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Research Progress of MOFs and Their Derivatives as Cathode Materials for Sodium-Ion Batteries

Xueyan Dong

School of Chemistry and Materials Science, Qinghai Minzu University, Xi’ning 810007, China

Abstract: ***Benefiting from the remarkable advantages of abundant sodium resource reserves and low cost, sodium-ion batteries (SIBs) have emerged as one of the most promising alternative technologies to lithium-ion batteries. However, SIBs still face several technical challenges in practical applications, particularly regarding the energy density and electrochemical performance of cathode materials. Metal-organic frameworks (MOFs), as materials with highly tunable pore structures, excellent specific surface areas, and favorable electrochemical properties, exhibit broad application prospects in batteries. MOFs materials not only possess unique advantages in energy storage but also can further enhance their electrical conductivity, structural stability, and cycling performance through strategies such as metal ion doping and carbon coating. This paper reviews the progress in the application of MOFs and their derivatives as cathode materials for sodium-ion batteries, elaborates on their structural characteristics, preparation methods, and electrochemical reaction behaviors, and focuses on analyzing the core advantages of such materials in improving the electrochemical performance of sodium-ion batteries. Finally, the paper prospects the future development directions and challenges of MOFs materials in sodium-ion battery cathode materials.***

Keywords: Metal-organic frameworks (MOFs), Sodium-ion batteries (SIBs), Cathode materials, Electrochemical performance, Energy storage materials.

1. INTRODUCTION

With the large-scale application of global renewable energy (such as wind power, solar power, hydropower, etc.), the rapid development of portable electronic devices, low-speed vehicles and distributed energy storage power stations, energy storage technology faces multiple requirements such as low cost, long cycle life, high safety and environmental adaptability [1-3]. Lithium-ion batteries (LIBs) have long dominated the secondary battery market due to their high energy density and long cycle life. However, lithium resources are scarce, with low abundance in the earth's crust (only 0.0065%), concentrated distribution, and large price fluctuations. In addition, the current collector of lithium batteries depends on high-priced copper foil, which may cause thermal runaway in high-temperature environments, limiting its application in cost-sensitive scenarios such as large-scale energy storage [4-5].

Sodium-ion batteries have become a strong competitor to lithium-ion batteries due to their similar electrochemical principles and the advantages of abundant sodium resources and low cost. Sodium is more than 300 times more abundant in the Earth's crust than lithium and is widely found in seawater. Sodium-ion batteries also exhibit superior low-temperature performance (capacity retention rate can reach more than 85% at 0°C) and higher thermal stability, and their safety is significantly better than that of lithium batteries [6]. Sodium-ion batteries are suitable for special scenarios such as energy storage in cold regions and outdoor backup power. In addition, sodium-ion batteries can use aluminum foil as a current collector, which further reduces production costs and has great potential for industrialization.

The cathode material of sodium-ion batteries directly affects the energy density, operating voltage and cycle stability of the battery. Currently, the cathode materials of sodium-ion batteries mainly include layered metal oxides (such as $NaNi_{0.5}Mn_{0.5}O_2$), polyanionic compounds (such as $NaFePO_4$) and Prussian blue analogs (such as $Na_2Mn[Fe(CN)_6]$)[7-11]. However, these materials generally suffer from poor conductivity, low ion diffusion rate and unstable structure, which limits the performance improvement of sodium-ion batteries.

Metal-organic frameworks (MOFs) are a new type of porous organic-inorganic hybrid materials with advantages such as large specific surface area, adjustable pore structure and controllable structure. With the core advantages of highly designable structure, ultra-high specific surface area, adjustable porosity and easy modification of surface chemical properties [12-13], metal-organic frameworks (MOFs) are called "molecular Lego bricks". They have gradually moved from laboratory research to industrial application, covering multiple core fields such as energy storage and conversion, gas storage and separation, catalysis, biomedicine, environmental governance and high-end chemical industry. They have also shown irreplaceable value in the national key development directions such as the "dual carbon" strategy, new energy and biomedicine [14-16].

