

Research Progress on the Preparation of Iron Molybdate Electrode Materials and Their Application in Supercapacitors

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Abstract. Against the background of rapid development of new energy technologies and growing demand for energy storage, supercapacitors have drawn extensive attention in fields including electric transportation, renewable energy regulation and backup power systems, thanks to their fast charge-discharge rate, high power density and long cycle life. As a core factor determining the performance of supercapacitors, electrode materials directly define the energy density and power output of devices via their structural composition and electrochemical activity. As a type of transition metal molybdate, iron molybdate (FeMoO_4) features multi-valence redox properties of Fe and Mo as well as favorable structural tunability, demonstrating certain application potential among pseudocapacitive electrode materials. This paper first introduces the basic structure, application characteristics and energy storage mechanisms of supercapacitors, then reviews the preparation and performance of FeMoO_4 -based materials, with focus on the preparation routes and performance differences of iron molybdate-based carbon composites, conductive polymer composites, metal oxide composites and nanostructure-regulated materials. The modification strategies and performance features of FeMoO_4 -based electrode materials for supercapacitors are summarized, which may provide reference for the design of high-performance iron molybdate-based energy storage materials in future studies.

Keywords: Iron molybdate, Supercapacitors, Electrode materials, Composite modification, Nanostructure regulation

1. Introduction

With the rising proportion of new energy utilization, energy storage technology plays an increasingly prominent role in power regulation, transportation electrification and portable electronic devices. Supercapacitors, characterized by fast charge-discharge speed, high power density and long cycle life, are applicable to scenarios that require rapid energy input and output [1]. However, compared with batteries, supercapacitors still suffer from relatively low energy density. Therefore, developing electrode materials with high activity and high stability has become the key to improving their overall performance.

Electrode materials are critical to the performance of supercapacitors. Their composition, conductivity and electrochemical activity are directly related to the specific capacitance, energy

density, power density and cycle life of devices. At present, material systems including carbon materials, conductive polymers and transition metal oxides/molybdates are major research directions for supercapacitor electrodes, among which nanostructure regulation and composite modification are common strategies to boost electrode performance [2, 3]. Among various transition metal molybdates, iron molybdate (FeMoO_4) has gained growing attention for its certain pseudocapacitive reactivity and structural design flexibility, as it contains two variable-valence metal elements Fe and Mo. Nevertheless, its poor intrinsic conductivity and limited utilization of active sites still restrict further improvement of its electrochemical performance.

To tackle the above issues, existing studies carry out optimization mainly from two aspects: one is to shorten ion diffusion paths and increase electrolyte contact area by constructing nanoparticles, nanosheets, porous or hollow structures; the other is to compound FeMoO_4 with rGO, carbon nanofibers, carbon black, PANI or other metal oxides, so as to improve electron transport and enhance multi-component synergistic energy storage [4-7]. On this basis, this paper first introduces the basic structure, application characteristics and energy storage mechanisms of supercapacitors, then summarizes and analyzes the classification, preparation methods and electrochemical performance of FeMoO_4 -based electrode materials. It focuses on comparing the performance differences of carbon composites, conductive polymer composites, metal oxide composites and nanostructure-regulated materials, aiming to provide reference for the design of high-performance iron molybdate-based supercapacitor electrode materials.

2. Supercapacitors and their devices

2.1. Introduction to supercapacitors

Supercapacitors are energy storage devices that lie between conventional capacitors and batteries. Their main merits include fast charge-discharge speed, high power density, long cycle life and good safety. Compared with batteries, they are more capable of delivering or absorbing large currents in a short time, and thus are often used in occasions requiring rapid energy exchange. In practical applications, supercapacitors are widely used for braking energy recovery and start-up assistance of new energy vehicles, as well as power fluctuation smoothing in wind and solar power generation systems. Besides, they can serve as backup power supplies for systems with frequent start-stop operations, such as electronic equipment, industrial facilities, rail transit and elevators. In brief, supercapacitors are not featured by "long-duration power supply", but by fast charging, fast discharging and reusability. Therefore, they are suitable to be used in combination with batteries to improve the stability and service life of energy storage systems.

