

Designing High-Performance Organic Electrode Materials for Sodium-Ions Batteries: Strategies and Insights

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Organic electrode materials, known for their abundance and environmental friendliness, have garnered significant attention as promising electrode materials of sodium-ion batteries (SIBs). Through the proposed structural design, organic compounds with flexible structure can meet the diverse demands of rechargeable SIBs. However, the practical applications of organic electrode materials are restricted by the intrinsic shortcomings, including poor conductivity and high solubility in organic electrolytes. This review summarizes recent advancements in organic electrode materials design strategies to overcome these limitations and enhance SIB performance. Key approaches include designing novel active centers and optimizing molecular structures in conjugated organic molecules, thereby maximizing theoretical specific capacity. Additional

strategies, such as chain extension and combination with the conductive substrate, improve ion/electron conductivity and reduce solubility in organic electrolytes. Furthermore, innovative structure designs, including non-conjugated materials and organic-metal composites, offer new pathways to achieve high-performance electrodes. These strategies provide foundational design principles for advancing organic electrode materials in SIB applications. Lastly, we address the challenges and opportunities in organic electrode materials research, highlighting their potential in promoting the development of next-generation, sustainable energy storage systems. High-performance organic electrode materials represent a critical step toward realizing the full potential of SIBs for large-scale energy storage solutions.

1. Introduction

With the rapid development of electronic devices and energy storage systems, lithium-ion batteries (LIBs) have received significant attention in recent years.^[1–3] LIBs with high energy density, elevated operating voltage, and excellent cycling stability are suitable for large-scale energy storage.^[4–6] However, the growing demand for LIBs has led to increased costs, and safety concerns arising from exothermic side reactions hinder their further application.^[7,8] Sodium-ion batteries (SIBs) have emerged as a promising alternative in recent years.^[9,10] SIBs have many distinct advantages. First, the abundance of sodium resources and compatibility with cost-effective aluminum current collectors lower overall costs,^[11] facilitating large-scale deployment. Second, excellent adaptability to a wide range of temperatures makes them suitable for diverse environmental conditions.^[12] Third, superior thermal stability would reduce safety risks, as SIBs do not exhibit the thermal runaway issues common in LIBs.^[13] However, despite these advantages, the larger ionic radius of sodium (1.12 Å) is incompatible with conventional electrode materials and high-performance electrode material is desired for advanced SIBs.^[14]

A variety of electrode materials, both organic and inorganic, have been explored for SIBs. Among them, organic electrode materials have gained raised focus in recent years. Unlike traditional layered transition metal compounds, organic compounds are not restricted by geographic resource availability.^[15] Additionally, organic compounds are composed primarily of non-metallic elements, such as C, H, N, O, making them environmentally friendly and biodegradable.^[16] Their flexible molecular structures allow customization through organic synthesis, enabling the design and introduction of functional groups such as C=N, C=O, and C-S- >C to increase theoretical capacity.^[17–19] Voltage regulation can be achieved by incorporating heteroatoms like nitrogen and sulfur into the organic framework, facilitating redox reactions within an appropriate voltage window.^[20,21] Different from intercalation reactions of conventional transition metal oxides, sodium-ion storage in organic electrode materials is realized through the attraction of sodium ions to the active functional groups. Meanwhile, the weaker interlayer forces between the organic molecules allows efficient ion insertion and release during the charge-discharge process.^[22] Hence, it is promising to design suitable organic electrode materials for widespread SIBs applications.

However, organic electrode materials face challenges such as low conductivity and high solubility in organic electrolytes, leading to reduced energy density and poor cycling stability.^[23–25] Significant research efforts have been directed toward addressing these issues. First, structural design for developing new active sites and constructing suitable internal frameworks for organic molecules. Maximizing the utilization of active groups within the molecule is also a key area of interest. Second, enhancing stability and conductivity through extending conjugation frameworks of the organic molecules. Combin-

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ing organic materials with conductive substrates such as carbon nanotubes or other carbon materials also improves ion and electron transport. Third, exploring novel organic systems, such as non-conjugated organic compounds and organic-metal materials. These materials often exhibit high conductivity and are readily available, making them attractive for electrode applications. This review summarizes the advancements in these areas, providing a detailed analysis of recent progress in organic electrode materials design. By addressing current challenges and exploring innovative solutions, we aim to guide future research and promote the practical application of organic electrode materials in high-performance SIBs.

