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Electrode Batteries: Organic Material's influence on Lithium-Ion Battery Performance

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Abstract

The integration of organic materials into electrode batteries, specifically lithium-ion batteries, has shown significant potential in enhancing battery performance. Organic materials offer several advantages, including abundant availability, environmental friendliness, and tunable properties, making them attractive alternatives to traditional inorganic materials. Research has demonstrated that organic compounds can improve the energy density, cycling stability, and overall efficiency of lithium-ion batteries. These materials contribute to better electrode flexibility, higher capacity retention, and faster charge-discharge rates. Innovations in the synthesis and design of organic electrode materials have led to the development of high-performance batteries with increased lifespan and reduced environmental impact. Challenges remain, such as ensuring long-term stability and addressing issues related to material degradation. However, ongoing advancements continue to drive the optimization of organic materials for lithium-ion batteries, promising a future with more sustainable and efficient energy storage solutions. This abstract highlights the transformative potential of organic materials in revolutionizing lithium-ion battery technology.

Keywords: Cycling Stability; Energy Density; Environmental Friendliness; Lithium-Ion Batteries; Organic Materials; Performance Enhancement

Abbreviations: DFT: Density functional theory, DNP-Li: Dimer electrode material, EELS: Electron Energy Loss Spectroscopy, NP-Li: Naphthazarin monomer, NMR: Nuclear magnetic resonance

1. Introduction

Lithium-ion batteries have revolutionized the energy storage landscape, powering a wide range of devices from smartphones to electric vehicles. At the heart of these electrode batteries lie intricate chemical reactions involving organic materials that play a crucial role in determining their performance and efficiency. As the demand for higher energy density and longer cycle life continues to grow, understanding the role of organic materials in lithium-ion batteries has become paramount. The evolution and progress of organic electrode materials, their redox mechanisms and charge storage capabilities, as well as their impact on energy and power density, cycle life, stability, gravimetric density, and electronic conductivity. It explores electrode engineering and optimization techniques, cost and resource availability considerations, and future prospects and challenges in the realm of organic materials for lithium-ion and solid-state batteries (Fig. 1) [1, 2, 3, 4, 5, 6, 7, 8].

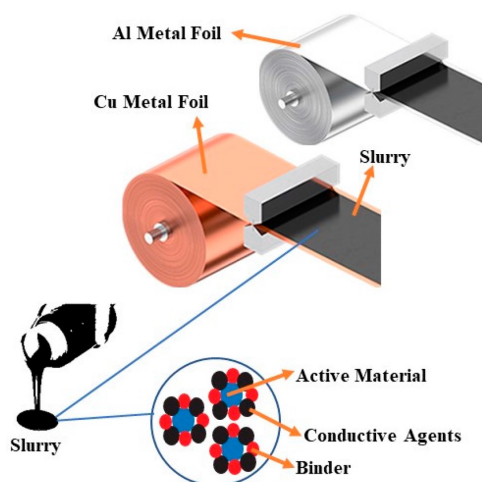


Figure 1. Illustration showcasing the trio of main elements within the LIB electrode.

2. Organic Electrode Materials: Evolution and Progress

The evolution of organic electrode materials for lithium-ion batteries has seen significant progress in recent years. One notable example is the study of naphthazarin (5,8-dihydroxy-1,4-naphthoquinone) as a high-capacity candidate material, with a theoretical capacity of 531 mAh/g based on a 4-electron transfer reaction. Initially, a naphthazarin monomer (NP-Li) was synthesized, delivering an impressive initial capacity of 403 mAh/g but suffering from poor cycle life. To address this issue, researchers developed a naphthazarin dimer (DNP-Li) fused by a dithiin ring, which exhibited an initial capacity of 416 mAh/g and maintained 300 mAh/g after 100 cycles, demonstrating improved cycle life [9, 10].

