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A Review of Cathode Materials for Lithium-ion Batteries and Future Prospects

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Abstract:

Lithium-ion battery (LIB) is widely used in electric vehicles, computers, mobile phones and other electronic products because of its high energy density, good rate performance and long life. This paper concentrates on the research of cathode materials for lithium-ion batteries. Through literature review and comparative analysis, four commonly used cathode materials for lithium-ion batteries are evaluated, including lithium cobalt oxide ($LiCoO_2$), lithium manganese oxide ($LiMn_2O_4$), Ferrous lithium phosphate ($LiFePO_4$) and emerging nickel-rich ternary materials (NCM and NCA) from the perspective of structure and performance. These materials are unique, but they all have some shortcomings and face some challenges. This paper further outlines the future research direction for cathode material, including the exploration and development of new materials and technology, as well as environmental and sustainability concerns. Finally, this paper concludes that the future development of cathode materials for lithium-ion batteries will depend on interdisciplinary cooperation and innovation to realize more efficient, safer and more environmentally friendly energy storage technology.

Keywords: lithium-ion batteries; cathode material; lithium storage.

1. Introduction

The world is experiencing an increasing energy crisis and environmental problems in the 21st Century. The traditional fossil fuel resources on the earth are limited and their large-scale usage has caused serious pollution and climate changes. In order to deal with these crises, countries all over the world have made many attempts and efforts, among which it is very important to actively promote electrification transformation. The development and application of advanced batteries is at the core of electrification. The storage and conversion of energy from diverse sources into the necessary forms for application in sectors including utilities, industry, buildings, and transportation are essential functions performed by battery systems [1]. The most common electrochemical secondary batteries include lead acid battery, nickel cadmium battery, nickel-hydrogen battery and lithium-ion battery. Lithium-ion battery (LIB) is widely used in electric vehicles, computers, mobile phones and other electronic products because of its high energy density, good rate performance and long life. In 1980, the scientist M. B. Armand introduced a revolutionary concept for a secondary battery design, known as the “Rocking-chair battery” [2]. The introduction of this concept has a far-reaching impact on the subsequent development of lithium-ion batteries. It also paved the way for the batteries’ widespread use and acceptance in other industries, like renewable energy storage and automotives.

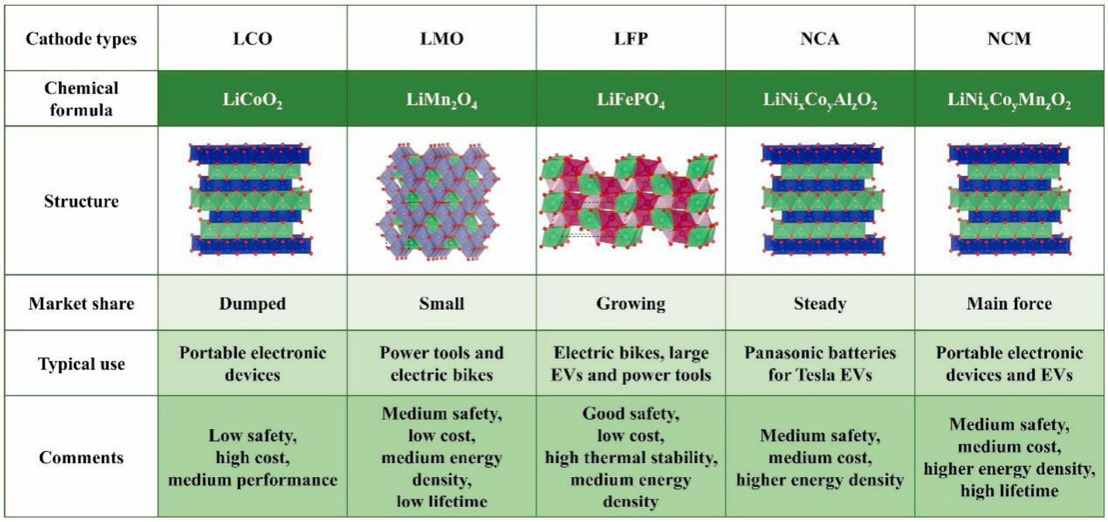
The structure of a lithium-ion batteries is composed of four main parts: cathode, anode, electrolyte and separator. Each component is essential to the normal operation and performance of lithium batteries. The anode, cathode, and other materials used in the battery should be able guarantee reversible intercalation and deintercalation of the lithium. This process is central to the process of battery charging and discharging. The choice of anode and cathode materials can make a big difference in battery performance and lifespan. The electrolyte must have a high ionic conductivity so that lithium ions are able to move quickly and smoothly between the cathode and anode when charging and discharging. The separator’s main purpose is to isolate the cathode and anode of the batteries, prevent electrons short-circuiting through the battery and ensure that lithium can freely pass through electrolyte.

