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Research article

Optimization of cobalt-based MOFs for super-capacitor electrode materials of new energy vehicle

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ABSTRACT

Super-capacitors (SCs), as new energy conversion storage elements, have attracted much attention, but there is still a research gap in the design of electrode materials. In this study, the optimization scheme of Metal-Organic Frameworks (MOFs) and cobalt-based MOF composites as electrode materials for SCs in new energy vehicles is explored, and a series of experiments are conducted to evaluate their performance. Scanning Electron Microscope (SEM) images reveal that the cobalt-based MOF composites have a surface morphology of particles with uniform distribution. The electrochemical performance test results show that the specific capacitance of the cobalt-based MOF composites is much higher than the sum of the two individual electrode materials and presents a remarkable increase with the scanning rate. Additionally, in the constant current charge-discharge test, cobalt-based MOF composites exhibit the longest charge-discharge time and good symmetry. Electrolyte particle contact tests for samples at different preparation temperatures display that high-temperature samples have better structural stability and electrolyte ion contact. In Cyclic Voltammetry (CV) and Galvanostatic Charge Discharge (GCD) tests, the 250 °C sample demonstrates the best electrochemical properties and the highest specific capacitance (269 F/g). Moreover, as the current density increases, the specific capacitance of the 600 °C sample decreases at a lower rate, showing stronger stability. However, the use of cobalt-based MOF materials may pose environmental and safety risks, such as the environmental impact of cobalt resource mining, instability under high-temperature conditions, and the possible production of hazardous substances. Therefore, these factors need to be fully considered when designing and using SCs to ensure the environmental friendliness and safety of cobalt-based MOFs. These results provide an important reference for selecting and optimizing electrode materials for SCs in new energy vehicles. Furthermore, this study offers research suggestions for improving new energy materials, filling the research gaps in related fields, and supporting the further development of SC technology.

1. Introduction

In recent years, there has been heightened global awareness of ecological crises, largely driven by the increased occurrence of natural disasters. This has elevated environmental pollution to a major global concern. One significant contributor to pollution and

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harmful gas emissions is the extensive use of fossil fuels. Effectively addressing the challenge posed by fossil energy necessitates proactive exploration of alternative energy sources and advancement of innovative energy storage techniques [1]. Despite their high energy storage capacity, traditional batteries fall short in terms of power density, thereby limiting their suitability for modern society. This limitation underscores the importance of seeking alternatives, and in this context, Super-capacitors (SCs) have emerged as promising solutions. SCs offer superior power density and an extended cycle life, making them a focal topic in energy storage research. In SCs' electrode reactions, electron transport and storage predominantly take place on the electrode's surface or nearby electrolyte. This emphasizes the pivotal role of the electrode material's microstructure in influencing the overall performance of the device [2].

The global restructuring of the power engineering sector has presented new challenges and opportunities for developing energy storage technologies in electric vehicles (EVs). Researchers have explored various approaches in this context. Wang et al. (2022) reported on a flexible zinc-ion hybrid supercapacitor (Zero-Halogen Supply Chain, ZHSC) utilizing porous bamboo charcoal as the cathode material. By controlling the porous structure, they achieved the construction of porous material with a high porosity rate. The SC exhibited a capacitance of 321.3 F/g at a power density of 800.0 W/kg^{-1} and 1 A/g^{-1}, showcasing significant cyclic stability [3]. Song et al. (2022) studied a three-terminal photoelectrochemical cell integrating perovskite solar cells and symmetrical SC units. They successfully improved the solar conversion storage efficiency of the photoelectrochemical cell to over 20 % by coordinating energy matching between the conversion and storage units [4]. Hassan et al. (2022) evaluated the application of SC modules as rapid-response storage units in solar systems. Their research demonstrated that using SC modules significantly enhanced the solar systems' energy self-consumption and self-sufficiency, thus improving overall system efficiency and reliability [5]. Zhou et al. (2022) proposed a strategy to design responsive spatial photonic barcodes, utilizing responsive dual-metal ($Tb3+/Eu3+$) MOF SC with a multicolored heterogeneous structure. This design offers new possibilities for advanced anti-counterfeiting applications, particularly in the information security field [6]. Boretti (2024) introduced the concept of using carbides as sustainable energy storage materials, creating outstanding SC materials by blending two-dimensional (2D) MXenes with Metal-Organic Frameworks (MOFs). The combination of MOFs and 2D MXenes holds enormous potential for enhancing SC performance, including increased charge storage capacity, accelerated ion diffusion kinetics, and improved long-term stability [7]. AlKawak et al. (2024) proposed a hybrid technology for managing Hybrid Energy Storage Systems, integrating batteries, SC, and integrated charging in EVs. Their hybrid approach utilized Nano Beetle Optimization (NBO) and Quantum Neural Network (QNN) technologies, referred to as the NBO-QNN method. This method optimized the power usage of energy vehicles and maximized battery life, achieving more efficient energy management. Results indicated that the NBO-QNN technology outperformed existing methods, displaying the best output among all existing Cooperative Search Algorithm, Latent Semantic Analysis, and Grasshopper Optimization Algorithm methods. It provided a new and effective method for energy management in energy vehicles [8].

The above research presents a series of latest research results in the field of current energy storage technology in new energy vehicles, involving the exploration of multiple materials and methods to improve the performance and application efficiency of energy storage equipment. However, some limitations and research gaps are worth discussing. (1) Standardization of material performance evaluation: Current research results usually focus on reporting synthesis methods and the performance of new materials, but lack consistency and standardization of performance evaluation criteria. Therefore, it is difficult to directly compare the properties of different materials and their application potential under various conditions. (2) Lack of long-term stability studies: Although some studies show good initial performance, there is a lack of systematic evaluation of long-term cyclic stability and reliability in practical applications. This is crucial for the reliability and durability of energy storage devices in practical applications. (3) Consistency of material synthesis and process control: In some reports, the material's synthesis method and process control may remarkably impact the final performance, but consistency and repeatability of these aspects are often not fully considered. (4) Lack of research on integrated systems and practical applications: Although some studies focus on the performance optimization of a single component, there is still a large research gap on the integrated design of the overall system and its performance in practical application scenarios.

Consequently, addressing the increasing demand for high-efficiency and long-lasting power in new energy vehicles has become a prominent research focus, necessitating the design and exploration of new electrochemical energy storage devices. Due to their relatively low cost and robust performance, batteries and capacitors dominate the market as available electrochemical energy storage devices. As electronic equipment undergoes continuous advancements, critical indicators for progress now include device size, mass, power supply, and charging/discharging speed. In this landscape of evolving demands, MOFs, a class of porous crystalline materials formed through the self-assembly of metal ions or clusters and organic ligands, are gaining increasing recognition. MOFs offer a unique specific surface area, regular pore size distribution, and controllable morphology and size, making them promising candidates for various applications, including gas separation, conversion, storage, and optics. With their high specific surface area and substantial internal pore volume, MOFs are drawing attention for potential use in SCs. However, due to their poor electrical conductivity, many MOFs require modification to enhance their electrochemical performance. Integrating MOFs with other functional materials has demonstrated potential in the fabrication of high-performance electrode materials. MOFs find application in SCs in three primary ways: direct use, conversion into metal oxide (MO) materials, or pyrolysis into porous carbon materials. Nevertheless, it is noted that these materials often possess insufficient electrical conductivity and have a limited structure, limiting their suitability for high-performance SCs [9].