2.2. Energy storage device of supercapacitors

As shown in Figure 1, a supercapacitor mainly consists of a power source, current collectors, polarized electrodes, electrolyte, a separator, and the Helmholtz double layer at the electrode/electrolyte interface. Current collectors connect the external circuit and conduct electrons; polarized electrodes provide interfaces for charge storage; positive and negative ions in the electrolyte migrate directionally under an electric field and adsorb onto electrode surfaces of opposite polarity, forming a double-layer structure. The separator is placed between the two electrodes to prevent direct contact of electrodes while allowing ion passage. This structure enables supercapacitors to realize rapid energy storage and release through interfacial charge separation.

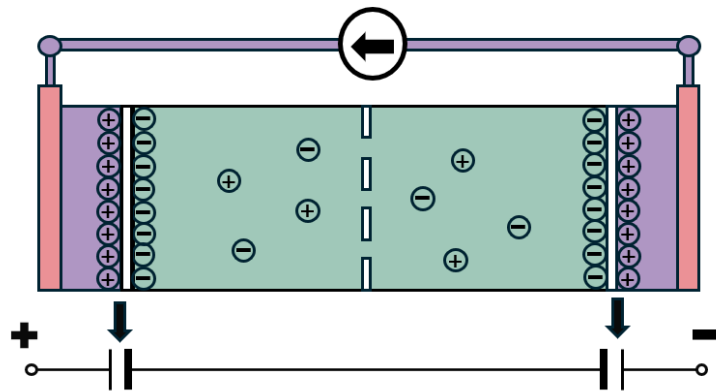


Figure 1. Schematic diagram of a supercapacitor device

3. Applications and working principles of supercapacitors

3.1. Electric double layer energy storage mechanism

As shown in Figure 2, under an applied voltage, polarized charges form on the electrode surface. Counter ions in the electrolyte migrate to the electrode/electrolyte interface and get adsorbed, forming a double-layer structure with separated charges. The distance between the ion layer adjacent to the electrode surface and the electrode charges is extremely small, endowing the interface region with high charge storage capacity. As the distance from the electrode surface increases, the electric potential decays gradually, and part of the ions distribute in the diffusion layer. This process relies mainly on electrostatic adsorption and desorption to store and release energy, and generally involves no obvious bulk chemical reactions, hence the characteristics of fast response and good cycling stability.

