charge regulation and solvation engineering. In the broader context of aqueous zinc batteries, polymer electrolytes have been demonstrated to suppress hydrogen evolution, reduce free water activity, and mitigate dendrite growth, thereby enhancing electrochemical stability. Ionic polymers are capable of modulating Zn²⁺ migration and deposition behavior through coordination interactions, electrostatic repulsion, and ion-selective transport mechanisms. In aqueous zinc-iodine batteries, polymer networks equipped with polar or ionic functional groups can chemically anchor polyiodide species while simultaneously buffering the volume fluctuations induced by iodine conversion and improving interfacial compatibility. More importantly, by designing polymeric frameworks with selective ion-transport capabilities, it is possible to suppress polyiodide migration in a directional manner while promoting efficient Zn²⁺ transport. Such dual-regulation functionality enables simultaneous optimization of mass transport and electrochemical stability within an integrated material system. Compared to conventional pore-structure engineering, ionic polymers introduce a different regulatory

framework that can be understood as distributed electrostatic field modulation. Through fixed ionic sites and dynamic chain motion, these macromolecular systems establish tunable local electric fields and solvation environments, enabling the active regulation of polyiodide speciation and Zn²⁺ flux. In this context, “polymer charge engineering” denotes the rational regulation of charged functional groups and the local charge distribution within polymer frameworks to modulate ion transport and interfacial reactions. Accordingly, “distributed electrostatic field modulation” and “reaction-environment regulation” refer to the ability of ionic polymers to reconstruct the local electric fields, solvation environments, and interfacial reaction conditions in AZIBs.

In this feature article, ionic polymers are defined as macromolecular materials containing fixed or mobile ionic groups within polymer chains, crosslinked networks, membranes, hydrogels, or ordered porous frameworks. The ionic polymer systems discussed herein mainly include linear polyelectrolytes, ion-exchange resins, polyionic liquids, charged hydrogels, polymer membranes, and porous ionic macromolecular frameworks. Ionic polymers provide a versatile platform for regulating polyiodide transport, Zn²⁺ migration, and interfacial stability in AZIBs. Their flexible chains, abundant polar/ionic groups, and tunable frameworks enable molecular-level control of polyiodide migration and interfacial ion transport. It should also be noted that electrostatic interactions are not the only origin of ionic polymer regulation. Depending on the polymer structure and working location, hydrogen bonding, Zn²⁺ coordination, hydrophobic effects, pore confinement, and local solvation regulation may also contribute to polyiodide stabilization, ion-selective transport, water activity reduction, and interfacial desolvation. To provide a clearer overview of the material scope and regulatory functions of ionic polymers in AZIBs, representative ionic polymer strategies are summarized in Table 1. Our research group has shifted from traditional physical confinement strategies to a polymer charge engineering approach for the design of high-performance ionic polymers. Such ionic polymers can stabilize polyiodide species and inhibit undesirable side reactions. This feature article summarizes the applications of ionic polymers in AZIBs, drawing on our studies of cathode regulation, electrolyte regulation, and anode protection. Accordingly, this feature article describes the coupled migration and interfacial evolution of iodine species and Zn²⁺ across the cathode, electrolyte/separator, and anode, emphasizing how ionic polymers regulate these interconnected processes at the system level.

Table 1 Representative ionic polymer regulation strategies in aqueous zinc–iodine batteries.

Polymer strategy	Working location	Main structural feature	Main regulation role
Cationic polymer hosts	Iodine cathode	Fixed cationic sites	Anchor polyiodides and regulate iodine conversion
Conductive/ionic polymer hosts	Iodine cathode	Conjugated backbones with polar/ionic sites	Improve charge transfer and iodine utilization
Polyanionic separators or interfacial layers	Separator/anode-facing interface	Negatively charged polymer networks	Block polyiodides and homogenize Zn ²⁺ flux
Polymer additives and gel electrolytes	Electrolyte	Polar chains or charged networks	Regulate solvation and stabilize iodine species
Polymeric zinc-anode coatings	Zn anode	Polar or ionic functional groups	Promote Zn ²⁺ desolvation and uniform deposition

2 POLYMERIC HOST MATERIALS FOR CATHODE REGULATION

Cathode-side regulation is essential for achieving stable cycling performance in AZIBs, whereas soluble polyiodide species are difficult to retain under prolonged operation. Additionally, the intrinsically low electronic conductivity of iodine further limits its reaction kinetics. In aqueous media, polyiodides are strongly stabilized by solvation and can therefore readily diffuse away from the cathode, resulting in self-discharge, gradual loss of active material, and deteriorated iodine utilization. The iodine redox process is often accompanied by large polarization and sluggish conversion kinetics under practical conditions. These cathode-side instabilities could propagate further through the full cell. Once polyiodides migrate to the Zn anode, they may perturb the interfacial environment, aggravate parasitic reactions, and indirectly worsen the Zn deposition behavior. The resulting interfacial instability, in turn, feeds back into the cathode by increasing the polarization and reducing the reversibility of iodine conversion during long-term cycling. In various cases, the origin of these problems lies in the limited ability of conventional hosts to simultaneously confine soluble iodine species and maintain efficient charge transport. Hosts that rely solely on pore confinement may temporarily retard polyiodide diffusion, but they often fail to provide durable retention because solvated polyiodides remain highly mobile in aqueous electrolytes. Moreover, iodine species undergo continuous changes in form and local volume during charge-discharge processes, which can disrupt the conductive network inside the cathode and hinder charge transfer. These limitations indicate that strategies based solely on physical confinement usually provide limited and short-lived improvements. Polymer-based hosts, particularly ionic polymers, offer distinct advantages. Owing to their fixed-charge groups and tunable framework structures, these materials can establish stronger interactions with polyiodides than those provided by simple physical adsorption while also regulating the local ionic distribution and solvation environment. In addition, certain polymer architectures can facilitate charge transport through conductive segments or continuous ion-rich phases, thereby enabling more controlled iodine conversion. In this section, we focus on the evolution of polymeric hosts from conventional conductive polymers to cationic and ordered ionic macromolecular frameworks for cathode regulation in AZIBs.

2.1 Conventional Polymers

In AZIBs, the iodine cathodes face two major problems. On the

one hand, I_2 has poor intrinsic electronic conductivity, which leads to high charge-transfer resistance and limited rate capability. On the other hand, polyiodide species, such as I_3^- and I_5^- , are highly soluble in aqueous electrolytes, causing severe shuttle effects and resulting in capacity decay, self-discharge, and low Coulombic efficiency.^[36] Conducting polymers are a class of π -conjugated macromolecules whose electronic conductivity can be enhanced through doping. For these hosts, the π -conjugated backbone is the primary structural feature for improving electronic transport, whereas heteroatom-containing units can provide additional interaction sites for iodine and polyiodide species. Representative conducting polymers include poly(3,4-ethylenedioxythiophene) (PEDOT), polypyrrole (PPy), polyaniline (PANI), poly(*p*-phenylene vinylene) (PPV), poly(*p*-phenylene) (PPP), and poly(phenylene sulfide) (PPS) (Fig. 1). Among them, PEDOT-based polymers, PPy, and PANI have been more frequently investigated in aqueous battery systems because of their conjugated backbones, tunable doping states, and relatively high electronic conductivity. Therefore, the following discussion mainly focuses on these representative conductive polymers and their roles in iodine cathode regulation.^[37]

On the cathode side, PPy, PANI, and poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) have been employed as representative conductive polymer hosts to promote iodine redox kinetics and mitigate polyiodide shuttling in AZIBs. Miao *et al.* compared electropolymerized PPy and PANI cathodes for iodine redox reactions in Zn- I_2 batteries (Fig. 2a).^[38] The results showed that the dense PPy layer mainly favored electron transport, whereas the porous PANI layer provided more accessible pathways for electrolyte penetration and iodine species conversion. This comparison indicates that the performance of conductive-polymer iodine cathodes is closely related not only to the intrinsic conductivity of the polymer backbone but also to the polymer morphology, chain packing, and ion/electron transport pathways. PANI is a commonly used conductive polymer. Its electrical conductivity originates from the charge carriers generated after doping, and its chemical structure contains abundant nitrogen-containing sites that can interact with iodine or polyiodide species. In PANI- I_2 composite cathodes, PANI has been demonstrated not only to provide continuous electron transport pathways but also to interact with polyiodides to suppress their dissolution and shuttling, thereby enhancing iodine-redox reversibility and reducing active-material loss

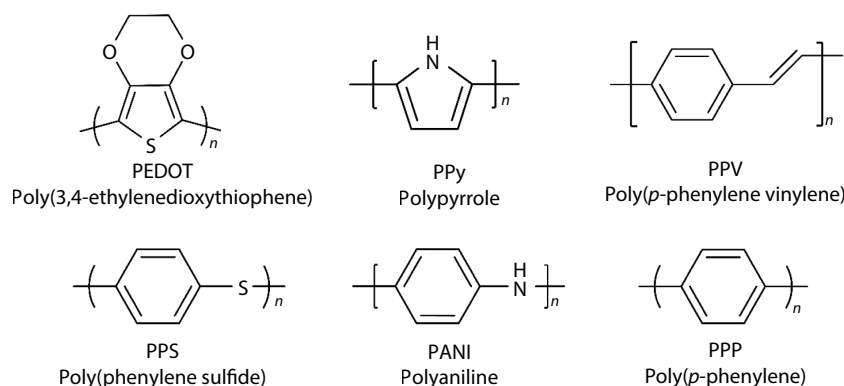


Fig. 1 Molecular structures of representative conducting polymers.

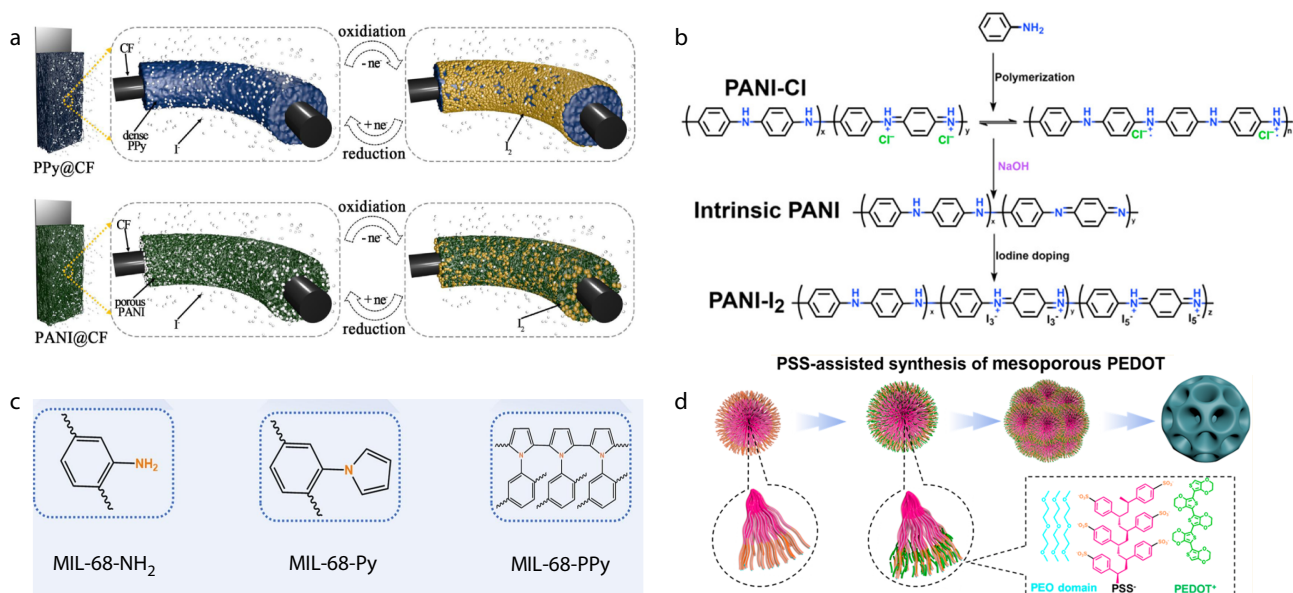


Fig. 2 Representative conventional conductive-polymer cathode hosts for iodine regulation in AZIBs. (a) Electropolymerized PPy and PANI cathodes for iodine redox reactions (Reproduced with permission from Ref. [38]; Copyright (2022), Elsevier); (b) PANI- I_2 composite cathode prepared through a dedoping/redoping strategy (Reproduced with permission from Ref. [39]; Copyright (2020), American Chemical Society); (c) MIL-68-PPy conductive host constructed by an interpenetrating PPy network (Reproduced with permission from Ref. [41]; Copyright (2025), Elsevier); (d) PSS-assisted synthesis of mesoporous PEDOT:PSS as an iodine host (Reproduced with permission from Ref. [42]; Copyright (2023), American Chemical Society).