This study aims to utilize the solvothermal method and high-temperature calcination decomposition method to prepare cobalt-based MOF materials and simulate the reaction of electrode materials for SCs of new energy vehicles, thereby optimizing these electrode materials. The research hypotheses are as follows: (1) Cobalt-based MOFs have the potential for excellent electrochemical properties and can be used as candidate materials for electrode materials of SCs in new energy vehicles. (2) The electrical conductivity and capacitance characteristics of cobalt-based MOFs can be improved through appropriate preparation methods and structural regulation, thereby enhancing the energy density and power density of SCs. (3) The composite of cobalt-based MOFs with other functional materials can further enhance the electrode performance and improve the overall performance of SCs. (4) Through systematic experimental research and theoretical simulation, the charge storage and transmission mechanism of cobalt-based MOFs in SCs can be deeply understood, providing theoretical guidance for their optimal design.

The novelty of this study lies as follows: (1) The application innovation of cobalt-based MOF materials in the SC field: This paper explores the application of cobalt-based MOFs in the electrode materials of SCs for new energy vehicles. Optimizing the performance of cobalt-based MOFs in SCs brings new ideas and possibilities for the development of the SC field. (2) In-depth study on the effect of preparation temperature: This study has explored the effect of preparation temperature on the properties of cobalt-based MOF materials and found that samples prepared at high temperatures have better electrochemical properties and stability. This discovery provides a new direction for the optimization of the preparation process and helps improve the performance and stability of SCs.

2. Experimental methods and procedures

2.1. Analysis of SCs

SCs are also called electrochemical capacitors or ultracapacitors. They are electrochemical devices designed to store charges at the interface between the electrode and the electrolyte solution by inducing polarization in the electrolyte [10]. Distinguishing themselves from batteries, SCs exhibit significantly higher power, reaching hundreds to thousands of times the power of conventional batteries. However, their storage capacity is relatively low. Consequently, SCs find suitability in electrical devices that prioritize instantaneous high-power output without the need for permanent discharge [11]. In the context of new energy vehicles, SCs represent innovative energy storage devices that amalgamate the strengths of batteries and traditional capacitors. They offer the advantages of high power density and extended charging and discharging time of conventional capacitors. New energy vehicles also have a high energy density of batteries, demonstrating superior charge-discharge speed and longer cycle stability compared to either component alone [12]. Additionally, the electrolyte of SCs used in new energy vehicles is generally a non-toxic and non-polluting water-based electrolyte, rendering SCs environmentally friendly [13]. Fig. 1 illustrates the specific classification of SCs applicable to new energy vehicles.

In Fig. 1, SCs used in new energy vehicles are categorized into Electrical Double-Layer Capacitors (EDLCs), pseudocapacitors, and hybrid capacitors [14]. The specific details are as follows.

(1) The first category is EDLCs. Within electrochemical solutions, solid surfaces often acquire a charge through surface group dissociation or the selective adsorption of specific ions from the solution. This results in the formation of an opposite charge layer distribution between the solid surface and the electrolyte. Consequently, an electric double layer is established, comprising a charge distribution layer with positive and negative charges arranged closely on the contact surface of the solid/liquid phases. When an external power source applies a charging voltage, directional alignment of electrons, ions, or dipoles occurs at the electrode/electrolyte interface. Simultaneously, a stable electric double-layer capacitance develops at this interface, serving the purpose of energy conservation. Therefore, the storage mechanism of the electric double layer resembles that of traditional electrostatic capacitors, which rely on electrostatic adsorption to store electricity. Electric double-layer SCs generally utilize nanoporous materials with large specific surface areas as active electrode materials. They can produce huge

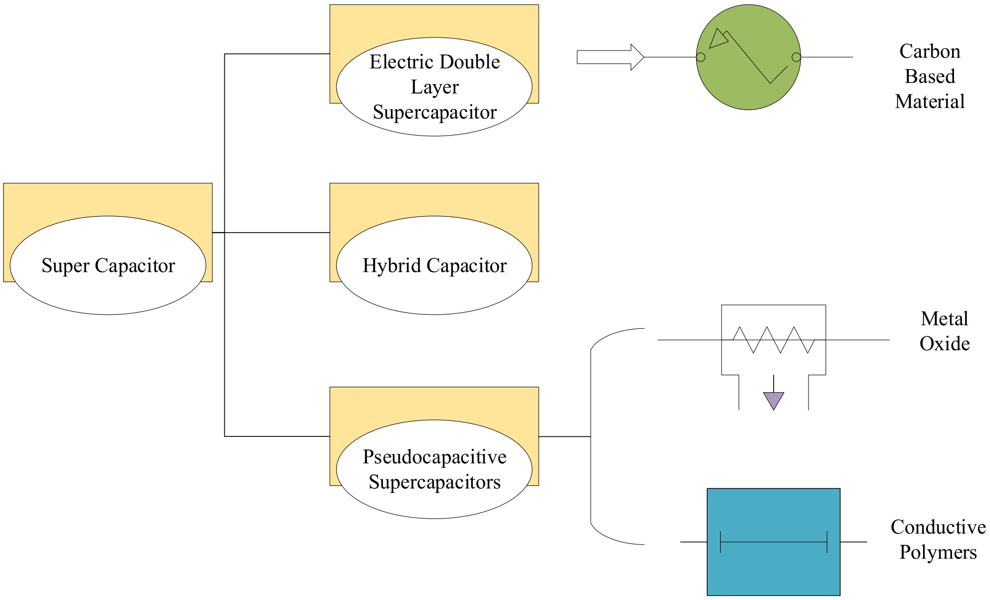


Fig. 1. Classification of SCs.

capacitances comparable to electrostatic capacitors. Notably, nanoporous carbon materials, a representative of such materials, known for their significant practicability, ease of industrial production, and relatively low cost, are favored by manufacturers of new energy vehicles [15]. Fig. 2(a and b) displays a schematic diagram illustrating the charging and discharging processes of EDLCs.

Fig. 2(a and b) illustrates that during the charging process of an EDLC electrode, the electrode surface charge in the ideally polarized electrode state attracts anisotropic ions in the surrounding electrolyte solution. These ions attach to the electrode surface, forming an electric double layer that constitutes the EDLC. The distance between the two charge layers is minimal, measuring less than 0.5 nm. Additionally, a specific electrode structure is employed, leading to a remarkable ten-thousand-fold increase in the electrode surface area, resulting in substantial capacitance [16].

The second category is the pseudocapacitor. Pseudocapacitors store charge through highly reversible redox reactions occurring on the surface and near-surface of electrode materials via ion intercalation or deintercalation. In principle, during the charging process of a pseudocapacitor, ions participating in the reaction within the electrolyte migrate toward the surface or into the corresponding electrode under the influence of an external electric field, compared to the electric double-layer capacitor. This process forms an interface between the electrode and the electrolyte. Simultaneously, a rapid and reversible redox reaction occurs with the electrode material at the interface, generating a reaction product that stores a substantial charge either on or inside the electrode. During discharge, the reaction product, laden with a significant electric charge, undergoes a swift and reversible redox reaction, releasing a large amount of electric charge. The charge is then transferred through an external circuit. This discharge process restores the corresponding electrolyte ions and electrode materials to their original states [17].