Organic electrode materials offer several advantages over conventional inorganic electrodes, including sustainable production, low cost, high flexibility, and resource sustainability. The history of organic electrode materials can be traced back to the 1960s, with various developments in the following decades. Carbonyl compounds, such as quinones, carboxylates, anhydrides, and imides, are widely used as organic electrode materials. Strategies to address the issues associated with these compounds include [11, 12, 13]:

- **Quinones:** Fusing multiple carbonyl groups, functionalization with ionic groups, and entrapping within insoluble substrates to address dissolution, low conductivity, and low voltage issues.
- **Carboxylates:** Tuning the position of the carbonyl group, using different processing methods, extending conjugation, and choosing appropriate counter ions to improve performance, especially for anode materials.
- **Anhydrides:** Increasing molecular weight, using 3D hybrid structures, and tuning substituents to address rapid capacity decay and high solubility issues, particularly for cathode materials like PTCDA.
- **Imides:** Tuning the active ion, using substituents and functionalization, and molecular engineering to shape the discharge curve and address decomposition, low voltage, and limited capacity challenges, especially for diimides as cathode materials.

Non-conjugated polymer forms of these carbonyl compounds can help suppress dissolution and improve cycling stability, although electrical conductivity may be a concern.

2.1 Redox Mechanisms and Charge Storage

The charge storage mechanism in organic electrode materials is primarily governed by redox reactions, where the charge state of the electroactive groups undergoes reversible changes. These redox mechanisms can be classified into three main categories based on the nature of the charge-state change [14, 15, 16, 17, 18, 19, 20]:

- **n-type:** In this mechanism, the active ion accepts electrons during the discharge process, resulting in a negatively charged species. Organic anodes typically exhibit this behavior.
- **p-type:** Conversely, the active ion loses electrons during discharge, leading to a positively charged species. Organic cathodes commonly follow this mechanism.
- **Bipolar:** Some organic materials can undergo both n-type and p-type redox reactions, enabling them to function as either anodes or cathodes depending on the operating conditions.

The specific redox mechanisms and charge storage processes in the organic materials discussed earlier can be further elucidated:

- **Naphthazarin Monomer (NP-Li) and Dimer (DNP-Li):** Ex-situ nuclear magnetic resonance (NMR) and electron energy loss spectroscopy (EELS) analyses confirmed the 4-electron and 8-electron redox mechanisms for the monomer and dimer, respectively, with Li⁺ as the charge carrier. The monomer undergoes a 4-electron transfer reaction, while the dimer facilitates an 8-electron process due to its extended conjugation system.
- **FBND:** Density functional theory (DFT) calculations were employed to elucidate the charge-discharge mechanism of FBND, which involves a reversible enolization process. During the discharge process, FBND undergoes a two-step reduction, with the first step involving the formation of a radical anion and the second step leading to the formation of an enolate dianion.

These redox mechanisms and charge storage processes play a crucial role in determining the electrochemical performance of organic electrode materials, including their capacity, voltage profiles, and cycling stability. Understanding and optimizing these mechanisms is essential for developing high-performance organic electrodes for lithium-ion and other battery technologies [21, 22, 23, 24, 25].

3. Energy and Power Density

The dimer electrode material (DNP-Li) exhibited a remarkable energy density of 1.1 Wh/g, showcasing its potential as a high-performance organic cathode. Additionally, it demonstrated good rate capability, maintaining an impressive capacity of 300 mAh/g even at a high current density of 400 mA/g (1C). This rate performance is crucial for applications that require rapid charge and discharge cycles [26, 27, 28, 29, 30, 31, 32, 33].

Furthermore, an electrode composition with 83 wt% active dimer material achieved a very high capacity of 374 mAh/g at a moderate current density of 20 mA/g. This highlights the potential of optimizing electrode formulations to maximize the energy density and capacity of organic electrode materials (Fig. 2) [34, 35, 36, 37, 38].

To gain insights into the cost and performance implications of using n-type organic materials in battery packs, a detailed analysis was conducted using the BatPaC 5.0 software. This analysis considered the influence of various electrode design choices on energy density and cost:

- **Conductive Carbon Content:** The amount of conductive carbon additives in the electrode can impact both energy density and cost. Higher carbon content can improve electronic conductivity but may dilute the active material content, affecting energy density.

