



Covalent organic frameworks as multifunctional platforms for durable and high-energy-density zinc–iodine batteries

Yixin Zhang¹, Fei Huang¹, Jujin Chai¹, Jinglun Yang², Dongdong Li¹, Boran Wang¹, Xucai Yin¹, Qichun Zhang² (✉), and Huibing He¹ (✉)

¹ University Engineering Research Center of Green Chemical New Materials, Guangxi Key Laboratory of Electrochemical Energy Materials, College of Chemistry and Chemical Engineering, Guangxi University, Nanning 530004, China

² Department of Materials Science and Engineering, City University of Hong Kong, Hong Kong 999077, China

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ABSTRACT

Aqueous zinc–iodine batteries (AZIBs) have attracted increasing attention as promising candidates for next-generation energy storage because of their intrinsic safety, low cost, and high theoretical capacity. However, their practical application is still limited by sluggish redox kinetics, the shuttle effect of soluble polyiodides, self-discharge, zinc corrosion, and dendrite growth. Covalent organic frameworks (COFs), featuring ordered pore channels, tunable pore-wall chemistry, designable active sites, and robust covalent skeletons, provide versatile platforms for addressing these issues. This review summarizes recent progress in COF-based materials for AZIBs from the perspective of structure–function relationships. After briefly introducing the working mechanisms and key challenges of AZIBs, the roles of COFs in iodine confinement, polyiodide adsorption, redox-kinetics regulation, and Zn²⁺ transport modulation are discussed, with attention to their application as cathode hosts, separator modifiers, and quasi-solid-state electrolytes. Representative COFs are further classified into neutral conjugated-framework COFs, 1,3,5-triformylphloroglucinol (Tp)/TpPa-type COFs (Pa denotes p-phenylenediamine), ionic COFs, and metallated COFs, with emphasis on how framework conjugated structures, polar or ionic sites, and metal centers influence electrochemical performance. In particular, ordered nanochannels and functional pore walls enable physical confinement and chemical regulation of iodine species, while catalytic or redox-active sites promote reversible iodine conversion and improve cycling stability. Finally, the remaining challenges and future design strategies of COF-based AZIBs are discussed.

KEYWORDS

aqueous zinc–iodine batteries, covalent organic frameworks, framework materials, iodine cathode hosts, polyiodide shuttle effects

1 Introduction

With the global energy structure shifting toward low-carbon and sustainable development, efficient, safe, and low-cost electrochemical energy storage technologies have become a major research focus [1]. Among various energy storage systems, lithium-ion batteries (LIBs) have dominated a wide range of energy storage devices because of their excellent performance and outstanding cycling stability, which have been widely used in portable electronics, medical technologies, and utility-scale applications [2]. However, the development of lithium-ion batteries still faces several challenges, such as the global scarcity of lithium resources and the rising cost of raw materials [3]. Moreover, spent lithium-ion batteries, if not properly treated, may contaminate soil and groundwater [4]. In addition, lithium plating and dendrite growth under improper operating conditions may induce short circuits and safety risks [5]. These issues severely

limit the further application and development of lithium-ion batteries, making it necessary to explore new battery systems that are safer, more stable, and more cost-effective.

Aqueous zinc–iodine batteries (AZIBs) are regarded as one of the most promising next-generation technologies for large-scale energy storage, owing to their relatively high theoretical capacity based on the iodine cathode (211 mAh·g^{−1} based on a two-electron reaction), high safety derived from the aqueous system, environmental friendliness, and the abundant availability of zinc resources [4–6]. However, their practical application is still hindered by several challenges, including severe self-discharge and Zn anode corrosion caused by polyiodide shuttling, as well as unavoidable dendrite growth and hydrogen evolution side reactions on the Zn anode side [7–9].

To address these issues, framework compounds have recently been introduced as functional materials for optimizing

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Address correspondence to Qichun Zhang, qiczhang@cityu.edu.hk; Huibing He, huibinghe@gxu.edu.cn



zinc–iodine batteries. An ideal material for zinc–iodine battery optimization should be rationally designed according to its specific application site. For iodine cathode hosts, abundant ordered pores, suitable electronic/ionic transport pathways, and strong adsorption or catalytic capability toward polyiodides are desirable for enabling efficient iodine confinement and reversible redox conversion [10, 11]. In contrast, materials used for separator modification, Zn anode protection, or quasi-solid-state electrolytes should primarily regulate ion transport, suppress polyiodide migration, and stabilize the Zn/electrolyte interface rather than simply pursue high electronic conductivity. Framework compounds commonly used in zinc–iodine batteries mainly include covalent organic frameworks (COFs) and metal–organic frameworks (MOFs). Compared with MOFs, COFs, which are constructed entirely from organic building units, generally exhibit lower material density as well as superior chemical and thermal stability [12, 13]. Therefore, they are more favorable for maintaining structural integrity and realizing high energy density under relatively harsh electrochemical environments.

As an emerging class of crystalline porous polymers, COFs possess unique advantages, including tunable porosity, abundant active sites, high synthetic controllability, and predesignable topological structures [14]. Therefore, they have been widely applied in catalysis, energy storage and conversion, gas adsorption, and optoelectronics [15, 16]. In zinc–iodine battery systems, the fine regulation of COF structures can fully exploit the physical confinement effect of their ordered nanopores toward iodine species. Meanwhile, predesigned active sites, such as nitrogen sites, can enhance the chemical adsorption and anchoring capability toward polyiodides, thereby effectively suppressing the shuttle

effect and improving the cycling stability and energy utilization efficiency of the batteries [17, 18].

This review systematically summarizes the recent advances on COFs for the applications in aqueous zinc–iodine batteries (Fig. 1). It first briefly introduces the working mechanism of aqueous zinc–iodine batteries and the key challenges that limit their practical application. Then, the optimization mechanisms of COFs are discussed from four aspects: structural confinement effects, functional group modification, heteroatom modification, and metal coordination. On this basis, representative COF materials reported in recent years are further classified into neutral conjugated framework COFs, 1,3,5-triformylphloroglucinol (Tp)/TpPa-type COFs (Pa denotes p-phenylenediamine), ionic COFs (iCOFs), and metal–covalent organic frameworks. Furthermore, their structure–function relationships in polyiodide confinement, iodine conversion kinetics regulation, and Zn^{2+} transport modulation are highlighted. Finally, the current limitations and future directions of COF-based materials for high-performance and long-life AZIBs are discussed.

2 The working mechanism and current challenges of zinc–iodine batteries

As a metal anode for batteries, Zn delivers a high theoretical specific capacity of $820 \text{ mAh}\cdot\text{g}^{-1}$ and a high theoretical volumetric capacity of $5854 \text{ mAh}\cdot\text{cm}^{-3}$ [19]. Common zinc-based batteries include zinc–manganese compound batteries, zinc–vanadium compound batteries, zinc–silver batteries, zinc–air batteries, zinc–nickel batteries, zinc–bromine flow batteries, and zinc–iodine batteries, etc. This review focuses on zinc–iodine batteries.

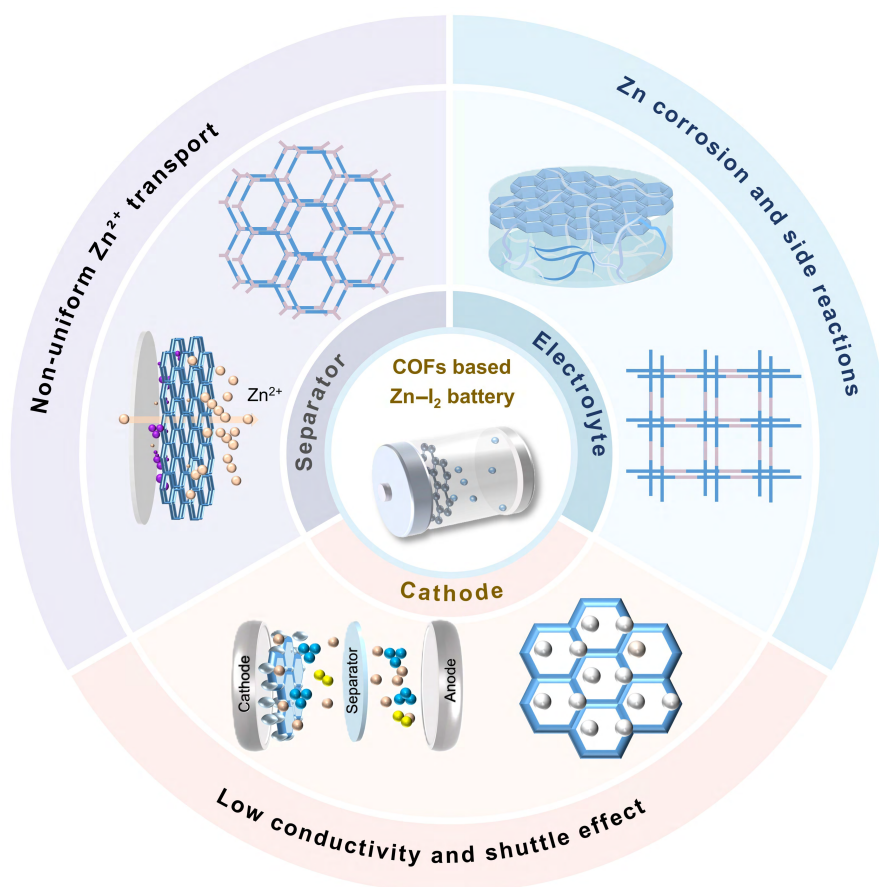


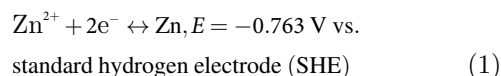
Figure 1 Schematic illustration of COF-based optimization strategies for zinc–iodine batteries.

2.1 Types and working principles of zinc-iodine batteries

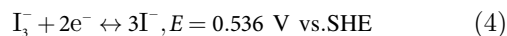
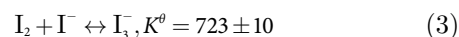
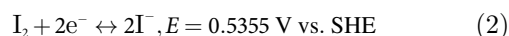
According to their configuration, zinc-iodine batteries can be classified into zinc-iodine redox flow batteries (ZIRFBs) (Fig. 2(a)) and static aqueous zinc-iodine batteries (SZIBs) (Fig. 2(b)). Compared with the former, the latter features a more simplified cell structure that can achieve higher energy density by immobilizing the cathode active materials, and therefore has attracted greater attention [20]. According to the iodine source, zinc-iodine batteries can be mainly divided into two categories (Fig. 2(c)). One type uses a zinc salt electrolyte and stores energy based on the reversible I_2 /iodide ions (I^-) redox reaction. The other type is similar to flow batteries, in which the electrolyte contains dissolved I^-/I_3^- species and is paired with high-surface-area carbon cathodes, such as Ketjen black and Super P [21, 22]. According to the number of electrons transferred during the redox process of iodine species, zinc-iodine batteries can be mainly classified into conventional two-electron systems and emerging four-electron systems (Fig. 2(d)). The two-electron system is based on the classical I_2/I^- redox couple, in which each iodine molecule undergoes a two-electron transfer process, featuring a stable voltage plateau and a well-established reaction mechanism. During discharge, oxidation occurs at the Zn anode, where metallic Zn releases two electrons to form Zn^{2+} ions, while iodine at the cathode accepts two electrons and is reduced to I^- . During charging, I^- at the cathode are oxidized back to I_2 with the release

of two electrons, whereas Zn^{2+} ions are reduced and deposited as metallic Zn on the anode [23]. The electrode reactions and overall reaction are as follows:

Anode



Cathode



Overall reaction



where E is the standard electrode potential, K^θ is the standard equilibrium constant, and E_{cell} is the standard cell potential.