Although lithium-ion batteries have good performance and are widely used at present, there are some problems in the actual situation. First, the electrolyte of lithium-ion battery is prone to spontaneous combustion, and it is prone to safety accidents when it is affected by the outside world. Especially for high-power lithium-ion batteries of electric vehicles, some cases of spontaneous combustion of electric vehicles have aggravated people’s concerns. At the same time, the performance of lithium-ion batteries will be affected to some extent at extreme temperatures, whether low or high. Finally, although the cost is gradually reduced by the improvement of technology, some key cathode metal materials (especially cobalt) in lithium-ion batteries are expensive, which increases the cost and limits its wider application. The research on cathode materials is one of the hot research directions of lithium-ion batteries. For example, in the study of fast charging, the specific step of the cathode material (CAM) is considered as the key factor to limit the rate [3]. New materials need to pursue lower cost and better performance at the same time. At present, lithium cobalt oxide ($LiCoO_2$), nickel cobalt manganese ternary material (NCM), lithium manganate ($LiMn_2O_4$), Ferrous lithium phosphate ($LiFePO_4$) and other cathode materials are extensively employed in commercial lithium-ion batteries. The batteries made of these materials have their own characteristics and are suitable for different application scenarios.

This review will report on various cathode material types since the development and introduction of lithium-ion battery technology. It will also analyze the advantages and disadvantages, and make suggestions and outline potential directions for the advancement of cathode material research and development in lithium-ion battery technology.

2. Cathode Material

Comparative analysis of the cathode materials used in common commercial LIBs is shown in Fig. 1. Next, this review will describe these materials in detail on the basis of these classifications and comparisons.



Cathode types	LCO	LMO	LFP	NCA	NCM
Chemical formula	$LiCoO_2$	$LiMn_2O_4$	$LiFePO_4$	$LiNi_xCo_yAl_zO_2$	$LiNi_xCo_yMn_zO_2$
Structure					
Market share	Dumped	Small	Growing	Steady	Main force
Typical use	Portable electronic devices	Power tools and electric bikes	Electric bikes, large EVs and power tools	Panasonic batteries for Tesla EVs	Portable electronic devices and EVs
Comments	Low safety, high cost, medium performance	Medium safety, low cost, medium energy density, low lifetime	Good safety, low cost, high thermal stability, medium energy density	Medium safety, medium cost, higher energy density	Medium safety, medium cost, higher energy density, high lifetime

Fig. 1 Comparative analysis of the cathode materials used in common commercial LIBs [4]

2.1 Lithium Cobalt Oxide ($LiCoO_2$)

The $LiCoO_2$ (LCO) cathode, first introduced by Goodenough, has become the most successful of the layered transition metal oxides in commerce [5]. LCO-based batteries dominate lithium-ion battery materials (LIBs) in computers, communication equipment, and consumer electronics because of their easy processing, good performance, and high energy density.

The different temperatures during production lead to the formation of two crystal types: lamellar and spinel. High-temperature melting forms a layered lithium cobalt material with a cubic spinel structure at low temperatures. However, the spinel structure of the lithium cobalt-cobaltate material is not conducive to de-embedding Li ions during charging and discharging, and its electrochemical performance does not meet the required standards for practical production [6]. Insertion cathodes store guest ions that can be reversibly inserted and removed from the host material. In a Li-ion battery, Li^+ acts as the guest ion while the host network is a compound. Co and Li are arranged in octahedral layers, alternating between them, and forming a hexagonal symmetry. The structure of lithium-cobaltate is a α-$NaFeO_2$ layered structure. The structural similarity is of great consequence for the maintenance of the stable electrochemical properties of lithium cobaltate, which is responsible for its performance in batteries. The oxygen atoms in the structure form a mixture between CoO_6 and LiO_6 atoms with the nearest Co atoms and Li atoms, respectively. An overall structure can be determined by observing how three atoms alternate; their arrangement in the crystal structure usually follows this rule and the de-embedding of lithium ions does not negatively impact overall structural stability. The theoretical specific capacity of lithium cobaltate material is 274 $mAh \cdot g^{-1}$. It is an ideal cathode material for lithium-ion batteries, and has also been widely used in reality. But lithium cobaltate has some disadvantages.