(2) The third type is the hybrid capacitor. The reaction mechanisms of the hybrid SC's two electrodes encompass both the electric double-layer mechanism and the pseudocapacitive mechanism, effectively amalgamating the advantages inherent in these two types of capacitors.

Additionally, the key to the chemical energy storage of SCs lies in the exchange and circulation of electrons. Therefore, the meticulous selection of electrode materials is crucial. An ideal electrode material must meet several criteria. The first is higher specific surface area. The second is controllable porosity. The third is good electrical conductivity. The fourth is electrochemically active sites. The fifth is long-term electrochemical structural stability [18]. Fig. 3 displays the conventional electrode materials used in SCs for new energy vehicles.

In Fig. 3, the electrode materials used in SCs for new energy vehicles include three points: (1) Carbon-based materials: Carbon materials have the advantages of high specific surface area, good thermal stability, high electrical conductivity, good corrosion resistance, wide availability, and cost-effectiveness. Electrodes using carbon materials as active materials predominantly rely on the electric double layer formed on the surface of the electrode to store charges. Hence, the carbon material possesses a high true specific surface area, and there should be effective electrical conductivity between the carbon particles. (2) MO materials: The Faraday pseudocapacitance exhibited by MOs can manifest at electrodes, solution interfaces, and inside electrodes. This significantly enhances the material's overall utilization, resulting in a larger specific capacitance. Consequently, MO materials are well-suited for constructing high-energy and power-density SC devices. (3) Conductive polymer materials: Conductive polymers are a class of electrode materials characterized by good electrical conductivity. Conductive polymers demonstrate voltage windows and electrical conductivity comparable to carbon-based materials. However, their conductivity tends to be related to the doping type. Undoped conducting polymers

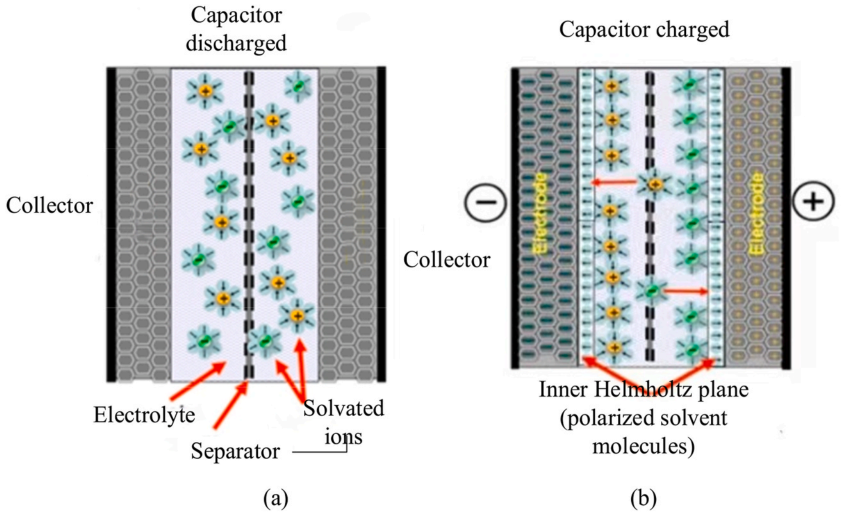


Fig. 2. Schematic diagrams of charging and discharging of EDLCs (a) charging state diagram; (b) discharging state diagram.

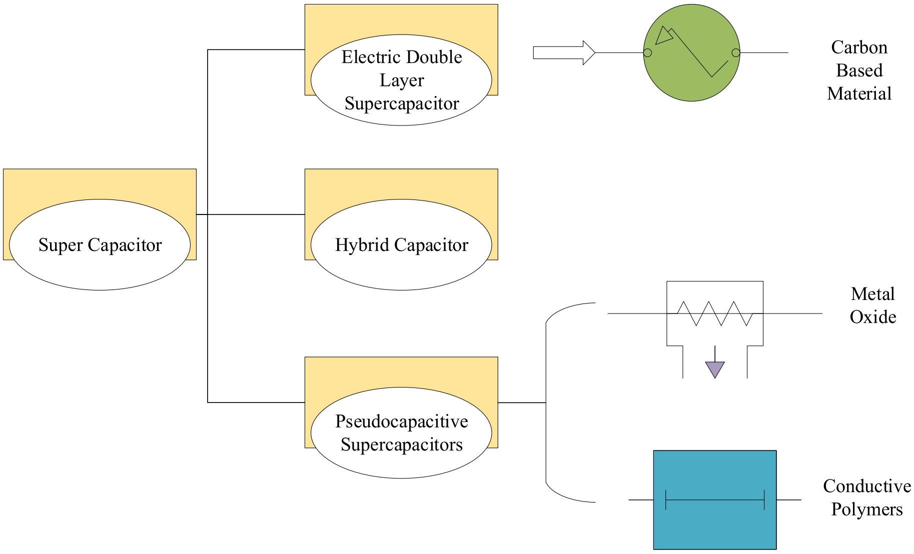


Fig. 3. Classification of SCs and their electrode materials.

have mediocre conductivity and cannot achieve optimal SC performance [19].

In summary, the existing traditional electrode materials for SCs in new energy vehicles are confronting a range of pressures and challenges. The demand for high battery life in new energy vehicles necessitates a departure from relying on a single electrode material. Consequently, there is a crucial need to transform and enhance traditional material synthesis concepts and methods. On the one hand, integrating different types of electrode materials, including two-phase, three-phase, and even multi-phase composite materials, can be explored and developed. On the other hand, there exists a pressing need for innovation in traditional SC materials, with concerted efforts directed toward the exploration and development of novel, cost-effective, and high-energy electrode materials. This pursuit of innovation has become an inevitable trend [20].

The mass capacitance of the capacitor can be determined according to Eq. (1).

$$C_s = I * \Delta t(\Delta V * m) \tag{1}$$

In Eq. (1), Δt represents the discharge time, ΔV represents the voltage window, I refers to the current, and m represents the mass of the sample in the electrode during the test. Energy and power density can be acquired according to Eq. (2) and Eq. (3):

$$E_s = 1/2 * Cs * \Delta V^2 \tag{2}$$

$$P_s = E_s * 3600/\Delta t \tag{3}$$

2.2. MOF material analysis

MOFs represent a novel class of materials characterized by periodic network structures formed through the self-assembly of multidentate organic ligands and metal ions. These materials amalgamate the features of organic porous materials and inorganic porous materials. Since the end of the 20th century, MOFs have developed rapidly. Research on MOFs covers various disciplines, such as organic chemistry, inorganic chemistry, crystallography, topology, and materials science [21].