The four-electron system activates higher-valence iodine species, such as I^+ , enabling a four-electron transfer per iodine molecule and thereby significantly increasing the energy density of

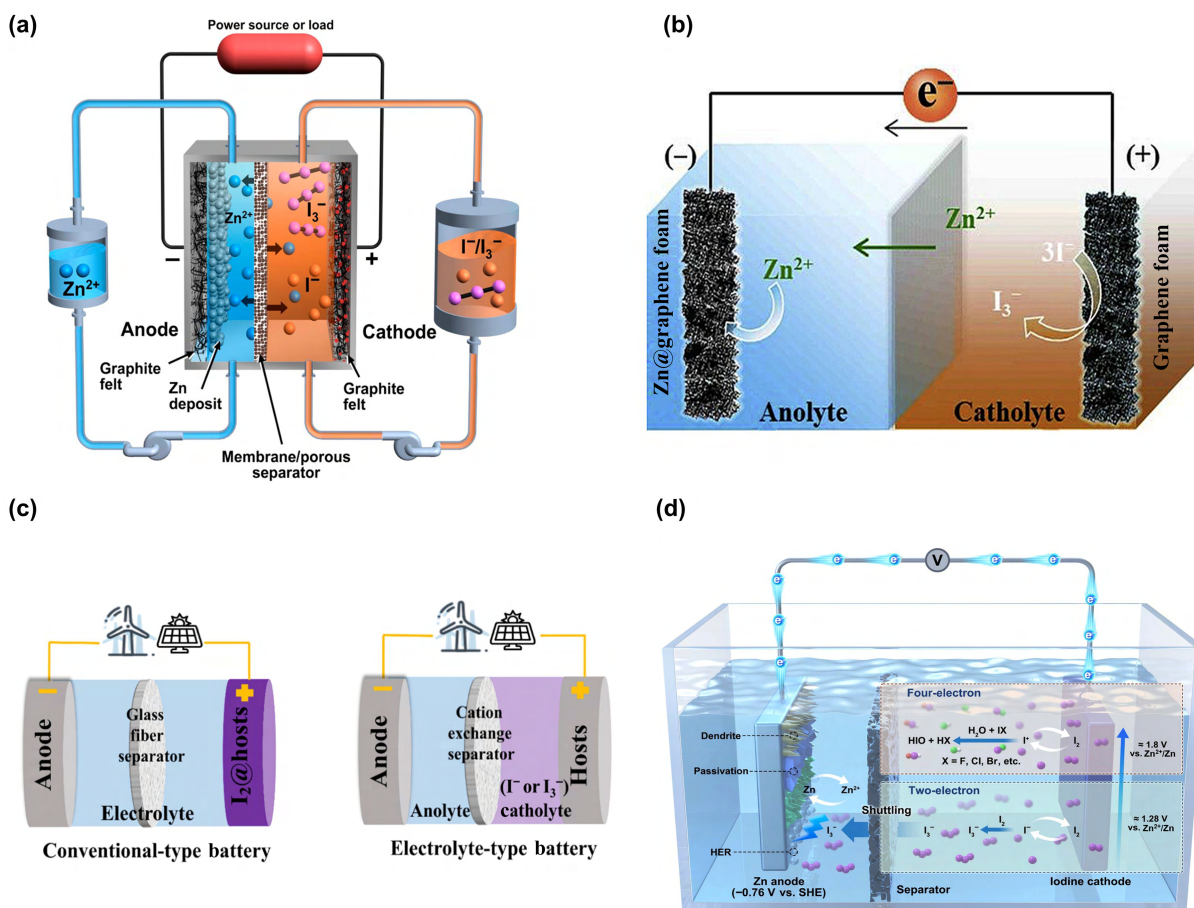


Figure 2 Common types of zinc-iodine batteries. (a) Schematic illustration of a ZIRFB device. Reproduced with permission from Ref. [23], © American Chemical Society 2016. (b) Schematic illustration of an SZIB device. Reproduced with permission from Ref. [24], © Elsevier Ltd. 2018. (c) Zinc-iodine batteries using different iodine sources. Reproduced with permission from Ref. [25], © Elsevier B.V. 2022. (d) Two-electron and four-electron systems in iodine batteries. Reproduced with permission from Ref. [26], © Wang, Y. et al. 2026.

the battery. However, this process also involves more complex reaction pathways and imposes more stringent requirements on material design. Many studies have realized four-electron iodine redox chemistry by introducing different intermediates, employing various electrolyte systems, and operating under different temperature conditions [27]. During charging, I_2 at the cathode undergoes oxidation to form I^+ , which subsequently reacts with halide ions to generate stable iodine halides. During discharge, I^+ species are successively reduced to I_2 and then further reduced to I^- [26, 28]. The corresponding electrode reactions and overall reaction are as follows:

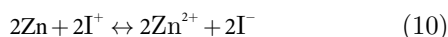
Anode



Cathode



Overall reaction



2.2 Advantages and challenges of zinc–iodine batteries

Zinc–iodine batteries commonly involve either two-electron or four-electron reaction systems. In the two-electron system, zinc–iodine batteries deliver a theoretical specific capacity of up to 211 mAh·g⁻¹, and exhibit a discharge plateau of approximately 1.3 V when assembled into a full cell with a Zn anode [8]. The liquid-phase conversion mechanism of the iodine cathode (I/I_3^-) endows zinc–iodine batteries with high rate capability [5, 29], allowing stable capacity retention over thousands of cycles. On this basis, the multivalent redox chemistry of iodine can be further exploited to construct a four-electron reaction system ($I/I_2/I^+$, 1.83 V vs. Zn^{2+}/Zn , theoretical specific capacity of 422 mAh·g⁻¹), thereby significantly improving the battery energy density [26].

However, the development of zinc–iodine batteries still faces numerous challenges. On the cathode side: (1) Elemental iodine exhibits an extremely low electrical conductivity of only 10^{-6} – 10^{-9} S·cm⁻¹ at room temperature, which severely limits the reaction kinetics and rate performance of the iodine cathode [30, 31]; (2) iodine suffers from poor thermal stability and is prone to sublimation, which can lead to the loss of active materials during electrode preparation [32]; (3) the redox reactions between iodide and polyiodide species involve multiple electron-transfer steps, resulting in sluggish reaction kinetics. Meanwhile, the shuttling of polyiodides toward the anode can induce self-discharge, which is further aggravated by the sluggish conversion kinetics; and (4) the limited loading of active materials compromises the energy density at the cell level [33, 34].

On the anode side: (1) The standard electrode potential of Zn/Zn^{2+} is lower than that of H_2/H^+ , making the Zn anode thermodynamically prone to the hydrogen evolution reaction (HER) in aqueous electrolytes. H_2 bubbles generated during HER can disrupt the electrode/electrolyte interfacial contact and thus compromise the stability of the Zn surface. Meanwhile, HER causes the local accumulation of OH^- on the Zn metal surface, leading to the formation of inactive $[Zn(OH)_4]^{2-}$ species, which consumes both the Zn anode and the electrolyte, resulting in decreased Coulombic efficiency (CE) and shortened cycling

life [35, 36]; (2) during the charge/discharge process of the Zn anode, Zn^{2+} undergoes dynamic deposition and stripping at the electrode/electrolyte interface. The uneven Zn^{2+} flux and electric-field distribution at the interface can readily induce the formation of Zn dendrites [37, 38]; and (3) Zn^{2+} exists in aqueous electrolytes as the solvated $[Zn(H_2O)_6]^{2+}$ structure, and its high desolvation energy barrier hinders the interfacial diffusion kinetics of Zn^{2+} , while also promoting hydrogen evolution and the formation of by-products [39].

3 Overview of COFs and their mechanism in zinc–iodine batteries

3.1 Introduction to COFs

COFs are a class of crystalline porous materials formed by linking organic molecular building units through covalent bonds, and are mainly composed of light elements, such as C, N, O, and B [16, 40, 41]. Benefiting from their highly ordered pore structures, tunable pore sizes, designable chemical compositions and functional sites, high specific surface areas, and excellent thermal and chemical stability, COFs have shown broad application prospects in the field of energy conversion and storage [15, 16, 42].

COF materials can be constructed from a variety of rigid organic building blocks with diverse structural configurations. The formation of strong covalent bonds through various organic synthesis reactions between these building blocks endows COFs with well-defined and predictable two-dimensional (2D) or three-dimensional (3D) crystalline structures [43–45]. Dimensionality is a key concept in COFs, which are generally classified into one-dimensional (1D), 2D, and 3D COFs. To date, many reported COFs are 2D structures. Topology is a major source of structural diversity in COFs; it governs in-plane charge transport and electronic coupling and introduces characteristic electronic band structures through linkage configurations [46]. So far, the main pore structures of covalent organic frameworks reported in the literature are triangular, tetragonal, pentagonal, or hexagonal pores (Fig. 3(a)). By integrating different types of pores into a single framework, heteropore COFs can be created through combinatorial effects (Fig. 3(b)) [47]. In addition, the stacking mode of 2D COFs plays an equally important role, modulating interlayer charge transport and electronic coupling through variations in relative slipping, interlayer spacing, and twisting between adjacent layers [46]. In 2D COFs, the stacking modes primarily include eclipsed (AA) stacking and staggered (AB) stacking [48, 49]. Compared with 2D COFs, the development of 3D COFs has been hampered by the limited range of elements accessible for constructing 3D structures and conducting intricate structural analysis [49]. In contrast, as low-dimensional organic systems, 2D COFs can rely on the tunability of their layers to achieve precise control over structure–property relationships [45, 46].

Wang et al. reported the excellent performance of 2D COFs with one-dimensional open channels in iodine capture, and pointed out that their high iodine loading capacity is primarily governed by accessible pore volume rather than by specific binding sites [50]. Some 3D COFs with conjugated structures have also shown high iodine adsorption capacity [51]. However, compared with 2D COFs, which can generally gain additional stabilization from interlayer π – π stacking, 3D COFs lack this interlayer stacking stabilization mechanism. As a result, their structural stability and crystallinity regulation are usually more challenging [52–54].

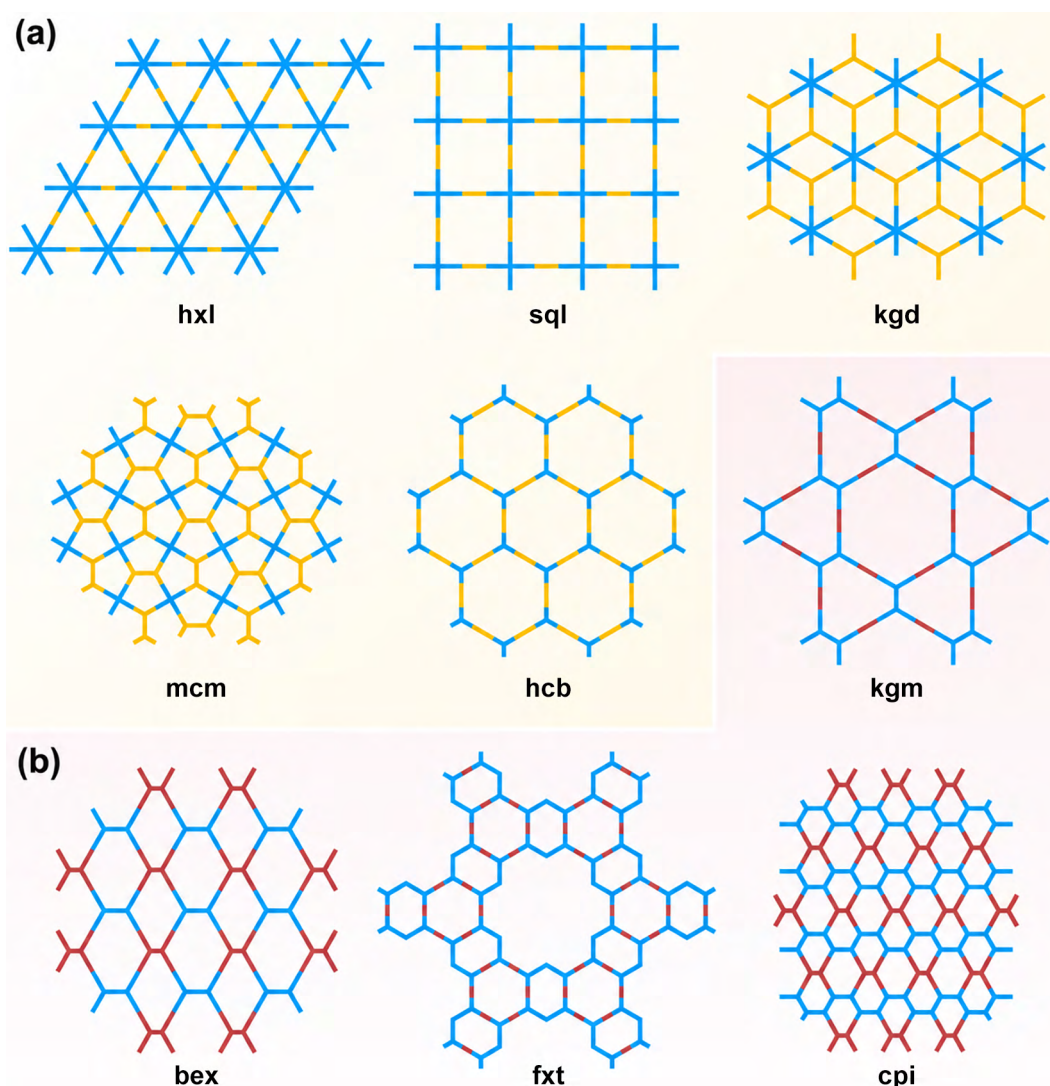


Figure 3 Reported topologies of 2D COFs. Reproduced with permission from Ref. [47], © American Chemical Society 2025.