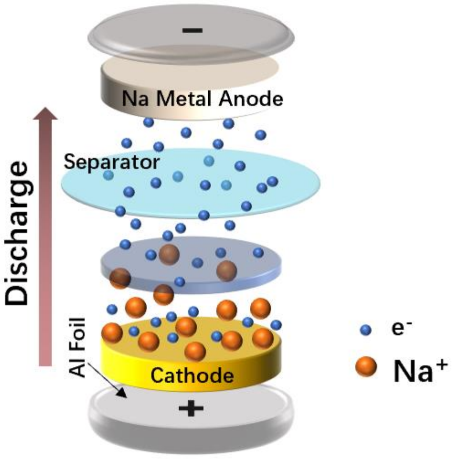

Figure 1: The composition of sodium ion battery and its ' rocking chair ' working principle [10]

In recent years, MOFs have made significant progress in the application of sodium-ion battery cathodes [17-18]. MOFs materials can not only be used directly as cathode materials, utilizing their high specific surface area and abundant active sites to achieve efficient storage and transport of sodium ions, but also as precursors to prepare composite materials such as metal oxides, sulfides, and phosphates through modification methods such as pyrolysis, sulfidation, and phosphating, which significantly improves electrochemical performance [19-20].

2. STRUCTURAL PROPERTIES OF MOF S MATERIALS

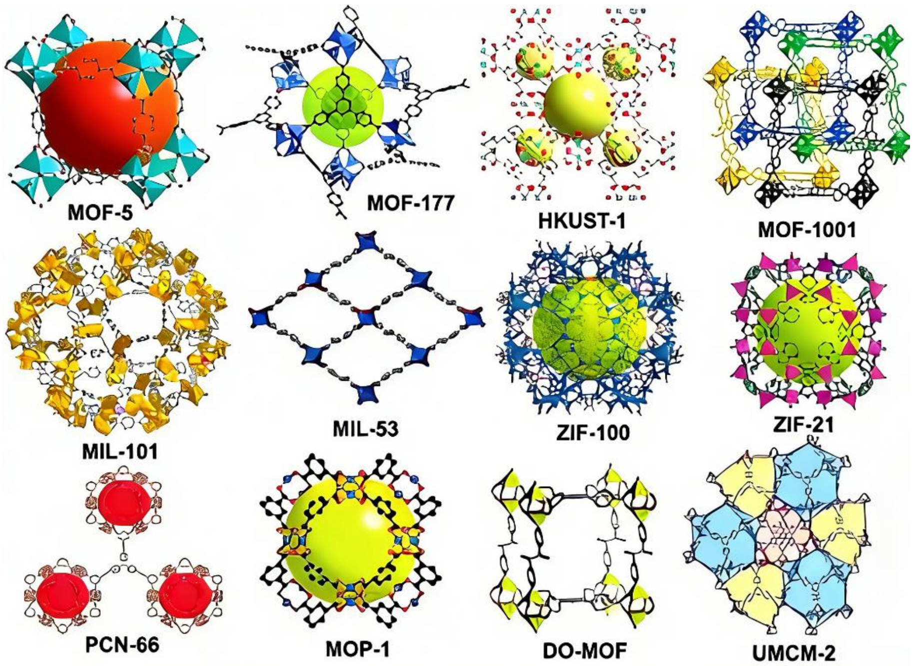

Figure 2: Structural diagrams of representative MOFs and their derivative structures

Metal-organic frameworks (MOFs) are widely used in energy storage due to their highly tunable structure, abundant porosity, and large specific surface area. MOFs form stable three-dimensional porous structures through the coordination of metal nodes and organic ligands. This structure not only provides ample space for sodium ion insertion/extraction but also improves the energy density and cycle stability of the battery. The porous structure and tunable chemical environment of MOFs give them unique advantages in sodium-ion battery applications. For example, the electrochemical activity of the metal center of a MOF can be modulated by changing the type of metal ion or ligand, thereby optimizing the performance of the sodium-ion battery. Furthermore, by finely controlling the structure and composition of MOFs, their conductivity can be effectively improved, and the diffusion rate of sodium ions in the electrode material can be enhanced, thus improving the overall performance of the battery.