The fundamental structure of MOFs can be conceptualized as an infinitely extended and uniform three-dimensional (3D) framework, created with metal ions or metal clusters as nodes and organic ligands as linkers. The precise selection of central metal ions or metal clusters and organic ligands facilitates self-assembly at the molecular level. The structure of MOFs is regulated through various means, such as temperature, solvent polarity, acidity, and alkalinity, resulting in the production of MOFs with diverse structures and unique functionalities. The design and synthesis of MOF materials have progressed rapidly, yielding tens of thousands of materials with different compositions and structures. Transition groups and rare earth elements are frequently chosen as central metal ions, while nitrogen-containing heterocycles and carboxylic acid ligands are commonly employed as organic ligands [22].

MOFs exhibit excellent properties, including good thermal stability, discrete ordered structure, ultra-low density, huge internal specific surface area, easy synthesis, and versatile properties, making them widely utilized in physics and chemistry. These exceptional characteristics have spurred the progress and development of MOF materials, giving rise to numerous new porous materials. Particularly noteworthy is the rapid advancement witnessed in MOFs over the past decade, attributable to their variable and flexible topology, as well as their extensive array of applications. It covers almost all fundamental application areas, especially gas storage and separation, catalysis, magnetism, optics, and drug storage and transport [23]. Moreover, MOFs possess suitable pores that facilitate the rapid passage of electrolyte ions during the charging and discharging process. Additionally, MOF materials are enriched with metal cations, contributing to high pseudo-capacitance [24]. However, pure MOFs exhibit poor electrical conductivity, necessitating efforts to enhance their electrochemical conductivity for direct application in SC electrodes to achieve remarkable performance. A viable solution involves selecting electrode materials with robust conductivity, such as carbon materials and conductive polymers, to compound with MOFs. This approach enhances capacitance, rate characteristics, and cycle stability [25].

The transition MOs obtained from MOFs retain a large internal specific surface area due to the preservation of the porous structure of the MOF material. These properties aid in the transport and diffusion of ions and improve the electrochemical activity of the electrodes. Therefore, experiments are conducted on cobalt-based MOFs for SCs intended for use in new energy vehicles.

The types of tools used to optimize MOFs and cobalt-based MOF composite materials in SC electrodes include the following: (1) Density Functional Theory (DFT): DFT is a quantum mechanical method for calculating electronic structure and related properties. It is used to simulate the electronic structure, structural stability, and adsorption properties of MOFs and cobalt-based MOF composite materials to optimize their performance in SC electrodes. (2) Molecular Dynamics (MD) Simulation: MD simulations can model atomic motion and structural dynamics, studying the stability, thermodynamic properties, and structural dynamics of MOFs and cobalt-based MOF composite materials to provide a foundation for material optimization. (3) First-Principles Calculations: This computational method is based on the principles of quantum mechanics and can predict the electronic structure, band structure, and adsorption properties of materials. It is utilized for the structural optimization and performance prediction of MOFs and cobalt-based MOF composite materials. (4) Density Functional Tight Binding (DFTB): DFTB is a simplified quantum chemistry method based on DFT, suitable for simulating the electronic structure and reaction dynamics of large systems. It offers advantages in simulating the structure and reaction performance of MOFs and cobalt-based MOF composite materials. (5) Multiscale Modeling: Multiscale modeling integrates diverse simulation techniques across varying scales, facilitating comprehensive simulation and optimization of materials from the atomic level to the macroscopic level. This approach aids in elucidating the performance of MOFs and cobalt-based MOF composite materials in SCs.

The integrated use of these tools allows for a comprehensive understanding of the structure, properties, and electrochemical behavior of MOFs and cobalt-based MOF composite materials, providing scientific and technical support for their optimization as electrode materials in EVSC [26].

The incorporation of cobalt-based MOFs brings substantial enhancements to existing SC electrode materials across multiple facets: (1) Increased Specific Surface Area and Active Sites: Cobalt-based MOFs exhibit high specific surface area and abundant active sites. This implies that the cobalt-based MOF material, with equivalent mass, can offer a larger surface area for the adsorption and desorption of electrolyte ions, thereby augmenting the energy storage capacity and facilitating the rapid charge-discharge performance of the capacitor. (2) Enhanced Structural Stability and Cycling Life: Cobalt-based MOF materials possess excellent structural stability, maintaining structural integrity during long-term cycling charge-discharge processes. In contrast, some traditional electrode materials are prone to structural fatigue and loss during the cyclic charge and discharge process, resulting in a shorter cycle life of the capacitor. Hence, utilizing cobalt-based MOFs can significantly enhance the cycle life of the capacitor. (3) Improved Charging Efficiency: Due to the rich active sites of cobalt-based MOFs, they can accelerate the adsorption and release rate of electrolyte ions, enhancing the charging efficiency of the capacitor. This means higher charging rates can be achieved in the same period, improving the overall efficiency of the capacitor. (4) High-Temperature Stability: Cobalt-based MOF materials maintain a stable structure and performance even after high-temperature treatment. This implies that cobalt-based MOFs can maintain good electrochemical performance in high-temperature environments, which is crucial for applications such as the power systems in EVs that operate in high-temperature conditions.

2.3. Research materials and containers

This study prepares a cobalt-based MOF composite material named Co-mo-74. The initially disordered one-dimensional and 2D morphologies undergo rearrangement and integration into a 3D flower-like structure by adjusting certain environmental factors during the growth process. The regular arrangement of pores in the structure improves the material's electrical conductivity and facilitates ion transportation and contact. The reagents, raw materials, and equipment types employed in the experiments are shown in Table 1 and Table 2, respectively.

Tables 1 and 2 display the types of reagents, raw materials, and equipment utilized in the experiments. The experimental procedures were streamlined as follows: First, for materials preparation, cobalt acetate tetrahydrate was dissolved in 25 mL of absolute

Table 1
Reagent raw materials.

Reagent	Purity	Manufacturer
2,5-Dihydroxyterephthalic acid	AR	Sinopharm Chemical Reagent Co., Ltd
Cobalt acetate tetrahydrate	AR	Sinopharm Chemical Reagent Co., Ltd
Potassium hydroxide	AR	Sinopharm Chemical Reagent Co., Ltd
Anhydrous ethanol	AR	Sinopharm Chemical Reagent Co., Ltd
Deionized water	–	Yangzhou University
Acetylene black	Battery level	Tianjin Ebory Chemical Co., Ltd
Isopropyl alcohol	AR	Sinopharm Chemical Reagent Co., Ltd
Polytetrafluoroethylene emulsion	Battery level	Shanghai 3F New Materials Co., Ltd
Nickel foam	–	Changsha Lyrun new material Co., Ltd

ethanol, while dihydroxy terephthalic acid was dissolved in 12.5 mL of deionized water to form two solutions. Solution A was prepared by dissolving cobalt acetate tetrahydrate in ethanol, and solution B was created by dissolving dihydroxy terephthalic acid in deionized water. These solutions were mixed, and the resulting solution was heated at 120 °C for 12 h, yielding the target product, a khaki-colored cobalt-based MOF composite. Afterward, the samples underwent washing and air drying. The preparation of the counter electrode involved selecting carbon fiber paper as the current collector, adding a 15 wt% conductive agent (conductive carbon black), and using a 10 wt% binder (polyvinylidene fluoride dissolved in methyl pyrrolidone). Mn-MOFs and -Mn-MOFs were used as active materials, and after adding conductive agents and binders, the electrode material was coated onto a 1 × 1 cm2 carbon paper. The electrodes were activated by immersing them in the electrolyte solution for 24 h, followed by the welding of nickel tabs, symmetric attachment to both sides of the ion separator, and sealing with hard plastic to obtain an SC device.