(Fig. 2b).^[39] Specifically, polyiodide-doped PANI cathodes were prepared through a dedoping/redoping process in which the iodine species mainly existed as I_3^- and I_5^- within the PANI backbone. *Ex situ* Raman and X-ray photoelectron spectroscopy analyses further revealed that the iodine redox process followed an $\text{I}_5^-/\text{I}_3^-/\text{I}^-$ conversion pathway rather than the conventional anion doping/dedoping mechanism of PANI, indicating that PANI can act as both a conductive skeleton and a molecular confinement matrix for iodine species.

PPy is another representative conductive polymer that has relatively high electrical conductivity and chemical robustness. Deng *et al.* reported an ion-selective PPy coating for aqueous zinc-ion battery cathodes, showing that conductive polymer layers can regulate the interfacial charge transport and ion migration.^[40] Owing to its facile *in situ* oxidative polymerization on various substrates, PPy has been widely explored for electrode construction and is generally employed as a conductive coating layer or host framework for iodine cathodes to promote electron transport and stabilize iodine species. The performance of iodine cathodes can be further improved by combining conductive polymers with porous frameworks. For example, PPy-containing porous or MOF-based materials can generate integrated hosts with high surface areas and continuous conductive pathways, which can accelerate the iodine conversion kinetics and improve the rate performance (Fig. 2c).^[41] In the MIL-68-PPy conductive host, the amino groups in MIL-68- NH_2 were converted into pyrrole-containing units, followed by oxidative polymerization to form an interpenetrating PPy network within the MOF framework. This design combines the porous iodine-accommodation capability of the MOF scaffold with the electron-transfer capability of PPy, leading to lower polarization and improved iodine redox kinetics. PEDOT:PSS is one of the most

widely used conductive polymers in aqueous batteries owing to its good processability, flexible film-forming ability, and relatively high conductivity. PEDOT:PSS can also be directly used as an iodine host. Wei *et al.* constructed mesoporous PEDOT:PSS nanospheres loaded with iodine (Fig. 2d).^[42] In this design, iodine species were uniformly dispersed in the polymer framework with improved electron transport and enhanced iodine utilization. The open mesoporous channels improved electrolyte accessibility and exposed more active sites, while the sulfur- and oxygen-containing groups in PEDOT:PSS interacted with I^- , I_2 , and I_3^- species, thereby suppressing polyiodide shuttling. The as-prepared cathode delivered a specific capacity of approximately $241 \text{ mAh}\cdot\text{g}^{-1}$ and maintained stable cycling for 2×10^4 cycles in AZIBs, demonstrating the structural advantages of PEDOT:PSS nanospheres as a conductive polymer host.

Overall, conventional conductive polymers, including PEDOT:PSS, PPy, and PANI, can enhance the charge-transfer kinetics of iodine cathodes by constructing continuous electron-transport pathways and stabilizing the cathode/electrolyte interface. They partially mitigate polyiodide shuttling by coupling conductive networks with heteroatom-assisted adsorption. Nevertheless, their confinement effect on polyiodides still mainly depends on reversible doping, weak adsorption, structural blocking, and limited charge interactions, and thus remains insufficient under high iodine loading and long-term cycling conditions. In aqueous electrolytes, these polymers may undergo chain rearrangement, framework distortion, and progressive interfacial degradation during repeated cycling.^[41,43] Therefore, the key design principle of conventional conductive polymer hosts is to couple the electronic conductivity with heteroatom-assisted iodine adsorption and accessible ion-transport pathways. However, because their

polyiodide confinement mainly relies on weak adsorption, reversible doping, and structural blocking, further improvements require the introduction of stronger and more controllable ionic interactions. Accordingly, recent efforts have increasingly shifted from conductive-network regulation to ionic-site regulation, in which a high density of fixed cationic sites is incorporated into polymer frameworks to strongly anchor polyiodide anions through electrostatic interactions.^[44]

2.2 Cationic Polymer Hosts

To achieve stronger confinement of polyiodides, the design of polymer hosts has gradually evolved from conventional adsorption and physical blocking to interfacial charge regulation based on ionic polymers. Unlike traditional conductive polymers or porous carbon hosts, which mainly depend on weak interactions and pore confinement, ionic polymers can incorporate a high density of fixed cationic sites into the framework, thus establishing a stable and tunable local charge environment at the interface between the iodine cathode and the aqueous electrolyte. In these systems, fixed cationic sites, such as quaternary ammonium or viologen units, are the most important structural motifs because they provide a strong electrostatic affinity toward polyiodide anions. As a result, these materials not only strengthen the electrostatic anchoring of polyiodide anions, such as I_3^- and I_5^- , but also regulate the local ion distribution, iodine conversion pathways, and interfacial charge-transfer kinetics. Along this line, we developed a quaternized *N,N'*-dimethyl-1,3-propanediamine-grafted and triethylenetetramine-crosslinked acrylic fiber/iodine (GC-PAN/I) cathode and proposed a double-layer concept for Zn- I_2 systems (Fig. 3a). In this design, the insoluble fibrous framework acted as a stable cationic scaffold, whereas the introduced quaternary ammonium groups generated a dense electrostatic coupling layer at the cathode surface to effectively constrain polyiodide migration. Notably, this strategy did not sacrifice reaction activity to improve stability. Rather, it suppressed polyiodide shuttling while retaining the solution-phase conversion process dominated by I^-/I_3^- and facilitating ion transport through the one-dimensional framework.^[45] Thus, effective polyiodide confinement and fast reaction kinetics were simultaneously achieved. The kinetic advantage of this quaternized framework was further supported by galvanostatic intermittent titration technique (GITT) measurements and normalized polarization analyses, which revealed a low polarization during charge/discharge. The ion transport and charge storage kinetics were further assessed using cyclic voltammetry (CV)-based kinetic analysis. These results indicate that cationic confinement did not simply block polyiodide migration but also helped maintain efficient ion transport and reaction reversibility. After establishing the electrostatic anchoring mechanism in the fibrous framework, the same strategy was further extended to a commercially available strong-base anion-exchange resin, D296, containing abundant quaternary ammonium groups. By directly soaking D296 in a polyiodide solution, a quaternized resin-D296/iodine (QR-D296/I) cathode was obtained to clarify the relationship between the strong polyiodide confinement and fast ion transport (Fig. 3b). The comparison showed that a denser framework suppressed polyiodide migration more effectively, but also increased the overall resistance to Zn^{2+} transport, leading to inferior rate performance. Nevertheless, this system exhibited more stable cy-

cling behavior during long-term tests. These results indicate that the polyiodide shuttling effect can be significantly suppressed by increasing the density of cationic sites, whereas the upper limit of the rate capability is mainly determined by the pore structure and ion flux. The preferential interaction between the quaternary ammonium sites and the different iodine species is further illustrated by the adsorption energy results in Fig. 3(c).

On this basis, we further recognized that the performance of ionic polymers is not solely governed by the number of cationic sites. The electronic structure surrounding these sites also plays an important role in the interfacial regulation. Thus, we constructed viologen-based dicationic conjugated polymers,^[46] which showed that the design of ionic polymers could be advanced beyond the simple introduction of fixed charges toward the regulation of the electronic structure and local electrostatic potential (Fig. 3d). The dicationic sites can strengthen electrostatic interactions with polyiodide anions, while the extended π -conjugated backbone facilitates electron transport, thereby integrating polyiodide confinement with electronic conduction within the same polymer framework. In other words, this strategy expands the function of ionic polymers from passive electrostatic hosts to reactive frameworks, integrating confinement and charge transport. From a broader perspective of research progress, the application of commercially available polycationic materials further demonstrates the universality of this strategy. For example, poly(quaternary ammonium iodide) (PDDA-I) was employed to construct a localized electric-field microenvironment at the cathode. The fixed quaternary ammonium sites regulate the spatial distribution of charged iodine-related species and promote the enrichment of nucleophilic species near the cathode, thereby stabilizing high-valence iodine intermediates while suppressing polyiodide migration (Fig. 3e).^[47] This result demonstrates that polycationic materials can function not only as electrostatic hosts but also as regulators of the local reaction environment. In a related strategy, poly(diallyldimethylammonium chloride) (PDDA) was employed as a bifunctional quaternary-ammonium binder in iodine-composite cathodes.^[48] Its fixed cationic sites electrostatically immobilize iodine species while the polymeric binder network contributes to electrode integrity and interfacial stability. The computational analysis reported in this work provides a more quantitative explanation of the role of the quaternary ammonium groups. The adsorption-energy comparison between carboxymethyl cellulose (CMC) and PDDA toward I_2 , I^- , and I_3^- shows that PDDA exhibits a stronger affinity toward iodine species, especially polyiodide anions, than the carboxylate-containing CMC binder. Charge-density-difference analysis further indicates charge redistribution between the quaternary ammonium sites and iodine species, suggesting that electrostatic interactions are accompanied by interfacial charge regulation. In addition, the calculated Gibbs free energy changes for consecutive iodine conversion steps indicate that PDDA can influence the thermodynamics of iodine species transformation rather than only immobilizing polyiodides. The PDDA binder not only immobilizes polyiodide anions through fixed cationic sites but also stabilizes the reaction interface through the regulated local microenvironment

