Research Progress of Covalent Organic Framework (COF) Based Anode Materials in Metal Ion Batteries

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Abstract. Covalent organic frameworks (COFs), characterized by structural designability, ordered porosity, and tunable redox activity, have emerged as promising electrode materials for metal-ion batteries (LIBs, SIBs, and ZIBs). This review summarizes the progress and challenges of COFs in lithium-, sodium-, and zinc-ion batteries. In LIBs, COFs like TRO-BT-COF offer high capacity via redox-active sites (e.g., C=O, C=N), but suffer from low conductivity, structural rigidity, and complex synthesis. For SIBs, materials such as BCOF-1 exhibit pseudocapacitive behavior and ordered pores, yet face sodium-ion induced swelling and redox site complexity. In ZIBs, COFs like HqTp-COF enable Zn^{2+} storage through coordination with functional groups, but hydrolytic instability in aqueous electrolytes and inefficient ion desolvation limit performance. Key challenges across systems include poor electrical conductivity, electrolyte compatibility issues, and scalability of synthesis. Potential solutions involve integrating conductive additives, engineering flexible/hierarchical structures, and developing hydrophobic or dynamic covalent frameworks. By addressing these limitations, COFs could unlock high-performance, sustainable metal-ion batteries for next-generation energy storage.

Keywords: Covalent organic frameworks; Lithium-ion batteries; Sodium-ion batteries; Zinc-ion batteries; Electrode materials.

1. Introduction

In the context of global warming and the ever - increasing energy demand, the development of new energy vehicles has become an essential strategy and trend to address sustainability and environmental issues for next generation. The new energy vehicle industry has experienced substantial and prosperous growth in recent years. According to the International Energy Agency (IEA), the number of electric vehicles on the road surpassed 16.5 million globally by the end of 2020, and this number is projected to reach 145 million by 2030 [1]. With the large scale development of new energy vehicles and the application of various new type batteries, there is an escalating demand for high performance batteries. These batteries are required to possess high energy density, safety, and economic efficiency [2].

Metal-ion batteries, including lithium ion batteries (LIBs), sodium ion batteries (SIBs) and zinc ion batteries (ZIBs), have been at the forefront of energy storage research. LIBs, which have been extensively used in consumer electronics and electric vehicles for numerous years, have achieved remarkable success[3]. However, the development of metal ion batteries still confronts a large number of challenges. The new pursuit of high-performance batteries necessitates high energy density, high cycle reliability, and low cost [4]. For example, the limited theoretical capacity of traditional graphite anodes in LIBs (372 mAh g^{-1}) restricts the further improvement of energy density [5]. In addition to LIBs, SIBs and ZIBs are also being actively explored. SIBs are regarded as a potential alternative to LIBs due to the abundant sodium resources. However, they also face issues such as the large ionic radius of sodium ions, which could cause significant volume changes during charge discharge processes, thereby affecting cycle stability [6]. ZIBs also encounter similar problems in electrode material design and electrochemical performance optimization [7]. Therefore, the development of novel electrode materials has been an ongoing pursuit.

Covalent organic frameworks (COFs), a class of porous crystalline organic materials discovered by Yaghi and his colleagues in 2005 [8], have emerged as promising candidates for electrode materials in metal - ion batteries. COFs are constructed by covalently linking organic building units, forming porous frameworks with periodic structures. They are composed of light elements such as H, B, C, N, and O, resulting in a low weight density [9]. The emergence of COF has significantly broadened the scope and boundaries of porous organic polymers and provided a new platform and opportunity for the development of materials science, particularly in the interdisciplinary and cross disciplinary field of materials. Compared with traditional graphite anodes, COFs possess several advantages. Their flexible structure can better accommodate the volume changes during the insertion and extraction of metal ions, enhancing the cycle stability. For instance, Zhang et al. reported that a specific COF-based anode material exhibited a significantly enhanced cycling stability in LIBs, maintaining a high capacity after 500 cycles [10]. COFs also show high energy density potential due to their unique porous structure and high surface area, which can provide more active sites for ion storage [11]. Additionally, their high stability, attributed to the strong covalent bonds in the framework, makes them suitable for long - term operation in batteries [12]. A recent review by Chen et al. summarized the applications of COF and its derivatives in lithium - sulfur batteries, highlighting the advantages of COF - based materials in improving battery performance [13]. Another study by Yu et al. discussed the synthesis methods of COFs and their applications in rechargeable zinc - ion batteries, further demonstrating the potential of COFs in metal - ion batteries [14].