2.4. *Material characterization test*

The study conducts various characterization analyses on the synthesized sample, including Powder X-ray Diffraction (PXRD) detection, Energy Dispersive X-ray Spectroscopy (EDX), and Fourier Transform Infrared Spectroscopy (FT-IR) tests. First, in the PXRD detection, the synthesized sample exhibits characteristic diffraction peaks consistent with the single crystal data of standard samples, indicating good crystallinity. The sharpness of the characteristic peaks suggests excellent crystallinity. Moreover, in the EDX analysis, the presence of cobalt, carbon, and oxygen elements is discerned, thus corroborating the chemical composition of the sample. Finally, in the FT-IR test, various characteristic absorption peaks are identified, including the binding water within the structure, skeletal vibration peaks of benzene rings, split peaks of conjugated structures, and C–O bonds. The appearance of these peaks indicates the presence of specific structural components in the sample, ensuring the successful synthesis of the sample.

In the process of material characterization, commonly used techniques include X-ray Diffraction (XRD), Scanning Electron Microscopy (SEM), EDX, FT-IR, and so on. These techniques can provide information about the crystal structure, surface morphology, elemental composition, and chemical bonding of materials. The XRD technique determines the crystal structure by irradiating the sample's surface and recording the scattered X-rays. By analyzing the X-ray diffraction pattern, the crystal structure and lattice parameters of the material can be determined, and the crystallinity and crystal size of the material can be evaluated. SEM uses a focused beam of electrons to scan the surface of the sample and generates a high-resolution image of the surface by collecting and analyzing the signals that interact with the sample surface. SEM can provide information about surface morphology, particle size distribution, particle shape, and others. EDX technology is used in conjunction with SEM to determine the composition and content of elements in a sample by measuring the X-rays generated on the sample surface. EDX can offer information about material composition, element distribution, and impurity content. FT-IR technology determines the chemical composition and structure of a material by recording its absorption and scattering spectra in the infrared region. FT-IR can provide information about functional groups, bonding conditions, and structural characteristics of materials.

2.5. *Electrochemical test*

When testing the prepared electrodes, a traditional three-electrode system is selected. Among them, the reference electrode is a saturated calomel electrode. The working electrode is prepared with cobalt-based MOF composite as the active material, while the counter electrode is a platinum wire electrode. The electrochemical test used the CHI660E electrochemical workstation from Shanghai CH Instruments Ins. For capacitor devices, their electrochemical performance is tested using a two-electrode system. In the two-electrode system, the newly prepared SC for energy vehicles is symmetric, with the cobalt-based MOF composite material serving as both the positive and negative electrode material.

(1) Cyclic Voltammetry (CV) testing: Analyzing the CV curve of the SC electrode material allows for an understanding of its electrochemical activity, charge-discharge cycling performance, reversibility of electrochemical reactions, specific capacitance of the electrode material, and electrochemical window [27–29]. The specific capacitance of the electrode material can be calculated based on the CV curve. The testing conditions for the electrode's CV curve are as follows: (I) the voltage window is −0.6 to 0.6 V; (II) the scan rate is 10–100 mV s^{-1}. Eq. (4) calculates the area specific capacitance under these conditions:

Table 2
Equipment details.

Equipment	Type	Manufacturer
Electric heating constant temperature blast drying oven	GFL-45	Labtech Laboratory Apparatus Company
Digital display constant temperature stirring water bath	HH-6	Jintan Xicheng Xinrui Instrument Factory
Centrifuge	TGL-16G	Shanghai Anting Scientific Instrument Factory
Electronic balance	GL224-ISN	Sartorius Scientific Instrument (Beijing) Co., Ltd
Electrochemical workstation	CHI660E	Shanghai CH Instruments Ins.
Field emission scanning electron microscope	Zeiss_Supra55	Carl Zeiss AG
High-resolution transmission electron microscope	Tecnai G2 F30	FEI Company
X-ray powder diffractometer	D8 Advanced	Bruker Corporation
Fourier transform infrared spectroscopy	TENSOR27	Bruker Optics Company
X-ray photoelectron spectrometer	ESCAL AB250Xi	ThermoFisher Scientific Company

$$C = \frac{I}{vS(V_b - V_a)} \int_{V_a}^{V_b} IdV \tag{4}$$

Here, C represents the area-specific capacitance (unit: mF·cm^{-2}); S is the effective area of each working electrode (unit: cm^2); v is the scan rate (unit: V·s^{-1}); V_b and V_a correspond to the high and low working voltages, respectively (unit: V); I is the instantaneous current (unit: A).

(2) Galvanostatic Charge/Discharge (GCD) testing

GCD testing can measure the change in specific capacitance of electrode materials or devices at different current densities and the relationship between power density and energy density [30,31]. The testing conditions for the electrode material's GCD curve are as follows: (I) the working voltage window is 0–1V; (II) the current density range is 5–20 mA cm^{-2}. The area specific capacitance value under these conditions can be calculated using the GCD method, represented by Eq. (5) and Eq. (6):

$$C_m = \frac{I \times \Delta t}{m \times \Delta V} \tag{5}$$

$$C_s = \frac{I \times \Delta t}{S \times \Delta V} \tag{6}$$

Here, C_m represents the specific capacitance by mass; I is the current (unit: A); Δt is the charging or discharging time (unit: s); ΔV is the voltage change during the charging or discharging time Δt (unit: V); m stands for the mass of the active material (unit: g).

Energy and specific power density can be obtained according to Eq. (7)-Eq. (8):

$$E = 1\Big/2C_{cell}(\Delta V)^2 \tag{7}$$

$$P = E/\Delta t \tag{8}$$

In Eq. (7)-Eq. (8), E is the specific energy density. P is the typical power density. C_{cell} is the specific capacitance of the symmetric SC device. The redox reaction with electrode material and carbon material is shown in Eq. (9).

$$Mn(C_8H_4O_4)(H_2O)_2 + C^+ + e^- = MnC(C_8H_4O_4)(H_2O)_2) \tag{9}$$

3. Experimental results and analysis

3.1. Characterization test results

Fig. 4 illustrates the characterization test results of the cobalt-based MOF composite material.

Fig. 4 indicates that the SEM images unveil a microparticle surface morphology of the MOF composite material, displaying uniformly distributed particles.

3.2. Electrochemical performance analysis of cobalt-based MOFs

The electrochemical properties of Carbon Nanotubes (CNTs), Manganese-based MOFs (Mn-MOFs), and cobalt-based MOFs are assessed in a three-electrode system. The electrochemical performance results are shown in Fig. 5(a–c).