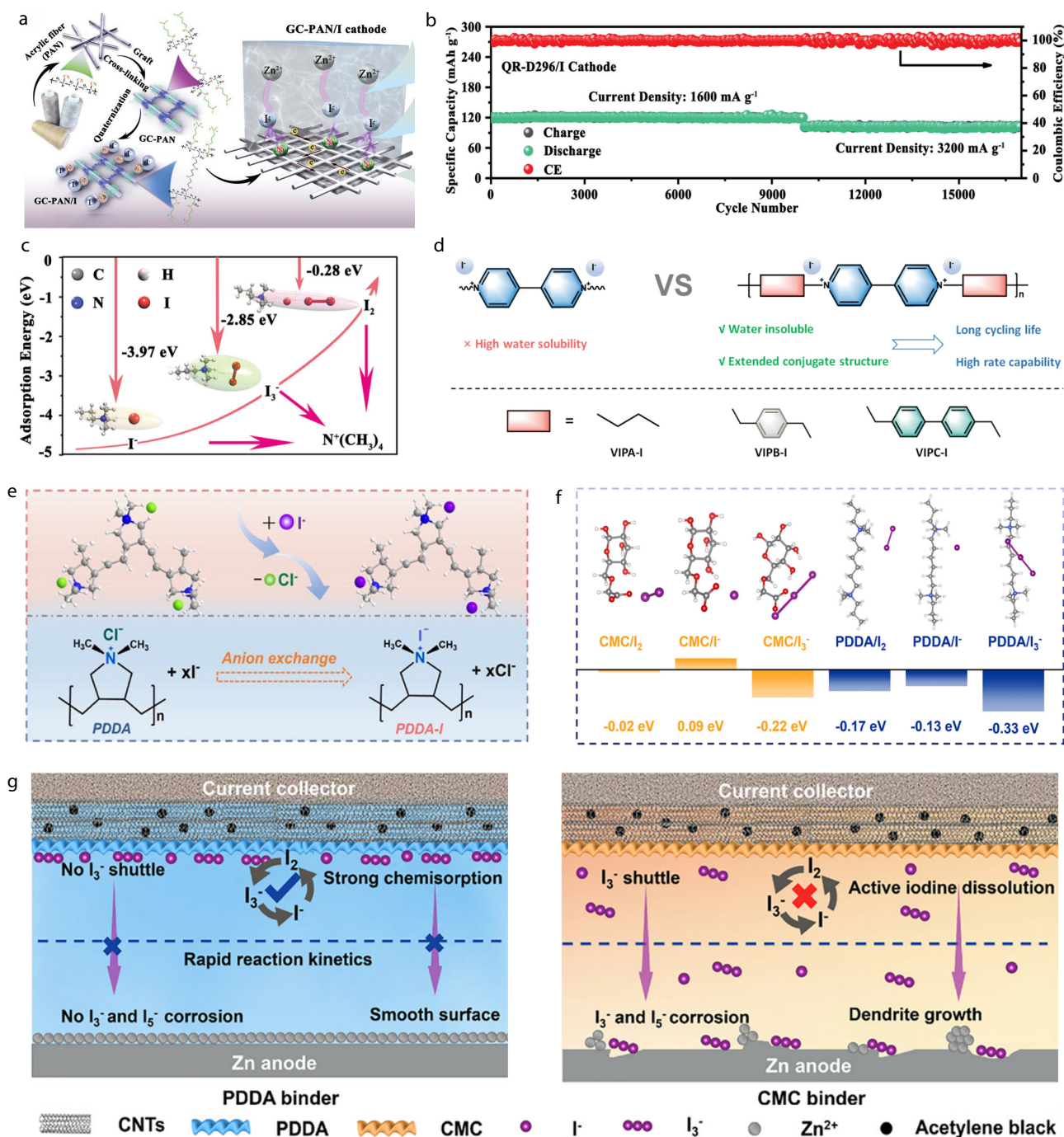


Fig. 3 Representative cationic polymer hosts for iodine-cathode regulation in AZIBs, highlighting fixed cationic sites and local electric fields for polyiodide immobilization. (a) Quaternized acrylic fiber (GC-PAN/I) cathode and its shuttling-suppression mechanism; (b) Cycling performance of QR-D296/I cathode; (c) Electrostatic interaction between polyiodide species and quaternary ammonium sites in the GC-PAN/I cathode; (a–c: reproduced from Ref. [45]; Copyright (2022), The Author(s). Published under a Creative Commons CC BY 4.0 license.) (d) Viologen-based conjugated polymer host (Reproduced with permission from Ref. [46]; Copyright (2025), Elsevier); (e) Localized electric-field microenvironment constructed by PDPA-I for regulating iodine species and stabilizing high-valence iodine intermediates (Reproduced with permission from Ref. [47]; Copyright (2025), Wiley-VCH GmbH); (f) Adsorption-energy comparison of iodine species on CMC and PDPA binders; (g) Comparison of PDPA and CMC binders in suppressing polyiodide shuttling and zinc-anode corrosion. (f, g: reproduced with permission from Ref. [48]; Copyright (2025), Wiley-VCH GmbH.)

and participates in regulating the thermodynamics and interfacial conversion of iodine species (Figs. 3f and 3g). This indicates that the role of ionic polymers in iodine cathodes has

gradually evolved from restricting polyiodide shuttling to actively shaping the interfacial reaction environment. However, these results also suggest that merely strengthening the elec-

trostatic confinement is not always sufficient for optimizing iodine cathodes. Although cationic sites can effectively strengthen the confinement of polyiodide anions, excessively strong interactions may also retard the redox conversion of iodine species. To address this trade-off, the design of ionic polymer hosts has gradually shifted from simple strong electrostatic confinement to coordinated regulation of polyiodide confinement and iodine conversion. Our group developed an ion-exchange-resin-based composite host containing both quaternary ammonium cationic sites and quaternary-ammonium-functionalized cobalt phthalocyanine (QA-CoPc) catalytic centers.^[49] The quaternary ammonium groups on the resin framework mainly provide electrostatic anchoring sites for polyiodide anions, whereas the introduced cobalt phthalocyanine units promote the interfacial redox conversion of iodine species. In this way, the host suppresses polyiodide shuttling while accelerating iodine species conversion. Catalytically active sites were integrated into the ionic polymer host, which stabilized the interfacial microenvironment and alleviated the kinetic sluggishness caused by overly strong host–guest interactions. These results indicate that ionic polymer hosts have evolved into multifunctional platforms that couple electrostatic anchoring, local electric field regulation, and catalytic iodine conversion. Through the cooperative effects of fixed charge sites, local electric fields, and catalytic units, ionic polymer hosts can effectively balance polyiodide confinement, ion transport, and rapid iodine conversion. Despite their strong polyiodide affinity, cationic polymer hosts must balance electrostatic confinement with reversible iodine conversion and efficient Zn^{2+} transport.

2.3 From Porous Crystalline Hosts to Ionic Macromolecular Frameworks

Before discussing the transition from porous crystalline hosts to ionic macromolecular frameworks, several carbon-based, MOF-based, and covalent organic framework (COF)-based iodine hosts are introduced as background and comparison. These materials are not typical ionic polymers but provide useful design references for pore confinement, polar-site recognition, catalytic iodine conversion, and ordered mass transport. Their discussion helps clarify the development from conventional porous confinement toward ionic polymer-based regulation with charged sites and macromolecular frameworks. In the study of cathode materials for AZIBs, the construction of ordered pores and functional adsorption sites has proven to be an effective strategy for suppressing polyiodide shuttling. However, the disordered pore structures of conventional carbon materials and amorphous polymers are often unable to maintain efficient polyiodide anchoring and fast charge transport under high active material loading and long-term cycling conditions. In this context, the role of pore structure is no longer limited to providing space for iodine storage. For porous and crystalline host systems, ordered channels, pore size compatibility, and polar or ionic pore-wall sites are key structural factors that determine polyiodide accommodation, retention, and transport. More importantly, the pore channels, interfacial sites, and electronic structure of the framework are considered together to regulate the confinement and conversion of polyiodide anions. Single-atom catalytic hosts represent an important strategy for integrating physical confinement with catalytic iodine conver-

sion. In a system based on Zn single-atom catalysts encapsulated in an edible-fungus-slag-derived carbon matrix (SAZn@CFS), the biochar framework first provided physical confinement and surface adsorption through its hierarchical porous framework (micropores and mesopores) and N-doped sites.^[50] Meanwhile, the Zn- N_4 -C sites stabilized the I_2 and polyiodide intermediates and promoted the I^-/I_2 conversion process. Therefore, the host participates directly in iodine redox conversion. In single-atom Fe-N-C encapsulated high-surface-area carbon (HC@FeNC) core-shell hosts (Fig. 4a), the high-surface-area carbon core and Fe- N_4 active shell allow the outer layer to participate in iodine conversion directly.^[51] This design can not only suppress the dissolution and diffusion of polyiodide anions but also regulate the reaction pathway among I^- , I_3^- , and I_2 . The bismuth hexacyanoferrate@zeolitic imidazolate framework-8-derived Bi/Zn/Fe multi-site co-doped hierarchical porous carbon (Bi-HCF@ZIF-8-derived BZPC) host prepared by Yang *et al.* further demonstrated the effect of multi-site regulation.^[52] Its hierarchical pore structure enables efficient mass transport and confinement, while the Bi/Zn/Fe multi-metal sites and N doping enhance the conversion kinetics of iodine species and suppress the formation of I_3^- . These results show that material design has moved beyond single-site adsorption and now involves integrated regulation of pore transport, catalytic sites, and interfacial processes in AZIBs. The use of B/N co-doped porous carbon is another important strategy that places more emphasis on electronic structure regulation.^[53] In this case, the key factor is the reconstruction of the local charge distribution and electronic state density by B-N pairs. This modified pore-wall environment can strengthen interactions with polyiodide species and maintain excellent framework conductivity. Thus, polyiodide confinement and conversion kinetics are coupled within the same electronically modulated pore-wall environment. In comparison, crystalline framework materials provide a more refined platform for structural regulation. As a related example beyond Zn- I_2 chemistry, similar barrier-trapping and electrocatalytic concepts have also been demonstrated in Li-S batteries, where redox-active COF frameworks combine physical blocking with catalytic conversion, further supporting the broader value of ordered porous frameworks in shuttle-suppression chemistry.^[54] In the case of the F-modified ZIF, a porous ZIF layer containing $-\text{CF}_3$ functional groups was introduced on the separator side to enhance the interfacial capture of I_3^- . This strategy directly regulates the migration pathways of the polyiodides.^[55] COF materials, such as TpPa- NO_2 , place more emphasis on the intrinsic effect of the framework structure on reaction behavior.^[56] For TpPa- NO_2 , the ordered channels provided well-defined confined spaces, while the NO_2 side groups tuned the electronic structure of the framework, reduced the band gap, enhanced polyiodide adsorption, and improved the charge transfer behavior. As a related example outside battery applications, Zhang *et al.* constructed nitrogen-rich clover-like COFs for iodine capture, further confirming the importance of nitrogen adsorption sites and ordered pore architectures for iodine species retention.^[57] The subsequent development of thiazole-linked covalent organic frameworks (TZ-COFs) further enabled the coordination of higher-valence iodine species and promoted multi-electron iodine conversion processes (Fig. 4b).^[58] Yao *et al.* further demonstrated that quaternary-ammonium-mediated I^+

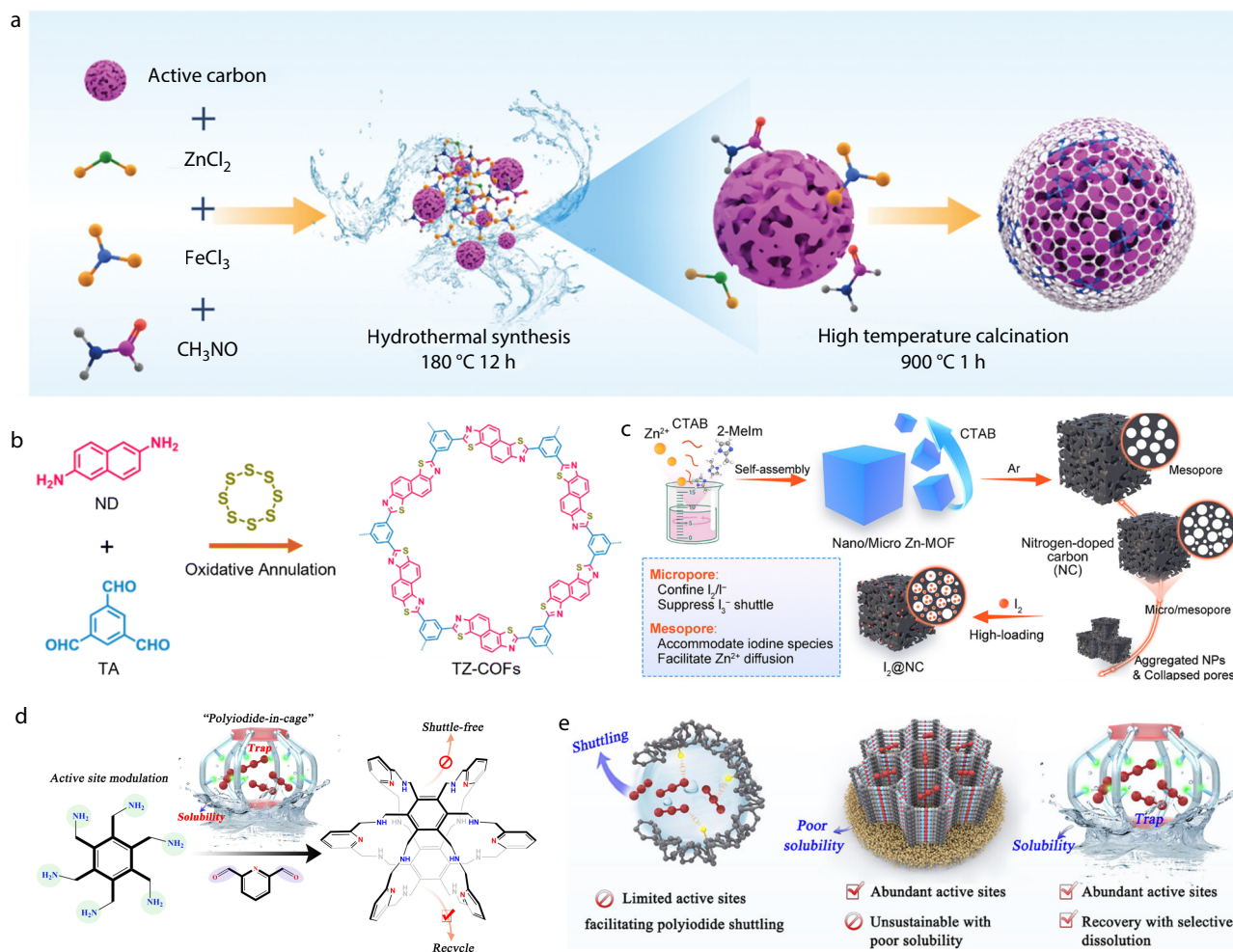


Fig. 4 Representative porous and crystalline host materials for polyiodide storage and transport. (a) HC@FeNC core-shell host with single-atom Fe-N-C catalytic sites (Reproduced with permission from Ref. [51]; Copyright (2024), Wiley-VCH GmbH); (b) Thiazole-linked covalent organic frameworks (TZ-COFs) (Reproduced with permission from Ref. [58]; Copyright (2025), The Royal Society of Chemistry); (c) Zn-MOF-derived N-doped porous carbon host (Reproduced from Ref. [61]; Copyright (2025), The Author(s). Published under a Creative Commons CC BY 4.0 license); (d) Molecularly engineered polyiodide reservoirs; (e) Comparison of carbon-based materials, organic frameworks, and covalent organic cages as polyiodide reservoirs. (d, e: reproduced with permission from Ref. [62]; Copyright (2025), Wiley-VCH GmbH.)