3.2 Optimization mechanisms of COFs in zinc–iodine batteries

To address the key challenges currently faced by zinc–iodine batteries, COF materials have demonstrated multidimensional and synergistic optimization effects by virtue of their unique structural and chemical advantages. Overall, their modification mechanisms are mainly reflected in structural confinement effects, functional group modification, heteroatom modification, and the metal coordination (Fig. 4). These factors are coupled with each other, together constituting the core mechanism to optimize the performance of zinc–iodine batteries.

Structural confinement is one of the most fundamental and universal mechanisms by which COFs optimize the performance of zinc–iodine batteries [58]. Its essence lies in the precise regulation of the distribution, confinement, and migration behavior of iodine species through their regular and ordered pore structures and tunable pore sizes. Although conventional porous carbons have been widely used as iodine hosts owing to their high specific surface areas and high electrical conductivity, they are still inherently limited by the weakness of physical adsorption [59, 60]. In comparison, COFs offer a high degree of structural design flexibility at the framework level [42]. This enables them to efficiently load iodine while effectively suppressing polyiodide

diffusion and optimizing ion transport pathways [61–64]. Ion conduction plays a crucial role in the charge/discharge kinetics of rechargeable batteries, mainly involving ion diffusion within the electrode, ion transport through the electrolyte, and ion permeation across the electrode/electrolyte interface [65, 66]. Generally, the kinetics of ion transport are much slower than those of electron transport, making ion transport a key rate-controlling step in electrochemical reactions [67]. Owing to the unique advantages of COF materials, such as highly ordered directional channels, functional diversity, and robust structural stability, which endow COF-based ion conductors with low ion diffusion barriers and excellent temperature tolerance [68]. Their overall performance is therefore significantly superior to that of conventional inorganic or polymeric ion conductors [69–71]. In addition, the tunable pore channels of COFs also endow them with excellent molecular sieving and confinement capabilities. Because of their precisely tunable pore sizes, COFs can exploit size effects to physically confine iodine species, including I^- (~ 0.21 nm), I_2 (~ 0.27 nm), and I_3^- (~ 0.58 nm), within their pore channels. This confinement restricts the diffusion of iodine species into the electrolyte, reduces the formation of undesirable iodine-related species, improves redox reversibility, and ultimately leads to enhanced electrochemical performance [61, 62]. Specifically, micropores (< 2 nm) can effectively confine iodine molecules and

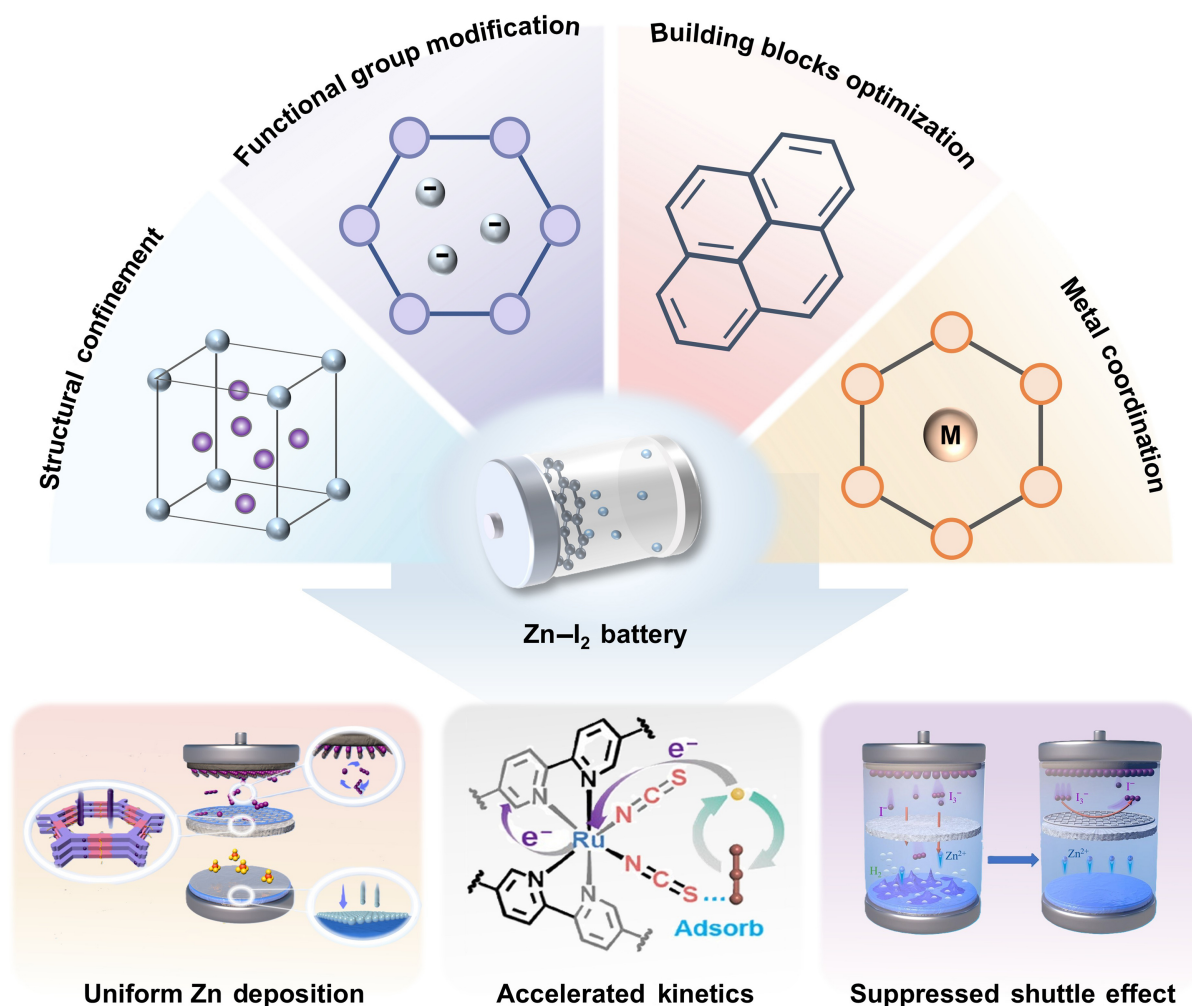


Figure 4 The optimization mechanism of COFs for zinc-iodine batteries. Reproduced with permission from Ref. [55], © Wiley-VCH GmbH 2025. Suppressed shuttle effect. Reproduced with permission from Ref. [56], © Elsevier Inc. 2025. Uniform Zn deposition. Reproduced with permission from Ref. [57], © American Chemical Society 2026.

polyiodides, whereas mesopores (2–50 nm) provide larger pore spaces that reduce the diffusion resistance of guest molecules. This allows iodine species to more readily access active sites, thereby improving mass transport and reaction kinetics [72–76].

In addition to structural confinement, the performance enhancement of zinc-iodine batteries by COFs is also closely related to their framework composition and pore-wall chemical environment. The core mechanism lies in the introduction of specific active sites, functional groups, and polar or ionic structural units into the framework, which regulates the electronic structure of the material and the local chemical environment within the pore channels. This regulation enhances the adsorption, confinement, and conversion control of iodine and polyiodide species, while further influencing Zn^{2+} transport and interfacial reaction behavior. The construction of active sites and functional groups in COFs mainly involves two strategies: bottom-up synthesis and post-synthetic modification [77]. The former refers to the direct selection of building units with specific functionalities during COF assembly, allowing functional sites to be incorporated into the framework simultaneously with framework formation. The latter involves the further introduction of desired functional units onto a preformed COF framework through chemical grafting, covalent reactions, or interfacial modification [78, 79]. Compared with the bottom-up predesign strategy, post-

synthetic modification offers greater flexibility, enabling more targeted regulation of the electronic structure, pore microenvironment, and surface properties of the material while preserving the original topology and porosity characteristics of the parent framework [77, 80].

4 Classification of COFs for zinc-iodine batteries

Because different COFs often possess multiple functions simultaneously, such as electron transport, polar adsorption, ion sieving, and catalytic conversion, classification based solely on electrical conductivity, redox activity, or catalytic activity may easily lead to overlap and logical ambiguity. Therefore, this review starts from the intrinsic framework structures of COFs and categorizes the related materials into four types: Tp/TpPa-based COFs, neutral conjugated-framework COFs, ionic COFs, and metallated COFs. The roles of different structural units and functional sites in polyiodide confinement, regulation of iodine conversion kinetics, and optimization of Zn^{2+} migration behavior are further discussed.

4.1 Neutral conjugated-framework COFs

Neutral conjugated-framework COFs mainly refer to a class of

COFs whose frameworks do not contain intrinsic charges or well-defined metal centers, but possess continuous π -conjugated characteristics. In studies on the optimization of zinc–iodine batteries, these COFs usually regulate the confinement, anchoring, and transport of iodine/polyiodide species through the synergistic effects of ordered π -conjugated frameworks, regular pore channels, and heteroatom-containing active sites. For ease of discussion, this review further classifies them into three categories: nitrogen-rich conjugated frameworks, extended π -conjugated frameworks, and neutral functional-group-regulated frameworks.

In this review, nitrogen-rich neutral-framework COFs refer to a class of COFs whose frameworks remain electrically neutral and are constructed from nitrogen-rich conjugated units. Their advantages mainly arise from the synergistic effect of “ordered channels + nitrogen-rich sites” [81]. On the one hand, the regular and continuous pore channels can provide uniform transport pathways for Zn^{2+} , thereby guiding homogeneous Zn^{2+} migration. On the other hand, triazine units in the framework can act as zincophilic groups and exhibit favorable adsorption toward Zn^{2+} ,

which promotes uniform Zn deposition on the Zn anode by preventing local ion accumulation [82, 83]. Meanwhile, the nitrogen-rich sites can also enhance the adsorption and confinement of I_2 and polyiodide species, which helps suppress the shuttle effect and improve the utilization efficiency of iodine species.