2. Sodium-Ion Storage Mechanisms in Organic Molecule

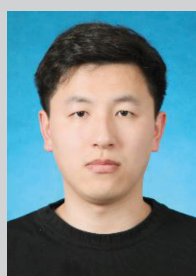
Different from traditional inorganic electrode materials, organic electrode materials stabilize sodium ions through unique interaction mechanisms.^[26,27] These mechanisms facilitate efficient sodium-ion storage and contribute to the high performance of SIBs. The first mechanism involves the stabilization of sodium ions by active groups such as C=O, C=N and N=N functional groups in conjugated system, as exemplified by sodium terephthalate (Figure 1a). Here, sodium ions are attracted to the active groups, and electrons are stabilized within the conjugated chain through bond breakage and recombination between single and double bonds.^[28,29] Additionally, the weak intermolecular forces in small organic molecules allow sodium ions to transfer easily within the system, resulting in excellent ion diffusion rates. A second storage mechanism relies on larger conjugated macrocycles (Figure 1b). The large conjugated molecule with abundant delocalized electrons usually exhibits a lower electronegativity and could attract sodium ions through electrostatic interactions.^[30] The conju-

gated state can switch between a stable locally aromatic and a stable globally aromatic state, thus demonstrating outstanding redox properties. A third mechanism is based on pore adsorption in frameworks such as covalent organic frameworks (COFs) and metal-organic frameworks (MOFs) (Figure 1c). Typical stabilization modes are still dominant in organic frameworks, sodium ions are attracted by active groups of conjugated system. However, these materials feature large pores that can accommodate additional sodium ions, contributing to a higher specific capacity.^[31,32] Finally, sodium-ion storage can also occur in non-conjugated organic materials (Figure 1d). Unique mechanisms, including hydrogen transfer and heteroatom-centered interactions, play key roles in stabilizing sodium ions in these systems.^[33,34] These approaches open new avenues for designing organic electrode materials with advanced storage capabilities. Together, these diverse mechanisms illustrate the potential of organic electrode materials to efficiently store sodium ions, paving the way for their application in high-performance SIBs

3. Optimization Strategies for Organic Electrode Materials

3.1. Structure Design

To date, numerous small organic molecules with functional groups, such as C=O, C=N, and N=N have been identified as electroactive for SIBs.^[35–37] Among these, organic compounds with C=O bonds, such as quinones, ketones, and carboxylates are the most extensively studied due to their high theoretical capacities and multi-electron reaction capabilities.^[38–40] To expand the range of viable organic materials, exploring new active centers is essential. Recent research on uric acid (UA) highlights innovative approaches to sodium-ion storage beyond



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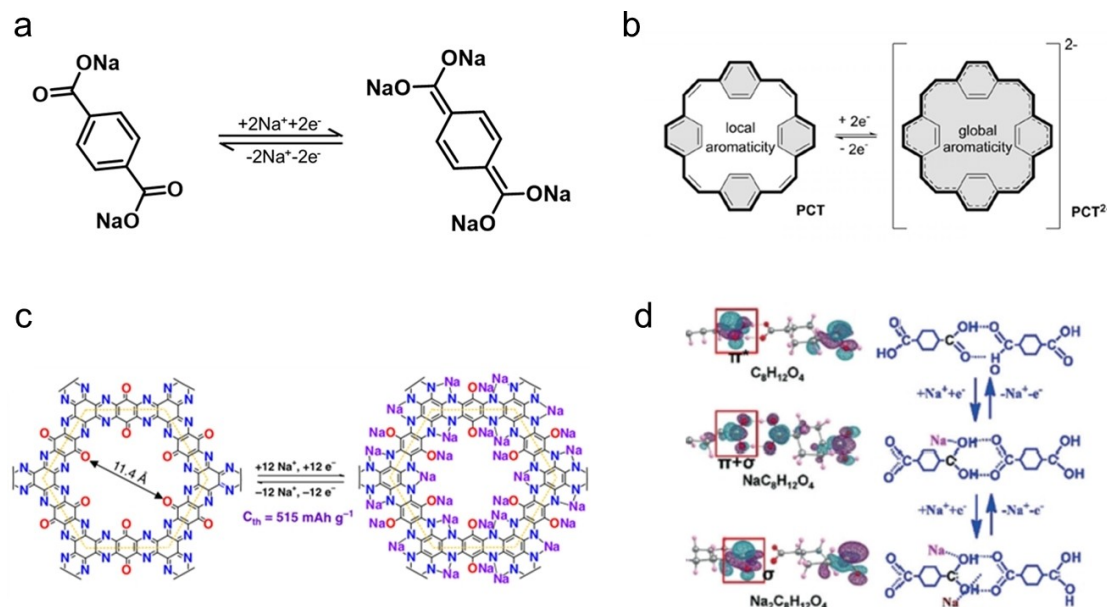


Figure 1. The storage mechanism of organic electrode materials: a) Conjugated materials with active groups, b) large conjugated ring. Reproduced with permission from ref [30]. Copyright 2020 Wiley-VCH, c) organic framework. Reproduced with permission from ref [31]. Copyright 2020 Springer Nature, d) non-conjugated compounds. Reproduced with permission from ref [33]. Copyright 2018 Wiley-VCH.

traditional conjugated bond reformation.^[41] In UA, additional storage activity arises from p- π conjugation stabilization of α -carbon radical intermediates and carbon-anion stabilization (Figure 2a, b). The first sodium ion (Na^+) typically interacts with C=O groups, electrons are stabilized within the conjugated system ($\text{C}=\text{C} \rightarrow \text{C}=\text{O}$), forming a unique carbon anion center. Subsequently, an extra Na^+ interacts with a carbon atom bonded to a nitrogen atom (Figure 2c). These findings confirm the sodium-ion storage activity of UA and underscore the potential of novel active centers in organic electrode materials.