complexation can stabilize high-valence iodine species in four-electron aqueous fiber Zn- I_2 batteries.^[59] Although the $\text{I}^-/\text{I}_2/\text{I}^+$ multielectron pathway can, in principle, increase the theoretical capacity and output voltage of Zn- I_2 batteries, its practical realization remains challenging in aqueous electrolytes. Recent ion-replenishing interlayer and tailored-electrolyte designs further demonstrate that the stabilization of high-valence iodine species requires the coordinated regulation of polyiodide transport, halide supply, and the local electrolyte environment.^[60] In particular, the stabilization of high-valence iodine species, such as I^+ , requires careful regulation of the local reaction environment to suppress hydrolysis, side reactions, and crossover-related instability. Taken together, the advantage of crystalline framework materials lies in the fact that pore channels, functional groups, and framework electronic properties can all be incorporated into molecular-level design for AZIBs. As another representative route, MOF-derived porous carbons combine hierarchical pore structures with adsorption sites containing het-

eroatoms. The Zn-MOF-derived N-doped porous carbon is a representative example (Fig. 4c).^[61] The advantage of this type of material is its hierarchical porous framework with both micropores and mesopores, which can simultaneously accommodate iodine species and facilitate electron and ion transport. In addition, graphitic N and pyridinic N sites convert the relatively inert carbon surface into a polar interface that can effectively recognize polyiodide species. Consequently, the confinement effect arises from the synergistic interplay between pore restriction and nitrogen site anchoring.

On this basis, we proposed porous crystalline ionic macromolecular hosts that integrate pore confinement, site interactions, and framework transport into the same ordered polymer framework (Figs. 4d and 4e). In this system, the molecular hosts provide well-defined internal cavities with sizes of approximately 0.96–1.07 nm, offering suitable confined spaces for accommodating polyiodide species.^[62] This indicates that the insertion, retention, and release of polyiodide

species during cycling occur within a well-defined local environment rather than in disordered or random pores. The confinement effect does not simply isolate polyiodides inside the cavities but further limits their spatial distribution and migration boundaries based on geometric restrictions. Polar or cationic sites on the pore walls enhance this confinement, turning the behavior of polyiodide anions from simple physical adsorption to a reversible capture-and-release process with site-recognition features. This site-recognition behavior is supported by adsorption-energy calculations in the reported molecularly engineered polyiodide reservoirs. The calculated interactions between polyiodide species and the cage-like framework indicate that the confinement effect arises not only from pore-size matching but also from favorable host-guest interactions between polyiodides and polar/cationic pore-wall sites. Therefore, the theoretical results help to distinguish simple geometric confinement from chemically assisted polyiodide capture in ordered macromolecular hosts. A cage-topology hybrid crystalline structure further integrates pore confinement and transport within the ordered framework. The key advantage of this cage-topology hybrid crystalline structure is the incorporation of fused-ring π -conjugated backbones, which enhance conductivity while integrating pore confinement and transport within the same crystalline macromolecular framework. Unlike traditional host systems, in which pore channels provide confinement while conductivity depends on the added components, this system maintains uniform channels. The π -conjugated backbone provides continuous electron transport pathways, allowing ion migration and electron transport to occur simultaneously. Mechanistically, this study couples polyiodide confinement, site recognition, and charge transport within an ordered framework. Overall, previous studies have advanced the development of Zn-I₂ cathode hosts from different perspectives, including pore confinement, catalytic-site construction, and fine regulation of crystalline frameworks. Our work further shifted the focus to an integrated design of polymer hosts. Unlike designs that consider polyiodide confinement, interfacial recognition, and framework transport as separate optimization issues, this strategy offers a synergistic approach to simultaneously stabilize polyiodide species, promote reversible conversion, and ensure continuous transport.

3 MACROMOLECULAR ION SIEVES FROM SEPARATORS TO ELECTROLYTES

Whereas cathode hosts primarily retain iodine species near the reaction sites, separators and electrolytes govern polyiodide crossover and Zn²⁺ flux across the cell. Conventional polyolefin and glass-fiber separators generally possess relatively large and weakly charged pores, which provide limited electrostatic exclusion or molecular sieving under concentration gradients.^[63] Although rigid framework membranes and highly concentrated electrolytes can improve ion selectivity,^[64,65] they may introduce interfacial incompatibility, increased viscosity, or additional ion-transport resistance. Therefore, flexible polymer frameworks with tunable fixed-charge sites provide an alternative approach for constructing selective mass transport pathways. To achieve molecular-level ion selectivity along mass-transfer path-

ways, researchers have used flexible polymer frameworks and tunable side-chain chemistry to regulate the interfacial charge distribution. Therefore, the construction of macromolecular ion sieves has emerged as an important strategy for regulating polyiodide crossover and Zn²⁺ flux across separator/electrolyte interfaces.

3.1 Electrostatic-shielding Polyanionic Separators

The electrostatic basis of such separator designs can be understood in terms of Donnan exclusion in charged polymer networks.^[66,67] In Zn-I₂ batteries, sulfonated polymer layers have subsequently been employed to suppress polyiodide crossover and regulate Zn²⁺ transport. This strategy relies on the formation of a stable electrostatic shielding layer at the solid-liquid interface. When a high density of anionic groups is introduced into the polymer side chains, the polymer framework generates a negative charge distribution inside the pores and near the interface. This negative electrostatic environment repels polyiodide anions, such as I₃⁻ and I₅⁻, and thus suppresses their migration (Figs. 5a and 5b). Qu *et al.* further developed a sulfonate-functionalization strategy for Zn-I₂ batteries, in which sulfonate groups served as both iodine-species limiters and Zn-plating guidance sites, thereby coupling polyiodide-shuttle suppression with anode-side deposition regulation.^[68] Thus, the charged polymer network can regulate the local ion distribution and mitigate polyiodide shuttling. From the viewpoint of ion transport theory, this electrostatic shielding can be understood as a redistribution of mobile ions in response to fixed charges within the polymer network. The fixed anionic groups generate a local electrostatic potential, which affects the concentration distribution of the polyiodides and Zn²⁺ near the separator/electrolyte interface. Therefore, models such as the Poisson-Nernst-Planck theory can be used to correlate the fixed charge density, ion concentration gradients, and interfacial potential distribution in charged polymer layers.^[69,70] However, conventional fluorinated ion-exchange membranes can face practical limitations associated with cost and membrane-structure control in Zn-I₂ systems.^[71,72]

In our earlier work, a sulfonate-rich ion-exchange layer based on sulfonated cross-linked poly(phenylene sulfide) (SC-PPS) was constructed.^[73] This work systematically revealed the corrosion process of zinc induced by polyiodides. Specifically, I₃⁻ spontaneously reacts with Zn to form soluble ZnI₂. This process triggers honeycomb-like corrosion, interfacial passivation, and dendrite growth. Therefore, a polymer protection layer should suppress polyiodide crossover and regulate Zn²⁺ transport and deposition (Fig. 5c). In the SC-PPS system, cross-linked PPS serves as an insoluble framework, whereas the ionized sulfonate groups act as negatively charged active sites. Collectively, they establish a stable ion-exchange environment at the interface. The high negative zeta potential arising from the negatively charged sulfonate groups provides strong electrostatic repulsion against polyiodide permeation. Meanwhile, the sulfonate groups coordinate Zn²⁺, facilitating the formation of a Zn-rich interfacial layer. This interfacial layer increases the Zn²⁺ transference number and suppresses the harmful two-dimensional surface diffusion. In addition, the polar sulfonate-containing sites interact with water molecules and reorganize the local hydration environment, thereby lowering the desolvation energy

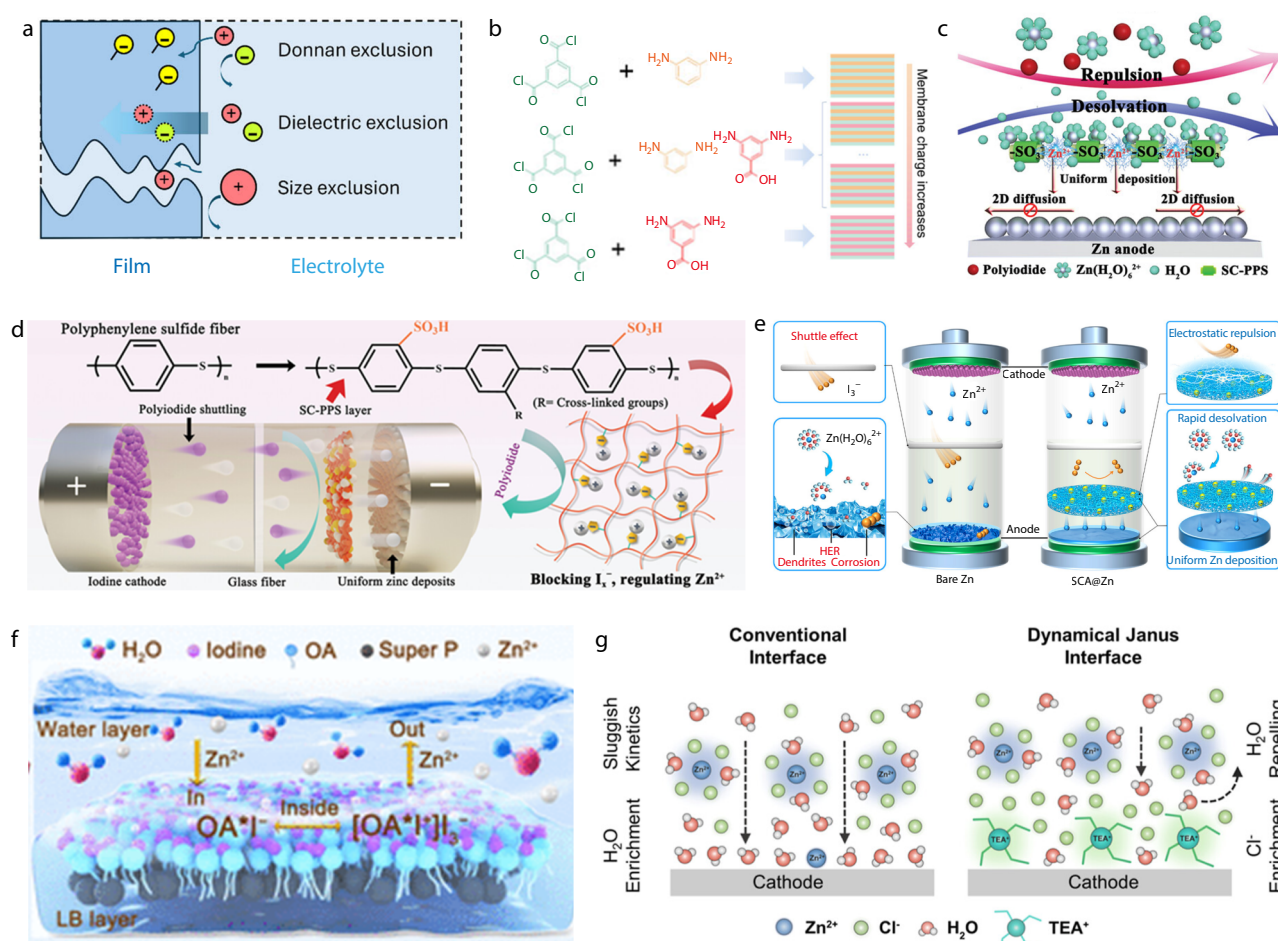


Fig. 5 Representative ion-sieving concepts and dynamic interfacial regulation strategies relevant to AZIBs, illustrating suppression of polyiodide crossover and regulation of Zn²⁺ flux. (a, b) Ion-selective transport in charged polymer films (Reproduced with permission from Ref. [69]; Copyright (2025), AIP Publishing); (c, d) Sulfonate-rich SC-PPS ion-exchange layer for polyiodide blocking and Zn²⁺ regulation (Reproduced with permission from Ref. [73]; Copyright (2023), Wiley-VCH GmbH); (e) Sulfonated cellulose acetate nanofiber protective layer (Reproduced from Ref. [74]; Copyright (2026), The Author(s). Published under a Creative Commons CC BY 4.0 license); (f) Langmuir-Blodgett oleylamine film (Reproduced with permission from Ref. [78]; Copyright (2024), American Chemical Society); (g) Dynamic Janus interface induced by tetraethylammonium cations (Reproduced with permission from Ref. [79]; Copyright (2024), American Chemical Society).

barrier of [Zn(H₂O)₆]²⁺. This mitigates water-induced corrosion and parasitic reactions. Because of this coupled mechanism, SC-PPS can block polyiodides while regulating Zn²⁺ transport and deposition (Fig. 5d). The SC-PPS-modified Zn-I₂ battery achieved stable cycling over 6000 cycles at 3.2 A·g⁻¹, with a capacity retention of 90.2% and an average Coulombic efficiency of 99.89%. It outperformed the bare Zn system in terms of cycling stability and Coulombic efficiency.