In a specific study, Huang et al. synthesized a neutral-framework TTA-DHTPA-COF (TTA denotes 4,4',4''-(1,3,5-triazine-2,4,6-triyl)trianiline and DHTPA denotes 2,5-dihydroxyterephthalaldehyde) rich in triazine and hydroxyl sites through Schiff-base condensation, and assembled it with a glass fiber (GF) substrate and a graphene layer via a two-step vacuum filtration method to construct a Janus separator, TTA-DHTPA-COF@GF@Gr, for separator optimization in zinc–iodine batteries (Fig. 5(a)). The one-dimensional ordered open channels and triazine/hydroxyl polar sites of TTA-DHTPA-COF can jointly construct a transport interface favorable for uniform Zn^{2+} migration and enhance the confinement of polyiodide anions, while the graphene layer further improves interfacial conductivity

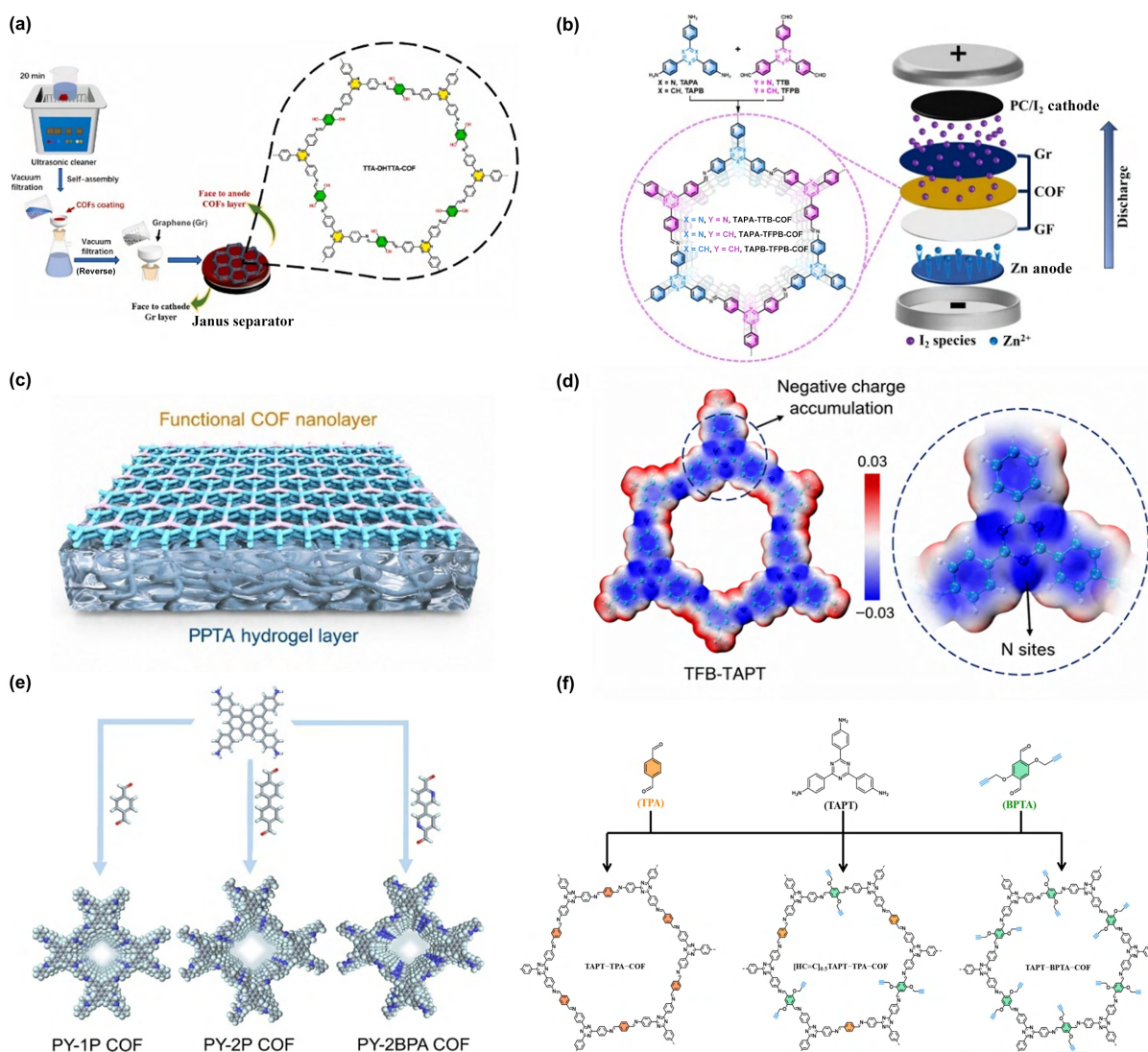


Figure 5 Summary of neutral conjugated framework COFs for zinc–iodine battery optimization. (a) The schematic diagram of the preparation of TTA-DHTPA-COF@GF@Gr Janus separator. Reproduced with permission from Ref. [84], © Elsevier B.V. 2024. (b) Schematic diagram of the assembly of Zn–I₂ battery with the proposed COF composite separators. Reproduced with permission from Ref. [56], © Elsevier Inc. 2025. (c) Schematic diagram of skin-QSSE. (d) ESP distributions of TFB-TAPT. ((c) and (d)) Reproduced with permission from Ref. [85], © The Royal Society of Chemistry 2025. (e) Synthesis of PY-1P, PY-2P, and PY-2BPA COFs. Reproduced with permission from Ref. [86], © Elsevier Inc. 2023. (f) Synthesis diagram of TAPT-TPA-COF (TPA denotes terephthalaldehyde), [HC≡C]_{0.5}TAPT-TPA-COF, and TAPT-BPTA-COF. Reproduced with permission from Ref. [57], © American Chemical Society 2026.

and iodine species conversion kinetics [84]. Building on this, Hu et al. further synthesized three nitrogen-rich neutral-framework COFs with similar topologies but different triazine contents, namely TAPA-TTB-COF (TAPA denotes 2,4,6-tris(4-aminophenyl)triazine and TTB denotes 4,4',4''-(1,3,5-triazine-2,4,6-triyl)tribenzaldehyde), TAPA-TFPB-COF (TFPB denotes 1,3,5-tris(4-formylphenyl)benzene), and TAPB-TFPB-COF (TAPB denotes 1,3,5-tris(4-aminophenyl)benzene), via solvothermal Schiff-base condensation. These COFs were then integrated with graphene/glass fiber through a two-step vacuum filtration method to construct Gr@COF@GF functional separators (Fig. 5(b)), enabling a systematic comparison of the influence of triazine site density on separator regulation. This work demonstrated that, for nitrogen-rich neutral-framework COFs, their role in separator optimization is not merely derived from the presence of nitrogen atoms. More importantly, the density of triazine sites can directly affect the zincophilicity of the framework and its ability to repel polyiodide species. Among them, TAPA-TTB-COF, with the highest triazine content, exhibited the most effective Zn^{2+} flux regulation and polyiodide anion suppression. Its ordered channels ensured uniform Zn^{2+} migration, while the abundant triazine sites further promoted Zn^{2+} diffusion and inhibited polyiodide anion shuttling [56]. Together, these two studies indicate that the essential role of nitrogen-rich neutral-framework COFs in separator optimization lies in the synergistic construction of an interfacial structure that integrates zincophilicity, iodine repellence, and ion-channel functionality through high-density triazine sites and ordered pore channels. This interfacial regulation effectively improves the cycling stability and areal capacity performance of zinc–iodine batteries.

In addition to their use in separators, nitrogen-rich neutral-framework COFs have also been employed to optimize cathodes and quasi-solid-state electrolytes. Cao et al. *in situ* integrated the triazine-rich neutral imine-linked COF TFB-TAPT (TFB denotes 1,3,5-triformylbenzene and TAPT denotes 4,4',4''-(1,3,5-triazine-2,4,6-triyl)trianiline) onto the surface of PPTA (PPTA denotes protonated poly(p-phenylene terephthalamide)) aramid nanofiber hydrogel in the form of nanosheets, thereby constructing a biomimetic skin-like quasi-solid-state electrolyte (skin-QSSE) (Fig. 5(c)). In this system, the role of TFB-TAPT as a nitrogen-rich neutral-framework COF is no longer limited to the simple confinement of iodine species. Instead, the negatively polarized electrostatic environment generated by the electron-rich N sites in the triazine framework can induce pronounced electrostatic repulsion and selective blocking toward iodide/polyiodide anions, such as I^-/I_3^- , thereby improving the utilization efficiency of active iodine and suppressing the shuttle effect (Fig. 5(d)). Meanwhile, the PPTA hydrogel layer reduces the Zn^{2+} desolvation energy barrier through its hydrogen-bonding network, promoting uniform Zn^{2+} migration and deposition, and thus mitigating Zn anode corrosion and dendrite growth. Benefiting from this asymmetric synergistic design of a “COF-based anion-sieving layer and hydrogel-based ion-regulating layer”, the assembled Zn– I_2 battery delivered a specific capacity of 201.5 $\text{mAh}\cdot\text{g}^{-1}$ at 0.5 C and maintained stable cycling for 45,000 cycles at 10 C, with a capacity decay rate of only 0.0018% per cycle. This work demonstrates that nitrogen-rich neutral-framework COFs are not only suitable for cathode and separator optimization, but also can serve as selective ion-regulating layers in quasi-solid-state electrolytes, enabling the simultaneous suppression of iodine shuttling and interfacial side reactions [85].

In addition to nitrogen-rich neutral frameworks, other

extended π -conjugated neutral frameworks and neutral functional-group-regulated COFs can also be used to optimize zinc–iodine batteries, although their application sites and functional emphases differ. For extended π -conjugated neutral frameworks, existing studies have mainly employed them as iodine hosts for cathodes. Li et al. constructed a semiconducting PY-2PBA (PY denotes 4,4',4''-(pyrene-1,3,6,8-tetrayl)tetraaniline and 2PBA denotes a bipyridine-based aldehyde building block) COF through a supramolecular mineralization strategy and employed it as a cathode host for Zn– I_2 batteries (Fig. 5(e)). In this COF, pyrene serves as an electron-rich building block and represents a commonly used structural unit for constructing semiconducting COFs [86]. Benefiting from its extended π -conjugated framework, hollow nanotubular structure, and pyridinic N sites, this material enables effective confinement of I_2 /polyiodide species and promotes polyiodide conversion, thereby alleviating the loss of active materials and shuttle-related issues on the cathode side. As a result, the battery retained a capacity of 156 $\text{mAh}\cdot\text{g}^{-1}$ after 2800 cycles at 2 $\text{A}\cdot\text{g}^{-1}$ [86]. In contrast, neutral functional-group regulation is not limited to the construction of cathode hosts, but also can be extended to separator interfacial optimization. Ren et al. developed an alkynyl-functionalized COF separator, TAPT-BPTA-COF (BPTA denotes 2,5-bis(2-propyn-1-yloxy)-1,4-benzenedicarboxaldehyde), to suppress polyiodide anion shuttling in zinc–iodine batteries (Fig. 5(f)). The authors pointed out that the electron-rich π system of terminal alkynyl groups can coordinate with the vacant 4s and 4p orbitals of Zn^{2+} , thereby facilitating rapid and homogeneous Zn^{2+} migration. Meanwhile, the introduction of alkynyl groups further improves the electrochemical stability of the COF coating. In addition, the ordered crystalline channels enable selective ion sieving. Therefore, this neutral functional-group regulation strategy mainly targets the synergistic mitigation of two separator-side issues, namely polyiodide shuttling and Zn^{2+} flux imbalance. As a result, the battery delivered an initial areal capacity of 8.4 $\text{mAh}\cdot\text{cm}^{-2}$ at 20 $\text{mA}\cdot\text{cm}^{-2}$ and retained 70.1% of its initial capacity after 1200 cycles [57].

In summary, the core design principle of neutral conjugated-framework COFs lies in the synergistic regulation of ordered conjugated structures and active sites, thereby achieving comprehensive optimization of electron transport, iodine species confinement, and Zn^{2+} migration behavior. These materials do not rely on charged frameworks or metal centers; instead, they mainly exploit their intrinsic electronic characteristics and pore-wall chemical environments to enhance the performance of Zn– I_2 batteries. Therefore, they possess unique advantages in balancing structural stability, electrical conductivity, and chemical confinement capability.

4.2 Tp/TpPa-based COFs

Tp/TpPa-based COFs represent one of the most commonly used COF structural backbones in current studies related to zinc–iodine batteries. These materials are typically constructed using Tp as the core aldehyde monomer, which condenses with various aromatic diamines or their derivative monomers to form stable β -ketoenamine-type frameworks [87–91]. Their common features are as follows: On the one hand, Tp units can provide high framework chemical stability and regular pore channels, thereby serving as stable hosts for the confinement of iodine species [88, 92]. On the other hand, the Tp/TpPa frameworks also possess high modifiability, allowing the pore-wall microenvironment, electronic structure, and Zn^{2+} transport behavior to be further

regulated by introducing electron-withdrawing groups, sulfonic acid groups, or metal/ion-exchange sites [30, 89–91]. Therefore, Tp/TpPa-based COFs not only appear frequently in the existing literature, but also have been developed as a relatively clear design pathway based on “framework backbone–functional site regulation”.