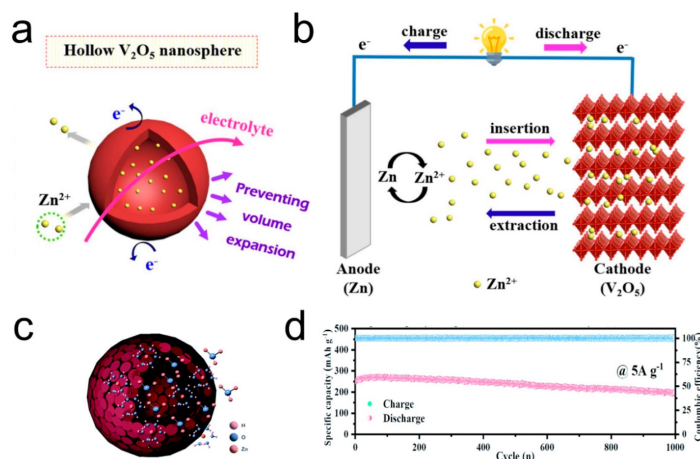


Figure 2. The fabrication of Hollow V2O5 nanospheres.

- **Active Material Mass Loading:** Increasing the mass loading of the active organic material in the electrode can enhance energy density but may introduce challenges related to ionic and electronic transport within the electrode.
- **Electrode Density:** Optimizing the electrode density can balance energy density and cost considerations. Higher density electrodes can improve volumetric energy density, but excessive compression may hinder ion transport and reduce capacity.

The analysis provided valuable insights into the trade-offs involved in electrode design and highlighted the importance of optimizing these parameters to achieve the desired balance between energy density, cost, and performance for organic electrode materials in battery applications [39, 40, 41].

4. Cycle Life and Stability

Evaluating the cycle life and stability of organic electrode materials is crucial for their practical implementation in lithium-ion batteries. The symmetric full cell utilizing the naphthazarin dimer (DNP-Li) as both the positive and negative electrode exhibited a promising capacity of 189 mAh/g, maintaining 122 mAh/g after 100 cycles. This demonstrates the potential of the dimer material to deliver stable cycling performance over an extended period (Fig. 3) [42].

The improved cycling stability of FBND compared to EFID can be attributed to its imide structure and extended π -conjugation. These structural features contribute to enhanced electrochemical performance and better capacity retention during cycling.

However, accurately determining the capacity of organic materials is a critical challenge. The measured capacity can be significantly lower than the theoretical capacity due to various factors, such as:

- Poor electrolyte accessibility
- High solubility of the organic material
- Side reactions occurring during cycling

To address this issue, rigorous testing procedures are necessary to subtract the contribution from conductive additives [43]. This ensures that the reported capacity accurately reflects the perfor-

