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# *A Systematic Review on the Research Progress of Three Mainstream Cathode Materials for Sodium-Ion Batteries*

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***Abstract.*** Driven by the global energy transformation and the "dual carbon" goal, the demand for large amount storage in renewable energy has expanded significantly. However, lithium-ion batteries can hardly satisfy such low-cost and high-safety requirements, due to limited amount and uneven distribution of lithium. Conversely, sodium ion batteries (SIBs) with advantages of sufficient sodium resources, low cost and excellent safety, have become a promising alternative technology. As one of SIBs' core, cathode materials directly determine key performances such as energy density and cycle life. Their performance weakness restrict the development of SIBs, which makes the research on cathode materials significant. This paper provides a review on the research progress of three mainstream cathode materials for SIBs, including layered oxides, polyanionic compounds, and Prussian blue analogs, and focuses on their crystal structures, electrochemical performances and existing deficiencies. Finally, this paper summarizes key modification strategies and outlines promising research directions, thereby offering insights for related studies and supporting the optimization of electrode materials as well as the advancement of their industrial application.

***Keywords:*** Sodium-ion batteries, Cathode materials, Layered oxides, Polyanionic compounds, Prussian blue analogs

## 1. Introduction

Global energy structure transformation is continuously deepening, and the realization of the "dual carbon" goal has become an urgent task. Moreover, secondary batteries are highly demanded in applications including large-amount energy storage, electric vehicles, and off-grid power supply in extreme environments, where low cost, high safety, and long cycle life are essential requirements. Lithium-ion batteries are widely commercialized in many fields, because of the high energy density and excellent cycle performance. However, lithium resources are scarce and unevenly distributed geographically, with approximately 70% of global lithium reserves located in South America [1]. This situation may lead to high costs and unstable supply chains, restricting the sustainable development of the energy storage and the security of the industrial chain.

Sodium-ion batteries (SIBs) emerge as a promising alternative to LIBs, gaining attention from both academia and industry. The key advantage is the abundant sodium resources. The abundance of sodium in earth is around 1300 times that of lithium, having lower extraction costs. In addition, SIBs

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exhibit excellent electrochemical performance, with much lower risk of lithium dendrite formation, thus demonstrating outstanding safety. Therefore, SIBs are particularly suitable for large-amount energy storage and low-cost power battery applications.

As a core component of SIBs, cathode materials determine the key electrochemical properties of batteries, including energy density, cycle life, rate capability, and operating voltage. The performance weaknesses of cathode materials have become the main bottleneck limiting the application of SIBs.

At present, there are mainly three types of cathode materials for SIBs. They are layered transition metal oxides, polyanionic compounds, and Prussian blue analogs. Each type of material has its own advantages and inherent weaknesses. Layered oxides have high energy density. But their structural stability is insufficient. Polyanionic compounds have excellent cycle stability. But their electronic conductivity is low. Prussian blue analogs have fast ion transport capability. But they suffer from crystalline water and structural defects. Numerous studies have been carried out on these three types of materials. However, there is still a lack of a systematic summary. The summary should cover their research progress, performance characteristics, and existing problems. This is not conducive to the in-depth research and performance optimization of SIBs cathode materials. Therefore, this review systematically summarizes the research progress of the three mainstream cathode materials for SIBs. It analyzes the crystal structures, electrochemical performances, and existing deficiencies of each type of material in detail. It also sorts out the common modification strategies. This work aims to provide a comprehensive reference for relevant researchers. It also promotes the in-depth study of SIBs cathode materials.

## 2. Introduction to sodium ion batteries

### 2.1. The basic structure and working process of sodium-ion batteries

A typical sodium-ion battery generally consists of the cathode, anode, electrolyte, separator, current collector and casing. Its core working mechanism involves the reversible insertion and extraction process of sodium ions ($Na^+$) between the cathode and anode materials, accompanied by the directional flow of electrons through the external circuit. As a result, SIBs achieve the conversion between chemical energy and electrical energy.

In the discharge process, the sodium-embedded cathode material undergoes an oxidation reaction, releasing $Na^+$ and electrons ($e^-$). $Na^+$ migrates through the electrolyte and the separator to the anode, with electrons flowing through the external circuit to the anode, driving the load to operate. At the anode, $Na^+$combines with electrons and is embedded in the anode material, forming a sodium-embedded anode. Conversely, during the charging process, under the drive of an external power source, the above process occurs in reverse. $Na^+$is released from the anode, migrating back to the cathode, while electrons flow from the anode back to the cathode. The specific process is shown in Figure 1.