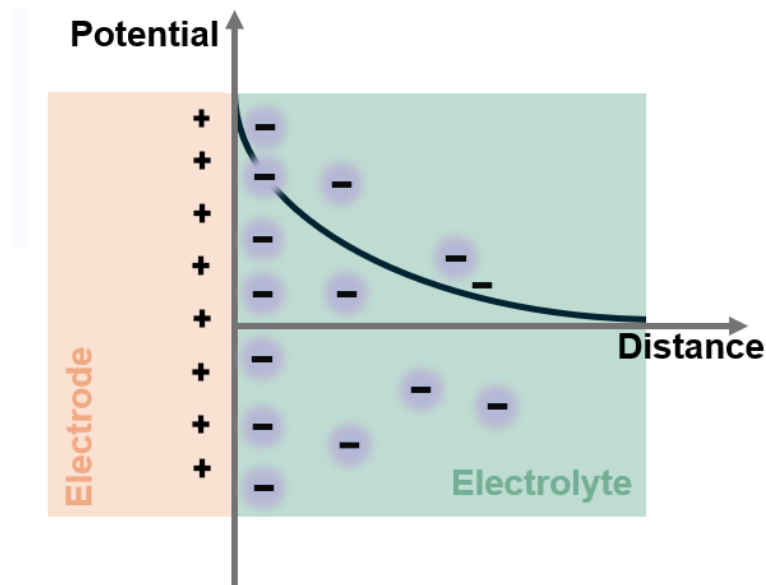


Figure 2. Schematic diagram of electric double layer energy storage mechanism

3.2. Pseudocapacitive energy storage mechanism

Pseudocapacitive energy storage originates from fast reversible Faradaic reactions on or near the electrode surface. As shown in Figure 3, electrolyte cations migrate toward the electrode under an electric field and undergo charge transfer with redox-active materials, causing reversible changes in the valence state of electrode materials. Charge storage in this process can be realized via surface adsorption/desorption or shallow ion intercalation. Compared with the electrostatic adsorption mechanism of electric double layer capacitance, pseudocapacitance delivers higher charge storage capacity while maintaining fast charge-discharge response.

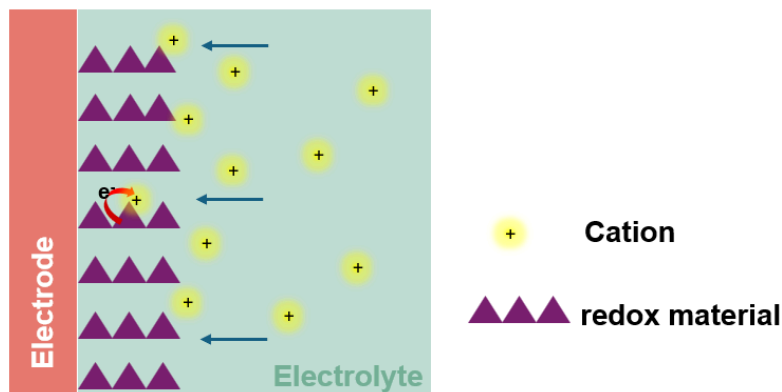


Figure 3. Pseudocapacitive energy storage principle

4. Preparation methods of iron molybdate materials

Iron molybdate (FeMoO_4) is a class of transition metal molybdate materials with multi-valence redox activity. Owing to its high electrochemical activity, tunable structure and great room for composite modification, it is regarded as a promising electrode material for supercapacitors. To improve its energy storage performance, current studies mainly focus on nanostructure regulation and composite structure design, including constructing nanoparticles, nanosheets, porous or hollow structures, and compounding with materials such as rGO, carbon nanofibers, carbon black and PANI, so as to enhance its conductivity, ion diffusion and pseudocapacitive reaction capability. In addition, $\text{FeMoO}_4/\text{rGO}/\text{PANI}$ composites have also been applied to uric acid detection, indicating that iron molybdate-based composites have certain application potential in electrochemical sensing as well [5].

In terms of preparation routes for iron molybdate-based electrode materials, existing studies can be roughly classified into three categories. First, hydrothermal method followed by calcination is adopted to regulate the nano-morphology of FeMoO_4 , such as nanosheets, nanoparticles and one-dimensional hollow structures, so as to improve electrolyte contact and ion transport [7-9]. Second, FeMoO_4 is compounded with conductive components like rGO, carbon nanofibers, carbon black or PANI to compensate for its poor intrinsic conductivity [4, 5, 8, 10, 11]. Third, metal oxide composite structures such as $\text{MoO}_2@\text{FeMoO}_4$ and $\text{FeMoO}_4/\text{Fe}_2\text{O}_3/\text{rGO}$ are constructed via precursor pyrolysis or solvothermal-calcination methods, to strengthen the synergistic effect among multiple components [6, 12]. These methods share the same goal: optimizing the conductivity, active site utilization and ion diffusion paths of FeMoO_4 , so as to improve its performance as supercapacitor electrodes.