Organosulfur compounds are usually designed as promising high energy density electrode materials. Sulfur-containing functional groups can undergo two-electron electrochemical oxidation/reduction reactions, delivering higher specific capacities.^[42] Sodium 2,5-thiophene dicarboxylate ($\text{Na}_2\text{C}_6\text{H}_2\text{O}_4\text{S}$, STDC), a thiophene derivative with a conjugated sulfur backbone, exemplifies this approach (Figure 2d).^[19] STDC containing backbone sulfur (C-S bonds) can not only take the advantage of the high capacity of sulfur and the good electronic conductivity of the conjugated system, but also avoid the direct involvement of element sulfur and the cleavage of covalent S-S bonds. Up to four Na^+ could be reversibly stored by one STDC molecule (Figure 2e). Two additional Na^+ are symmetrically stored adjacent to the backbone sulfur atoms of the conjugated five-membered thiophene ring. Used as an anode for SIBs, the STDC electrode achieves a high specific capacity of 394 mAh g^{-1} (Figure 2f). Thiophene derivatives with expanded thiophene rings are constructed to further enhance theoretical capacity of organic electrode materials (Figure 3a). Considering the balance of solubility and capacity, sodium thieno[3,2-b]thiophene-2,5-dicarboxylate (STTDC), which features two thiophene backbone rings, has emerged as a promising electrode material.^[43] The larger conjugated thiophene backbone system in STTDC not

only improves sodium-ion storage, allowing up to six Na^+ per molecule (Figure 3c), but also improve the charge transfer of organic materials, benefiting to rate performance and cycling stability. The STTDC electrode demonstrates exceptional electrochemical performance, achieving a high theoretical specific capacity of 591 mAh g^{-1} . It retains remarkable reversible capacities after long-term cycles (Figure 3b).

Polymeric Schiff bases with low operating voltages are considered promising anode materials for SIBs.^[35,44] Sodium ions are stored within the conjugated system of Schiff base groups and a phenyl ring ($-\text{N}=\text{CH}-\text{Ar}-\text{HC}=\text{N}-$, where Ar represents an aromatic group). However, the specific capacity of these materials is limited by the non-active mass of their large conjugated systems. To address this issue, polymeric Schiff bases incorporating thiophene rings (PSB-2) were developed as high-performance anode materials (Figure 3c).^[45] The introduction of thiophene rings reduces the band gap of the polymeric Schiff material, corresponding to enhancement of the electronic conductivity (Figure 3d). When employed as an anode for SIBs, PSB-2 demonstrates excellent specific capacity and cycling stability (Figure 3e). Another effective way to increase the capacity is activation of the inactive functional groups for organic materials. Sodium phthalate with two active groups (C=O) and conjugated system has shown electrochemical activity for SIBs, but the low reversible capacity caused by steric hindrance limits practical applications.^[46] Sodium 5,5'-carbonylbis (isobenzofuran-1,3-dione) (SCID) is synthesized by linking two sodium phthalate molecules via a bridging carbonyl group (Figure 3f). This design activates adjacent carbonyl groups in the sodium phthalate units, leading to a higher specific capacity for SIBs (Figure 3g).

Organic framework with orderly structure and wide interior spaces have been proposed as electrode materials, such as