On this basis, we further developed an electrospun nanofiber protection layer based on sulfonated cellulose acetate (SCA).^[74] Compared with the above ion-exchange layer, this system places more emphasis on the synergistic regulation of the chain polarity, nanofibrous channels, and interfacial wettability (Fig. 5e). The SCA nanofiber protective layer forms a uniform negative electric field through the —SO₃⁻ groups on the side chains. This effect strengthens electrostatic shielding and effectively repels linear I₃⁻ species. As a result, I₃⁻ transport toward the Zn anode was significantly suppressed. Meanwhile, the polar groups on the SCA backbone, including the —OH and —C=O groups, reduce the activity

of interfacial water through dipole-dipole interactions. This process further promotes Zn²⁺ desolvation. Theoretical calculations and finite element simulations further clarified this ion regulation mechanism. For the SCA nanofiber protective layer on the Zn anode, the polyanionic network redistributed the local electric field and Zn²⁺ flux through the combined effects of physical pore restriction and electrostatic shielding. In this case, the —SO₃⁻ groups do not simply act as static anionic sites for repelling I₃⁻; they also participate in constructing a zincophilic interface that promotes Zn²⁺ desolvation and homogenizes Zn deposition. Therefore, this study provides a representative example in which theoretical analysis links fixed anionic groups, local electric-field redistribution, ion-flux homogenization, and dendrite suppression within an anode-facing interfacial design. In addition, its three-dimensional channel structure improves the uniform deposition of Zn, prevents local Zn²⁺ accumulation, and reduces zinc dendrite growth. These coupled effects homogenize the Zn²⁺ flux and suppress parasitic reactions, thereby improving cycling stability and Coulombic efficiency. When SCA@Zn was used as the

anode, the battery delivered an ultralong cycling life of more than 1.3×10^4 cycles, with a capacity retention of 96.74% and a high Coulombic efficiency. These examples indicate that electrochemical kinetic parameters should be analyzed together with long-term cycling data. Electrochemical impedance spectroscopy (EIS) can provide information on changes in the interfacial and charge-transfer resistances, whereas GITT can be used to evaluate the apparent chemical diffusion behavior of the overall electrochemical process. These parameters should be interpreted together with polyiodide-permeability tests, ion-transference measurements, and Zn-deposition analyses to distinguish the contributions of iodine conversion kinetics, polyiodide crossover, and Zn^{2+} flux regulation. Although electrostatic-shielding separators can effectively retard polyiodide crossover and regulate Zn^{2+} transport, their action relies on a relatively static solid barrier. As a result, further improvement requires moving from passive ion blocking toward more adaptive interfacial regulation at ultrathin solid-liquid boundaries.

3.2 Two-dimensional Self-assembled Dynamic Interfacial Layers

In AZIBs, the major failure processes, including polyiodide shuttling, sluggish iodine conversion, and imbalanced Zn deposition, are ultimately manifested at the ultrathin solid-liquid boundary. Compared with separator-side charge shielding, two-dimensional self-assembled interfacial layers provide a more localized and dynamic method for regulating the ion distribution, interfacial electric fields, and reaction microenvironments. Recent electrolyte-level studies have shown that reorganizing phase behavior and solvation networks can simultaneously influence halogen conversion and Zn-anode stability.^[75,76] Building on this reaction-environment perspective, localized interfacial regulation offers a shorter transport length scale and more direct control over species migration. For example, a dynamic cathode interlayer formed *in situ* can suppress polyiodide dissolution and migration and substantially reduce self-discharge.^[77] Positively charged biomacromolecules can also build selective interfacial layers on the cathode surface to confine iodine species, whereas zwitterionic additives can regulate iodine species and Zn^{2+} at both electrode interfaces.^[33,63]

In this context, a two-dimensional interfacial assembly can be regarded as a precursor model and boundary expression for polymer charge engineering. Its key feature is the establishment of strong local chemical potential gradients and ion-sieving effects over an ultrashort transport distance. Following this concept, the Langmuir-Blodgett (LB) technique was used to construct an ultrathin preorganized interface based on oleylamine (OA) molecules.^[78] As shown in Fig. 5(f), the self-spreading of amphiphilic molecules in a two-dimensional plane enabled the OA film to form a high-density array of amine head groups at extremely low mass loadings. Through Lewis acid-base interactions, this interface forms an *in situ* charge-transfer complex with iodine, denoted as $[\text{OA}^+\text{I}^-]_3$. This interfacial layer maintains fast mass transport and precisely traps polyiodide anions through an in-plane high-density charge network, similar to that in ionic polymers.

Interfacial charge regulation does not rely solely on pre-formed static films. *In situ* reconstruction driven by an applied electric field and concentration gradient endows the ul-

trathin interfacial layers with enhanced environmental adaptability. Based on this idea, tetraethylammonium (TEA^+) cations were introduced. These cations underwent spontaneous enrichment and phase separation at the electrode boundary, leading to the formation of a dynamic Janus soft interface with water-deficient and Cl^- -rich characteristics (Fig. 5g).^[79] Maintained by noncovalent interactions, this dynamic boundary layer suppresses local water activity and stabilizes high-valence halogen intermediates such as ICl without the support of a covalent framework. As a result, the interfacial Zn plating/stripping kinetics and halogen redox kinetics were simultaneously improved. The design principles of two-dimensional supramolecular interfaces clearly show that the effectiveness of an interfacial layer is closely related to the local density of the charged units and their dynamic response. Thus, local charge density and dynamic responsiveness are as critical as the covalent chain architecture for interfacial ion regulation. From static LB molecular films to dynamically reconstructed Janus boundary layers, these ultrathin systems demonstrate that effective ion regulation can be achieved even without bulk polymer networks, provided dense and responsive charged units are established at the interface. Extending this concept from nanometer-thick boundary layers to the entire electrolyte phase has motivated the design of electrolyte-phase reaction environments.

3.3 Ion Sieving and Reaction-environment Regulation in Electrolytes

Beyond separator-side ion sieving and ultrathin interfacial field regulation, recent studies have increasingly extended macromolecular ion-sieving concepts to the electrolyte phase, where ion distribution, solvation structure, and species partitioning can be regulated on a much broader scale. Therefore, a full-cell perspective on electrolyte function, rather than isolated local interfaces, is essential for achieving high reversibility and stable operation in high-loading Zn-I_2 systems. In recent years, electrolyte design has evolved from conventional salt-solution optimization to a wider range of strategies, including the use of functional additives, polymer/gel electrolytes, biphasic electrolytes, and solid composite electrolytes. Despite their different forms, these approaches share a common objective of deliberately regulating ion distribution, solvation structure, and interfacial chemistry, thereby constructing a more selective and better-defined reaction environment.^[63,80,81]

From the perspective of research development, early studies on functional additives first identified the local interactions that play a decisive role in Zn-I_2 systems, thereby providing an important chemical basis for the subsequent design of ionic polymer electrolytes. Yin *et al.* reported a multifunctional additive for Zn-ion batteries, showing that interfacial adsorption and water-shielding effects can regulate hydrogen evolution and dendrite growth on Zn anodes.^[82] Such additive studies highlight the importance of interfacial adsorption, water shielding, and local solvation regulation in suppressing parasitic reactions, thereby providing guidance for incorporating these interaction modes into polymer chains. Soluble polymer additives further highlight the unique advantages of polymer segments in regulating the reaction medium. Taking poly(vinyl alcohol) (PVA) as an example, the abundant hydroxyl groups along the polymer chains can in-

teract with I_3^- , thereby lowering the effective activity of the polyiodide species in the bulk phase (Fig. 6a), while simulta-

neously disrupting the hydrogen-bonding network of water and reducing the activity of free water. This mechanism was

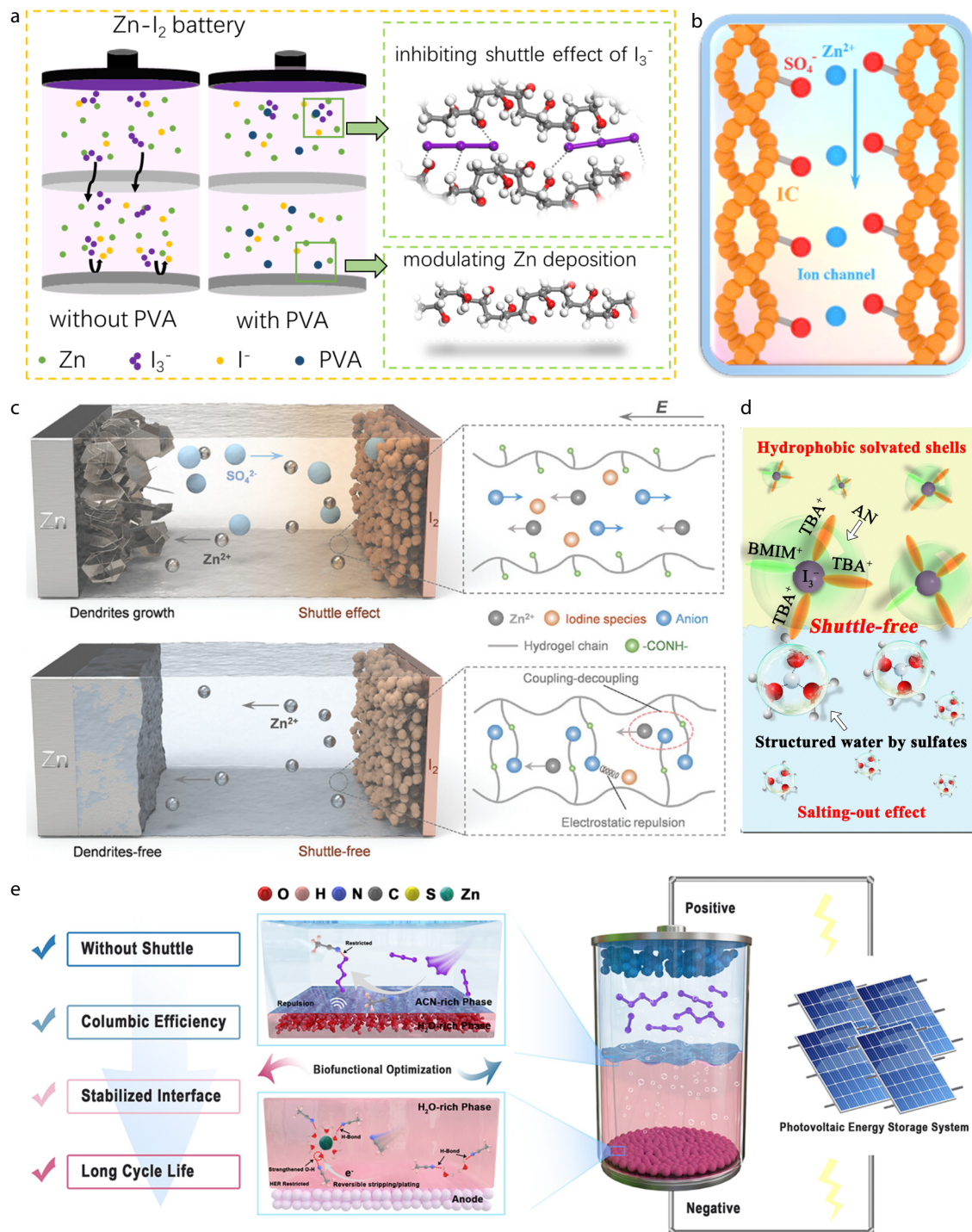


Fig. 6 Representative electrolyte-regulation strategies of ionic polymers and biphasic systems in AZIBs, highlighting solvation modulation, polyiodide partitioning, and ion-transport control. (a) PVA additive suppressing polyiodide shuttling and regulating Zn deposition (Reproduced with permission from Ref. [83]; Copyright (2024), The Royal Society of Chemistry); (b) IC gel electrolyte with Zn²⁺-transport channels (Reproduced with permission from Ref. [84]; Copyright (2023), The Royal Society of Chemistry); (c) Hetero-polyionic hydrogel for coupled shuttle inhibition and dendrite suppression (Reproduced with permission from Ref. [86]; Copyright (2023), Wiley-VCH GmbH); (d) Liquid-liquid biphasic electrolyte for efficient polyiodide extraction and shuttle suppression (Adapted with permission from Ref. [88]; Copyright (2025), The Royal Society of Chemistry); (e) Self-segregated biphasic electrolyte for optimizing polyiodide partitioning and Zn-anode interfacial stability (Reproduced from Ref. [89]; Copyright (2024), The Author(s). Published under a Creative Commons CC BY 4.0 license).