Main constraints of this technology include its expensive nature, poor thermal stability, and quick capacity deterioration during high current rates or deep cycles [7]. Lithium cobaltate is a costly material, largely due to the volatility of its primary raw material, cobalt, and its scarcity. Lithium cobalt oxide also has poor thermal stability at high temperatures, reacting with oxygen to release toxic gases and cause battery overheating. Lithium cobaltate batteries must be operated within 4.25V of their working voltage for optimal performance. Exceeding this range may result in irreversible phase change processes that impede battery performance and energy density enhancement efforts. At high voltages, lithium cobaltate will gradually transition from its hexagonal state into monoclinic form; an irreversible phase transition may then take place at this point as well.

Modification of lithium cobalt dioxide is one approach to enhancing the performance and durability of lithium-cobalt oxide materials. Modification is primarily achieved through two methods: surface coating and doping modification. Surface coating typically employs conductive materials, whereby conductive materials, such as carbon, are introduced to the surface of lithium cobaltate. This significantly enhances the electron transfer rate and decreases the thermal resistance of the battery, thereby improving the cycle life of the material. The doping of lithium cobaltate in the bulk phase has been demonstrated to markedly enhance the stability and cycling performance of the material by the introduction of specific elements. For example, doping with aluminum ions improves cycling performance under high pressure and reduces costs.

2.2 Lithium Manganate ($LiMn_2O_4$)

Lithium manganate (LMO) cathode materials are predominantly spinel structures with tetragonal symmetry, situated within the Fd3m space group and belonging to the cubic crystal system. Manganese and oxygen atoms are intertwined to form the mesh structure of Mn_2O_4, allowing lithium ions to travel freely through it. The specific capacity of lithium manganate material is 148 $mAh \cdot g^{-1}$ in theory.

Lithium manganate has the advantage of a low price and abundant resources, which makes it a cost-effective material that may be used in a variety of applications. At the same time, lithium manganate has better safety characteristics. It operates at a lower voltage, reducing the risk of thermal runaway, and such batteries have better safety. Moreover, lithium manganate is among the first cathode materials found in commercially available lithium-ion batteries. It has a well-established manufacturing process and finds extensive use in various applications..

However, lithium manganate has also revealed a lot of problems in the course of years of use. Lithium manganate exhibits pronounced capacity deterioration during prolonged cycling, particularly at elevated temperatures. This phenomenon is primarily attributable to the dissolution-migration-deposition (DMD) process of manganese ions (Mn), which results in the deterioration of the cathode material's structure and the loss of active substance [8]. Lithium manganate batteries generate less heat overall but are inherently less thermally stable. At higher temperatures, lithium manganate will react with its electrolyte faster, producing greater amounts of acid production and potentially leading to runaway batteries.

Similarly, lithium manganate can be modified to enhance

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the functionality of the electrode. The doping modification of materials can be achieved by the introduction of metal cations (e.g. Co and Cr) to increase the particle radius of the material and reduce contact with the electrolyte. This results in a decreased decomposition rate of the electrolyte and electrodes. Anionic doping (e.g. F and S) improves crystal stability and inhibits capacity degradation, especially at high temperatures. Composite doping further enhances battery performance by combining the advantages of cations and anions. An oxide layer, such TiO_2, on the cathode material's surface can be covered using the cladding modification. This serves to isolate the direct contact between the material and the electrolyte, reduce the dissolution of manganese, and simultaneously neutralise the HF in the electrolyte. The result is an enhancement in the electrochemical stability and cycling performance of the battery.