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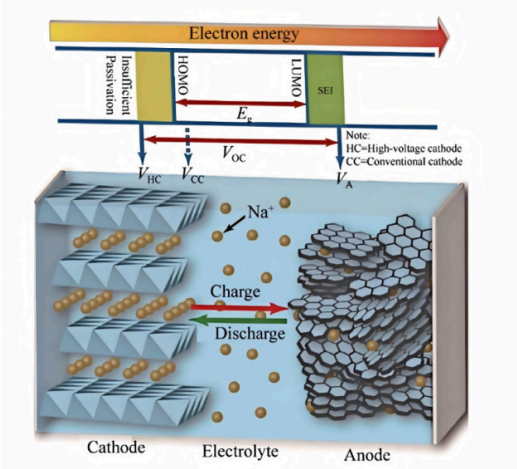



Figure 1. The working principle of SIBs [2]

Throughout the entire process, the electrode materials are the site where the electrochemical reactions occur and as the carriers for sodium ions. Their properties directly determine the core performance of the battery.

## 2.2. Key performance parameters

The comprehensive performance of sodium-ion battery electrode materials is determined by multiple interrelated parameters. Among them, energy density and power density are the core indicators that attract attention, as they are directly influenced by the key characteristics of the electrode materials.

A battery's energy density is its stored energy per unit mass or volume, and it is a key factor in evaluating the battery's endurance. The mass energy density (E, Wh/kg) can be defined by formula 1:

$$E = \frac{C_{cell} \times V_{avg}}{m_{cell}} \approx \frac{1}{\frac{1}{C_+} + \frac{1}{C_-} + m_{inactive}} \times (V_+ - V_-) \quad (1)$$

In the formula, $C_{cell}$ and $V_{avg}$ represent the total capacity of the entire battery and the average operating voltage, respectively; $C_+$ and $C_-$ represent the reversible specific capacity (mAh/g) of the cathode and anode materials, respectively; $V_+$ and $V_-$ represent the average operating voltages of the cathode and anodes (V), respectively; $m_{inactive}$ represents the mass proportion of all non-active components except for the active substances.

Thus, the theoretical maximum energy density is defined by the combined contribution of the specific capacity and the voltage platform. Improving the capacity and voltage of the cathode material, or using a anode material with a lower voltage and higher capacity, are direct ways to increase the energy density.

Power density (rate performance) reflects the battery's ability to rapidly charge, and is mainly limited by the dynamics of ion and electron transport. Among these, the ionic diffusion coefficient and electron conductivity of the electrode material determine the transmission speed of sodium ions and electrons within the electrode; the ionic conductivity (S/cm) of the electrolyte determines the migration speed of $Na^+$ in the electrolyte. Additionally, the impedance to charge transfer at the electrode/electrolyte interface is equally crucial.

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## 3. Cathode materials

Generally, there are three cathode materials for sodium-ion batteries, such as layered oxides, poly-anionic compounds, and prussian blue-like compounds. Layered oxides have a mature preparation process and high energy density, but their structural stability is insufficient. By doping with rare earth elements, their electrochemical performance can be improved. Poly-anionic compounds have stable structures and good cycling performance, but their conductivity is poor. Ion doping or carbon coating is often used to enhance their capacity and sodium ion diffusion rate. Prussian blue-like materials have an open framework structure, which is conducive to sodium ion migration, and have excellent rate performance. However, they have problems of crystalline water and vacancy defects. This section focuses on the performance and research progress of each type of cathode materials for SIBs.

### 3.1. Layered oxide

In SIBs, the layered oxides are usually combined with transition metals, represented by $Na_xMO_2$, in which M are transition metals. Research indicates that $NaxMO_2$ tends to form a layered structure when the value of x is between 0.5 and 1; if x is lower than 0.5, the material is more likely to exhibit a three-dimensional tunnel structure. Due to the different crystal structures, the various crystal forms of $Na_xMO_2$ show significant differences in electrochemical performance [3]. Based on different occupation modes of $Na^+$ and the stacking sequences of $O^{2-}$ layers, these layered materials can be further classified into phases like O3, O2, P3, and P2. Currently, iron-based and manganese-based layered oxides are the main research objects in this field. The performance of some materials are shown in Table 1.