This review aims to comprehensively discuss the current implementation schemes of COF electrode materials in various metal - ion battery anodes. In the following sections, different cases of COF - based electrode materials in LIBs, SIBs, and ZIBs will be presented. Then, an in - depth analysis of the existing problems in the application of COF electrode materials, such as low electrical conductivity, partial dissolution in electrolytes, complex synthesis processes, and limited understanding of redox mechanisms, will be carried out. Based on the analysis, corresponding suggestions for improving the performance of COF electrode materials will be proposed. Finally, a conclusion will be drawn to summarize the research status and future development trends of COF electrode materials in metal ion batteries.

2. Case Description

2.1. COFs in LIBs

Lithium - ion batteries (LIBs) are widely used in modern electronic devices and electric vehicles. COF materials suitable for LIBs generally possess abundant redox - active sites, such as carbonyl (C=O) and imine (C=N) groups. These sites can reversibly undergo redox reactions during the charging and discharging processes, enabling the storage and release of lithium ions, thus providing a relatively high capacity. Additionally, a large specific surface area and ordered pore structures are common features of such COFs. The large specific surface area increases the contact area with the electrolyte, facilitating ion transport, while the ordered pores serve as fast - diffusion channels for lithium ions, enhancing the rate performance of the battery. For example, some two - dimensional COF materials with an appropriate interlayer spacing in their layered structures can ensure both high-efficiency ion transport and structural stability, demonstrating excellent electrochemical performance in LIBs.

2.2. COFs in SIBs

Sodium-ion batteries (SIBs) have attracted significant attention due to the abundance of sodium resources. COFs suitable for SIBs usually have large pore sizes and adjustable pore structures to accommodate the relatively large ionic radius of sodium ions (1.02 Å). These COFs often contain heteroatoms like nitrogen and oxygen, which can enhance the adsorption and storage of sodium ions through interactions with them. Moreover, COFs with flexible framework structures show advantages in SIBs, as they can better buffer the volume changes during the insertion and extraction of sodium ions, improving the cycling stability of the battery. Some COFs with multiple active groups and a certain degree of flexibility in their frameworks have exhibited high capacities and good cycling performance in SIBs.

2.3. COFs in ZIBs

Zinc-ion batteries (ZIBs) have received much attention for their high safety and low cost. COFs suitable for ZIBs usually contain abundant groups that can coordinate with zinc ions, such as carbonyl and amino groups, enabling the reversible storage of zinc ions through coordination. Furthermore, the chemical stability of COF materials in aqueous electrolytes is of great significance, as they need to maintain structural stability in complex aqueous environments. COFs with rigid and porous structures can meet this requirement. The rigid structure ensures that it is not easily deformed during the charging and discharging processes, and the porous structure provides space for the diffusion and storage of zinc ions. Some COFs based on benzoquinone and phenanthroline have demonstrated good electrochemical performance and cycling stability in ZIBs.

3. Analysis and Challenges

3.1. COFs as anode in LIBs

In the study by Wang et al. [15], a highly crystalline 2D truxenone - based COF (TRO - BT - COF) was synthesized for LIBs cathode. TRO and BT monomers were used, with acetic acid as the catalyst and 1,4-dioxane and mesitylene as solvents. The synthesis was carried out via a solvothermal stirring method, and the reaction conditions like atmosphere, time, temperature, catalyst concentration, and post - processing were optimized to enhance crystallinity. The design, synthesis process and obtained COF was shown in Fig. 1.