In this type of system, an important strategy is to regulate the electronic structure of the framework through neutral electron-withdrawing groups. Sun et al. used the TpPa framework as a platform and constructed TpPa, TpPa-SO₃H, and TpPa-NO₂ through side-group engineering, which were employed as iodine cathode hosts for zinc–iodine batteries. This work demonstrated that TpPa-based COFs can serve as iodine hosts by virtue of their stable β -ketoenamine frameworks and regular pore channels. Meanwhile, it further regulates the electronic structure of the framework through side-group modification. Among them, nitro-functionalized TpPa-NO₂ exhibited the best performance (Fig. 6(a)). This improvement was mainly attributed to the

introduction of electron-withdrawing nitro groups, which reduced the band gap of the framework (Fig. 6(b)), enhanced its electron affinity, and strengthened its adsorption capability toward polyiodide anions, thereby simultaneously improving iodine species confinement and redox kinetics. After electronic structure regulation, TpPa-NO₂ could more effectively suppress the dissolution of polyiodide anions, indicating that side-group engineering is an effective strategy for TpPa-based COFs to achieve both electronic structure optimization and chemical confinement of polyiodide anions [91].

Similarly, Tan et al. constructed a trifluoromethyl-functionalized Tp-based COF, namely F-COF (Fig. 6(c)), as an iodine cathode host. By introducing strongly electron-withdrawing trifluoromethyl groups into the framework, the authors regulated the electronic distribution and electrostatic potential (ESP) environment of the COF. Compared with the control sample TpBD-COF (BD denotes benzidine), F-COF exhibited stronger adsorption capability toward polyiodides

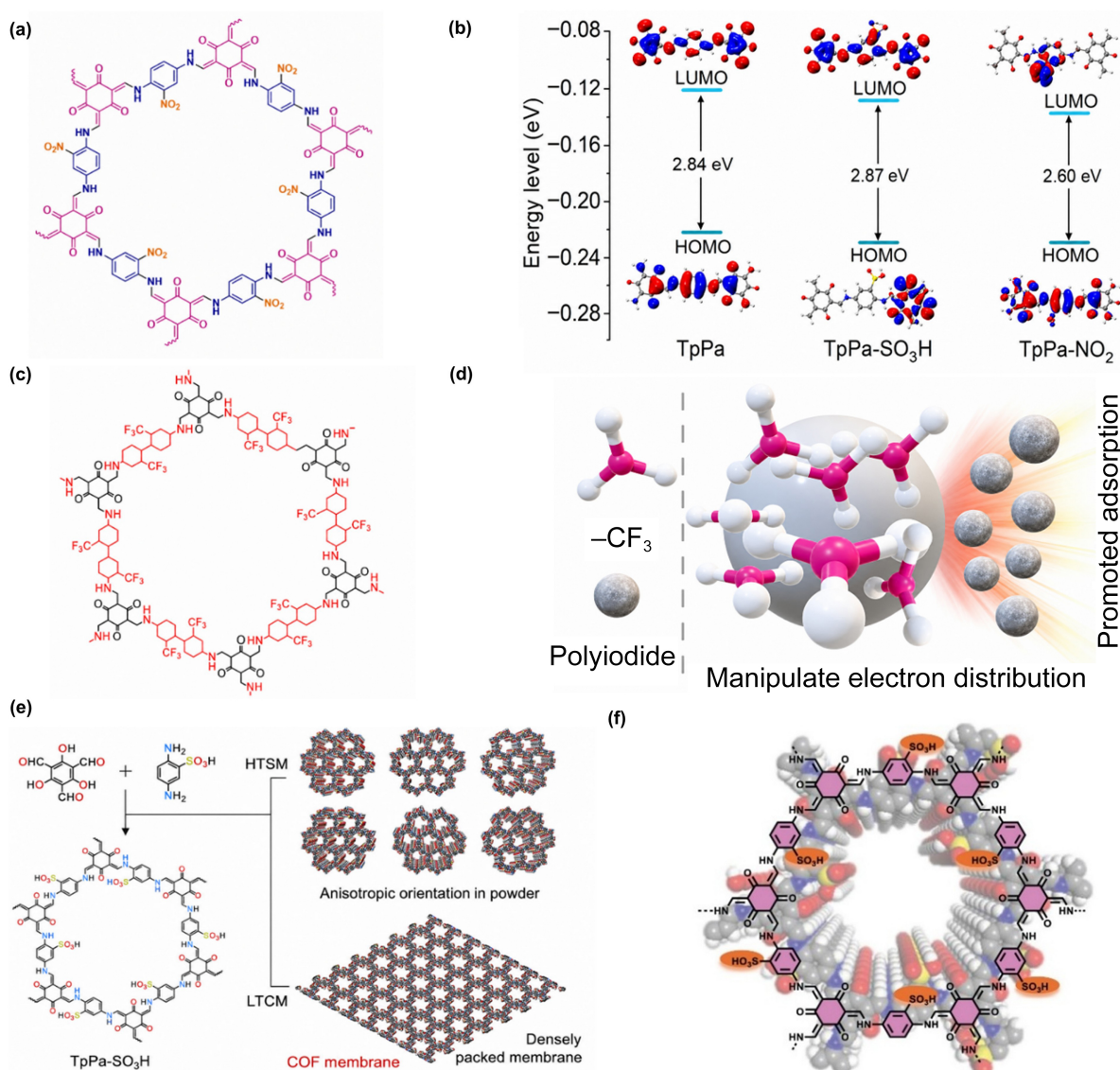


Figure 6 Summary of Tp/TpPa-based COFs for zinc–iodine battery optimization. (a) Molecular structure of TpPa-NO₂. (b) Illustrations of the energy levels, along with the corresponding optimized molecular structures. ((a) and (b)) Reproduced with permission from Ref. [91], © Wiley-VCH GmbH 2025. (c) Structure of F-COF. (d) Simulation of F-COF adsorption process. ((c) and (d)) Reproduced with permission from Ref. [90], © Wiley-VCH GmbH 2025. (e) Schematic diagram illustrating the preparation process of TpPa-SO₃H COFs. Reproduced with permission from Ref. [88], © Elsevier Ltd. 2025. (f) Structure of DMSBA-Tp-COF. Reproduced with permission from Ref. [92], © Institute of Process Engineering, Chinese Academy of Sciences 2025.

(Fig. 6(d)). This enhancement was mainly attributed to the electron-withdrawing effect of the trifluoromethyl groups, which increased the electrostatic potential of the framework and strengthened its affinity for polar iodine species, thereby more effectively suppressing polyiodide shuttling. Meanwhile, the electronically regulated F-COF also displayed more favorable charge-transport characteristics, which contributed to improved iodine redox reaction kinetics. These studies indicate that, for Tp-based COFs, regulating the framework electronic structure by introducing electron-withdrawing groups is an effective strategy to achieve both chemical confinement of polyiodides and optimization of reaction kinetics [90].

Another commonly adopted strategy is sulfonation regulation. Li et al. prepared TpPa-SO₃H (Fig. 6(e)) as a separator material for zinc-iodine batteries, demonstrating that Tp/TpPa-based COFs can acquire stronger ion-sieving capability after the introduction of sulfonic acid groups. This work showed that TpPa-SO₃H retains the regular microporous channels of the TpPa framework, and endows the pore walls with pronounced negative charge and a polar environment through -SO₃H groups. In particular, the densely packed and vertically aligned ordered channel structure significantly strengthens channel continuity and directional transport characteristics, allowing the framework to simultaneously achieve size-sieving and electrostatic-repulsion effects at the structural level. These structural features help restrict the migration of polyiodides, such as I₃⁻, while promoting rapid Zn²⁺ transport within the channels [88].

Huang et al. also developed a Tp/TpPa-based COF material for separator optimization (Fig. 6(f)), namely the sulfonic-acid-functionalized Tp-based COF, DMSBA-Tp-COF (DMSBA denotes 2,5-diaminobenzenesulfonic acid), further highlighting the structural modifiability of Tp-based frameworks. The authors pointed out that DMSBA-Tp-COF was constructed through the condensation of Tp units with a sulfonic-acid-containing diamine monomer, enabling abundant -SO₃H groups to be incorporated into the pore walls while maintaining the stability of the Tp-based β -ketoenamine framework. The resulting structure integrates ordered channels, negatively charged functional sites, and strong zincophilic characteristics. On the one hand, -SO₃⁻ sites can repel polyiodide anions through electrostatic interactions, thereby enhancing the restriction of intermediate migration within the framework. On the other hand, the uniform channels and abundant sulfonic acid groups facilitate homogeneous Zn²⁺ migration along the channels and further regulate the solvation structure and interfacial behavior of Zn²⁺. These studies demonstrate that introducing sulfonic acid groups into Tp/TpPa-based COFs is an effective structural design strategy for optimizing the pore-wall electrostatic environment, ion transport pathways, and polyiodide confinement [92].

4.3 Ionic COFs

iCOFs are a class of COFs featuring repeating units with ionic groups. They are fabricated either by assembling monomers bearing ionic groups or via post-functionalization of pre-synthesized neutral COFs. Based on the properties of the ionic groups, ionic COFs can be categorized into anionic and cationic types [93]. Previous reviews have pointed out that when charged groups are distributed along the inner walls of COF pores, their ion-regulation capability can be significantly enhanced: Anionic sites are favorable for cation migration, whereas cationic sites are more suitable for the adsorption, sieving, or electrostatic confinement of anionic species. For zinc-iodine batteries, this

structural feature of “charged pore walls plus ordered channels” is particularly important because it enables ionic COFs to strengthen the enrichment, anchoring, and confinement of polyiodide anions, such as I₃⁻ and I₅⁻, through electrostatic interactions. Furthermore, to some extent, it guides Zn²⁺ migration along ordered channels, thereby simultaneously suppressing the shuttle effect and regulating ion transport [70, 94]. Unlike neutral conjugated-framework COFs, which mainly rely on conjugated structures and neutral polar sites to regulate interfacial behavior, the core advantage of ionic COFs lies in their ability to directly regulate the transport and distribution of Zn²⁺ and polyiodides through electrostatic interactions. This feature is particularly critical in zinc-iodine battery systems, because polyiodides, such as I₃⁻ and I₅⁻, possess high solubility and mobility, which readily leads to the shuttle effect, self-discharge, and Zn anode corrosion [8, 95, 96]. Therefore, directly incorporating charged sites into COF frameworks, thereby endowing them with both ordered channels and electrostatic confinement capability, has become an important strategy for improving the cycling stability and Coulombic efficiency of Zn-I₂ batteries.

From a structural perspective, the design of ionic COFs can be mainly summarized into two strategies. The first strategy is the direct construction of cationic frameworks [97, 98]. Das et al. directly constructed a bipyridinium-based ionic COF, TAB-DNP-BP COF (TAB denotes 1,3,5-tris(4-aminophenyl)benzene, and DNP-BP denotes 1,1'-bis(2,4-dinitrophenyl)-4,4'-bipyridinium dichloride) (Fig. 7(a)), via the Zincke reaction and employed it as an iodine cathode host. Compared with conventional neutral COFs, the most prominent structural feature of this material is the incorporation of abundant viologen-type cationic sites into the framework, which endows the entire structure with stable positive charges. TAB-DNP-BP COF possesses regular hexagonal channels and good chemical stability, and more importantly, its cationic framework can generate strong electrostatic adsorption toward negatively charged polyiodides, such as I₃⁻ and I₅⁻, enabling rapid enrichment and stable confinement of polyiodides through an ion-exchange mechanism (Fig. 7(b)) [99]. Similarly, Zhao et al. constructed an ionic covalent triazine framework, CCTF-TPMB (CCTF-TPMB denotes a cationic pyridine-rich covalent triazine framework prepared from 1,2,4,5-tetrakis(4-cyanopyridinium-1-methylene)benzene-4Br⁺), rich in cationic pyridinium sites, and employed it as an iodine cathode host. This design reflects a synergistic strategy based on a nitrogen-rich triazine framework with cationic pyridinium sites (Fig. 7(c)). By simultaneously incorporating triazine units and abundant cationic pyridinium sites into the framework, a pronounced electrostatic potential gradient was established within the ordered porous structure, thereby strengthening the adsorption of polyiodides [100].