also supported by EIS analysis, in which the PVA-containing electrolyte showed reduced interfacial resistance compared to the polymer-free electrolyte. This result suggests that suppressing polyiodide shuttling through polymer–polyiodide interactions does not necessarily sacrifice reaction kinetics; instead, the regulated solvation and interfacial environment can facilitate charge transfer and improve electrochemical reversibility.^[83] Mechanistically, the value of PVA does not simply arise from the introduction of functional groups capable of interacting with polyiodides. More importantly, these sites are continuously distributed along the polymer backbone, allowing discrete weak interactions to be amplified into segment-level effects that can persist throughout the medium. As a result, polymer additives transform local coordination and hydrogen-bond rearrangement at the molecular scale into a more stable and sustained mode of electrolyte–environment regulation. Polymer and gel electrolytes further immobilize these local interactions within stable macromolecular networks, enabling the electrolyte to evolve into a reaction medium with structural continuity and ion selectivity. Tian *et al.* reported an ι-carrageenan (IC) gel electrolyte in which the double-helical natural polysaccharide framework and sulfate functional groups collectively constructed a continuous ion transport network (Fig. 6b). The polar polymer segments and fixed anionic groups are beneficial for organizing Zn^{2+} migration pathways and improving the uniformity of interfacial deposition, whereas the confinement of water molecules within the gel network reduces the activity of free water, thus alleviating corrosion and side reactions.^[84] Such systems integrate segmental polarity, fixed-charge sites, and confined aqueous environments into a continuous macromolecular phase. A similar concept was further extended by constructing a cation-conduction-dominated hydrogel electrolyte. Yang *et al.* covalently anchored anions to the polymer chains, allowing Zn^{2+} to become the predominant mobile charge carrier. The fixed-anion framework increased the Zn^{2+} transference number, regulated the Zn^{2+} solvation environment and hydrogen-bonding network, and promoted more uniform interfacial Zn deposition.^[85] This work demonstrates that hydrogel electrolytes can actively impose transport asymmetry through fixed ionic sites, rather than merely serving as passive liquid-retaining matrices.

Another representative strategy is the construction of hetero-polyionic hydrogel electrolytes, in which differently charged polymer networks perform spatially differentiated electrochemical functions (Fig. 6c). On the cathode side, an iodophilic polycationic network confines the polyiodide species and accelerates the iodine redox kinetics. On the anode side, the polyanionic network homogenizes the Zn^{2+} flux and electrostatically retards the migration of polyiodide anions, thereby protecting the Zn anode. This spatially differentiated architecture generates a functional ionic gradient tailored to the distinct reaction requirements of the two electrodes, enabling simultaneous regulation of polyiodide shuttling, iodine conversion, and Zn deposition.^[86] Consequently, the hydrogel electrolyte evolved from a passive liquid-containing medium to a structured reaction environment capable of actively organizing local ionic fields. Recent studies on polycationic hydrogel electrolytes have further expanded the

functional scope of ionic polymers in AZIBs.^[87] These studies indicate that polycationic chain segments may not only stabilize halogen-related intermediates and alleviate shuttle effects but also participate in the regulation of cascade conversion processes. This observation suggests that polymeric ionic environments have begun to influence the reaction pathways of iodine/halogen species as well as the stability of the reaction intermediates. For polymer electrolytes, this means that the functions of fixed-charge sites and segmental polarity have progressed from simple ionic confinement to the integrated regulation of the reaction sequence, local charge distribution, and interfacial chemical potential. At this level, ionic polymer electrolytes highlight the capability of polymeric ionic environments to simultaneously coordinate the mass transport and reaction pathways through structured media.

Although the above polymer and ionic polymer electrolytes have shown a pronounced capability to regulate local solvation structures and ionic migration behavior, further optimization is still required to regulate the thermodynamic partitioning of polyiodide species throughout the bulk phase. Thus, biphasic electrolytes shift the electrolyte design from regulating homogeneous continuous phases to selectively partitioning species between distinct phases. The self-stratified aqueous biphasic Zn-I and Zn-Br batteries demonstrated that ionic-liquid-induced spontaneous phase separation, together with halogen extraction effects, can achieve the spatial separation of active species under membraneless conditions.^[75] Our group developed a liquid-liquid biphasic electrolyte with self-sieving characteristics and integrated it with a cell-structure design to achieve efficient polyiodide extraction, low self-discharge, and long-term cycling stability through the combined effects of a hydrophobic solvation sheath and salting-out behavior (Fig. 6d).^[88] In this system, polyiodide confinement is elevated from local adsorption or electrostatic shielding to bulk-phase sequestration, allowing the migration pathways of polyiodides, reaction regions, and electrolyte recovery to be coordinated within a unified design framework. A related study on self-segregated biphasic electrolytes further showed that phase differentiation and solvation-environment reconstruction can simultaneously suppress polyiodide shuttling, reduce water activity, and stabilize the Zn anode and electrolyte interface (Fig. 6e).^[89] From a mechanistic point of view, these systems extend ion selectivity from the segmental scale to the phase separation scale. In the former case, ionic migration is organized through fixed-charge sites and segmental polarity, whereas in the latter, the distribution of active species is governed by phase contrast and solvation-environment differences. Both strategies embody the fundamental principle of preorganizing the reaction medium. Along this line, the development of biphasic electrolytes may be regarded as a further extension of the ionic polymer reaction-medium design to the bulk-phase scale. In terms of reaction-environment regulation, this type of work remains intrinsically consistent with the ionic polymer strategies discussed above, as both are directed toward the management of ion distribution, mass-transport behavior, and interfacial reactions through structured media. Overall, electrolyte research in Zn-I₂ batteries has progressed from identifying molecular-level interactions to the design of struc-

tured reaction media. Small-molecule additives have revealed key local interactions that govern polyiodide stabilization, Zn^{2+} solvation, and parasitic reactions. Polymer additives and gel electrolytes have subsequently immobilized these interactions within the continuous polymer chains and network structures. Ionic polymers have further enhanced the transport selectivity and controllability of the reaction environment through the introduction of fixed-charge sites, whereas biphasic electrolytes have extended this concept to the regulation of species partitioning in the bulk phase. Accordingly, the focus of electrolyte design has increasingly shifted toward the ability of the medium structure itself to organize the ion distribution, solvation states, and reaction pathways. For aqueous Zn-I_2 batteries, this evolution also positions ionic polymer-based strategies as a key material platform for constructing stable reaction environments.

4 POLYMERIC INTERFACE ENGINEERING FOR ZINC ANODE REGULATION

The effects of polyiodide crossover, Zn^{2+} flux imbalance, and interfacial water activity ultimately converge at the Zn anode, where they induce corrosion, parasitic reactions, and non-uniform Zn deposition. Therefore, anode-side regulation should not be considered independent of the cathode and electrolyte/separator processes. Instead, polymeric interface engineering at the Zn anode is required to mitigate polyiodide-induced passivation, regulate Zn^{2+} desolvation and deposition, and suppress water-related side reactions. Zhang *et al.* constructed an ultrathin hydrophobic anode/electrolyte interphase, showing that water exclusion and selective Zn^{2+} coordination can be combined to suppress dendrite formation and hydrogen evolution, while maintaining Zn^{2+} transport.^[90] Compared to inorganic solid-electrolyte interphase (SEI) layers, which are usually mechanically brittle, macromolecular interfacial layers with mechanical flexibility can form more continuous physical barriers and better accommodate volume changes during Zn plating/stripping. More importantly, the abundant polar or charged groups on the polymer frameworks can actively regulate the hydration structure, distribution of cations and anions, and their migration pathways at the interface, thereby reconstructing the local reaction environment. For example, Shen *et al.* constructed a self-assembled electrode/electrolyte interphase through amphiphilic molecule adsorption and interfacial coordination, whereas Meng *et al.* introduced zincophilic cucurbituril supermolecules with abundant lone-pair electrons to guide Zn^{2+} migration through molecular channels.^[91,92] In these interfacial layers, carboxylate, hydroxyl, sulfonate, amino, and zwitterionic groups can serve as coordination or dipolar sites for modulating Zn^{2+} desolvation, interfacial wettability, and nucleation.

4.1 Interfacial Desolvation Driven by Polar Segments

Hydrated Zn^{2+} possesses a stable primary-solvation shell. As a result, desolvation at the electrode surface usually involves a relatively high energy barrier. When hydrated Zn^{2+} reaches the metal interface, its coordinated water molecules can promote hydrogen evolution and corrosion. To address this issue, polar polymer protection layers have been designed to regulate the interfacial coordination environment through heteroatom-con-

taining segments, thus altering the solvation state of Zn^{2+} near the electrode surface. Xie *et al.* employed poly(acrylic acid) (PAA) as an electrolyte additive to form a Zn-rich polymeric SEI *in situ* on the Zn surface.^[93] The key feature of this system is the continuous coordination ability of carboxyl-containing segments toward Zn^{2+} . The PAA-derived interfacial layers isolated the Zn surface from direct contact with the electrolyte (Fig. 7a). In addition, the carboxyl groups provided uniformly distributed adsorption and nucleation sites for Zn^{2+} and lowered the activation energy for interfacial desolvation and cross-interface migration (Fig. 7b). As a result, the coordination chemistry of Zn^{2+} and its initial nucleation behavior at the interface were improved. The ultrathin poly(sulfobetaine methacrylate) (PSBMA) molecular brush reported by Hao *et al.* represents another effective interfacial strategy.^[94] In this system, the dense brush conformation governs the interfacial Zn^{2+} flux by forming nanoscale ion transport channels (Figs. 7c and 7d). A molecular brush with a thickness of approximately 10 nm was firmly anchored on the Zn surface through covalent grafting. This structure avoids the delamination and cracking commonly observed in conventional coated films. The highly ordered sulfobetaine side chains create uniform Zn^{2+} transport channels at the nanoscale. As a result, the Zn^{2+} flux was homogenized at the interface, while SO_4^{2-} and other side-reaction species were prevented from approaching the metal surface. This feature helps to maintain low polarization and uniform deposition under high-rate conditions.

In comparison, Geng *et al.* proposed a glutamic acid additive that further extends polymer protection toward *in situ* generation and dynamic renewal during cell operation.^[95] In this system, the additive did not form a static film in advance. Instead, radical-initiated electropolymerization proceeded at the cathode, whereas polycondensation occurred at the anode. These processes produced bilateral SEI layers composed mainly of poly(glutamic acid) (PGA). The resulting interfacial layers exhibited strong *in situ* adhesion to the electrode surface and could be continuously repaired and renewed during cycling (Figs. 7e and 7f). Moreover, the carboxylic acid sites along the PGA backbone could reorganize the local solvation environment of Zn^{2+} . These carboxyl-containing segments also improved the interfacial wettability and reduced the activation barriers for Zn^{2+} diffusion and desolvation. Consequently, interfacial Zn^{2+} diffusion and desolvation kinetics were simultaneously enhanced.