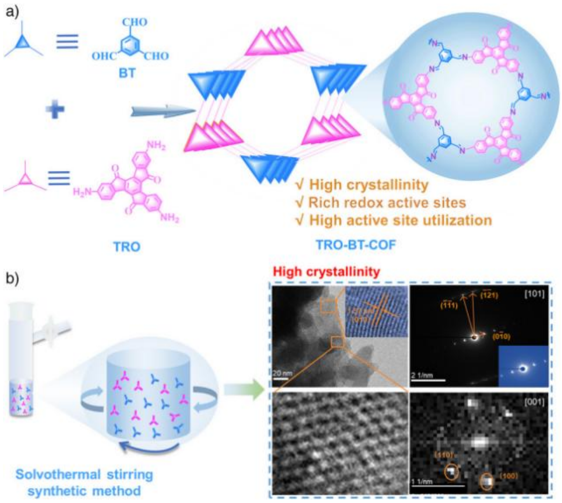

Fig. 1 Design and synthesis of TRO-BT-COF. a) Structures of TRO, BT, and TRO-BT-COF. b) Schematic diagram of solvothermal stirring synthetic method and corresponding HRTEM, SAED, and FFT of the TRO-BT-COF[15].

The obtained TRO - BT - COF was characterized by TEM, SEM, HRTEM, XRD, FTIR, ^{13}C NMR, XPS, N_2 adsorption - desorption, and TGA. In - situ FTIR, in - situ Raman microscopy, EPR, ex-situ XPS, and in-situ UV-vis spectroscopy were used to explore the charge - discharge mechanism. Electrochemical performance was tested in a battery with Li metal anode and 1.0 m LiTFSI in 1,3 - dioxolane and 1,2 - dimethoxyethane (1:1) electrolyte and the result showed in Fig. 2. The TRO - BT - COF showed a lamellar morphology with high crystallinity, good thermal and chemical stability. In - situ characterizations and DFT calculations revealed that C═O and C═N groups were redox active sites. Its electrical properties can be well observed from Fig. 3. As a cathode, it delivered a high discharge capacity of 435 mAhg^{-1} in the third cycle and maintained 206 mAhg^{-1} after 400 cycles. The rate performance at 2 C was 114 mAh g^{-1}[15]. The researchers concluded it was a promising cathode but needed performance improvement.

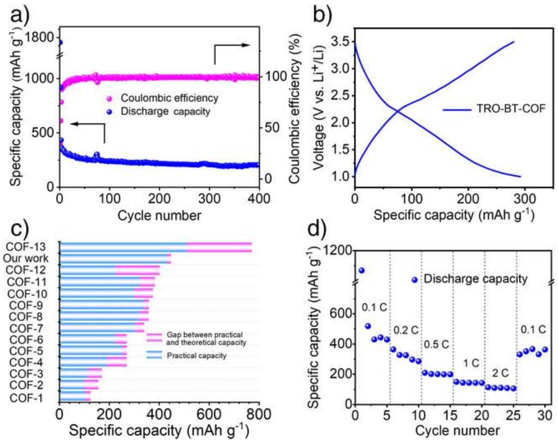

Fig. 2 Electrochemical performance of TRO-BT-COF as cathode material in lithium batteries. a) Cycling performance and b) typical discharge/charge curves of TRO-BT-COF. c) Comparison of the practical capacity and the gap between practical and theoretical capacity of TRO-BT-COF and previously reported representative COF cathode materials for lithium batteries. d) Rate performance of TRO-BT-COF[15].