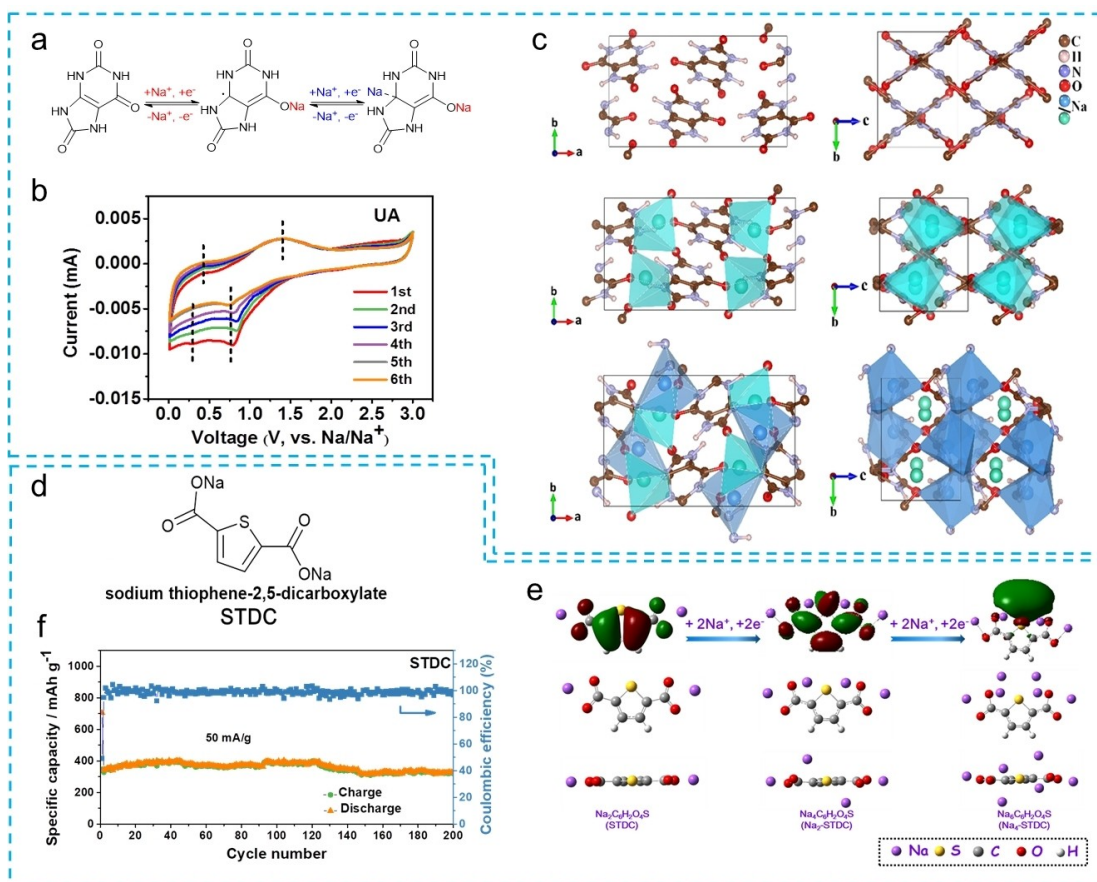


Figure 2. a) The storage process of uric acid for sodium ions and b) CV curves of UA, c) Optimal structure of $\text{Na}_x\text{C}_5\text{H}_4\text{N}_4\text{O}_3$ ($x = 0, 1, 2$). Reproduced with permission from ref [41]. Copyright 2017 American Chemical Society, d) Molecular structure of STDC, e) Calculated HOMO plots and spatial structure of molecule STDC with different Na^+ , and f) Cycling performance of STDC electrode. Reproduced with permission from ref [19]. Copyright 2016 Royal Society of Chemistry.

covalent organic frameworks (COFs) and metal organic frameworks (MOFs).^[47] The rational design of organic ligands would endow high theoretical capacity of organic-metal framework, the porous framework facilitates electrolyte diffusion and improves access to electroactive sites.^[31] A highly crystalline, porous polyimide COF (COF-TPDA-PMDA) is presented as a typical example for high-performance organic framework (Figure 3h).^[48] COF-TPDA-PMDA operates via a dual-ion storage mechanism, the larger anions are absorbed by C–N redox groups within the mesopores and the cations are attracted to the active groups C=O (Figure 3i). This dual-active center COFs material highlights an effective strategy for designing high-performance organic electrode materials in SIBs.^[49] By simultaneously optimizing the molecular structure and composition of polyimide COFs, these materials show great promise for next-generation rechargeable batteries.^[50]

3.2. Improving Stability and Conductivity

The electrochemical performance of small organic molecules is often limited by high dissolution in organic electrolytes and low conductivity. A promising approach to address these challenges

involves constructing larger, more stable conjugated systems.^[51] For instance, a triaminophthalocyanine derivative (TPCDS) with an extended non-localized π -electron system has been designed by covalently binding small organic molecules (Figure 4a).^[52] The enlarged conjugated system significantly reduces the solubility of 4-aminophthalonitrile monomer in electrolytes while enhancing conductivity (Figure 4b). The high theoretical capacity is also maintained by covalent ligation to avoid the introduction of inactive mass. Consequently, TPCDS electrodes exhibit high specific capacity and excellent long-term cycling stability in SIBs (Figure 4c). Structural stability is another critical factor influencing cycling performance. MOFs with π -d conjugated systems have demonstrated high electroactivity for sodium-ion storage. However, their poor structural stability often leads to rapid capacity degradation during cycling, as observed in materials such as MIL-88 A and UIO-66.^[53,54] A promising solution to this issue involves generating surface polymer coatings to stabilize the MOFs structure.^[55] For instance, catalyzed by surface Fe^{3+} ions of MIL-88 A, p-phenylenediamine are polymerized and coated onto the surface of MOF nanorods, generating a MOF-polymer composite PPA@MIL-88 A (Figure 4d). The polymer coating significantly enhances the structural stability of MIL-88 A, resulting in exceptional