4.2 Ion Flux Homogenization Guided by Polyanion Networks

In AZIBs, the polymeric interfacial layers on the anode side regulate Zn deposition by coupling electrostatic repulsion, Zn^{2+} coordination, and interfacial structural optimization. Our group developed a highly sulfonated poly(ether ether ketone) (H-SPEEK) coating that optimized the electrolyte/electrode interface and improved the stability of Zn anodes (Fig. 8a).^[96] The function of the H-SPEEK coating arises from its oriented sulfonic groups and high local charge density, which generate a strong interfacial electric field for regulating Zn^{2+} migration and desolvation. The ionized $-\text{SO}_3^-$ groups provide negatively charged coordination sites for Zn^{2+} and lower the desolvation energy barrier, thereby suppressing hydrogen evolution, non-uniform Zn deposition, and capacity decay. Electrochemical tests further showed that the H-SPEEK coating could prevent excessive local

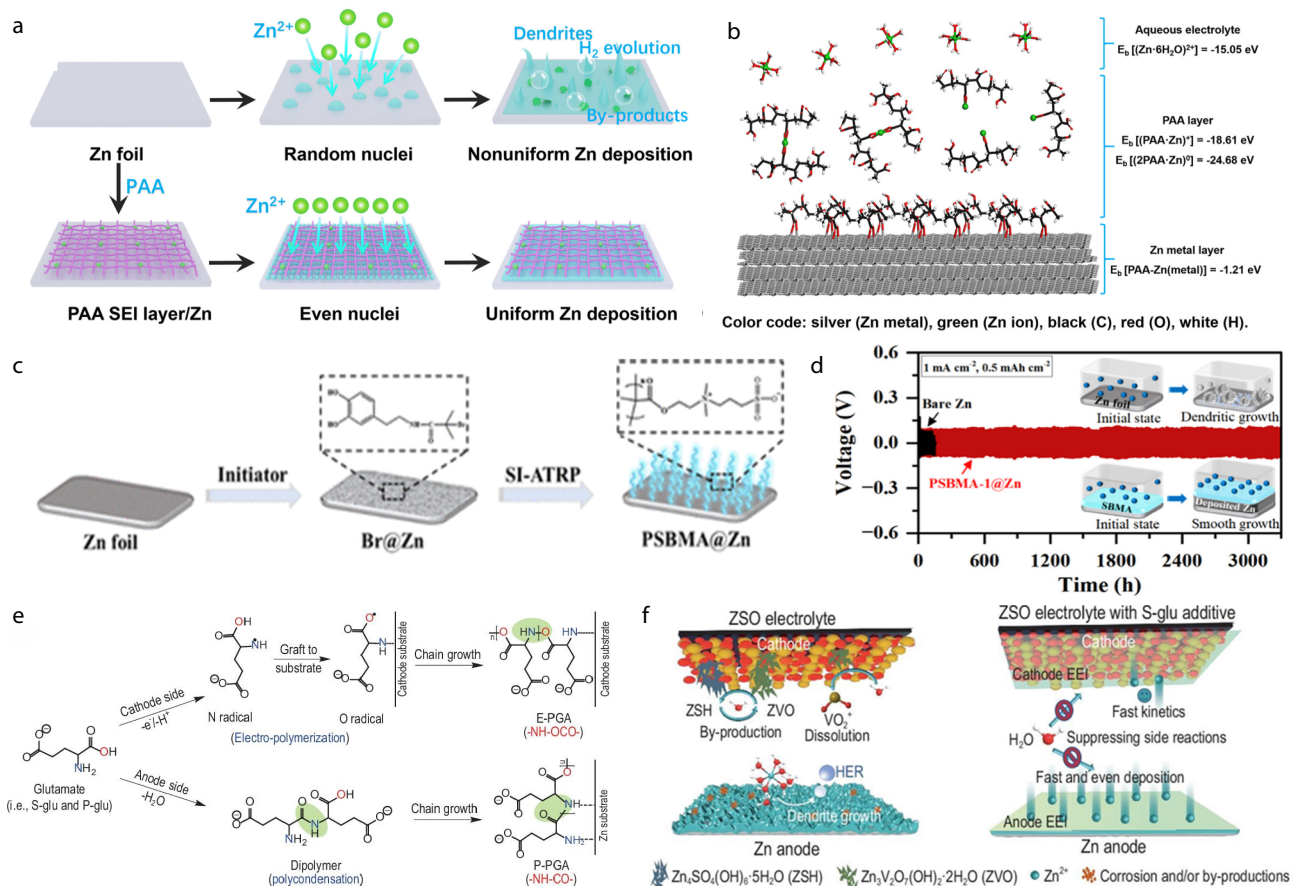


Fig. 7 Representative polymeric interfacial layers for Zn-anode protection in AZIBs. (a, b) *In situ* formed Zn-rich polymeric SEI derived from poly(acrylic acid) (Reproduced with permission from Ref. [93]; Copyright (2023), The Authors. Published by Elsevier B.V. on behalf of Nankai University); (c, d) Ultrathin PSBMA molecular brush grown on Zn (Reproduced with permission from Ref. [94]; Copyright (2023), American Chemical Society); (e, f) Bilateral electrode/electrolyte interphases derived from glutamate self-polymerization (Reproduced from Ref. [95]; Copyright (2025), The Author(s). Published under a Creative Commons CC BY 4.0 license).

solvation and dendrite growth at the electrode surface by optimizing the electrolyte interface, thereby enhancing long-term Zn-anode reversibility. The cell operated stably for 1230 h at 2 mA cm^{-2} and 2 mAh cm^{-2} without soft short-circuiting, demonstrating excellent long-term stability. These results indicate that the H-SPEEK coating enhances Zn^{2+} transport, desolvation kinetics, and deposition behavior at the microscopic interface. In addition to strongly acidic sulfonic systems, natural polymer frameworks and weakly acidic polyanionic networks have shown distinct interfacial functions. Wang *et al.* developed a topological chitosan framework for reversible columnar-array Zn anodes and showed that natural polymer frameworks with abundant polar groups and continuous macromolecular architectures can guide reversible Zn deposition.^[97] This example also illustrates that the Zn anode regulation mechanism strongly depends on the functional group chemistry: amino and hydroxyl groups mainly influence interfacial hydration and coordination, whereas carboxylate, sulfonate, and zwitterionic groups regulate Zn^{2+} coordination, electrostatic interactions, and ion-flux distribution through different mechanisms. The key quantitative descriptors for anode-side ionic polymers are different from those for iodine cathode hosts. Instead of the polyiodide adsorption energy, the Zn^{2+} coordination strength, desolvation

energy barrier, nucleation overpotential, and interfacial ion-flux distribution are more relevant for evaluating functional groups such as $-SO_3^-$, $-COO^-$, $-OH$, and zwitterionic moieties. Sulfonate-containing polymers mainly regulate Zn^{2+} migration and interfacial electric fields, whereas carboxyl-containing polymers are directly involved in Zn^{2+} coordination, desolvation, and nucleation. Therefore, combining coordination/desolvation calculations with electrochemical analysis is essential for comparing the Zn anode regulation ability of different ionic polymers. For example, CMC-based interphases regulate the flux and local concentration of Zn^{2+} through carboxyl groups, thereby promoting uniform Zn dissolution/deposition and suppressing interfacial corrosion by reducing water activity.^[98] Such CMC-based interphases consequently enhance Zn plating/stripping reversibility (Fig. 8b).

The fluorapatite aerogel (FAG) interfacial layer further demonstrated the importance of interfacial ion transport regulation.^[99] The internal nanochannels of FAG and the abundant Zn^{2+} adsorption sites generated by Ca^{2+} and Zn^{2+} ion exchange can significantly accelerate Zn^{2+} migration and homogenize the ion flux at the electrode interface. This design promotes fast ion transport and guides more ordered Zn deposition, thereby suppressing dendrite growth caused by the

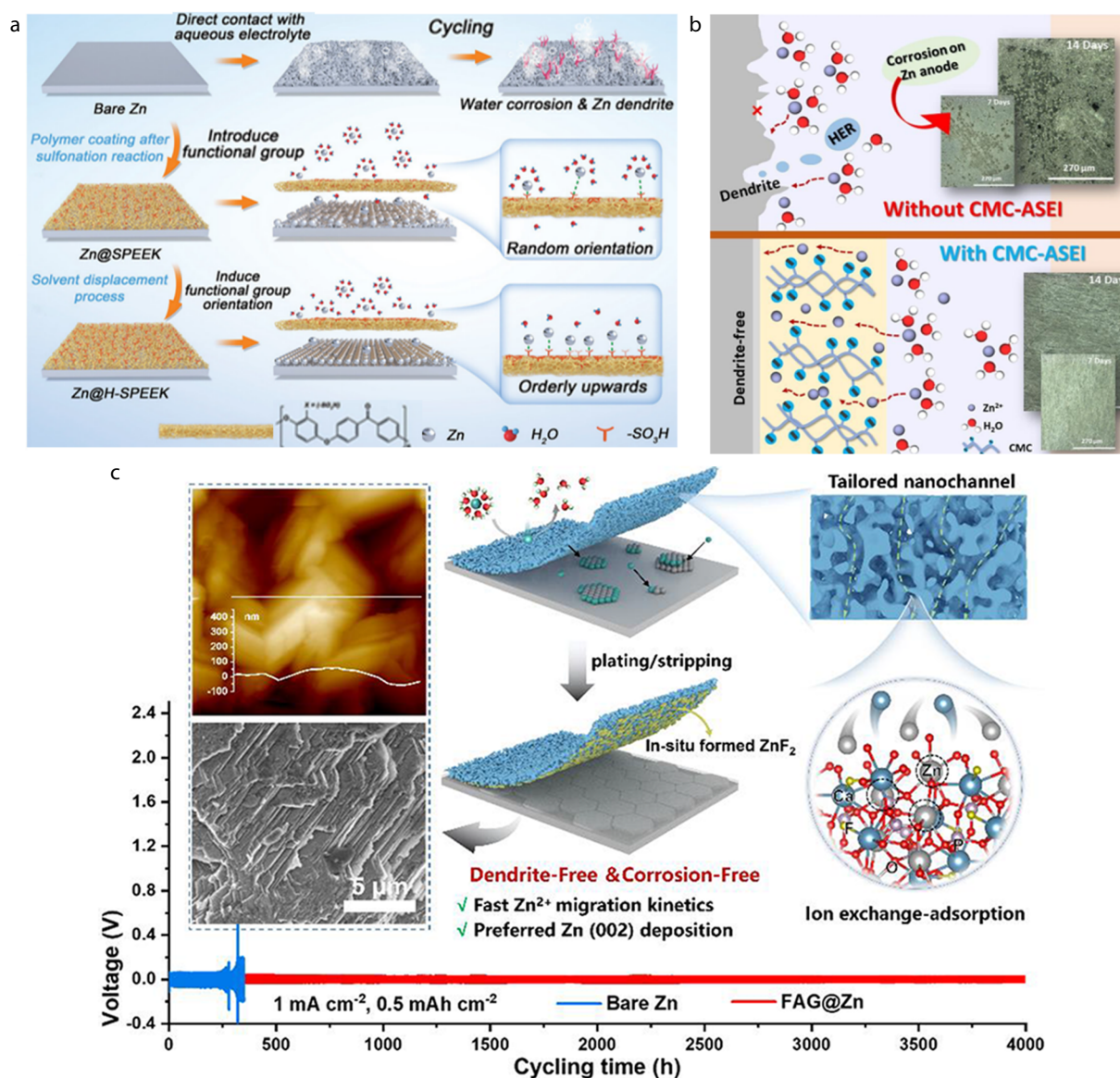


Fig. 8 Representative interfacial layers for Zn-anode protection in AZIBs. (a) H-SPEEK coating with induced anionic sulfonate-group orientation (Reproduced from Ref. [96]; Copyright (2024), The Author(s). Published under a Creative Commons CC BY 4.0 license); (b) CMC-based artificial solid electrolyte interphase (Reproduced from Ref. [98]; Copyright (2023), The Author(s). Published under a Creative Commons CC BY 4.0 license); (c) Fluorapatite aerogel interfacial layer with tailored nanochannels and Zn^{2+} adsorption sites (Reproduced with permission from Ref. [99]; Copyright (2023), American Chemical Society).

local deposition inhomogeneity. In addition, the fluorine-containing species in FAG promoted the *in situ* formation of ZnF_2 during cycling. The ZnF_2 layer can further stabilize Zn deposition and induce preferred growth along the Zn (002) crystal plane. Electrochemical tests showed that the FAG interfacial layer enabled the modified Zn electrode to cycle stably for more than 4000 h at 1 mA cm^{-2} , significantly extending the cycle life (Fig. 8c). Taken together, these interfacial strategies primarily improve Zn-anode reversibility by regulating Zn^{2+} transport, desolvation, nucleation, and deposition. An additional benefit of charged polymer layers that also repel polyiodide anions is the mitigation of polyiodide-induced corrosion and passivation at the Zn surface. Future polymer interface designs will place greater emphasis on building tun-

able ion-transport channels, optimizing local reaction kinetics, and guiding ordered deposition behavior. This multidimensional synergy is expected to provide new design principles for efficient and stable operation of AZIBs. Different ionic polymer strategies have distinct advantages and limitations in AZIBs. Cationic polymer hosts effectively anchor polyiodides, but overly strong binding may slow the iodine conversion. Polyanionic separator layers suppress polyiodide crossover and regulate Zn^{2+} flux, whereas densely charged networks may increase the ion-transport resistance. Polymer additives and gel electrolytes regulate the solvation structure and water activity; however, their conductivity and mechanical stability need to be balanced. Polymeric anode coatings promote Zn^{2+} desolvation and uniform deposition, although

long-term adhesion and interfacial stability remain important concerns.