While TRO-BT-COF demonstrates promise for LIBs, a notable limitation is its low intrinsic electrical conductivity, which restricts electron transport and leads to performance decline at high current densities. The material's reliance on π-π stacking for conductivity results in insufficient charge transfer kinetics, evident in its rate performance dropping to 114 mAhg^{-1} at 2 C. Additionally, the structural rigidity of its crystalline framework hinders efficient lithium-ion diffusion, causing capacity decay during prolonged cycling due to limited interlayer ion accessibility.

3.2. COFs in SIBs

Shehab and El-Kaderi [16] prepared BCOF-1 for SIBs by reacting HATNHA with TA in 1,4 - dioxane and mesitylene, using acetic acid as a catalyst at 120 °C for 5 days. The product was purified through a series of washings. To analyze BCOF-1, they employed PXRD, HRTEM, nitrogen adsorption/desorption isotherms, ATR-IR, and ^{13}C SS-NMR. Electrochemical measurements such as CV, galvanostatic charge-discharge, rate capability, cycling stability tests, and EIS were carried out in a sodium-metal half - cell with BCOF-1 as the working electrode. The synthesis process was shown in Fig. 3.

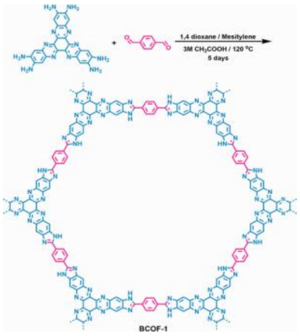

Fig. 3 Synthesis of BCOF-1 by the Condensation Reaction between HATNHA and Terephthalaldehyde[16].

BCOF-1 demonstrated excellent performance in SIBs. From Fig. 4, a high theoretical specific capacity of 392 mAhg^{-1} could be achieved, and in the first charge at 0.1 C, it reached 387mAh g^{-1}. The charge/discharge capacities were highly reversible from the second cycle. The material also showed good rate capabilities. At 15 C, it still had a discharge capacity of 50 mAhg^{-1}. Over 400 cycles at 3 C, BCOF-1 had a Coulombic efficiency close to 100% and retained ~77% of its capacity[16]. Its pseudocapacitive-like behavior was beneficial for high-power applications. The researchers concluded that BCOF-1 was a promising electrode material for SIBs.

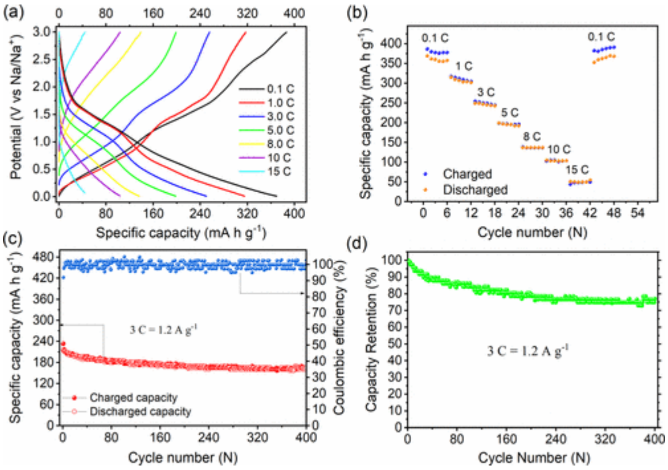

Fig. 4 (a) Galvanostatic charge/discharge plots at different C rates. (b) Rate capability at different current rates varies from 0.1 to 15 C. (c) Cycling stability and Coulombic efficiency over 400 cycles. (d) Capacity retention at 3 C over 400 cycles[16].

BCOF-1's application in SIBs is challenged by sodium-ion induced framework swelling, which disrupts the ordered pore structure and reduces cycling stability. The material's pseudocapacitive storage mechanism, while enabling high initial capacity 387mAhg^{-1}, is accompanied by irreversible structural rearrangement during Na^{+} insertion/extraction, leading to a 23% capacity loss after 400 cycles. Furthermore, the complex interplay between multiple redox sites (C=O, C=N) complicates kinetic analysis and limits the optimization of ion storage pathways.