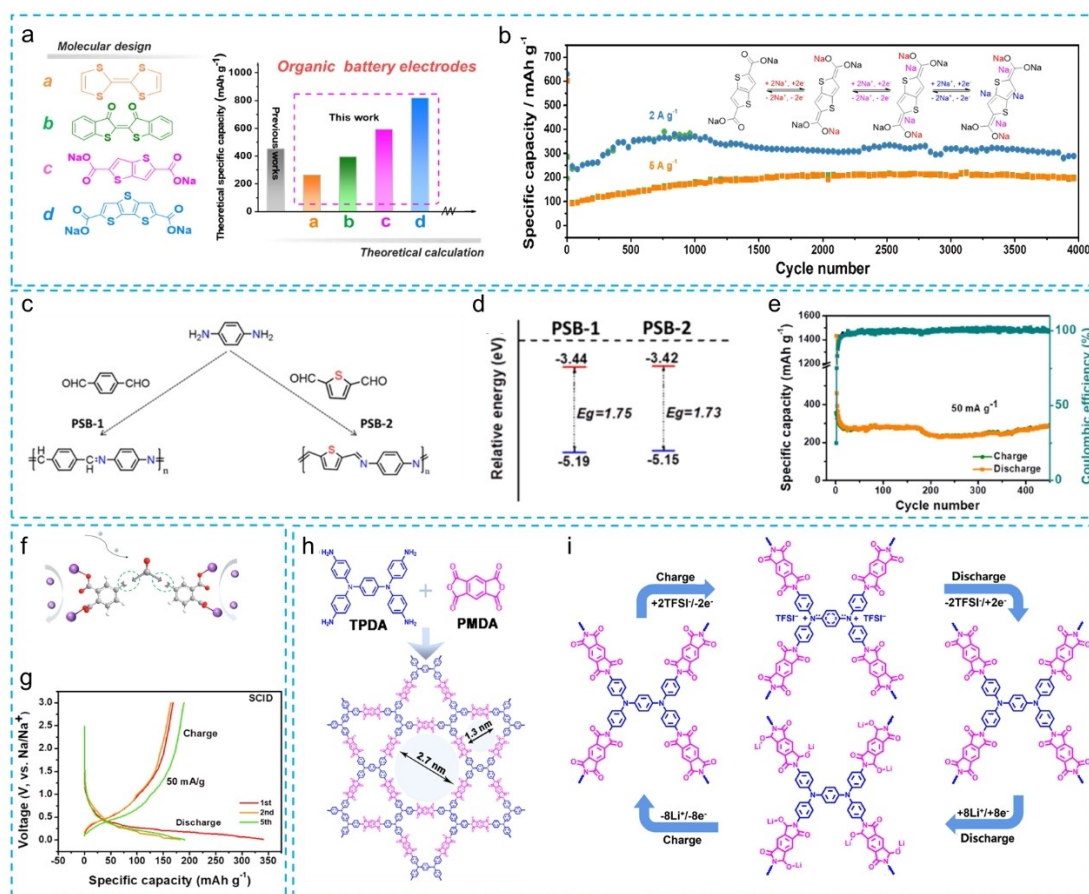


Figure 3. a) Design strategy of the thiophene derivatives, b) Long-term cycling stability of STTDC, the insertion image is the sodiation progress of STTDC molecule. Reproduced with permission from ref [43]. Copyright 2021 Royal Society of Chemistry, c) Synthesis schematic of polymeric Schiff bases from aldehydes and amines, d) Calculation of bonding gap, e) Cycling performance of PSB-2. Reproduced with permission from ref [45]. Copyright 2022 American Chemical Society, f) Design strategy of SCID molecule, g) The charge/discharge curves of SCID electrode. Reproduced with permission from ref [46]. Copyright 2020 Royal Society of Chemistry, h) Synthesis of crystalline COFs, i) Proposed redox mechanism of COFs during the charge/discharge process. Reproduced with permission from ref [48]. Copyright 2022 American Chemical Society.

long-term cycling performance (Figure 4e). In addition, coupling MOF-polymer composites with Prussian blue analogues (PBG) offers further improvements (Figure 4f). PPA@MIL-88 A-PBG full cells achieve high operating voltage (>3.0 V) and specific capacity (Figure 4g). These results highlight the potential of MOF-polymer composites for high-performance sodium-ion full cells, demonstrating their viability for practical applications.

Incorporating organic electrode materials into suitable conductive networks is a practical strategy to enhance their electrochemical performance.^[56] Conductive substrates, such as multi-walled carbon nanotubes (MWCNTs) and graphene, not only improve electronic conductivity but also provide a large surface area for active material attachment, reducing dissolution in organic electrolytes.^[24] One effective example is the SCID@CNT composite, designed to improve the performance of SIBs.^[46] In this composite, SCID nanoparticles are uniformly distributed on the surface of MWCNTs, significantly increasing the surface roughness of the MWCNTs (Figure 4h). This design enhances specific capacity of composite by the construction of conductive network and loading substrate for organic materials (Figure 4i). A similar approach has been applied to COF_{TPDA-}

PMDA^[48] where charge transfer is significantly improved upon integration with CNTs (Figure 4j). The strong π - π interactions between COF_{TPDA-PMDA} and CNTs facilitate the heterogeneous nucleation and growth of COF_{TPDA-PMDA} on the CNT surface, forming a core-shell, cable-like morphology (Figure 4k, 4l). This unique architecture, with COF_{TPDA-PMDA} shells on the CNT surface, enhances electronic conductivity and optimizes charge transfer. As a result, the COF_{TPDA-PMDA}@50%CNT electrode achieves the best rate capacity among comparable COF-based electrode materials (Figure 4m) and exhibits outstanding long-term cycling stability (Figure 4n). This performance ranks as one of the highest reported in the literature for COF-based electrodes, highlighting the potential of conductive networks to advance the application of organic materials in SIBs.