The second strategy involves first constructing a neutral framework and then introducing charged sites through post-synthetic modification, thereby converting the material into a genuine ionic COF [97, 98, 102]. Huang et al. constructed an ion-enriched triazine-based two-dimensional COF, DHTPA-TTA-DHMA-PSM-COF (DHMA denotes 2,5-dimethoxyterephthalaldehyde and PSM denotes post-synthetic modification) (Fig. 7(d)), through a post-synthetic modification strategy. This approach represents another typical design route for ionic COFs: A neutral imine-linked framework, DHTPA-TTA-DHMA-COF, is first constructed, followed by the grafting of quaternary ammonium cationic groups onto the pore walls through post-synthetic modification, thereby converting the material into a genuine ionic COF. In this work, DHTPA-TTA-DHMA-PSM-

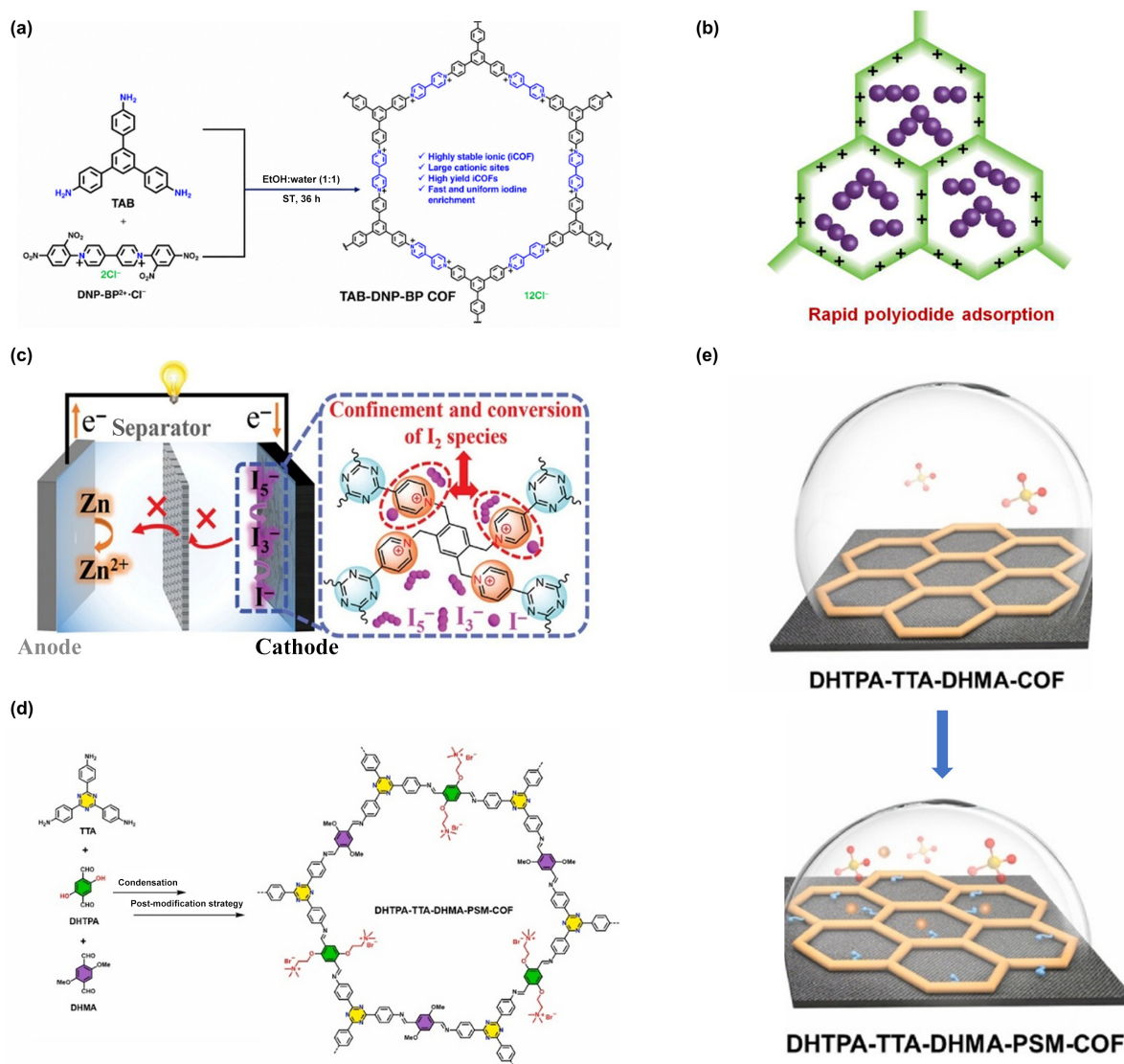


Figure 7 Summary of Ionic COFs for zinc–iodine battery optimization. (a) Schematic synthesis of TAB-DNP-BP COF. (b) Rapid polyiodide adsorption. ((a) and (b)) Reproduced with permission from Ref. [99], © Wiley-VCH GmbH 2025. (c) Schematic operation mechanisms of a ZIB with I₂@CCTF-TPMB cathode and Zn flake anode. Reproduced with permission from Ref. [100], © Wiley-VCH GmbH 2024. (d) Synthesis of DHTPA-TTA-DHMA-PSM-COF. (e) Schematic illustrations of the COFs accelerated Zn²⁺ transport and enhanced oxidation-reduction kinetics. ((d) and (e)) Reproduced with permission from Ref. [101], © Elsevier B.V. 2025.

COF was employed as an iodine cathode host for zinc–iodine batteries. After ionization, DHTPA-TTA-DHMA-PSM-COF retained its two-dimensional ordered open channels and high specific surface area, while high-density cationic sites were further introduced onto the pore walls, endowing the framework with both physical confinement and chemical adsorption capabilities. Compared with the non-ionized neutral precursor, the post-modified ionic framework exhibited higher iodine adsorption capacity and stronger polyiodide suppression. Meanwhile, the more hydrophilic and ion-conductive surface environment formed after pore-wall ionization facilitated rapid Zn²⁺ migration and improved the redox kinetics of iodine species (Fig. 7(e)) [101].

Notably, ionic COFs have so far been mainly employed for the construction of iodine cathode hosts, while their use in separator or electrolyte modification remains relatively limited. This is because polyiodides, such as I₃⁻ and I₅⁻, are initially generated and converted at the cathode/electrolyte interface, where the charged pore walls and ion-exchange sites of ionic COFs can more directly enable their electrostatic enrichment, anchoring, and confinement. Compared with neutral frameworks, ionic COFs provide stronger

regulation of polyiodides and are therefore more suitable as iodine cathode hosts, contributing to improved active iodine utilization and enhanced reaction reversibility.

4.4 Metallated COFs

MOFs are constructed by connecting metal ions or metal–oxo clusters with organic linkers through coordination bonds [103, 104]. Although MOFs offer a wide range of possible combinations of metal ions and organic linkers, their relatively weak coordination bonds still limit their practical applications [105, 106].

COFs are entirely composed of light elements, typically H, B, C, N, O, and Si, which are connected through covalent bonds [15]. The strength of covalent bonds is generally higher than that of coordination bonds, which endows COFs with highly stable structures under harsh chemical environments, including boiling water, strong acids and bases, as well as oxidative and reductive conditions [107–109]. However, COFs lacking metal centers are somewhat limited in functionality and may not fully meet the demands of more complex practical applications. The emergence

of metallated COFs has addressed this limitation by expanding their functional diversity. Metallated COFs are COF-based crystalline porous frameworks in which metal centers are deliberately incorporated into the framework architecture, pore-wall coordination sites, or metal-containing building units through direct synthesis, subcomponent self-assembly, or post-synthetic metalation [110]. Metallated COFs can integrate the respective advantages of MOFs and COFs, exhibiting a balanced combination of crystallinity, porosity, stability, and tunability, thereby achieving complementary features between these two classes of materials [110, 111].

The core feature of metal-covalent organic frameworks lies in their ability to construct multifunctional active sites through metal centers, which can simultaneously participate in adsorption, electron transfer, and catalytic conversion [110–113]. In zinc-iodine battery applications, after transition metals with multivalent characteristics, such as Co, Fe, and Ru, are introduced into COF frameworks, these metal centers can serve as efficient

“electron-transfer hubs” during redox processes, promoting iodine redox reaction kinetics through their reversible valence-state transitions [30, 55, 89, 114]. The introduction of metal centers enables COFs to function not merely as porous hosts, but further as reaction platforms integrating adsorption, catalysis, and transport regulation.

From a synthetic perspective, metallated COFs are mainly prepared through two strategies. The first strategy focuses on post-metalation or metal introduction to reconstruct the electronic structure of the framework. Feng et al. adopted a post-metallation strategy to coordinate Co ions into a pre-synthesized TAPT-Tp-COF framework, thereby constructing the metallated COF, Co-TAPT-Tp-COF, as an iodine host (Fig. 8(a)). This work reflects a triple-synergistic design concept of “conductivity–confinement–catalysis”: d–p orbital hybridization between the Co metal centers and the COFs framework reconstructs the electron-transport pathways and significantly enhances electrical conductivity. Meanwhile, it creates a more positive electrostatic potential