3.3. COFs in ZIBs

In the research on COFs for ZIBs, various synthesis methods were employed. HqTp-COF was synthesized through a reaction between 1,3,5 - triformylphloroglucinol (Tp) and 2,5 - diaminohydroquinone dihydrochloride (Hq) [17]. The reaction conditions were precisely controlled to ensure the formation of the desired COF structure. The design, synthesis process, and partial charge discharge performance were shown in Fig. 5. To characterize the synthesized COFs, techniques like powder X-ray diffraction (PXRD) were used to analyze the crystal structure, and scanning electron microscopy (SEM) was applied to observe the morphology. Fourier - transform infrared spectroscopy (FTIR) was also utilized to identify functional groups, confirming the successful formation of the target COF.

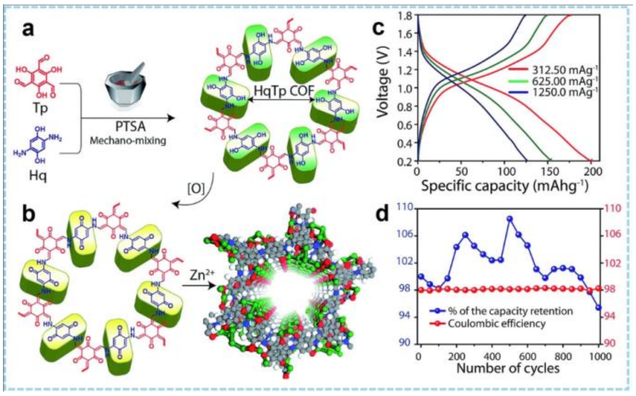

Fig. 5 a) Schematic synthetic pathway of HqTp COF. b) The process of electrochemical oxidation of hydroquinone to C2 symmetric C=O group to store Zn^{2+}. c) Charge–discharge curve of Zn//HqTp COF battery. d) Cycle stability test plot of HqTp COF at 3.75 A g^{-1}[17].

COFs demonstrated excellent performance in ZIBs. HqTp - COF, as a cathode material, exhibited a specific capacity of 276 $mAhg^{-1}$ at 125 mAg^{-1}. Even at a high current density of 3750 mA g^{-1}, it still had a specific capacity of 85 $mAhg^{-1}$, and maintained 95% of its initial capacity after 1000 cycles [17]. Tp - PTO - COF, another cathode material, achieved a reversible capacity of 301 mAh g^{-1} at $0.2Ag^{-1}$ and retained a high reversible capacity of 193 $mAhg^{-1}$ at 5 Ag^{-1}. In anode applications, DIP - D COF, used as a protective layer, enabled the symmetric DIP D@Zn//Zn cell to achieve ultra - long recycling in excess of 420 h at 1 $mAcm^{-2}$, with a Coulombic efficiency of 99.95% [17]. These results indicate that COFs have great potential in enhancing the performance of ZIBs.

In ZIBs, COFs face a critical challenge in aqueous electrolyte compatibility, as the hydrolytic instability of common linkages (e.g., imine bonds) leads to gradual degradation in Zn^{2+}-rich environments. For example, HqTp-COF exhibits pH-dependent capacity fade in acidic electrolytes, with a 15% loss in specific capacity after 500 cycles. Additionally, the inefficient Zn^{2+} desolvation kinetics at COF interfaces slow down charge transfer, limiting rate performance even in highly porous structures. The lack of selective ion transport channels also allows competitive adsorption of water molecules, exacerbating hydrogen evolution side reactions and reducing Coulombic efficiency.