Table 1. Performance of some layered oxide cathode materials

| Chemical formula | Operating voltage (V) | Reversible specific capacity (mAh/g) |
|---|---|---|
| O3-$NaFeO_2$ | 3.4 - 4.5 | 80 |
| O3-$Na[Fe_{1/2}Co_{1/2}]O_2$ | 3.0 - 4.0 | 167 |
| P2-$Na_{2/3}Mn_{1/3}Co_{2/3}O_2$ | 1.5 - 4.5 | 125 |

Rong Qiang et al. [4] have conducted research indicating that the electrochemical reversibility of single-phase O3-type $NaFeO_2$ is highly sensitive to the charging termination conditions. If the charging voltage exceeds 3.5V, the reversible capacity will significantly decrease; while when the termination voltage is controlled at 3.4V, the material exhibits excellent reversibility and relatively small polarization. Moreover, in layered materials containing three-dimensional transition metal ions, O3-$Na[Fe_{0.5}Co_{0.5}]O_2$ demonstrates excellent electrode performance. At a current density of 7.2 A/g and a temperature of 30°C, it can still maintain over 60% of the reversible capacity.

### 3.2. Polyanionic compound

Usually, $Na_xM_y(X_aO_b)_z$ represents polyanionic cathode materials, in which M are transition metals and x represents one or more elements such as S, P, Si, and B [4]. Polyanionic polyhedra and transition metal polyhedra are interconnected by covalent bonds, creating a stable and open 3D network structure. This structure can effectively buffer the structural stress caused by sodium ion insertion and extraction, prevent material collapse, and thereby ensure the structural stability of the battery during the cycling process.

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Based on these characteristics, this type of material exhibits advantages such as high safety and long cycle life, which meets the requirements of modern secondary batteries for high-performance cathode materials. Currently, the poly-anionic type materials that have been studied extensively mainly include phosphate ($NaFePO_4$), pyrophosphate ($Na_2XP_2O_7$, where X represents Fe, Mn, etc.) and sodium fast ion conductor (NASICON) type ($Na_xM(PO_4)_3$, where M represents V, Fe, Mn, Cr, Ni, Co, etc.).

$NaFePO_4$ can be classified into two types, the olivine phase and the phosphorodicalcite phase. Research [5, 6] has shown that the olivine phase $NaFePO_4$ can provide a reversible specific capacity more than 120 mAh/g, and its discharge curve exhibits a two-platform characteristic. However, due to the low diffusion coefficient of $Na^+$, the reaction kinetics is slow. In contrast, the phosphorodicalcite phase $NaFePO_4$ was initially regarded as electrochemically inert, but subsequent studies have found that through nanomization and amorphization treatment, the ion diffusion path can be shortened and random transport channels can be formed, thereby activating its electrochemical activity. Nano-sized phosphorodicalcite phase $NaFePO_4$ can be prepared using the solid-phase method. After the first charge and discharge, the structure of this material transforms into amorphous α-$FePO_4$. Its extremely low $Na^+$ migration barrier significantly enhances the discharge rate, and it can release 142 mAh/g of specific capacity, which is 92% of the theoretical value, at the first cycle. Furthermore, it exhibits good cycle stability, with nearly no capacity loss after 200 cycles.

$Na_2XP_2O_7$-type sodium cathode materials can effectively raise the operating voltage by introducing highly electronegative pyrophosphate anions$(P_2O_7)^{4-}$. Among them, $Na_2FeP_2O_7$, based on the $Fe^{3+}/Fe^{2+}$ redox couple, can achieve an average voltage of 3.0V and a discharge specific capacity over 80 mAh/g [7]. Its unique three-dimensional ion diffusion channels and stable structure give the material excellent rate performance and cycle stability. The discharge curve presents two platforms at 2.5V and 3.0V, corresponding to the ordered rearrangement of different $Na^+$ occupancy. For $Na_2MnP_2O_7$, although its theoretical capacity and energy density are higher, it is limited by the poor intrinsic electron conductivity and manganese dissolution problems, resulting in suboptimal electrochemical performance. Through graphene layer coating modification [8], the electron conductivity and structural stability can be significantly improved, allowing it to release approximately 95% of the theoretical capacity at 0.2C, with a specific energy exceeding 300 Wh/kg, and maintaining 83% capacity after cycling 600 times at 2C.