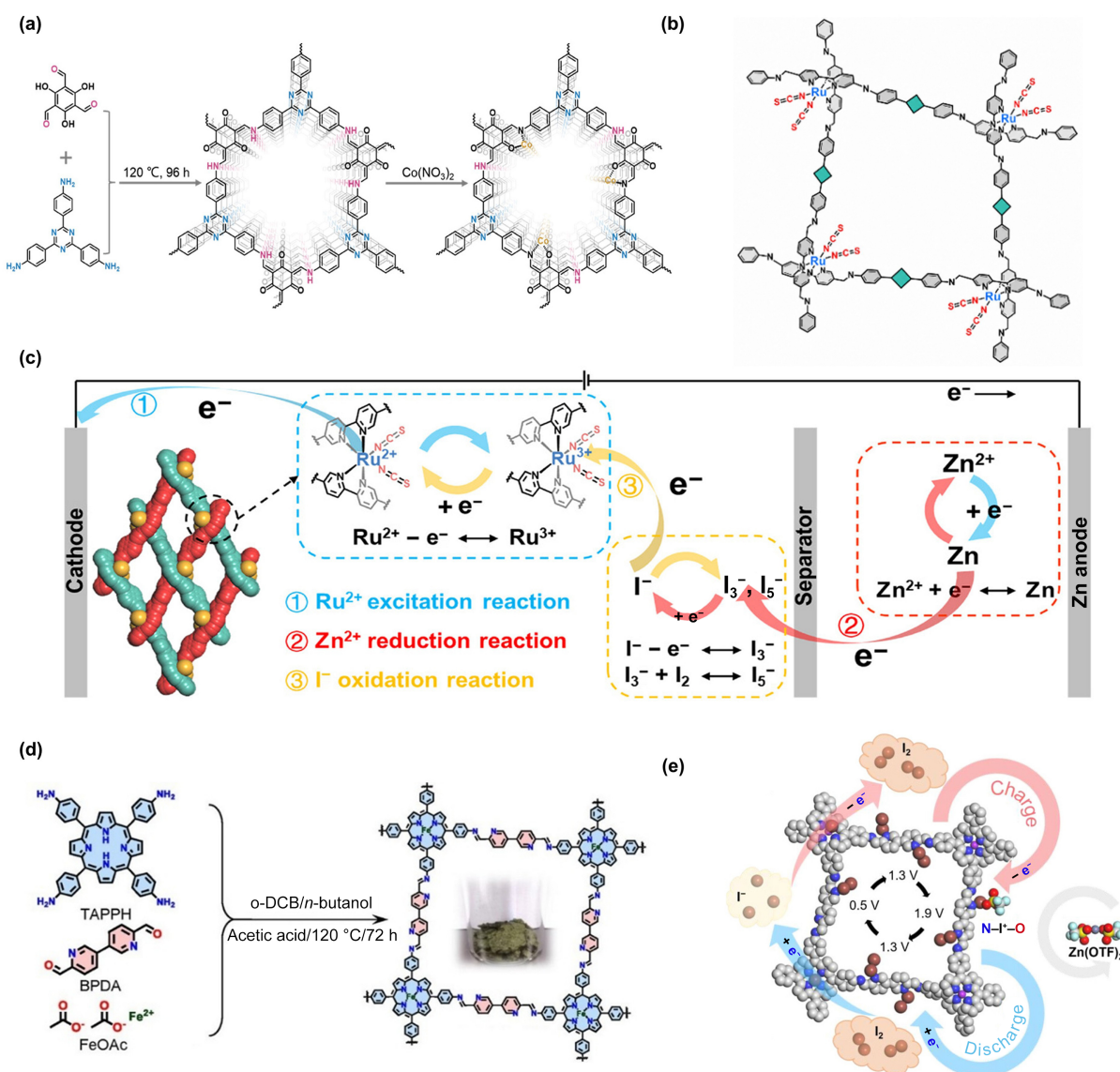


Figure 8 Summary of metallated COFs for zinc-iodine battery optimization. (a) Schematic illustrating the synthesis route of Co-TAPT-Tp-COF. Reproduced with permission from Ref. [30], © Wiley-VCH GmbH 2025. (b) Structure of COF-RuNCS-X. (c) The charging and discharging mechanism of the COF-RuNCS-X $\text{Zn}-\text{I}_2$ battery. ((b) and (c)) Reproduced with permission from Ref. [55], © Wiley-VCH GmbH 2025. (d) Schematic illustration of the synthesis of PPBY-Fe-COF. (e) Schematic diagram of iodine species conversion mechanism based on PPBY-Fe-COF. ((d) and (e)) Reproduced with permission from Ref. [114], © American Chemical Society 2025.

distribution on the framework surface, strengthening the chemical adsorption of polyiodides. In addition, the Co sites themselves serve as catalytic centers to lower the energy barrier of iodine redox reactions, thereby accelerating conversion kinetics [30].

The second strategy involves the preconstruction of metal-coordinated macrocyclic frameworks, in which the metal centers naturally become an integral part of the COF backbone. Feng et al. employed this strategy by directly weaving predesigned tetrahedral aldehyde complex monomers containing Ru(II) redox centers and thiocyanate (NCS) adsorption sites, thereby constructing a series of woven metallated COFs, COF-RuNCS-X (X denotes the serial number of the COF-RuNCS series, ranging from 1 to 6), as cathode materials for iodine redox regulation (Fig. 8(b)). This work reflects a synergistic design strategy by coupling Ru redox centers with S adsorption sites (Fig. 8(c)). Within the ordered porous framework, multivalent Ru(II) redox centers and abundant sulfur-based adsorption sites were simultaneously integrated: The sulfur sites enable efficient and selective adsorption of I_3^- and I_5^- through ion-dipole interactions, effectively suppressing the polyiodide shuttle effect; meanwhile, the reversible Ru(II)/Ru(III) valence transition serves as an additional redox-active center, significantly accelerating the conversion kinetics of iodine species [55, 115].

In addition to the above framework- or pore-wall-coordinated metallated COFs, metal-macrocycle-based metallated COFs are also important in zinc-iodine batteries. In this type of material, the metal center is not simply introduced into a preformed COF skeleton. Instead, it is located in a macrocyclic building unit, such as a metallophthalocyanine (MPc) or metalloporphyrin, and the metal-containing macrocycle further participates in the construction of the COF framework. Therefore, metallophthalocyanine- and metalloporphyrin-based COFs should also be considered as metallated COFs, because the metal centers are structurally integrated into the framework through well-defined M-N₄ (M denotes the metal center) coordination environments rather than physically loaded onto the COF surface. Guo et al. adopted an *in situ* metallation strategy, in which an iron source was directly introduced during the condensation synthesis of a porphyrin-bipyridine COF, enabling the one-pot construction of an Fe-coordinated COF, PPBY-Fe-COF (PPBY denotes porphyrin bipyridine), as an iodine host (Fig. 8(d)). From the viewpoint of metal location, PPBY-Fe-COF can be regarded as a metalloporphyrin-based metallated COF because the Fe centers are coordinated within porphyrin macrocycles. This work reflects a synergistic design concept involving Fe redox sites with conjugated C=N anchoring sites. Two types of functionally distinct active sites for iodine species were simultaneously integrated into the ordered porous framework: The cationic Fe sites served as catalytic centers for the I⁻/I⁰ redox reaction, accelerating iodine conversion kinetics through the Fe(II)/Fe(III) valence transition; meanwhile, the conjugated C=N sites, assisted by trifluoromethanesulfonate anion (OTF⁻), stabilized I⁺ species through the formation of N-I⁺-O bonds and suppressed their hydrolysis, thereby activating the high-valence I⁰/I⁺ redox couple and enabling continuous four-electron I⁻/I⁰/I⁺ conversion (Fig. 8(e)) [114].

Zhang et al. further reported two conductive COFs based on metal phthalocyanine/naphthalocyanine macrocycles, namely poly-zinc phthalocyanine (pZnPc) and poly-zinc naphthalocyanine (pZnNpc), and employed them as iodine-host cathode materials for aqueous Zn-I₂ batteries. From a structural perspective, these materials represent typical metal-macrocycle-based metallated

COFs, in which the Zn centers are embedded within the Zn-N₄ coordination environments of phthalocyanine or naphthalocyanine macrocycles, while the metal macrocyclic units are further interconnected through fully aromatic conjugated linkages to form ordered COF frameworks. The key feature of this design is not merely the confinement of iodine species within the pores, but also the enhancement of intrinsic charge transport through the extended π -conjugated structure and metal macrocyclic building blocks. This work proposed a host-guest donor-acceptor synergistic strategy. Upon incorporation of I₂ into pZnPc and pZnNpc, pronounced charge-transfer interactions were established between the COF framework and iodine molecules, leading to a significant increase in electrical conductivity. In particular, the conductivity of pZnPc increased by nearly two orders of magnitude after I₂ doping, reaching 1.03 S·m⁻¹. Meanwhile, the donor-acceptor interactions between the COF framework and I₂ lowered the energy barrier for iodine redox reactions, enhanced the electrochemical activity of iodine, and suppressed the shuttle effect of polyiodide intermediates. Electrochemical measurements demonstrated that I₂@pZnPc delivered a high specific capacity of 234.4 mAh·g⁻¹ in aqueous Zn-I₂ batteries, approaching the theoretical capacity of iodine, and maintained stable cycling performance for more than 40,000 cycles at 10 A·g⁻¹. These results indicate that metal phthalocyanine-based metallated COFs can serve as ordered porous iodine hosts and effective platforms for enhancing framework conductivity, iodine activation, and polyiodide confinement through electronic coupling between the π -conjugated metal macrocycles and iodine molecules, thereby providing a new design strategy for highly conductive and electrochemically active COF-based iodine cathodes [116].

In addition to being mainly used as iodine cathode hosts, metallated COFs have also shown noteworthy potential in separator functional layers. Overall, current studies on metallated COFs are still largely focused on the cathode side, where metal centers are mainly employed to enhance active iodine utilization and iodine conversion kinetics through polyiodide adsorption and catalytic regulation. However, the polyiodide shuttle in zinc-iodine batteries is essentially an interfacial and cross-interface process. It not only affects the stability of active species at the cathode side but also further interferes with Zn²⁺ transport and induces zinc anode corrosion and dendrite growth. Therefore, introducing metallated COFs into the separator interface is expected to simultaneously regulate polyiodide migration, iodine redox kinetics, and zinc deposition behavior at the transport-channel level, thereby enabling more comprehensive interfacial optimization. In this regard, Chen and co-workers developed separators modified with MPc-based COFs (MPc-COFs, M = Co, Ni, and Cu), and found that CoPc-COF exhibited the best zinc-iodine battery performance among them. The key advantage lies in the pronounced synergy between the Co active centers and the ordered COF framework. On the one hand, Co sites can provide stronger chemical adsorption toward polyiodides, such as I₃⁻, and promote the redox conversion of iodine species, thereby simultaneously suppressing the shuttle effect and improving reaction kinetics. On the other hand, the regularly arranged nanochannels in CoPc-COF offer more uniform transport pathways for Zn²⁺, alleviate local ion concentration inhomogeneity, and thus induce more uniform zinc deposition. In other words, this type of metallated COFs-modified separator not only serve as a simple physical blocking layer, but also establishes a cross-interface synergy between cathode-side polyiodide confinement

and anode-side deposition homogenization through a dual mechanism of “metal-site catalysis/adsorption + ordered pore-channel transport regulation.” This work demonstrates that the role of metallated COFs is not limited to the construction of iodine cathode hosts [117]; they can also function as multifunctional interfacial regulators through separator engineering. Compared with previous studies that mainly emphasized the role of metallated COFs in promoting iodine conversion at the cathode side, this separator-modification strategy further broadens the application scenarios of metallated COFs in zinc–iodine batteries. It also suggests that coupling metal active centers with the ordered channels of COFs is a promising route to simultaneously optimize polyiodide confinement, iodine redox kinetics, and Zn^{2+} transport behavior.

To provide a clearer comparison of the reported COF-based materials for aqueous $\text{Zn}-\text{I}_2$ batteries, representative examples discussed above are summarized in Table 1, including their structural features, application sites, dominant optimization mechanisms, and electrochemical performances.

Although considerable progress has been made in the optimization of zinc–iodine batteries by COFs in terms of adsorption/confinement, electronic structure regulation, and catalytic conversion, most existing systems still mainly focus on only one or two of these functions, and have not yet fully realized multifunctional synergy. The targeted introduction of metal sites into COF frameworks can endow the materials with integrated catalytic, adsorption, and confinement capabilities, representing a promising direction for the future development of zinc–iodine batteries. The four types of COFs discussed above have all shown beneficial effects in optimizing zinc–iodine batteries. In the era of rapid advances in artificial intelligence, machine learning (ML) can be introduced as an auxiliary tool to guide the optimization of COFs for zinc–iodine batteries. Starting from the key challenges of zinc–iodine batteries, machine learning may help screen suitable

building units, active sites, and metal sites for improving battery performance. In fact, the “materials genome” paradigm, in which ML accelerates COF development, has already been validated in multiple fields. Wang et al. systematically summarized the key roles of ML in COF screening, design, and performance prediction. By training models based on high-throughput computational or experimental databases, ML can rapidly predict the CO_2 capture capacity, CH_4 storage capability, gas separation selectivity, and catalytic activity of COFs, thereby guiding the rational design of high-performance COFs [118].

Similarly, ML can also be employed to computationally predict the adsorption capability of COF materials toward polyiodides. By taking iodine species adsorption/confinement capability, Zn^{2+} conduction efficiency, and redox catalytic activity as target properties, ML can efficiently screen optimal structures from large libraries of COF building units and metal-site combinations, thereby significantly accelerating the discovery and optimization of COFs for zinc–iodine batteries.