4. Summary and Suggestion

Table 1. Comparison of advantages and disadvantages of COFs in three types of batteries

COFs in LIBs

Type	Advantages	Disadvantages
COFs in LIBs	-High capacity -Structural designability -Good cycling stability in some cases	-Poor productivity -Low intrinsic electrical conductivity -Structural rigidity hindering ion diffusion
COFs in SIBs	-Pseudocapacitive behavior for high initial capacity -Potential for ion - binding site design -Ordered pore structure	-Sodium - ion induced framework swelling -Irreversible structural rearrangement during cycling -Complex redox site interplay
COFs in ZIBs	-Unique features for Zn^{2+} storage -Ability to address some ZIB challenges -Good performance in certain aspects	-Aqueous electrolyte compatibility issues -Inefficient Zn^{2+} desolvation kinetics -Lack of selective ion transport channels

As shown in Table. 1, COFs have their advantages and disadvantages in different metal-ion batteries. COFs in Lithium - ion batteries (LIBs) offer notable advantages. Their high capacity enables substantial lithium - ion storage, crucial for energy - storage applications like electric vehicles. Structural designability allows customization of pore size, shape, and connectivity to optimize ion diffusion and conductivity. Some COFs also show good cycling stability, ensuring long - term reliability. However, they face challenges. Poor productivity restricts large - scale production due to complex and costly synthesis. Low intrinsic electrical conductivity hampers fast charging and high-rate performance. Structural rigidity impedes efficient lithium-ion diffusion, causing capacity fade. Future research on COFs for LIBs should prioritize three strategic improvements. First, develop efficient, scalable synthesis methods-such as continuous-flow techniques or eco-friendly protocols—to overcome current production bottlenecks and reduce costs. Second, enhance electrical conductivity by integrating conductive additives like graphene or carbon nanotubes into COF matrices or via heteroatom doping (e.g., N/S) to create electron-transport pathways. Third, engineer hierarchical pore structures or introduce flexible linkages to improve lithium-ion diffusion kinetics, addressing structural rigidity. Additionally, design protective surface coatings or electrolyte additives to stabilize redox-active sites (e.g., C=O/C=N groups) against organic electrolyte degradation, ensuring long-term cycling stability and capacity retention. These measures will enhance COFs' viability for high-performance LIB applications.

COFs in SIBs offer distinct advantages, including pseudocapacitive behavior for high initial capacities (e.g., BCOF-1's 387mAh g−1 at 0.1 C), ordered pore structures for rapid ion transport, and ion-binding site designability (e.g., C=N groups) to optimize Na^+ storage. However, critical limitations persist: sodium-ion induced framework swelling causes ~14% capacity loss at higher rates due to poor Na^+ penetration, irreversible structural rearrangement (e.g., rigid imidazole linkages) leads to 23% capacity decay after 400 cycles, and the complex interplay of redox sites (e.g., C=O/C=N) complicates kinetic analysis. These challenges underscore the need for structural flexibility and mechanistic clarity to enhance SIB performance. To address these challenges, future research should focus on two key directions. First, atomic-level design of ion-binding sites—such as introducing Lewis base groups (e.g., amine, carboxyl) with tailored Na^+ affinity—can enhance intercalation efficiency and reduce reliance on surface adsorption. For example, integrating flexible, Na^+-responsive moieties into COF backbones could improve deep-pore ion accessibility. Second, developing dynamic covalent frameworks (e.g., using reversible imine or boronate linkages) can mitigate swelling by accommodating Na^+-induced structural changes. Such frameworks demonstrated 77% capacity retention in BCOF-1, but further optimization is needed to enhance cyclability. Kinetic studies using in-situ techniques (e.g., NMR, Raman) should also be prioritized to decouple the contributions of multiple redox sites, enabling rational design of high-performance SIB cathodes. By balancing structural flexibility with ion-binding specificity, COFs can overcome current limitations and unlock their full potential in sustainable SIBs.