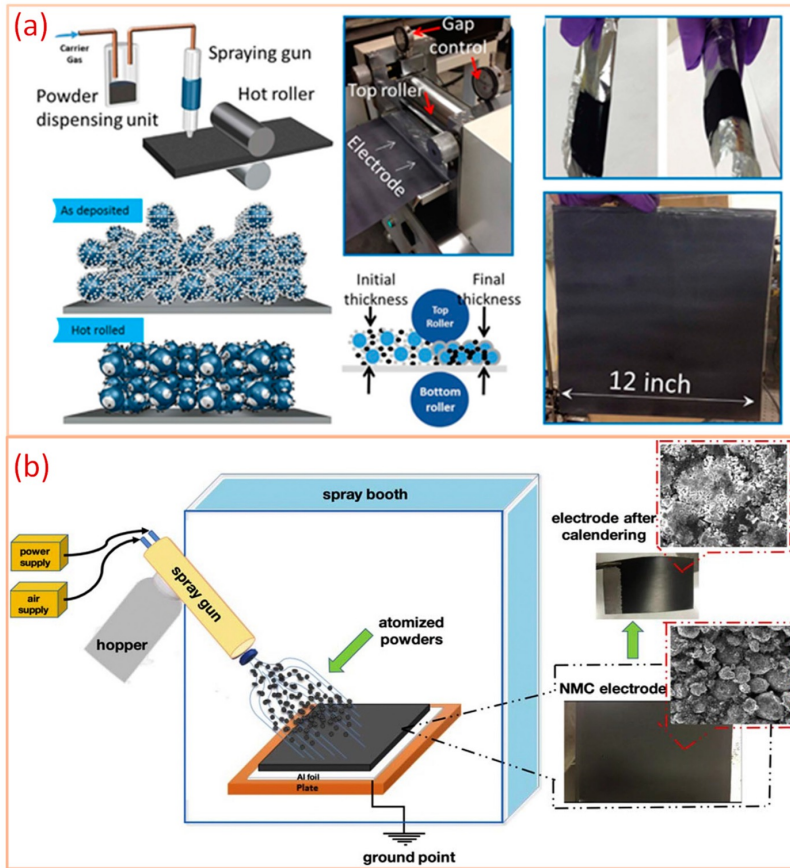


Figure 3. The dry spray deposition process for electrode fabrication.

mance of the organic material itself.

Furthermore, evaluating the obtained electrochemical capacity is essential. If the capacity is lower than the theoretical value, it may be due to:

1. Low utilization of the active material
2. Rapid capacity fade during cycling
3. Suboptimal electrode engineering

Addressing these challenges through advanced electrode engineering techniques, such as optimizing the electrode composition, mass loading, and density, can lead to improved cycle life and stability for organic electrode materials [30].

5. Gravimetric Density and Electronic Conductivity

Gravimetric density and electronic conductivity are crucial factors that influence the performance of organic electrode materials in lithium-ion batteries. These properties play a significant role in determining the energy density, power density, and overall efficiency of the battery system [31]. Gravimetric density, also known as specific density, refers to the mass of the active material per unit volume. Organic electrode materials typically exhibit lower gravimetric densities compared to their

inorganic counterparts. For instance, the gravimetric density of the naphthazarin dimer (DNP-Li) is approximately 1.5 g/cm^3 , while the gravimetric density of conventional inorganic cathode materials like lithium cobalt oxide (LiCoO) is around 5.1 g/cm^3 (Fig. 4).

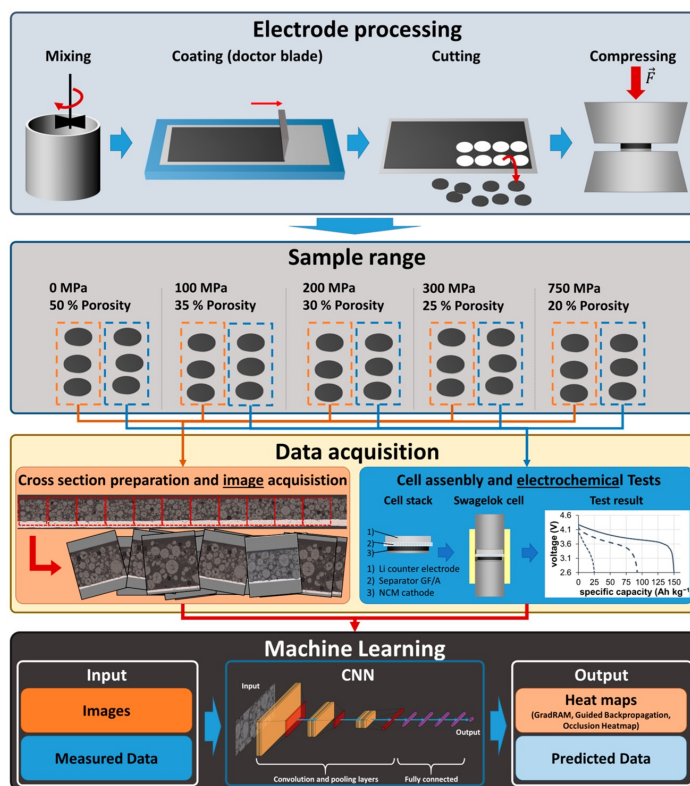


Figure 4. Methodical workflow showing electrode preparation.