5 SUMMARY AND OUTLOOK

The challenges in AZIBs do not stem from the failure of a single component. Instead, they arise from the continuous coupled migration of polyiodide species, Zn^{2+} , and solvent molecules among the cathode, electrolyte, separator, and anode. These processes are amplified at the interface, leading to a series of interrelated issues, such as shuttle effects, self-discharge, side reactions, and deposition imbalance.^[83,100] Thus, effective design shifts from optimizing individual material parameters to systematically regulating the charge distribution, ion selectivity, and interfacial reaction environment. The value of ionic polymers in this system lies in their ability to combine high-density charged sites, continuous macromolecular frameworks, and tunable transport channels.^[100,101] In particular, polymer binders can strengthen iodine-species confinement through charge-transfer complexation or chemisorption, thereby suppressing polyiodide shuttling and stabilizing high-loading iodine cathodes.^[102,103]

Our studies have gradually established a research route for AZIBs that extends from physical confinement to charge engineering. In the early stages, our work mainly relied on the pore structure and surface adsorption of carbon materials to slow down the diffusion of iodine species. This stage established the initial understanding of polyiodide shuttling and host-guest confinement. We then shifted our attention from retaining iodine species in pores to regulating the local environment surrounding iodine species and Zn^{2+} . Through the design of biphasic electrolytes and dynamic interfacial regulation strategies, we gradually identified the decisive roles of the solvation structure, local charge distribution, and interfacial microstructure in Zn-I_2 systems.^[79] On this basis, we further incorporated these regulatory mechanisms, which were originally dependent on small molecules and local environments, into more stable macromolecular frameworks. This effort has led to a series of studies on cationic polymer hosts, supramolecular ionic phases, polyanionic ion-exchange layers, and dynamic macromolecular interfacial layers. In this way, our research ultimately progressed toward ionic polymer networks. The LB molecular film showed how a two-dimensional molecular layer can build an effective regulatory environment for iodine species at an ultrathin interface.^[78] The sulfonate-rich ion-exchange layer and dynamic Janus interface further demonstrated that once charged units are stably organized within macromolecular or quasi-macromolecular frameworks, they can simultaneously regulate polyiodide migration, interfacial Zn^{2+} transport, and electrode reaction pathways.^[73,79] Taken together, these studies represent a clear shift in the research logic. The focus has shifted from passive confinement based on the pore structure and surface adsorption to active charge engineering based on fixed charge sites, ion selectivity, and interfacial microenvironment design. Small-molecule models helped identify the local interactions that govern polyiodide stabilization and interfacial Zn^{2+} transport. Ionic polymer networks provide a way to stabilize, extend, and amplify these interactions in practical devices. As a result, ionic polymers in AZIBs are now understood

to be more than a single host material, barrier layer, or additive. They have become a unified design framework that connects the cathode, separator, electrolyte, and anode.

However, several issues remain to be resolved. From the perspective of practical applications, the adaptability of ionic polymer strategies under large-capacity and pouch-cell conditions should also be carefully considered. Most reported AZIBs are evaluated in coin-cell configurations with moderate iodine loading and a relatively large amount of electrolyte. Under more practical conditions, such as high areal iodine loading, lean-electrolyte conditions, thick electrodes, temperature fluctuations, and enlarged electrode dimensions, ionic polymers must simultaneously maintain polyiodide confinement, Zn^{2+} transport, electrolyte wettability, mechanical integrity, and interfacial adhesion. Excessive polymer content or overly dense charged networks may increase ion-transport resistance, reduce active material utilization, or complicate slurry processing and membrane fabrication. In addition, polymer swelling, long-term chemical stability, scalable coating or membrane fabrication processes, and material costs should be considered for large-capacity and pouch-cell configurations. Therefore, future studies should evaluate ionic polymer strategies under high-loading cathodes, limited electrolyte dosage, wide-temperature operation, and pouch-cell-level validation. Although many existing ionic polymer systems already feature a high density of functional sites, achieving a balance between ion selectivity, interfacial dynamics, and long-range transport efficiency remains challenging. The ionic polymers used in different regions of the battery are still relatively scattered in terms of their chemical structure and functionality, and a unified material platform spanning the cathode, separator, electrolyte, and anode remains to be established. Moreover, although fixed charge sites can enhance interfacial control, maintaining the structural stability, process adaptability, and interfacial continuity under long-term cycling still requires a more refined polymer chemical design. Existing studies have pointed to two directions that deserve further investigation: first, the development of ionic polymers with single-ion-conduction-like characteristics through more precise charge-site distribution to improve Zn^{2+} migration selectivity; second, the development of continuous material systems from the same polymer chemical framework to unify mass transport pathways and reaction environments across the cathode, separator, and anode. Overall, our studies on AZIBs have systematically demonstrated the unique value of ionic polymers in this system. Earlier molecular and interfacial studies helped identify key local interactions governing iodine-species stabilization and interfacial ion regulation.^[78,79] Subsequent ionic-polymer studies translated these interactions into stable macromolecular architectures and long-term device-level regulation.^[46,73,74] With the continued development of polymer chemistry and interfacial characterization, ionic polymers are expected to evolve beyond isolated functional layers and become an integrated materials platform for regulating the overall reaction environment in AZIBs. Achieving this transition requires future research to move from the separate optimization of individual battery components to the construction of all-in-one ionic-polymer systems based on unified chemical frame-

works. Such systems should integrate polyiodide confinement, Zn^{2+} transport regulation, electrolyte environment control, and Zn anode protection while maintaining sufficient ionic conductivity, mechanical stability, and processability under practical operating conditions. *In situ* characterization techniques, including *in situ* Raman spectroscopy, *in situ* X-ray analysis, and interfacial microscopy, should be combined with theoretical calculations and data-driven screening to clarify the relationships among the polymer structure, charge distribution, ion transport, and interfacial reaction pathways. The development of self-healing or dynamically adaptive polymeric interfaces may further help to maintain interfacial continuity during repeated Zn plating/stripping and iodine conversion. In addition, these design principles can be extended to other aqueous halogen batteries, such as Zn-Br_2 , Zn-I/Br , and related mixed-halogen systems, which face similar challenges associated with shuttle effects, halogen speciation, and interfacial corrosion. By connecting unified material design, mechanism-resolved characterization, and adaptive interfacial regulation, ionic polymers may provide a broadly applicable framework for the development of high-performance aqueous halogen batteries. We anticipate that this feature article will provide useful guidance for the rational design of ionic polymers in AZIBs, particularly for polyiodide confinement, ion-selective transport, electrolyte reaction-environment regulation, and zinc-anode protection.

BIOGRAPHIES

Jiajia Huang received his Ph.D. degree in Polymer Chemistry and Physics from Sun Yat-sen University in 2014. He joined the School of Chemical Engineering at Zhengzhou University in 2014 and is currently a Professor at Zhengzhou University. He was a postdoctoral researcher at Zhengzhou University from 2015 to 2017 and a visiting scholar at the University of New South Wales, Australia, from 2019 to 2020. His research focuses on functional polymer and carbon-based materials for adsorption and separation, electrochemical energy storage, and energy conversion.

Feili Lai received his B.S. degree from Donghua University in 2014, his M.S. degree from Fudan University in 2017, and his Ph.D. degree from the Max Planck Institute of Colloids and Interfaces/Potsdam University in 2019. He conducted postdoctoral research from 2019 to 2023 at KU Leuven, Belgium, as well as visiting scholars at Max Planck Institute for Polymer Research and Harvard University. In 2023, he joined the School of Materials Science and Engineering at Shanghai Jiao Tong University, where he is currently an Associate Professor. His research interests include polymer nanocomposites for energy-storage materials and devices, electrocatalysis, and soft robotics.

Conflict of Interests

The authors declare no interest conflict.

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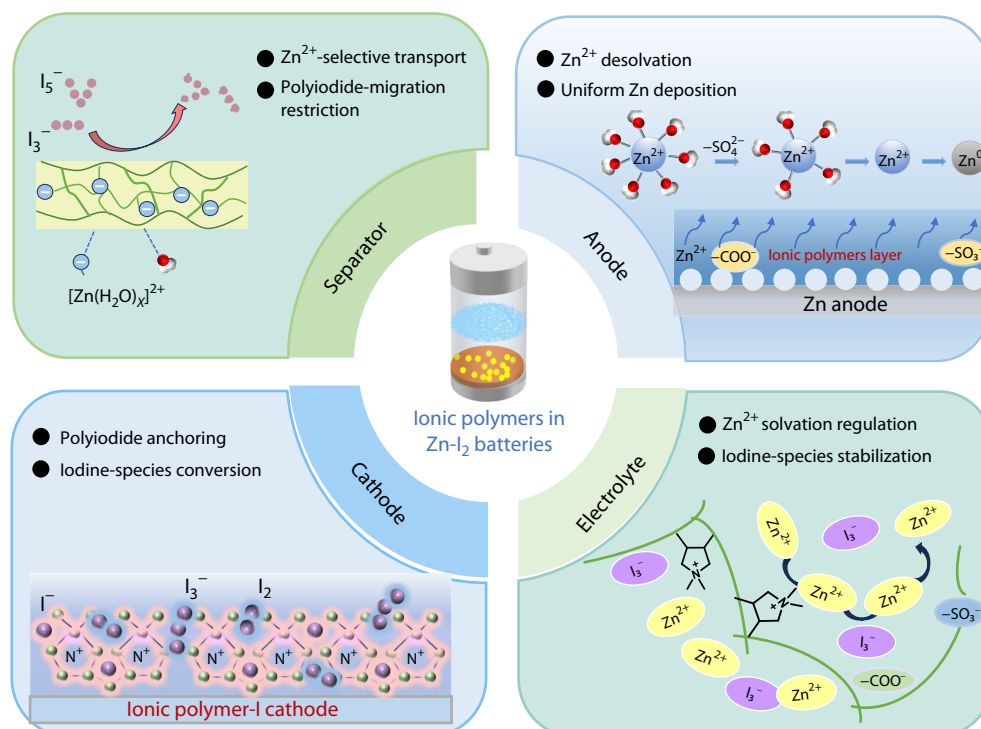
Graphical Abstract

Ionic Polymers in Aqueous Zinc–Iodine Batteries

Qian Chen, Jonas Wilhelm, Jean-François Gohy, Jiajia Huang, and Feili Lai

Zhengzhou University, China; RWTH Aachen University, Germany; Université Catholique de Louvain (UCL), Belgium; Shanghai Jiao Tong University, China

Ionic polymers regulate polyiodide anchoring, ion-selective transport, electrolyte solvation, and zinc-anode interfaces in aqueous zinc–iodine batteries, highlighting the evolution from physical confinement to polymer charge engineering.



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