3.3. Exploring Novel Organic Systems

Traditional organic electrode materials for SIBs are typically based on conjugated compounds (Figure 5a), where sodium ions are attracted by electroactive functional groups and

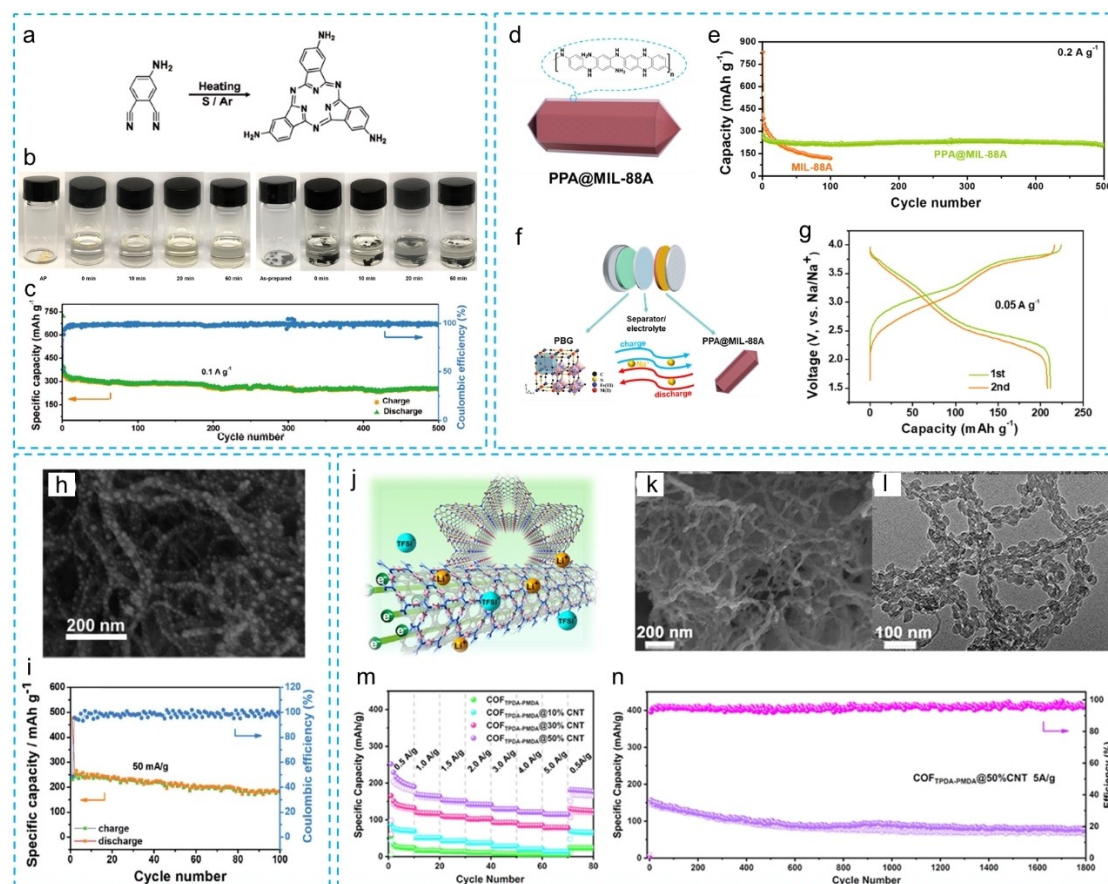


Figure 4. a) Schematic illustration of the polymerization process of 4-aminophthalonitrile molecules, b) Dissolution test of 4-aminophthalonitrile and TPCDs, c) Long-term cycling of TPCDs electrode. Reproduced with permission from ref [50]. Copyright 2022 Wiley-VCH, d) Schematic illustration of the PPA@MIL-88 A nanorod, e) Cycling performance of PPA@MIL-88 A and MIL-88 A, f) Full cell of the PPA@MIL-88 A-PBG, g) Charge/discharge curves of the full cell. Reproduced with permission from ref [53]. Copyright 2023 American Chemical Society, h) SEM images of the SCID@CNT, i) Cycling performance of SCID@CNT electrode. Reproduced with permission from ref [46]. Copyright 2020 Royal Society of Chemistry, j) Schematic illustration of COF material, k) SEM and l) TEM images of COF@50%CNT, m) Rate capacity of the COF@CNT composites, n) Long-term cycling of COF@50%CNT electrode. Reproduced with permission from ref [48]. Copyright 2022 American Chemical Society.

electrons are added to low-electronegativity atoms and stabilized through the breaking and reformation of double bonds.^[57] However, these materials often face limitations, including low capacity, uncontrolled operating voltages, and a narrow range of viable organic compounds.^[58] To overcome these challenges, non-conjugated organic compounds have been explored as potential electrode materials. One promising candidate, 1,4-cyclohexanedicarboxylic acid ($C_8H_{12}O_4$, CHDA), features a non-conjugated six-membered ring with two carboxylate groups in opposing positions.^[33] The hydrogen bonds between CHDA molecules establish a robust framework (Figure 5b). During the insertion of sodium-ion, hydrogen transfer occurs between two CHDA monomers. The sodium ion occupies the symmetrically positions of near the carboxylate groups, interacting with oxygen atoms (Figure 5c). The weak intermolecular forces within CHDA facilitate easy sodium-ion migration to the carboxylate positions. Impressively, CHDA exhibits a high reversibility and specific capacity (249 mAh g^{-1} after 100 cycles) (Figure 5d, e), underscoring its excellent cycling stability and potential for practical applications.