MOFs materials have highly designable and tunable structural characteristics. Their basic structural units consist of metal nodes and organic ligands, which self-assemble into a three-dimensional porous network through coordination bonds [21-22]. The type of metal nodes directly determines the electronic structure and chemical stability of MOFs materials, while the length, functional groups and spatial configuration of organic ligands directly affect the pore structure, specific surface area and functional properties [23-26]. The pore types of MOFs materials are tunable, including micropores (<2 nm), mesopores (2~50 nm) and macropores (>50 nm), which can provide sufficient space for the insertion and extraction of sodium ions, optimize ion transport efficiency and enhance the electrochemical performance of the materials. In addition, MOFs materials have rich functional groups, and their electronic structure and electrochemical performance can be precisely controlled by introducing different ligands or metal nodes, so that they can show broad application prospects in energy storage, catalysis and other fields.

3. PREPARATION METHODS OF MOF MATERIALS

MOFs materials are mainly based on the coordination reaction between metal ions and organic ligands. The core is to achieve the ordered self-assembly of metal nodes and organic ligands by controlling the reaction conditions, so as to obtain MOFs crystals with uniform structure and stable performance. At present, there are various methods for preparing MOFs materials, including solvothermal method, hydrothermal method, sol-gel method, etc. [27-32]. When preparing sodium-ion battery cathode materials, solvothermal method and hydrothermal method are usually used to synthesize stable crystal structures under high temperature and high pressure environment. These methods can ensure the high specific surface area and ideal pore size of MOFs materials. In addition, in recent years, new MOFs preparation methods such as template method, electrochemical deposition method and mechanochemical method have been developed, providing a new path for the large-scale industrial preparation of MOFs materials. To improve the conductivity of MOFs, researchers have also employed techniques such as metal ion doping and carbon coating to enhance their electrochemical performance. For example, doping with metals such as cobalt, nickel, and manganese can improve the electronic conductivity and structural stability of MOFs, while carbon coating helps to further improve their conductivity and enhance their cycling performance [32].

4. APPLICATION OF MOFS MATERIALS IN SODIUM-ION BATTERIES

MOFs materials and their derivatives can be applied to sodium-ion battery cathode materials in two ways due to their unique structural and performance advantages: one is to use them directly as cathode materials, utilizing their high specific surface area and abundant active sites to achieve efficient storage and transport of sodium ions; the other is to use them as precursors, through pyrolysis, sulfidation, phosphating and other modification methods, to prepare metal oxides, sulfides, phosphates and their composite materials, effectively solving the problems of poor conductivity and insufficient structural stability of pure MOFs materials, while retaining the porous structure advantages of MOFs materials, and further optimizing the electrochemical performance of cathode materials [33-34].

For example, using MOF - derived sodium nickel cobalt oxide ($NaNiCoO_2$) as a cathode material can significantly improve the capacity and cycle stability of sodium-ion batteries. MOF materials not only provide a large surface contact area but also improve the diffusion efficiency of sodium ions. Furthermore, by combining carbon materials with MOFs, the conductivity of sodium-ion batteries can be further optimized, enabling them to maintain good performance during high-rate charge and discharge processes.

Despite the great potential of MOFs in sodium-ion batteries, some challenges remain: Poor conductivity: MOFs materials themselves have poor conductivity and usually need to be combined with other conductive materials to improve performance. High synthesis cost: The synthesis process of MOFs materials is relatively complex and requires high-temperature treatment, which increases their production cost. Volume expansion problem: During the charging and discharging process of sodium-ion batteries, the electrode materials will undergo volume expansion, which may affect the stability of MOFs materials [35-37].

Table 1: Summary of material parameters of MOF s -based materials and their performance as electrodes for SIBs