The NASICON-type materials possess a three-dimensional structure formed by the co-angle connection of $MO_6$ octahedra and $PO_4$ tetrahedra, providing stable ion transmission channels and high structural stability. These materials exhibit excellent electrochemical performance. Among them, $Na_3V_2(PO_4)_3$ based on the $V^{3+}/V^{4+}$ redox couple can provide a specific capacity of approximately 120 mAh/g and a stable voltage platform of 3.4V [9]. By substituting the transition metal part (Mn, Fe, Ni), the working potential can be further regulated. $Na_3(VO)_2(PO_4)_2F$ (NVPOF) as a representative of this system, combines high specific capacity (about 130 mAh/g) and high operating voltage (>3.9V), and has extremely small volume change (< 2%) during charging and discharging, with outstanding structural stability. Its porous micro-nano structure design is conducive to shortening the ion/electron transmission path, thereby demonstrating excellent capacity retention rate and stable redox reaction characteristics in long cycles, maintaining 92.1% of its initial capacity after 1000 cycles.

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### 3.3. Prussian blue analogues

The Prussian blue analog ($Na_xM[Fe(CN)_6]_y.nH_2O$) is a type of sodium-ion battery cathode material with an open cubic three-dimensional structure. Its large ion channels (about 0.46 nm) facilitate a rapid transmission speed of $Na^+$, supporting high-rate charging, and the theoretical specific capacity can achieve 170 mAh/g. Chen et al. [10] developed $Na_{1.63}Fe_2(CN)_6.1.75H_2O$, which can provide 130 mAh/g of specific capacity at room temperature. Even in an environment of 80 ℃, it can still maintain a capacity of 120 mAh/g. Moreover, after cycling 500 times at 80 ℃ and 2C rate, its reversible specific capacity can still reach 57 mAh/g. Meanwhile, the sodium-rich iron-manganese-based material ($Na_{1.72}MnFe(CN)_6$) presents a reversible specific capacity of approximately 130 mAh/g, and its operating voltage in average can reach 3.2 V [11].

## 4. Conclusion

This review systematically summarizes the research progress of three mainstream cathode materials for sodium-ion batteries. These materials are layered oxides, polyanionic compounds, and Prussian blue analogs. It focuses on their crystal structures, electrochemical performances, inherent deficiencies, and modification strategies. Layered transition metal oxides are represented by $Na_xMO_2$. They have the advantages of mature preparation process and high energy density. Their electrochemical performance can be effectively improved by rare earth element doping. The crystal forms of these materials are determined by the sodium content and ion stacking mode. The main crystal forms are O3, O2, P3, and P2. These crystal forms have significant impacts on their reversible capacity and cycle stability. Iron-based and manganese-based layered oxides are current research hotspots. Their operating voltage and reversible specific capacity can meet the basic requirements of low-cost energy storage applications. However, structural instability under high voltage remains an unsolved problem. Polyanionic compounds have the general formula $Na_xM_y(X_aO_b)_z$. They have a stable 3D network structure. This structure is formed by covalent bonding. It endows the materials with excellent safety and long cycle life. Phosphate, pyrophosphate, and NASICON-type materials are the main research directions in this category. Among them, olivine-phase $NaFePO_4$, pyrophosphate $Na_2FeP_2O_7$, and NASICON-type $Na_3V_2(PO_4)_3$ and $Na_3(VO)_2(PO_4)_2F$ have shown excellent electrochemical performance. Their conductivity and ion diffusion rate can be significantly improved. The improvement methods include ion doping, carbon coating, and nanomization modification. However, some polyanionic compounds have low specific capacity. This limits their application in high-energy-density SIBs. Prussian blue analogs ($Na_xM[Fe(CN)_6]_y \cdot nH_2O$) have an open cubic 3D structure. This structure provides large ion channels. The channels allow rapid sodium ion transmission. Thus, they show excellent rate performance. Sodium-rich iron-based and iron-manganese-based Prussian blue analogs have high reversible specific capacity and operating voltage. They also have good high-temperature stability. However, these materials have problems of crystalline water and vacancy defects. These problems still affect their cycle stability and practical application. In summary, the three types of cathode materials each have their own advantages and deficiencies. Modification strategies are effective ways to optimize their electrochemical performance. Common modification strategies include element doping, carbon coating, and structural design. This review systematically sorts out the research status of the three mainstream cathode materials. It provides a comprehensive reference for the subsequent research on performance optimization of SIBs cathode materials.

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