The lower gravimetric density of organic materials can be advantageous in terms of reducing the overall weight of the battery pack, which is crucial for applications such as electric vehicles. However, it may also lead to lower volumetric energy densities, necessitating careful electrode engineering and optimization. Electronic conductivity is another critical factor that influences the performance of organic electrode materials. Many organic compounds exhibit relatively low electronic conductivity, which can hinder the efficient transport of electrons during the charge and discharge processes. This limitation can be mitigated through various strategies:

- **Conductive Additives:** Incorporating conductive additives, such as carbon black or carbon nanotubes, into the electrode formulation can enhance the overall electronic conductivity of the electrode composite.
- **Doping:** Introducing dopants, such as ionic liquids or redox-active polymers, can improve the electronic conductivity of organic electrode materials by facilitating charge transfer processes.
- **Structural Modifications:** Modifying the molecular structure of organic compounds through strategies like extending conjugation or introducing functional groups can enhance their intrinsic electronic conductivity.
- **Nanostructuring:** Designing organic electrode materials with nanostructured architectures, such as nanoparticles or nanofibers, can increase the surface area and facilitate efficient charge transport

pathways.

It is essential to strike a balance between the gravimetric density, electronic conductivity, and other performance parameters when designing and optimizing organic electrode materials for lithium-ion batteries. Advanced electrode engineering techniques, such as tailoring the electrode composition, mass loading, and porosity, can help achieve the desired trade-offs and maximize the overall performance of the battery system [31, 33].

6. Electrode Engineering and Optimization

Computational analysis provided valuable insights into the improved cycling stability of the naphthazarin dimer (DNP-Li) compared to the monomer (NP-Li). The dimer exhibited stronger intermolecular interactions and lower solvation energy, which contributed to its enhanced cycle life performance [34].

However, organic cathodes often face challenges such as low conductivity, leading to poor rate performance, and high solubility in traditional carbonate-based electrolytes. To address these issues, researchers have explored various electrode engineering and optimization strategies (Fig. 5):

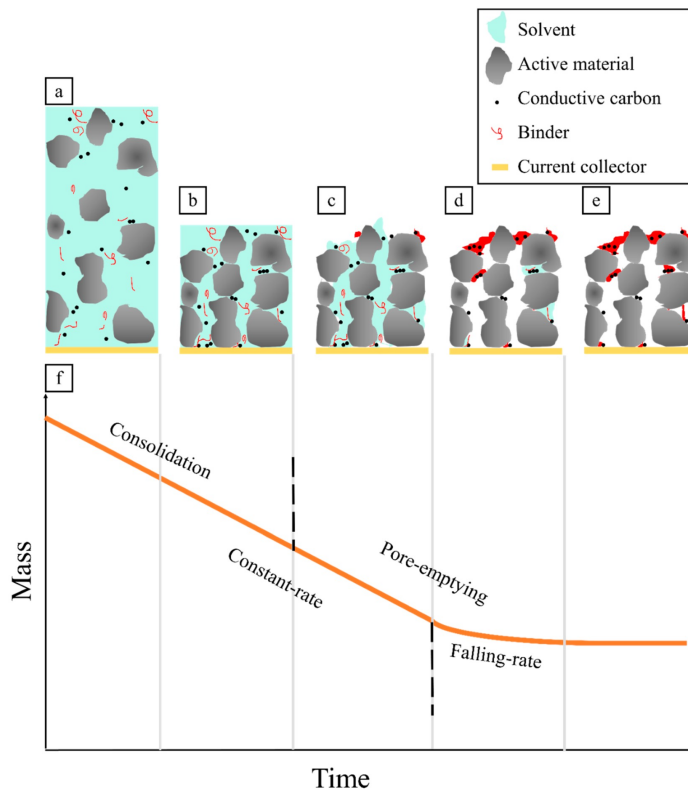


Figure 5. Illustration of the electrode-film drying mechanism

1. Electrolyte Optimization:

- Ether-based electrolytes, specifically 1-4 M lithium bis(trifluoromethanesulfonyl)imide in 1,3-dioxolane/dimethoxyethane, were found to improve the solubility of organic electrode materials compared to traditional LiPF₆ in diethyl carbonate electrolyte.