2.3 lithium Iron Phosphate ($LiFePO_4$)

In 1997, Goodenough and Padhi, among others, proposed the use of $LiFePO_4$ material with an olivine structure ($LiFePO_4$) as a cathode material for lithium-ion batteries [9]. $LiFePO_4$ has an ordered-olivine structure and is a member of the space group Pnma. The arrangement of the oxygen atoms is hexagonal close packed and slightly distorted. The iron and lithium atoms are situated at the 4a and 4c sites of the octahedral structure, while the phosphorus and iron atoms are in the tetrahedral position. The theoretical specific capacity of $LiFePO_4$ is 170 $mAh{\cdot}g^{-1}$. Lithium Iron Phosphate batteries ($LiFePO_4$) stand out from other battery technologies because of their high safety, high cycle life, high power density, fast charging ability, environmental friendliness, recyclability and low manufacturing costs. Lithium iron phosphate batteries are highly stable, less susceptible than other battery types to overheating and combustion, and provide safety and performance even under extreme conditions. As such, they're the ideal choice for electric cars, energy storage systems, renewable energy storage solutions. And early research into electric vehicles frequently used this cathode material as their cathode material for batteries.

As it stands, lithium iron phosphate materials also face a number of challenges. The theoretical density of $LiFePO_4$ powder is $3.6g{\cdot}cm^{-3}$, which is significantly lower compared to that of $LiCoO_2$ ($5.01g{\cdot}cm^{-3}$), $LiNiO_2$ ($4.88g{\cdot}cm^{-3}$), and $LiMn_2O_4$ ($4.20g{\cdot}cm^{-3}$). Furthermore, $LiFePO_4$'s tap density is poor. [10]. A further disadvantage is the poor performance exhibited by the battery at low temperatures. At lower temperatures (e.g. -20°C), the diffusion rate of lithium ions in the electrolyte is significantly impeded, resulting in a battery with diminished low-temperature tolerance.

The properties of the lithium iron phosphate crystal structure, including polarisation phenomena and defects in the crystal structure, can give rise to a number of issues. These include slow diffusion in ion channels of lithium ions and low electronic conductivity. The modification of lithium iron phosphate represents a current area of research. The primary methods employed are surface coating and ion doping. The application of a carbon capping layer can facilitate the formation of a carbon cladding over the surface of the $LiFePO_4$ particle. This process has been observed to enhance the electronic conductivity of the particle, facilitate improved electrolyte contacts, and reduce the interfacial resistance between the electrode and the electrolyte. Additionally, the carbon capping layer has been shown to enhance the conductive ability of the particle. The application of a coating to some metal oxides on the surface (such as Ga) can be combined with electrodes to form a network with enhanced conductivity and conductivity itself increased. In practice, metal and carbon coatings are employed in conjunction with one another. The doping of ions within the $LiFePO_4$ crystal lattice allows for the replacement of Li, Fe, and O. The various doping elements can be classified as either metallic or non-metallic. The doping of these elements can result in enhanced stability of the $LiFePO_4$ crystal and an enhancement in the performance of electrodes.

2.4 Nickel-rich Cathode Materials (NCA/NCM)

Dyer et al. first discovered the layered $LiNiO_2$ material in 1954, from which nickel-rich cathode materials are derived [11]. A substantial body of evidence from numerous studies indicates that $LiNiO_2$ exhibits a notable deficiency in structural stability. Nevertheless, the incorporation of additional metal elements may prove an effective means of addressing this challenge. The substitution of cobalt, manganese and aluminium among all metal elements is of particular importance in order to enhance the performance of the nickel-rich cathode material. The nickel-rich lithium ternary battery was produced on this basis. Ternary lithium battery is a kind of lithium-ion battery with nickel, cobalt, manganese (NCM) or nickel, cobalt, aluminium (NCA) as the cathode material. The layered hexagonal structure of a-NaFeO2 with space group R-3m is present in these materials. In this structure, the lithium (Li), occupies the position 3a. Transition metals (Ni or Co, Mn or Al), in disorder, occupy position 3b. And oxygen (O) occupies 6c. This forms an octahedral MO_6 structure. These materials have comparable operating voltages and achievable specific capacities to those of LCOs, but are more suitable for widespread use because they are multi-metal blends with reduced Co content and lower prices.

At the same time changing the ratio of different metals can optimise the performance of ternary materials to meet the needs of different application scenarios. For instance, several studies have substantiated the observation that an elevated Ni content is associated with augmented storage capacity but diminished electrode stability. In particular, an increase in the Ni ratio results in the formation of high-valent Ni ions (Ni^{3+} and Ni^{4+}), which contribute to an enhanced capacity [12].