Organometallic compounds consist of organic ligands and transition metal ions, forming unique π -d conjugated systems.^[59] The metallic-carbon (M–C) bonds in organometallic compounds exhibit significant covalent properties, enhancing the stability of the coordination framework.^[60] This contributes to improved rate performance and cycling stability. Fe-PDA-220, an iron-based organometallic compound, has recently been identified as a promising anode material for SIBs.^[61] The poly-phenylenediamine organic ligands provide abundant active sites (Figure 5f), while the Fe ions link the polymer chains, creating a multi-atom $Fe-C_{12}N_8$ sites with robust coordination structure. Fe-PDA-220 adopts a microsphere morphology and the surface contains numerous micropores and mesopores, facilitating electrolyte infiltration (Figure 5g). The C=N groups along the polymer chain also readily interact with the weak C–H groups of adjacent chains through hydrogen bonding, resulting in a two-dimensional supramolecular framework (Figure 5h). When used as an anode for SIBs, Fe-PDA-220 demonstrates impressive electrochemical reversibility (Figure 5i). Long-term cycling tests further highlight its remarkable cycling stability and rate capability (Figure 5j), with reversible capacities

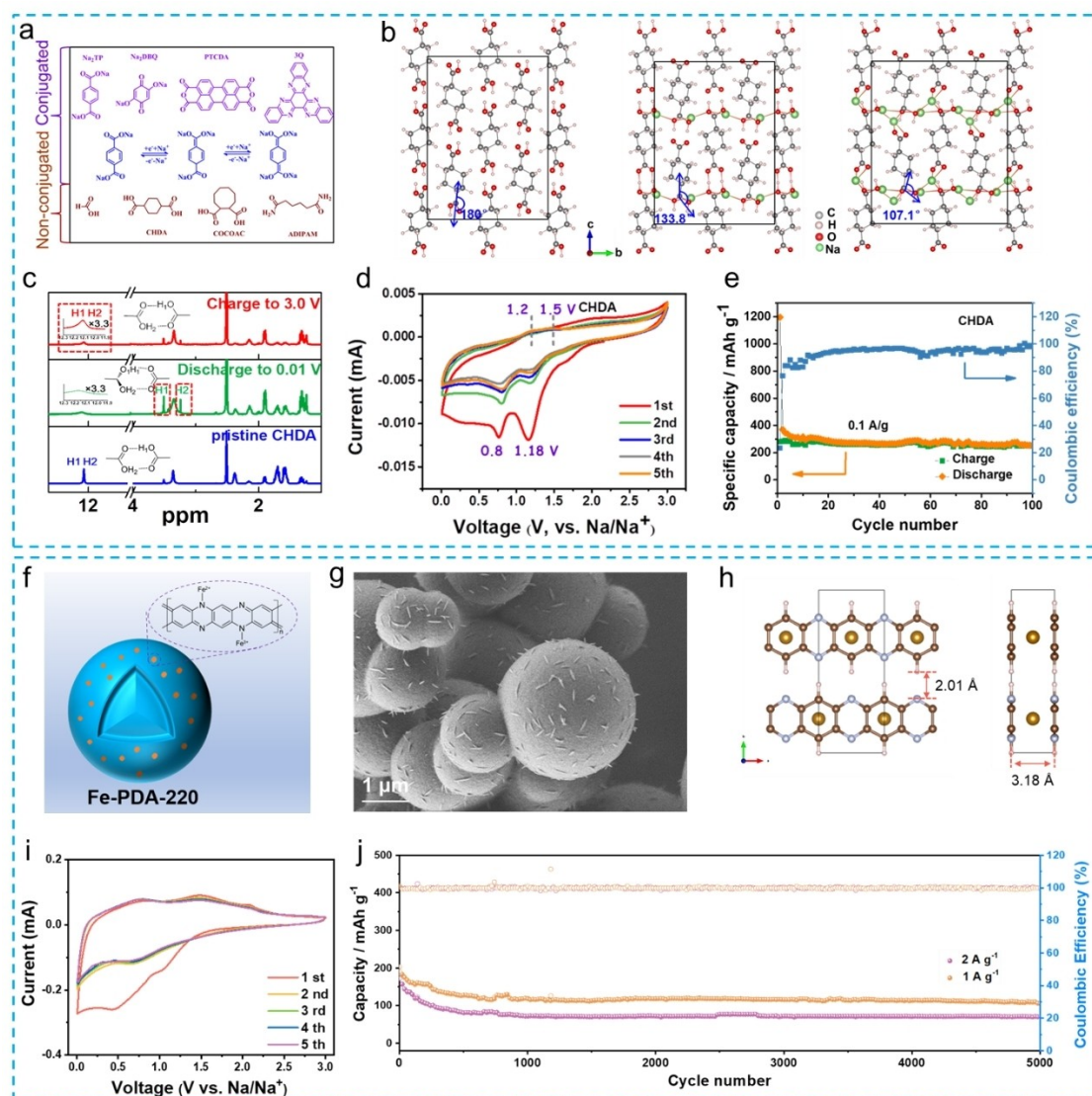


Figure 5. a) The typical conjugated, non-conjugated compounds and the electronic stabilization mechanism in conjugated compounds, b) Structural evolution during the discharge, Relaxed CHDA, NaCHDA, and Na₂CHDA crystal structures along the (100) direction, c) ¹H NMR of the CHDA electrode and those discharged to 0.01 V and recharged to 3.0 V, d) CV curves and e) Cycling performance of CHDA electrode. Reproduced with permission from ref [33]. Copyright 2018 Wiley-VCH, the structural feature and electrochemical performance of Fe-PDA-220, f) Schematic illustration, g) SEM image, h) Optimization structure, i) CV curve and j) Long-term cycling at high current density. Reproduced with permission from ref [59]. Copyright 2024 Wiley-VCH.

of 106 and 71 mAhg⁻¹ maintained even after 5000 cycles at high current densities of 1.0 and 2.0 Ag⁻¹, respectively.

4. Summary and Perspectives

In recent decades, organic electrode materials have attracted significant attention for advanced rechargeable batteries, driven by the increasing demand for high energy density. Despite their promise, several challenges remain, including issues with intrinsic conductivity and material dissolution. This review outlines key design strategies for enhancing the performance of organic electrode materials, focusing on three main approaches. First, the development of new active centers can expand the range of organic electrode materials and improve their

theoretical capacity. Extending the molecular framework introduces additional active groups, which facilitate better sodium ion storage. Second, the use of larger molecular structures and polymer coatings helps mitigate dissolution and improve the structural stability of organic materials, enhancing their cycling performance in organic electrolytes. When combined with conductive carbon substrates, both conductivity and material stability are further improved. Finally, exploring non-conjugated systems and designing organometallic polymers broadens the potential applications of organic materials. These strategies provide valuable guidance for the future design of high-performance organic electrode materials.