Fig. 5(a–c) illustrates that the specific capacitance of the cobalt-based MOF composite material exhibits a substantial increase at varying scan rates compared to the individual electrode materials. Notably, it surpasses the combined capacitance values of the two constituent materials by a significant margin. These findings suggest that the cobalt-based MOFs are not just externally mixed; rather,

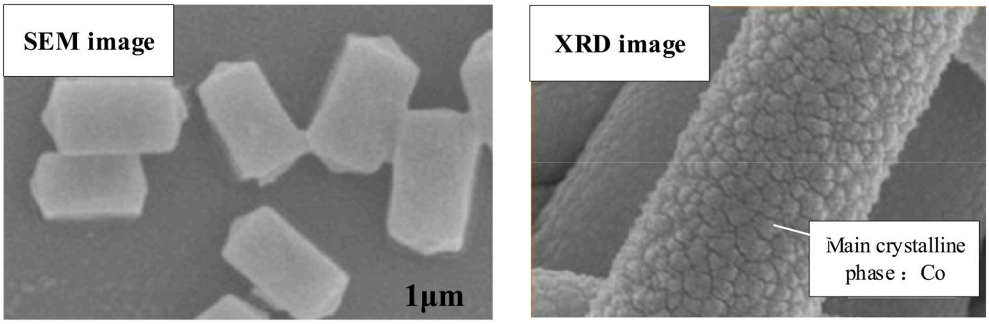
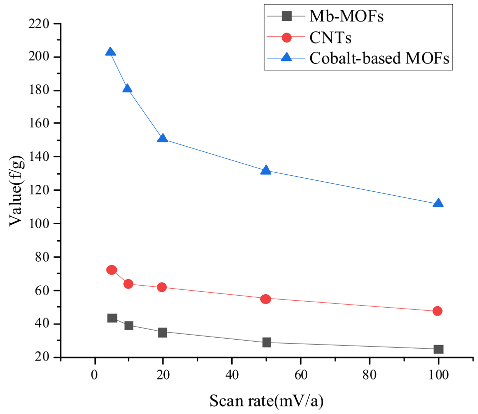
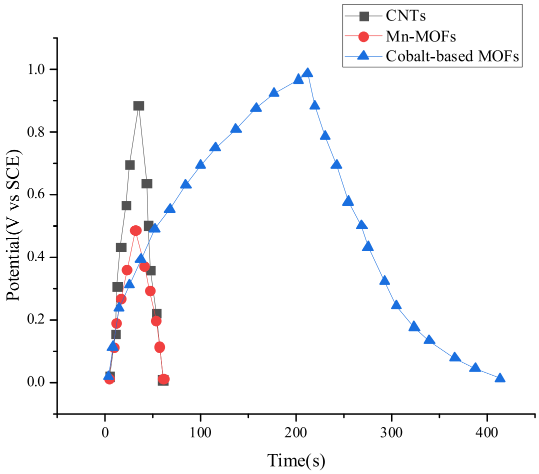


Fig. 4. Characterization test results.

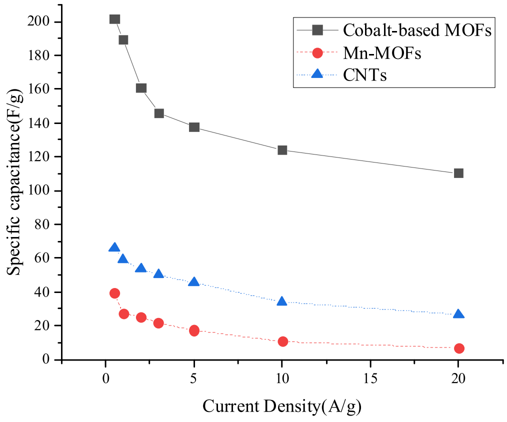
the structure of the composite material undergoes internal modifications within the MOFs, resulting in the formation of a unique conductive network. This distinctive feature contributes to the exceptional electrochemical performance of the cobalt-based MOF composite material. Additionally, in constant current charge-discharge tests, the cobalt-based MOF composite material presents the longest charge-discharge time and shows good symmetry.



(a)



(b)



(c)

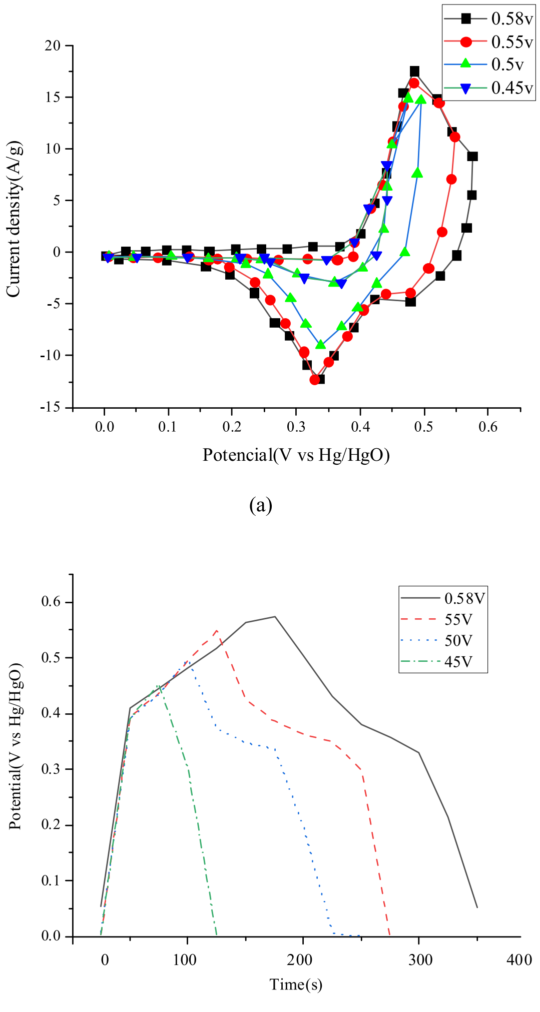
Fig. 5. Electrochemical performance results (a) Capacitance results; (b) Galvanostatic curve results; (c) Capacitance results at different current densities.

3.3. Electrode performance analysis of cobalt-based MOFs for SCs

The cobalt-based MOF material samples prepared at 400 °C, 500 °C, and 600 °C are used for experiments employing experimentally prepared electrode materials and SC accessories. The material samples undergo testing in a 3 mol L-1 KOH electrolyte solution with different voltages. The results of the test voltage and selected current are shown in Fig. 6(a and b).

Fig. 6(a and b) reveals the CV curves acquired under various voltage conditions. The symmetric oxidation-reduction peaks observed indicate the Faradaic behavior of the electrode, demonstrating the high reversibility of the electrode reaction. As the voltage increases, the area under the curve increases, indicating the excellent charge storage performance of the electrode material. Moreover, the appropriate voltage window is explored based on the GCD curves obtained under different voltage conditions. Ultimately, integrating specific capacity data and Coulombic efficiency, along with the two test results, 0.5V is chosen as the test voltage for all three electrodes. Fig. 7(a–c) illustrates the GCD curve results obtained for different sample temperatures.