COFs in ZIBs offer unique advantages for aqueous energy storage, with materials like HqTp-COF delivering 276mAh g−1 at 125mA g−1 via pseudocapacitive mechanisms. Their ordered pores and redox-active sites (e.g., C=O, N-heteroatoms) enable efficient Zn^{2+} storage and mitigate dendrite growth. However, critical limitations include poor aqueous electrolyte compatibility—hydrolytic instability of linkages (e.g., β-ketoenamine) causes ~5% capacity loss per 100 cycles in $ZnSO_4$—inefficient Zn^{2+} desolvation kinetics, and a lack of selective ion channels leading to side reactions. Most COFs are 2D lamellar, lacking 3D porosity for high-areal-capacity electrodes. To overcome these barriers, research should focus on three key strategies. First, hydrophobic surface functionalization—such as introducing fluoroalkyl or alkyl chains can shield COF backbones from water-induced degradation while preserving ion conductivity. For example, fluoroalkyl-modified COFs demonstrated 20% improved cyclability in acidic electrolytes. Second, kinetic optimization via Zn^{2+}-specific binding sites (e.g., Lewis base groups) can enhance desolvation efficiency, reducing the energy barrier for ion insertion. In situ spectroscopy studies to monitor Zn^{2+}-COF interactions will guide this design. Third, developing scalable synthesis of 3D COFs—using roll-to-roll fabrication or templated growth—will address the lack of thick, freestanding electrodes. 3D COFs with hierarchical pores could increase Zn^{2+} storage capacity while maintaining structural integrity. By integrating these approaches, COFs can unlock stable, high-performance ZIBs for next-generation aqueous energy storage.

In conclusion, COFs have shown great potential in metal - ion batteries, but each application in LIBs, SIBs, and ZIBs faces distinct challenges. Future research should focus on addressing these specific drawbacks through innovative material design, synthesis optimization, and in - depth understanding of electrochemical mechanisms. By doing so, COFs can be further developed to realize their full potential in high - performance metal - ion batteries, paving the way for more efficient and sustainable energy - storage solutions.

5. Conclusion

Covalent organic frameworks (COFs) have emerged as a versatile class of materials with significant potential in metal-ion batteries (LIBs, SIBs, and ZIBs), leveraging their structural designability, ordered porosity, and tunable redox activity. Across different battery systems, COFs demonstrate distinct advantages but also face unique challenges that require tailored solutions.

In lithium-ion batteries (LIBs), COFs such as TRO-BT-COF offer high capacity and structural customizability, enabling efficient lithium-ion storage. However, their low intrinsic electrical conductivity, structural rigidity, and complex synthesis processes limit practical scalability. Key improvements include integrating conductive additives, engineering hierarchical pores for better ion diffusion, and developing protective coatings to stabilize redox sites against electrolyte degradation.

For sodium-ion batteries (SIBs), COFs like BCOF-1 exhibit promising pseudocapacitive behavior and ordered pore structures, but suffer from sodium-ion induced framework swelling, irreversible structural rearrangements, and complex redox site interactions. Addressing these requires dynamic covalent framework design to accommodate ion-induced swelling and atomic-level optimization of ion-binding sites to enhance intercalation efficiency.

In zinc-ion batteries (ZIBs), COFs such as HqTp-COF show robust Zn^{2+} storage via pseudocapacitive mechanisms, but aqueous electrolyte instability, inefficient Zn^{2+} desolvation kinetics, and the predominance of 2D lamellar structures hinder performance. Strategies like hydrophobic surface functionalization, kinetic optimization through Zn^{2+}-specific binding motifs, and scalable synthesis of 3D COFs with hierarchical pores are critical to overcoming these barriers.

Overall, COFs hold transformative potential for next-generation metal-ion batteries, but their commercialization hinges on addressing material-specific limitations through interdisciplinary approaches. Future research must prioritize scalable synthesis methods, mechanistic insights into ion-COF interactions, and the development of multifunctional frameworks that balance stability, conductivity, and ion selectivity. By addressing these challenges, COFs can unlock new frontiers in sustainable energy storage, driving the transition toward high-performance, eco-friendly battery technologies.

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