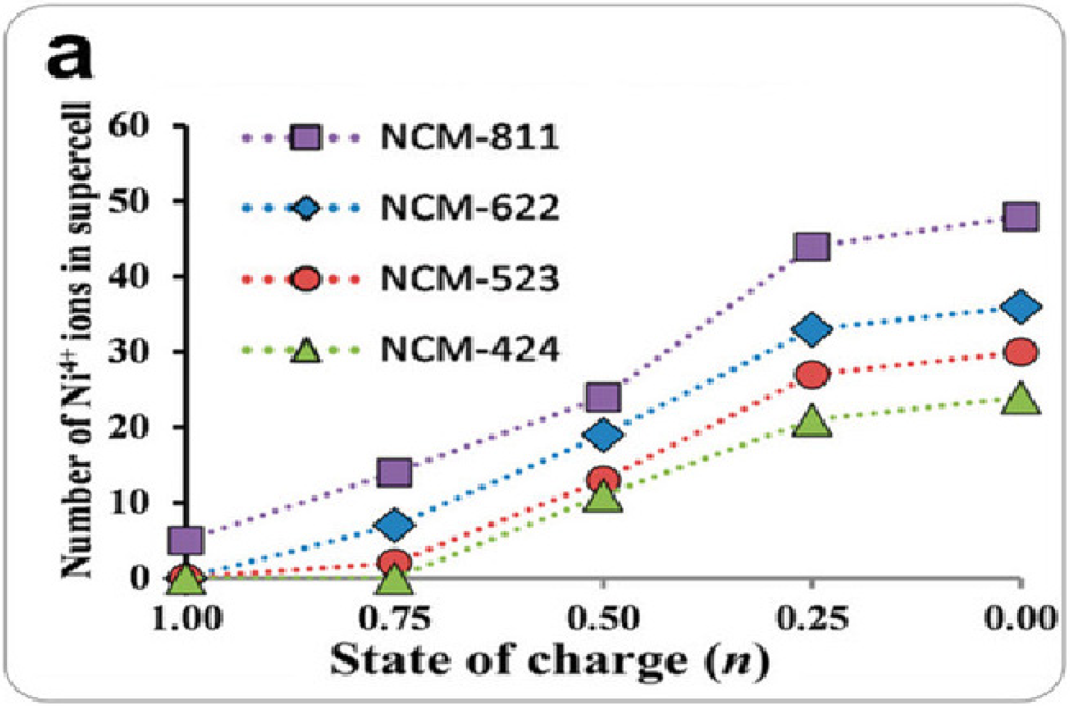

Fig. 2 Calculation of DFT to determine the amount of Ni^{4+} in the supercells of different NCMs [13]

The high valence nickel ion (Ni^{4+}) in NCMs is shown in Figure 2. It can be observed that the concentration of Ni^{4+} progressively rises from NCM with a reduced nickel content to NCM with a high nickel content, accompanied by an enhancement in energy density. However, high nickel content will also bring risks. Ni^{4+} and Ni^{3+} are easy to react with electrolyte, resulting in the decline of battery stability and non-cycling.

The primary distinction between NCM and NCA is the final element employed: NCM utilises Mn, whereas NCA employs Al. The use of Mn can assist in stabilising the stability of nickel-rich batteries, thereby extending their lifespan and enhancing safety. Similarly, aluminium can enhance battery capacity; however, this may entail additional costs and necessitate more sophisticated technology. It can be concluded that both batteries have a place in the electric vehicle market at the present time, with each having its own advantages.

3. Prospects

Every time the performance and longevity of lithium-ion battery (LIBs) are improved, this has a profound effect on the sustainability of the society. The development and innovation of cathode material will be key in the advancement of battery technology as we strive for higher energy density, faster charge speeds, and longer battery lives. And when developing the cathode material for lithium-ion batteries, three factors will be taken into consideration.

3.1 Exploration of New Cathode Materials

Future research should concentrate on developing new cathode materials to overcome the energy density limitations of conventional ones. In recent years, lithium-sulfur battery technology has attracted a lot of attention. And in these batteries, the cathode is made of sulfur, while the anode is usually made up of lithium or a Lithium alloy. Sulfur offers a high theoretical energy density and is cost-effective. However, lithium-sulfur batteries currently face several challenges, including rapid capacity degradation, low conductivity of the sulfur cathode, the polysulfide shuttle effect, lithium-ion deposition, and structural changes due to volume fluctuations during charging and discharging. These issues hinder the large-scale commer-

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cial production of lithium-sulfur batteries [14]. Other lithium-ion battery types utilizing novel materials have also attracted significant attention. Rechargeable lithium-air battery, for example, has the highest theoretical specific power of all rechargeable batteries. Additionally, green, sustainable, and versatile organic cathode materials are being explored.