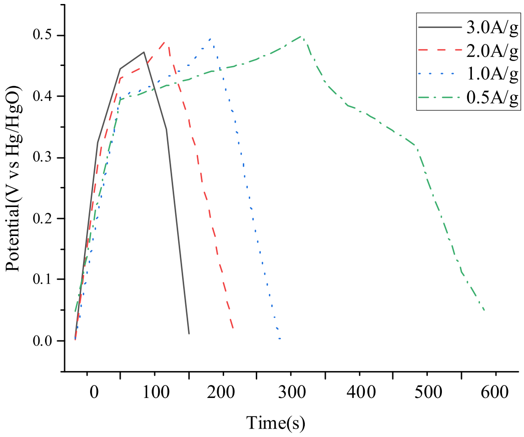
Fig. 7(a–c) demonstrates that among the three samples, the one processed at 250 °C exhibits the longest discharge time, corresponding to the highest specific capacity. At a current density of 0.5 A/g, a charge storage capacity of 269 F/g can be achieved, possibly attributable to the enhanced conductivity of the sample at 250 °C. Under the same current density, the sample at 300 °C only shows a charge capacity of 115 F/g. This discrepancy may be attributed to the partial sintering of the sample's calcined oxide at 300 °C, leading to a slight collapse of the microstructure and a slightly lower specific surface area compared to the other materials. Upon reaching 350 °C, the sample undergoes complete calcination due to the elevated temperature, resulting in increased crystallinity, smaller grain size, and a larger specific surface area, providing more reaction sites.



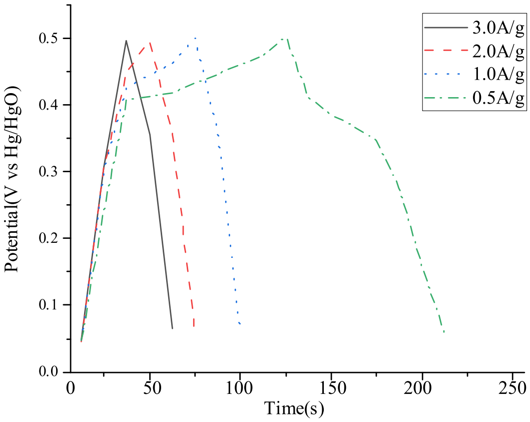
(a)

(b)

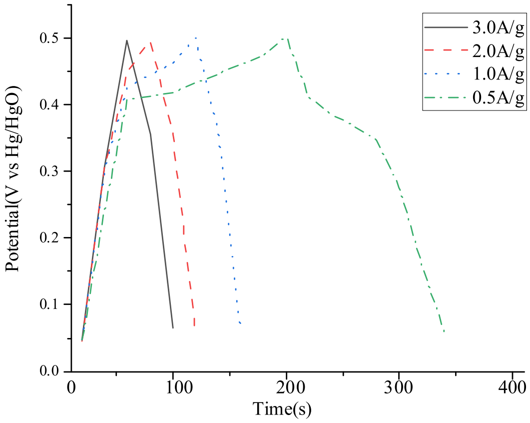
Fig. 6. Results of test voltage and current selection (a) Voltage results; (b) Current influence results.



(a) Results for the sample prepared at 200°C



(b) Results for the sample prepared at 300°C



(c) Results for the sample prepared at 350°C

Fig. 7. GCD curve results.

3.4. *Electrolyte particle contact test*

Based on the data collected from the experiment, the alternating current impedance of the electrode material designed here is analyzed. Fig. 8 presents the experimental results.

Fig. 8 illustrates that as the current density increases gradually, the specific capacities of all three types of SC electrodes, prepared based on cobalt-based MOFs, show a decreasing trend. This phenomenon occurs because, with the increase in current density, the corresponding electrochemical reaction rate accelerates. The accelerated electrochemical reaction rate may exceed the effective diffusion rate of electrolyte ions entering the internal pores of the electrode material, thereby limiting the actual utilization of the electrode material. However, it is noteworthy that among the samples prepared under three different conditions, the specific capacity decay rate of the cobalt-based MOF electrode material treated at 600 °C is relatively low. This suggests that the structure of cobalt-based MOFs prepared at higher temperatures exhibits higher stability and can interact more effectively with electrolyte ions. Even under drastic changes in current density, it can maintain higher specific capacity performance. Hence, it can be preliminarily concluded that compared to samples prepared under other temperature conditions, the cobalt-based MOF SC electrode material treated at 600 °C demonstrates stronger electrochemical stability and adaptability when facing various current density application requirements.

One of the primary reasons for the decrease in specific capacity due to the increase in current density is the accelerated electrochemical reaction rate. With the rise in current density, the electrochemical reaction rate on the surface of the electrode material also increases, leading to more charge storage and release processes occurring per unit time. However, the diffusion rate of electrolyte ions into the internal pores of the electrode material is relatively slow, particularly at high current densities. This results in the electrochemical reaction rate accelerating beyond the increase in the diffusion rate of electrolyte ions, thereby limiting the actual utilization of the electrode material. Moreover, the relatively low specific capacity decay rate of the cobalt-based MOF electrode material treated at 600 °C indicates that the material prepared at this processing temperature exhibits higher stability. High-temperature treatment may help enhance the stability of the material's crystal structure and reduce defects and imperfections in the material, thus slowing down the rate of specific capacity decay. Additionally, high-temperature treatment promotes more effective interaction between the material and electrolyte ions, enhancing the material's electrochemical performance and stability.

3.5. *Environmental and safety impacts of cobalt-based MOFs*

Cobalt is a vital component of cobalt-based MOFs, and its mining and extraction processes can have adverse environmental effects, including land disruption, water pollution, and ecosystem destruction. In SCs, cobalt-based MOFs may interact with other materials, potentially leading to the release or dissolution of components. These releases could adversely impact water bodies, soil quality, and ecosystems, necessitating thorough assessment and monitoring of their environmental effects. Additionally, cobalt-based MOFs may undergo decomposition or unstable reactions under high-temperature conditions, compromising battery performance or posing safety hazards such as fires. Therefore, evaluating the thermal stability of cobalt-based MOFs is essential to ensure safety under diverse environmental conditions. Moreover, cobalt-based MOFs may react with electrolytes or other materials, generating harmful substances or degradation byproducts. These reactions may affect SC performance and pose risks to both human health and the environment. Therefore, assessing and monitoring their chemical stability are imperative. Furthermore, cobalt-based MOFs and their degradation products may exhibit toxicity or trigger allergic reactions in living organisms. Hence, evaluating their biocompatibility within living systems is crucial to ensure safety. In the design and use of SCs, comprehensive consideration of these factors is necessary to ensure the environmental friendliness and safety of cobalt-based MOFs.