- The optimal electrolyte concentration was determined to be 2 M.
2. **Anode Protective Additive:**
 - The addition of 1% LiNO_4 additive to the electrolyte formed a protective film on the lithium metal anode, improving cycling performance.
 3. **Cathode Nanocomposite:**
 - The organic anthraquinone (AQ) cathode was impregnated into mesoporous carbon at a ratio of 30 wt% carbon, significantly improving performance at high currents by reducing ion transfer impedance.
 4. **Performance Metrics:**
 - The fully optimized cell, comprising a lithium anode, 30 wt% carbon AQC cathode, and 1% LiNO_4 2M-DD electrolyte, demonstrated 85% capacity retention after 100 cycles, a significant improvement over previous organic cathode cells.
 - This optimization strategy has also been applied to other biphenyl quinone-based cathodes, achieving over 97% of theoretical capacity and up to 84% capacity retention after 100 cycles.

These electrode engineering and optimization techniques have proven effective in enhancing the performance of organic electrode materials, addressing challenges related to conductivity, solubility, and cycling stability. By combining computational analysis, electrolyte optimization, protective additives, and nanocomposite architectures, researchers have made significant strides in improving the viability of organic materials for lithium-ion and other battery technologies [35, 37].

7. Cost and Resource Availability

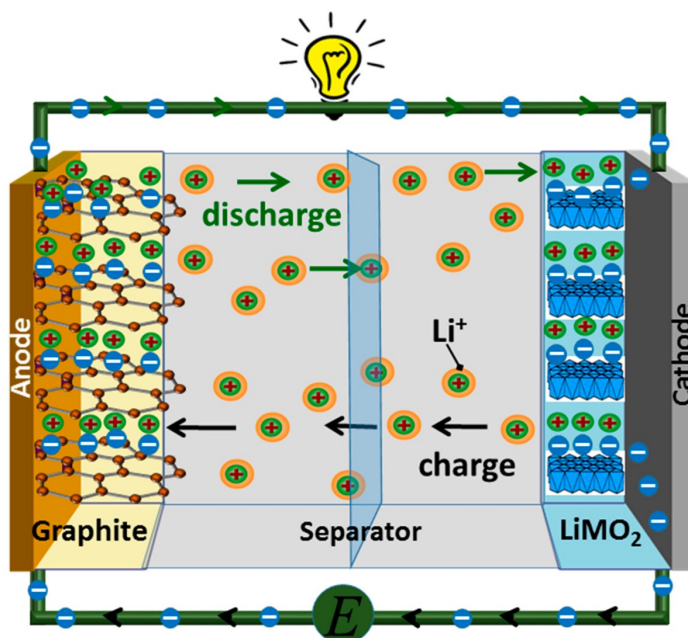


Figure 6. Schematic of the operation principles for rechargeable lithium ion batteries.

The high demand for critical minerals like lithium, copper, nickel, and cobalt required for lithium-ion batteries has raised questions about maintaining a steady and affordable supply of raw materials. As the adoption of electric vehicles and renewable energy storage systems continues to grow, the pressure on these finite resources is expected to intensify. Here are some key considerations regarding

the cost and resource availability of organic electrode materials (Fig. 6):

- **Sustainable and Abundant Resources:** Organic electrode materials are typically derived from carbon-based compounds, which can be obtained from renewable and abundant sources such as biomass, agricultural waste, or even carbon dioxide. This presents a significant advantage over traditional inorganic materials that rely on scarce and geographically concentrated mineral resources.
- **Cost-Effective Production:** Many organic compounds can be synthesized using relatively inexpensive and scalable chemical processes, potentially reducing the overall manufacturing costs of organic electrode materials compared to their inorganic counterparts.
- **Recycling and Reuse:** The organic nature of these materials may facilitate easier recycling and reuse processes, further contributing to cost savings and resource sustainability. However, research is still needed to develop efficient recycling methods for organic electrode materials.
- **Scalability Challenges:** While organic materials offer promising prospects in terms of resource availability and cost, scaling up their production to meet the rapidly growing demand for energy storage solutions remains a challenge. Significant investments in research, development, and manufacturing infrastructure may be required to achieve the necessary economies of scale.