Materials	Testing potential vs. Na/Na^+	Reversible capacity (mA h g-1)	Initial / cycled CE (%)	Retention (%) / cycles	Ref
MOF-199	-	-	76.8	-	[38]
u-CoOHtp	0.01-3.0	450 @ 0.05 A g^{-1} (0.11 C) 215 @ 2 A g^{-1} (4.44 C)	~75.3 / ~100	82.4 / 50	[39]
$Fe_2(DOBPDC)$	2.0-3.65	108 @ 0.05 C 77 @ 2 C	~98 / >99	91 / 50	[40]
$FeFe(CN)_6$/carbon cloth	2.0-4.0	82 @ 0.2 C 50 @ 10 C	98.7 / ~100	75 / 200	[41]
Co-HAB	0.5-3.0	82 @ 0.2 C 50 @ 10 C	− / ~100	76 / 50	[42]
Zn-NDC	0.05–3.0	-	-	-	[43]
Zn-PTCA	0.01-2.0	357 @ 0.05 A g^{-1} (0.14 C) 256 @ 1 A g^{-1} (2.8 C)	45.3 / ~100	75.5 /	[44]
K4Na2 $[Fe(C_2O_4)_2]_3 \cdot$ 2H2O	1.6-4	45 @ 0.02 C	-	91 /	[45]
Ni-DTA	0.01–2.8	-	˜68	-	[46]

5. SYNERGISTIC MECHANISM

The superior electrochemical performance of MOFs (Metal-Organic Facility Materials) stems from the synergistic effect of metal ions and organic ligands in their structure. Metal nodes participate in electrochemical reactions, contributing to increased battery capacity, while organic ligands improve the stability of MOFs and provide more electrochemical active sites. Furthermore, the porous structure of MOFs provides sufficient space for the insertion and extraction of sodium ions, a characteristic that results in smaller volume changes during cycling, thereby enhancing material stability.

MOFs materials is mainly reflected in three aspects: First, their high specific surface area and rich pore structure can provide a large number of adsorption sites, enabling sodium ions to achieve rapid physical adsorption and desorption in the pores, thereby improving the reaction efficiency and rate performance of the battery; Second, the transition metal nodes in MOFs undergo reversible redox reactions during charging and discharging, promoting the insertion and deintercalation of sodium ions and bringing additional capacity contributions; In addition, some organic ligands with conjugated structures can also participate in the energy storage process through interaction with sodium ions, further improving the sodium storage capacity of the material. Therefore, MOFs materials can achieve relatively excellent electrochemical performance through the combined effects of pore adsorption, metal active site reaction and ligand-assisted energy storage [47].

MOFs and their derivatives mainly comes from the synergistic effect between porous structure, conductive network and component regulation: their high specific surface area and rich pore structure can provide a large number of active sites and build continuous ion transport channels, thereby enhancing the adsorption capacity of sodium ions, reducing diffusion resistance and improving rate performance; at the same time, by combining with conductive materials such as graphene and carbon nanotubes or generating carbon matrix through pyrolysis, the conductivity of the material can be significantly improved, and the volume expansion during charging and discharging can be alleviated by the coating and support of active components by the carbon skeleton, inhibiting particle aggregation and structural collapse, thereby improving cycle stability; in addition, by regulating metal nodes and organic ligands, and by adopting strategies such as doping and composite, the electronic structure and active site distribution of the material can be further optimized, enhancing ion transport efficiency and electrochemical activity, thereby achieving a comprehensive improvement in capacity, rate performance and cycle life [48-50].

6. SUMMARY

Although MOFs and their derivatives have shown unique advantages in sodium-ion battery cathode materials and significant progress has been made in related research, they still face many core bottlenecks in practical application and industrialization, mainly in terms of synthesis cost, electrochemical performance, large-scale preparation and stability, as detailed below.

Future research on MOFs materials in sodium-ion battery cathodes will focus on three key directions: First, developing low-cost MOF synthesis technologies, optimizing production processes, and promoting large-scale preparation technologies to reduce production costs and increase yields; second, optimizing the electrochemical performance of MOFs materials by doping with heteroatoms and composite conductive materials to alleviate volume expansion problems and improve ion transport efficiency, thereby enhancing the battery's specific capacity and cycle stability; and finally, strengthening the industrial application of MOF - based cathode materials, promoting their use in large-scale energy storage, low-speed electric vehicles, and portable electronic devices, while exploring their potential in novel batteries such as solid-state sodium-ion batteries and sodium-ion hybrid supercapacitors. Future research should focus on optimizing the pore structure and conductivity of MOFs materials to improve their performance during high-rate charge-discharge processes. With the continuous development of new synthesis methods, the application prospects of MOFs materials in sodium-ion batteries will be even broader.

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