4. Discussion

Through characterization tests of cobalt-based MOF composite materials, this study discovers that the composite material exhibits a

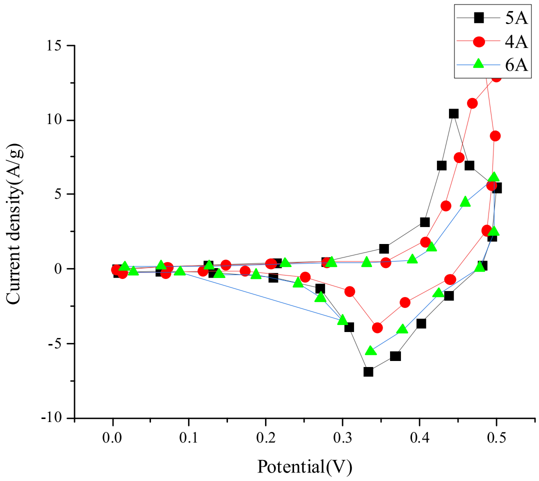


Fig. 8. Effect of electrochemical properties of samples: CV curve result.

particle surface morphology with uniform particle distribution. X-ray photoelectron spectroscopy (XPS) results indicate that cobalt is the main element in this composite material. In terms of electrochemical performance analysis, the study investigates the electrochemical properties of CNTs, Mn-MOFs, and cobalt-based MOFs. The results reveal that the specific capacitance of cobalt-based MOF composites is notably higher than the sum of the two individual electrode materials, and it increases significantly with increasing scan rate. The electrode performance analysis of cobalt-based MOF materials prepared at diverse temperatures reveals that samples prepared at 250 °C exhibit the longest discharge time and the highest specific capacitance, reaching 269 F/g. Electrolyte particle contact tests demonstrate that as the current density increases, the specific capacitance of all three materials decreases. However, the decrease rate is relatively slow in the high-temperature samples, illustrating better structural stability and electrolyte ion contact in the high-temperature samples. Shahzad et al. (2024) investigated the potential applications of MOF-derived MO composite materials in lithium-ion batteries, sodium-ion batteries, potassium-ion batteries, zinc-ion batteries, and SCs. They emphasized the advantages of these composite materials in improving the performance of batteries and SCs, encompassing increased reversible capacity, cycling performance, and significant rate performance [32]. Khan et al. (2024) focused on the development of SCs and reviewed the progress of SC materials, including the development of new electrode materials, heat-resistant separators, flexible SCs, and high-conductivity electrolytes. The research also detailed the latest developments in high-temperature capacitive energy storage, various industrial applications of SC battery systems, SC design, and the challenges involved [33]. Combining the contents of the two reviews, it can be found that they both emphasized the importance of MOF-derived MO composite materials and the field of SCs. Compared with these two reviews, this study more specifically explores the application of cobalt-based MOFs in the electrode materials of SCs for new energy vehicles and concludes the effects of preparation temperature on electrochemical performance and stability through experimentation and analysis.

In summary, cobalt-based MOF composites exhibit excellent electrochemical properties in the electrode materials of SCs for new energy vehicles. By forming a special conductive network, this composite material can improve charge transfer efficiency and enhance electrochemical reaction activity, thus achieving superior performance compared to individual materials. Additionally, this study indicates that the preparation temperature significantly impacts the electrochemical performance and stability of cobalt-based MOF materials. A higher preparation temperature helps improve the material's crystallinity and conductivity, thereby enhancing electrochemical activity and stability. These findings provide important references and guidance for further optimizing electrode materials for SCs, contributing to the development of new energy vehicle technologies.

In the application of cobalt-based MOFs in SCs, it is vital not only to focus on their excellent electrochemical performance but also to recognize the potential risks that their environmental and safety impacts may bring. Using cobalt-based materials may involve environmental issues such as cobalt resource mining, energy consumption in the preparation process, and waste disposal. Additionally, cobalt is toxic, posing potential hazards to both human health and the environment during preparation, use, and disposal. To address these potential issues, the following mitigation strategies or recommendations for future research can be made: (1) Material design and preparation optimization: Optimize the design and preparation of cobalt-based MOF materials to reduce the demand for hazardous substances, decrease energy consumption, and minimize waste generation in the production process. (2) Resource recovery and reuse: Conduct research on the recovery and reuse of cobalt-based materials to decrease the consumption of limited resources such as cobalt and mitigate environmental impact. (3) Environmental monitoring and assessment: Strengthen the monitoring and assessment of the impact of cobalt-based MOF materials on the environment throughout their life cycle – from preparation to use and disposal – to promptly identify and address potential environmental problems. (4) Research on alternative materials: Promote research and development of alternative materials to cobalt-based MOFs, aiming to identify more environmentally friendly and safer alternatives, thus reducing environmental and safety risks. (5) Full life cycle assessment: Perform a full life cycle assessment of cobalt-based MOF materials, comprehensively analyzing the entire process from resource extraction, production, use, to disposal. This approach aids in fully understanding its environmental and safety impacts and provides a scientific basis for sustainable development.

By adopting these mitigation strategies and recommendations for future research, the environmental and safety risks associated with the use of cobalt-based MOFs in SCs can be minimized, promoting their sustainable development and widespread application.

5. Conclusion

The study initially discusses SCs for new energy vehicles and MOF materials, establishing a theoretical foundation for further preparation of cobalt-based MOF materials. Simultaneously, relevant materials, electrode materials, and SC components are prepared and subjected to simulation experiments. In this study, the optimization of cobalt-based MOFs' application in the electrode materials of SCs for new energy vehicles is successfully achieved. Electrochemical performance analysis of cobalt-based MOF materials prepared at various temperatures reveals that samples processed at higher temperatures exhibit superior performance and stability. Therefore, it is suggested to consider employing cobalt-based MOF composite materials in the design of SC electrode materials, with a preference for samples prepared at higher temperatures to achieve better electrochemical performance. While this study has made significant findings, some limitations need to be addressed. For instance, the sample size used in this study is relatively small, and only cobalt-based MOFs at different preparation temperatures have been investigated. A broader range of samples and additional preparation conditions may provide a more comprehensive understanding and more accurate assessment of the material's performance. Furthermore, the experimental conditions utilized here may be limited; for example, uncertainties may exist in the material preparation process, and certain interfering factors may affect electrochemical performance tests. Modifications to these conditions could potentially influence the final experimental results.

The main contributions of this study are as follows: (1) Optimization of cobalt-based MOFs application in electrode materials for SCs of new energy vehicles: Through experiments and analysis, the researchers have successfully optimized the application of cobalt-based MOFs in electrode materials for SCs of new energy vehicles. The samples prepared using cobalt-based MOF composites at different temperatures exhibit excellent electrochemical properties and stability, providing a crucial experimental basis for enhancing the performance of SCs. (2) Proposal of the significant influence of preparation temperature on material properties: Through electrochemical performance analysis of cobalt-based MOF materials prepared at various temperatures, it is found that samples processed at higher temperatures demonstrate better performance and stability. This discovery offers essential theoretical guidance for the design of electrode materials for SCs, suggesting a preference for higher-temperature conditions in the preparation process to achieve superior performance.

These contributions have advanced the current state of knowledge in the field and provided a crucial experimental and theoretical basis for the development of SCs for new energy vehicles. By optimizing the application of cobalt-based MOF composites, this study is expected to propel SC technology forward, enhancing its performance and stability, and fostering the application and sustainable development of clean energy. Additionally, by identifying existing limitations and proposing future research directions, this study offers significant guidance and insight for further exploration in this field.

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Availability of data and materials

All data generated or analyzed during this study are included in this published article [and its supplementary information files].

CRediT authorship contribution statement

Xinjun Jin: Writing – review & editing, Writing – original draft, Validation, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Zhiyu Jiang:** Writing – review & editing, Writing – original draft, Visualization, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Yunhe Feng:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Formal analysis, Data curation, Conceptualization. **Xiaofen Fang:** Writing – review & editing, Writing – original draft, Resources, Project administration, Methodology, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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