Table 1. Resources and significances

Resource	Advantages	Challenges
Organic Electrode Materials	Sustainable and abundant sources, cost-effective production, potential for recycling and reuse	Scalability challenges, need for efficient recycling methods
Inorganic Electrode Materials	Well-established production processes, high energy density	Reliance on scarce mineral resources, higher production costs, recycling complexities

It is important to strike a balance between the performance, cost, and resource availability of electrode materials to ensure the long-term sustainability and affordability of lithium-ion and other battery technologies as given in Table 1. Ongoing research and development efforts are crucial to address the scalability challenges and unlock the full potential of organic electrode materials in meeting the growing energy storage demands [38].

8. Future Prospects and Challenges

The potential of n-type organic materials as a low-cost and sustainable solution for energy storage is highlighted, while emphasizing the need for further advancements. The following points underscore the future prospects and challenges in this domain (Fig. 7):

- **Scalability and Manufacturing:** While organic electrode materials offer cost-effective production and abundant resource availability, scaling up their manufacturing to meet the rapidly growing demand for energy storage solutions remains a significant challenge. Substantial investments in research, development, and manufacturing infrastructure are necessary to achieve the required economies of scale.
- **Performance Optimization:** Despite the promising attributes of organic materials, their electrochemical performance, including energy density, power density, and cycling stability, often lags

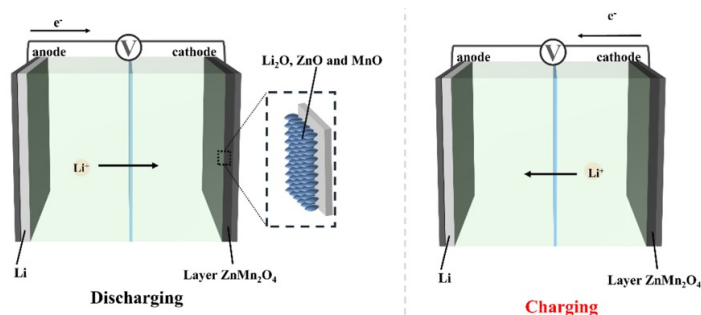


Figure 7. Charge and discharge processes.

behind conventional inorganic counterparts. Ongoing research efforts are crucial to optimize the molecular design, electrode engineering, and electrolyte formulations to bridge this performance gap.

- **Electrolyte Compatibility:** The solubility of organic electrode materials in traditional carbonate-based electrolytes can lead to capacity fade and degradation. Developing electrolyte systems that are compatible with organic materials, while maintaining high ionic conductivity and electrochemical stability, is a key challenge that needs to be addressed.
- **Structural Stability and Degradation Mechanisms:** Understanding the structural changes and degradation mechanisms that organic materials undergo during cycling is essential for improving their long-term stability and cycle life. Advanced characterization techniques and computational modeling can provide valuable insights into these processes and guide the design of more robust organic electrode materials.

Table 2. Typical Prospects and Challenges

Prospects	Challenges
Cost-effective production	Scalability and manufacturing
Sustainable resource availability	Performance optimization
Potential for recycling and reuse	Electrolyte compatibility
	Structural stability and degradation mechanisms

From the Table 2, while the use of n-type organic materials in lithium-ion batteries holds significant promise, overcoming these challenges through continued research and development efforts is crucial for realizing their full potential as a sustainable and cost-effective energy storage solution.

9. Conclusion

The exploration of organic materials for lithium-ion batteries has unveiled promising prospects for sustainable, cost-effective, and high-performance energy storage solutions. These materials, derived from abundant and renewable resources, offer advantages such as low production costs, potential for recycling, and compatibility with sustainable practices. However, challenges remain in optimizing their electrochemical performance, ensuring structural stability, and scaling up manufacturing processes to meet the growing demand. While significant progress has been made in understanding the redox mechanisms, charge storage capabilities, and electrode engineering strategies for organic

electrode materials, further research and development efforts are crucial. Addressing issues such as electrolyte compatibility, degradation mechanisms, and performance optimization will pave the way for the large-scale adoption of organic materials in lithium-ion and next-generation battery technologies. The future holds exciting opportunities to harness the unique properties of these materials and contribute to a more sustainable and affordable energy landscape.

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