Looking ahead, several key areas will shape the future development of organic electrode materials for next generation SIBs:

- High-voltage organic materials:** Organic compounds typically exhibit lower operating voltages compared to other materials. While the electrochemical voltage of organic materials can be increased through strategies such as introducing electron-withdrawing groups like C=O, but poor stability caused by these modifications often limits cycling performance.^[54,62] Therefore, there is a strong need for the development of new high-voltage organic materials for practical applications. Recent research has focused on quinones, ketones with hydrogen bonding, and organic composites with active metal ions.^[63–66] These materials exhibit both high operating voltage and excellent cycling stability, making them promising candidates for large-scale application.
- Mechanism illustration:** Advanced analytical techniques, including in situ Fourier transform infrared (FTIR) spectroscopy, Raman spectroscopy, X-ray absorption fine structure (XAFS), X-ray diffraction (XRD), electron paramagnetic resonance (EPR), and nuclear magnetic resonance (NMR) spectroscopy, enable the monitoring of intermediate products and verification of reversibility during battery operation.^[67–69] These methods provide valuable insights into the electrochemical processes of sodium ion insertion and extraction. Additionally, theoretical simulations are increasingly used to predict optimal material structures, accelerating the development of organic materials. Therefore, understanding the storage mechanisms of sodium ions is essential for developing high-performance organic electrode materials.
- Organic full-cell development:** While many organic electrode materials have been designed for rechargeable batteries, only a limited number have been explored in organic full-cell configurations.^[70] The primary challenge in using organic electrode materials for full cells lies in their low active material loading and difficulty in pairing with traditional inorganic electrodes.^[71] Recently, all-organic rechargeable batteries have been developed, demonstrating high electrochemical performance.^[72] The development of all-organic full cells is critical for advancing the application of organic electrode materials. Additionally, the unique behavior of sodium ions offers a high theoretical specific capacity, meaning organic full-cells could provide higher energy density compared to traditional inorganic batteries.^[73] With continued progress in all-organic full cells, the practical potential of organic electrode materials is expected to be further realized.

In summary, organic electrode materials have seen rapid advancement in recent years, with a diverse range of materials developed. Researchers are focusing on structural design, mechanism analysis, electrochemical performance testing, and full-cell construction to unlock the potential of organic materials in rechargeable batteries. More importantly, identifying suitable application scenarios for organic batteries and accelerating the development of all-organic cells will be crucial for the commercialization of this technology, bringing it closer to widespread market adoption.

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Conflict of Interests

The authors declare no conflict of interest.

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