3.2 Safety and Reliability

Lithium-ion batteries are widely used in daily life, so it is critical that any study of them prioritize safety and dependability. The most frequent occurrence in lithium-ion battery technology, thermal runaway, has raised concerns about new energy vehicles' spontaneous combustion. Recent advancements have been made in the research on thermal runaway. The team led by Ou Yang utilized an accelerated adiabatic calorimeter to perform multiple thermal runaway tests on various lithium-ion battery systems. They identified three key characteristic temperatures, designated as T1, T2, and T3 [15]. These temperatures are defined as follows: T1 is the self-heating onset temperature (where the temperature rise rate reaches 0.02℃/min), T2 is the thermal runaway trigger temperature (where the temperature rise rate reaches 1℃/s), and T3 is the maximum thermal runaway temperature. Investigating these temperatures provides deeper insights into the thermal runaway process, enabling more effective accident prevention. The most frequent issues with cathode materials are lattice structure collapse and lattice oxygen release. Lattice structure collapse occurs due to irreversible lattice distortion during phase transitions, while lattice oxygen release is driven by oxygen acting as an electron donor in an active state. Currently, the primary approach to enhancing cathode material stability involves modifying the materials themselves.

3.3 Environmental and Sustainability Considerations

As lithium-ion battery technology advances, concerns regarding the environmental impact and recycling of spent lithium-ion cells are growing. Over its lifecycle, the environmental impact of a lithium-ion battery is generally positive, as it contributes to reducing carbon emissions and replacing some fossil fuel consumption, particularly in the automotive industry. Waste lithium-ion batteries contain various toxic electrolytes and flammable organic solvents, which are highly corrosive and pose significant environmental hazards. If improperly recycled, harmful substances from the battery cathode materials can leach into the environment, leading to heavy metal pollution and posing serious risks to human health. Materials like ferrous lithium phosphate, lithium cobaltate, and lithium manganate in the anodes of waste lithium-ion batteries can react with the electrolyte. If not properly managed, these reactions can lead to heavy metal pollution, causing significant environmental harm. Therefore, the recovery of these materials is essential. Recycling battery materials is a vital part of the lithium-ion battery lifecycle, enabling the extraction of valuable metals from waste batteries and helping to mitigate the resource shortages and mining pressures on rare metals like nickel, cobalt, and lithium [19]. Currently, various reagents are primarily used for the wet recovery of materials. Since the reaction characteristics of waste lithium-ion batteries differ based on the cathode materials, it is crucial to fully comprehend the redox mechanisms for efficient recycling management. Additionally, developing greener and non-toxic reagents for recycling is a key focus for environmental protection.

4. Summary

This review examines the current status and future trends of cathode material for lithium-ion battery, highlighting the crucial role of material innovation to enhance battery performance. Tracing the evolution from the initial discovery of lithium cobaltate ($LiCoO_2$) to the advanced development of widely-used nickel-rich cathode materials (NCA/NCM), this review underscores the unique advantages and diverse applications of each material, demonstrating the versatility of lithium-ion batteries. Additionally, it is observed that enhancements in these cathode materials are primarily achieved through modifications. Therefore, advancing various modification techniques remains a crucial approach to further improving lithium-ion battery performance. This review highlights the critical role of ongoing innovation in lithium-ion battery cathode materials in improving energy density, safety and sustainability. Consequently, future research should prioritize the development of novel materials, safety enhancement strategies, and the integration of environmentally-friendly recycling technologies. These recommendations can guide future researchers in identifying key research directions, thereby contributing to the diversified advancement of lithium-ion batteries. While this review provides a comprehensive overview of current research on cathode materials for lithium-ion batteries, certain gaps in the existing research remain. Future studies should place greater emphasis on harnessing large-scale models and AI technologies, which have already made substantial contributions to materials science but remain insufficiently explored in the investigation of cathode materials for lithium-ion batteries.

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