5 Summary and outlook

In summary, COF materials, owing to their highly designable framework structures, well-ordered pore architectures, and tunable pore-wall chemical environments, have demonstrated unique and multifaceted advantages in aqueous zinc–iodine batteries. Existing studies have shown that COFs can not only effectively confine iodine species and polyiodide intermediates through ordered nanoporous channels and tunable pore sizes, thereby suppressing the shuttle effect and active-material loss, but can also regulate the adsorption, conversion, and transport behavior of iodine species through framework active sites, polar/ionic groups, and local microenvironments, thus improving cathode reaction reversibility, enhancing the Coulombic efficiency, and prolonging cycling life. Meanwhile, some COF systems can

Table 1 Summary of electrochemical performance of COF-based materials for $\text{Zn}-\text{I}_2$ batteries

Compound	Application site	Reversible capacity (mAh·g ⁻¹)/current density (A·g ⁻¹)	Number of cycles/current density (A·g ⁻¹)/retention	High rate capability (mAh·g ⁻¹)/current density (A·g ⁻¹)	References
Skin-like QSSE	Quasi-solid-state electrolyte	201.5/0.5 C	45,000/10 C/99.998%	190.1 mAh·g ⁻¹ /0.5 C after 25 C	[85]
I_2 @CCTF-TPMB	Cathode	243/0.2	30,000/5/93.9%	180/5	[100]
I_2 @HPCOF@GO-22	Cathode	191.2/0.5	20,000/3/154.8 mAh·g ⁻¹ retained	125/5	[119]
F-COF- I_2	Cathode	197/1 C	40,000/50 C/99.7%	121/50 C	[90]
I_2 @Co-TAPT-Tp-COF	Cathode	221.6/1	130,000/15/82%	133.1/30	[30]
I_2 @PY-2PBA/CNT	Cathode	192.3/0.5	2800/2/156 mAh·g ⁻¹ after cycling	146.3/5	[86]
TpPa-SO ₃ H FSF COF	Separator	140.3/0.2	1200/0.2/82.2%	121.7/5	[88]
I_2 @N ₄ -COF	Cathode	348/1 C	7000/10 C/> 80%	272/10 C	[17]
COF-RuNCS-6	Cathode	395.8/1	5000/3/97.1%	208.1/5	[55]
I_2 @PPBY-Fe-COF	Cathode	240/1	8000/2/90.9%	173/10	[114]
Gr@DMSBA-Tp-COF@GF	Separator	3.2 mAh·cm ⁻² /20 mA·cm ⁻²	400/20 mA·cm ⁻² /2.7 mAh·cm ⁻² retained	4.3 mAh·cm ⁻² /20 mA·cm ⁻²	[92]
TpPa-Zn@GF	Separator	204.2/0.5 C	3000/10 C/86.1%	143.1/20 C	[89]
I_2 @TpPa-NO ₂ -COF	Cathode	183/0.1	10,000/5/84.3%	113/5	[91]
Gr@CoPc-COF@GF	Separator	208.6/0.571	1000/20 mA·cm ⁻² /99.948%	7.2 mAh·cm ⁻² /20 mA·cm ⁻²	[117]
I_2 @pZnPc	Cathode	234.4/0.5	40,000/10/108.6 mAh·g ⁻¹ retained	148.9/10	[116]

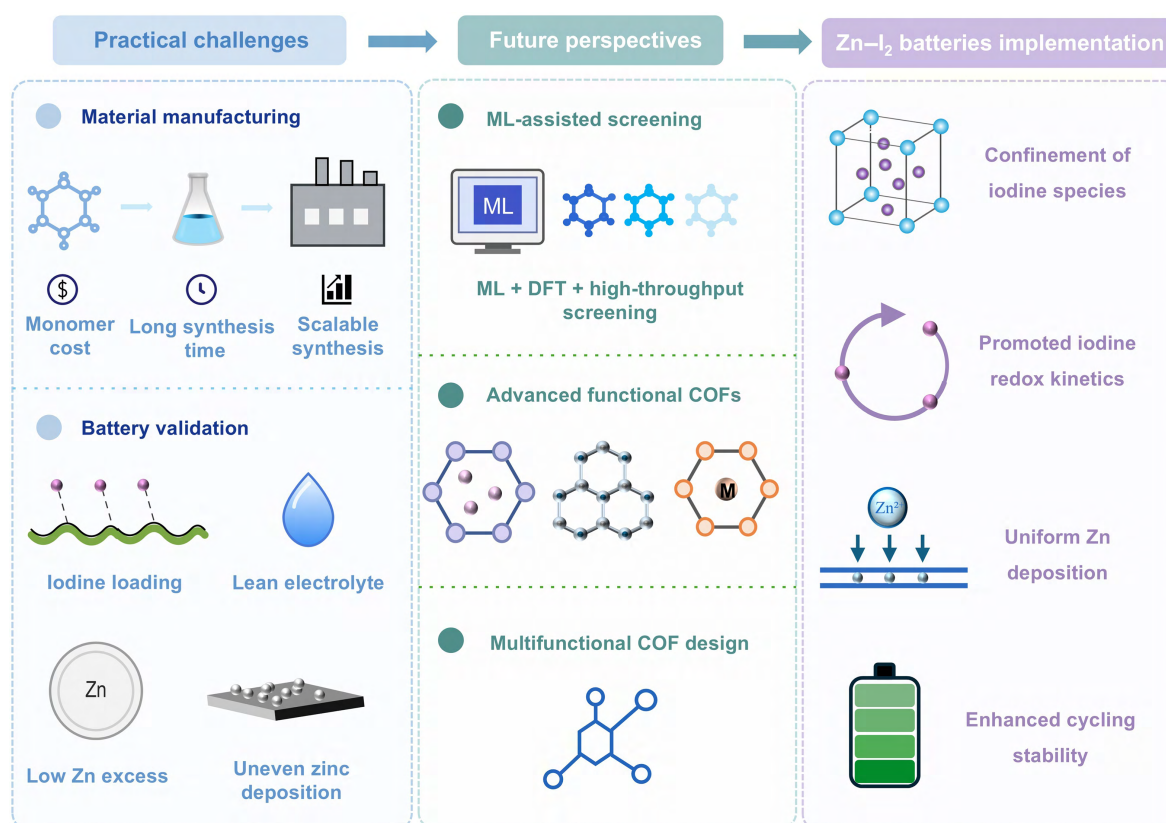


Figure 9 Summary and outlook.

also alleviate zinc dendrite growth and side reactions on the anode side to a certain extent by optimizing Zn^{2+} migration pathways, regulating interfacial ion flux, and promoting uniform deposition.

From the current state of research, the application of COFs in zinc-iodine batteries is still mainly focused on iodine cathode hosts and functional separator layers. Among them, ionic COFs show prominent advantages in the electrostatic confinement of polyiodides, Tp/TpPa-type COFs and neutral conjugated framework COFs exhibit great potential in pore engineering, electronic-structure regulation, and interfacial chemical adsorption, while metallated COFs have begun to show unique advantages in promoting iodine redox kinetics. Overall, COF materials have gradually evolved from early iodine hosts relying solely on physical confinement into a class of key framework materials integrating multiple functions, including confinement, adsorption, catalysis, and transport regulation, thereby providing important guidance for the construction of high-performance aqueous zinc-iodine batteries. In addition, to more intuitively present the recent research progress of COF materials in aqueous zinc-iodine batteries, representative materials and their electrochemical performances reported in recent studies are summarized in Table 1. This table helps to further reveal the differences among various COF systems in terms of capacity, rate capability, and cycling stability from a comparative perspective.

Nevertheless, research on COF materials for applications in zinc-iodine batteries is still at a developing stage, and several key issues remain to be addressed before practical application can be realized. First, most current COFs still mainly rely on pore confinement and framework polar sites to regulate polyiodides. However, under complex electrochemical environments, a single mode of action is often insufficient to simultaneously meet the multiple requirements of high iodine loading, strong adsorption,

fast conversion, and long cycle life for AZIBs. Therefore, one important future direction is the introduction of metal sites into COF frameworks to construct metal-coordinated/metallated COFs. The incorporation of metal centers is expected to endow COFs with stronger multifunctional synergy. On the one hand, metal sites can serve as catalytically active centers to accelerate the reversible conversion among iodine species, such as $\text{I}_2/\text{I}_3^-/\text{I}^-$, lower reaction barriers, and improve rate performance. On the other hand, the synergistic interaction between metal sites and framework heteroatoms or organic ligands may further enhance the adsorption and anchoring of polyiodides, optimize the local electronic structure, and regulate Zn^{2+} migration behavior within pores or at interfaces. In other words, metallated COFs are expected to realize multifunctional coupling of “adsorption–confinement–catalysis–transport regulation”, thereby overcoming the limitations of traditional non-metallated COFs in promoting reaction kinetics and regulating electrochemical interfaces.

In addition, as COF structures become increasingly complex, it is becoming difficult to efficiently identify optimal material systems solely through empirical trial-and-error design. In the future, greater efforts should be devoted to applying ML and high-throughput computation in COF design, so as to predict and screen candidate structures suitable for zinc-iodine batteries on a broader scale. By incorporating key descriptors into predictive models, including framework topology, pore-size parameters, specific surface area, heteroatom distribution, metal-site type, electronic-structure features, as well as the adsorption energies, diffusion barriers, and reaction barriers of iodine species/ Zn^{2+} , it should be possible to establish quantitative structure–property–performance relationships. With the assistance of ML, optimal COF systems that combine high iodine-loading capability, strong polyiodide confinement, excellent Zn^{2+} transport performance,

and rapid iodine redox kinetics could be rapidly identified from a vast space of building blocks, functional groups, and metal-site combinations, thereby significantly improving material design efficiency and reducing blind synthesis and screening costs. Furthermore, by combining density functional theory (DFT) calculations, molecular dynamics simulations, and *in situ* characterization techniques, a closed-loop research paradigm integrating theoretical prediction, material construction, and mechanistic validation may be achieved in the future.

Overall, the development of COF materials for aqueous zinc–iodine batteries should evolve from single-function optimization toward multifunctional synergistic design, and from experience-driven approaches toward the parallel advancement of data-driven and intelligent design strategies (Fig. 9). In particular, endowing COFs with stronger catalytic and interfacial regulation capabilities through metalation strategies, together with achieving precise structural prediction and efficient screening via ML, is expected to promote COF-based materials from laboratory research toward practically applicable aqueous zinc–iodine battery systems with high performance and long service life.

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Declaration of conflicting interests

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

Data availability

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

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Yixin Zhang received her B.S. in Chemistry from Hefei Normal University in 2025. She is currently a Master’s student in Chemical Engineering at Guangxi University. She has co-authored one research paper as second author in *eScience*. Her current research interests focus on COF-based aqueous zinc-ion batteries, particularly zinc–iodine systems, zinc anode interface protection, and functional electrolyte additives.



Qichun Zhang received his B.S. degree from Nanjing University in 1997, an M.S. from the Chinese Academy of Sciences in 2000, an M.S. from the University of California, Los Angeles in 2003, and a Ph.D. from the University of California, Riverside in 2007. He then worked as a postdoctoral researcher at Northwestern University before joining Nanyang Technological University as an Assistant Professor in 2008, where he later became an Associate Professor. He is currently a Professor in the Department of Materials Science and Engineering at City University of Hong Kong. He serves as an Associate Editor of *Materials Chemistry Frontiers* and is on the editorial boards of several journals. He has received awards, including the Mitsui Chemicals Catalysis Science Award, and has been recognized as a Highly Cited Researcher. He has published over 650 papers in journals indexed by the Science Citation Index (SCI), such as *J. Am. Chem. Soc.*; *Angew. Chem., Int. Ed.*; and *Adv. Mater.* He has an h-index of > 126. His current research interests include organic functional materials, conjugated small molecules and polymers for optoelectronics, and energy storage devices.



Huibing He received his Ph.D. degree from Wuhan University in 2016. He worked at Tianjin EV Energies Co., Ltd. (2017–2019) and then as a Postdoctoral Fellow at the University of British Columbia (2019–2020). In 2020 he joined Guangxi University, where he is currently an Associate Professor and Doctoral Supervisor in the College of Chemistry and Chemical Engineering, and a Visiting Scholar at City University of Hong Kong (2025–2026). He serves as an Associate Editor of *Battery Energy* and a Youth Editorial Board Member for *iScience* and *eScience*. He has led over 10 research projects, including the National Natural Science Foundation of China (NSFC) General and Youth Programs. He has published over 90 papers in journals indexed by the Science Citation Index (SCI), such as *Angew. Chem., Int. Ed.*; *Adv. Mater.*; and *Adv. Energy Mater.* He has authored one book chapter on lithium-ion batteries. His current research interests focus on electrochemical energy storage and catalysis.