Among them, the hydrothermal method is most widely used. It is suitable for preparing FeMoO_4 and its composites with various morphologies, and features relatively mild reaction conditions and strong morphology controllability [4, 7, 9, 11]. However, from the perspective of specific preparation procedures, some systems still require combination with calcination, polymerization or multi-step compounding, so the preparation complexity rises with the increase of composite components [5, 6, 8, 11, 12]. The electrospinning-hydrothermal method can construct self-supported FeMoO_4 /carbon nanofiber electrodes, which helps form a continuous conductive framework. But its process involves steps including electrospinning, stabilization, annealing and hydrothermal growth, posing high requirements for equipment and heat treatment conditions [8]. The metal oxide composite method can build a synergistic system with multiple active components, but it usually involves precursor design, high-temperature pyrolysis or solvothermal-calcination treatment, leading to relatively complicated process flow [6, 12]. Overall, the preparation of iron molybdate-based materials requires a balance between process simplification, composite uniformity and structural stability, as detailed in Table 1.

Table 1. Types and preparation methods of iron molybdate materials

No.	Material type	Raw material/composition	Preparation method	References
1	Iron molybdate-based carbon composites	FeMoO_4 and r-GO; FeMoO_4 nanosheets and carbon nanofibers(CNF); FeMoO_4 porous nanospheres and carbon black (CB); FeMo(x)@MgO/CNTs containing iron molybdenum oxide components such as FeMoO_4 and $\text{Fe}_2(\text{MoO}_4)_3$	Hydrothermal method-calcination treatment; electrospinning-hydrothermal method-calcination treatment; hydrothermal synthesis of FeMoO_4 followed by composite modification with carbon black; combustion synthesis of FeMo(x)@MgO catalyst, two-step pyrolysis-catalysis to prepare CNTs and coat into electrodes	[4, 8, 10, 13]
2	Iron molybdate-based conductive polymer composites	FeMoO_4 , rGO and PANI; FeMoO_4 and PANI	Hydrothermal method; hydrothermal synthesis of FeMoO_4 , oxidative polymerization of PANI, followed by hydrothermal compounding	[5, 11]
3	Iron molybdate-based metal oxide composites	$\text{MoO}_2@\text{FeMoO}_4$; $\text{FeMoO}_4/\text{Fe}_2\text{O}_3/\text{rGO}$	MOF precursor pyrolysis method; solvothermal method-calcination treatment	[6, 12]
4	Iron molybdate nanostructure-regulated materials	$\beta\text{-FeMoO}_4$ nanoparticles; one-dimensional hollow FeMoO_4 structure	Hydrothermal method; hydrothermal method-calcination treatment	[7, 9]

5. Performance study of iron molybdate materials for supercapacitors

Different types of iron molybdate-based electrode materials have different performance strengths. Iron molybdate-based conductive polymer composites and metal oxide composites show outstanding performance in specific capacitance, cycle stability and energy density. Though with relatively low energy density, iron molybdate nanostructure-regulated materials deliver high power density, reflecting good fast charge-discharge capability. Iron molybdate-based carbon composites are mainly used to improve the conductivity and electrode structure of FeMoO_4 . Different structural designs correspond to different performance improvement paths, as detailed in Table 2.

The main advantage of iron molybdate-based carbon composites lies in conductivity improvement, rather than achieving the highest specific capacitance or energy density. Carbon materials can build an electron transport network for FeMoO_4 , raise the utilization rate of electrode materials, and improve the output stability of devices [4, 8, 10]. However, judging from the device performance in Table 2, simple carbon compounding brings limited improvement to energy density and specific capacitance, and further combination with pseudocapacitive components or stable composite structures is still needed.

Iron molybdate-based conductive polymer composites exhibit the most excellent comprehensive performance. Such materials generate a synergistic effect through the multi-valence redox reactions of FeMoO_4 , the pseudocapacitive contribution of PANI and the conductive function of rGO, hence the high specific capacitance, energy density and cycle retention rate shown in Table 2 [5, 11]. Their strength is that they can simultaneously enhance charge storage capacity and device energy output; their drawback is the relatively complex material composition, which puts forward high requirements for composite structure uniformity and interface stability.

Iron molybdate-based metal oxide composites present balanced performance in energy density and cycle stability. By introducing other oxide components, such materials add more sources of redox reactions and form a composite energy storage structure with FeMoO_4 , thus improving the energy output and long-cycle performance of devices [6, 12]. Their strength is stable comprehensive performance; their drawback is that the preparation process usually involves precursor construction, heat treatment and multi-step compounding, with relatively high process complexity.

Iron molybdate nanostructure-regulated materials are distinguished by high power density and steady cycle stability. Nanoparticles and one-dimensional hollow structures can improve electrolyte contact, shorten ion diffusion paths, and provide more favorable transport channels for fast charge-discharge processes [7, 9]. Therefore, although their specific capacitance and energy density are lower than those of composite materials, they have advantages in rate performance and fast energy delivery. Their limitation is the lack of additional conductive or pseudocapacitive components, resulting in limited improvement of overall energy density.

In conclusion, the modification strategy for iron molybdate-based electrode materials should be selected according to target performance. For high specific capacitance and energy density, conductive polymer composites and metal oxide composites are more advantageous; for fast charge-discharge and structural stability, nanostructure-regulated materials have higher application value; for solving the problem of poor conductivity of FeMoO_4 , carbon composites are a direct and effective modification approach. On the whole, multi-component compounding is more conducive to improving the comprehensive energy storage performance of FeMoO_4 -based materials, while nanostructure regulation has unique advantages in rate performance and structural stability. The details are shown in Table 2.

Table 2. Performance of iron molybdate materials

Material category	Specific capacitance (F/g)	Cycle stability (%)	Energy density (Wh/kg)	Power density (W/kg)	References
Iron molybdate-based carbon composites	121	87	43	790	[4, 10, 14]
Iron molybdate-based conductive polymer composites	295	96	110	507.4	[5, 11]

Table 2. (continued)

Iron molybdate-based metal oxide composites	237.3	95.0	82.0	831.1	[6, 12, 15]
Iron molybdate nanostructure-regulated materials	96–101	89.0–89.62	29.89–40.75	850–1001.58	[7, 9, 16, 17]

6. Conclusion

Studies have shown that with the multi-valence redox activity and structural tunability of Fe/Mo, iron molybdate (FeMoO_4) can be fabricated into various types of supercapacitor electrode materials through nanostructure regulation and composite modification. Different modification paths have different focuses: conductive polymer compounding and metal oxide compounding are more effective in improving specific capacitance, energy density and cycle stability; nanostructure regulation excels in fast charge-discharge and structural stability; carbon compounding mainly serves to improve the conductivity and electrode structure of FeMoO_4 . Overall, FeMoO_4 -based materials possess favorable electrochemical activity and modification potential, but issues such as complex preparation, composite uniformity and long-term stability still need to be further addressed. Focusing on the application of iron molybdate (FeMoO_4)-based materials in supercapacitors, this paper systematically reviews the research progress of FeMoO_4 as a pseudocapacitive electrode material, and analyzes the impacts of different preparation routes on material structure and energy storage performance. Combined with the device structure of supercapacitors, the electric double layer energy storage mechanism and the pseudocapacitive energy storage mechanism, the performance characteristics of four typical systems, namely iron molybdate-based carbon composites, conductive polymer composites, metal oxide composites and nanostructure-regulated materials, are summarized and compared, revealing the rule of how different modification strategies improve the performance of FeMoO_4 -based electrode materials. In summary, as a transition metal molybdate with multi-valence redox activity and structural tunability, FeMoO_4 shows good application potential as supercapacitor electrode materials. Through classification and performance analysis of existing studies, this paper clarifies the relationship between structural design, composite modification and electrochemical performance, and provides reference for the subsequent design of high-performance and stable iron molybdate-based energy storage materials.

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