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Performance Optimization Strategies for Porous Carbon Supercapacitors

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Abstract: Porous carbon materials are carbon-based materials with well-developed pore structures and high specific surface areas. This unique structure endows porous carbon with excellent physical properties, adsorption capacity, and catalytic activity, making it a popular material for supercapacitor electrodes. However, low energy density has become a major bottleneck that limits its development. This article reviews the optimization methods from the two perspectives of the template method and the heteroatom doping method by sorting out its energy storage mechanism. The results show that the hard template method can accurately regulate the pore size, but the process of removing the template involves strong corrosive reagents; The soft template method does not need to use acid/base etching, but it is difficult to regulate the pore size accurately; The salt template method can remove the template by washing, which is the most environmentally friendly strategy among the three; Heteroatom doping is mainly represented by nitrogen doping, which can effectively improve the electrochemical properties of porous carbon materials, but the underlying mechanism still needs to be studied. Finally, the possible development prospects of porous carbon supercapacitors are discussed at the end.

Keywords: Porous carbon; Supercapacitors; Template method; Heteroatom doping

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1. Introduction

A supercapacitor is a new type of energy storage device with the characteristics of high power density and long cycle life, so it has attracted much attention in energy storage. However, compared with commercial lithium-ion batteries, their energy density is lower and their endurance is poor, which seriously limits the development of supercapacitors [1].

In order to overcome these limitations, researchers tried to introduce porous materials into the electrode materials of supercapacitors, and porous carbon has attracted much attention because of its low cost, simple preparation process, and excellent stability [1]. The researchers used the hard template method to prepare and successfully achieved the regulation of the pore size, but the preparation process involves the use of strong corrosive acids or strong alkalis to remove the template, which does not meet the requirements of green chemistry [1]. The emerging salt-template method mainly uses water for cleaning, which is cheaper and more

environmentally friendly [2]. Although heteroatom doping can effectively improve the conductivity of carbon materials, thereby improving the energy density, the underlying mechanism has not been fully exposed and hinders its practical application [3].

This article will start with the energy storage mechanism of supercapacitors, review the two processes of the template method and heteroatom doping to optimize porous carbon materials, and compare and analyze the advantages and disadvantages of different strategies in terms of pore size control, green preparation, performance improvement, etc., which can provide literature support and theoretical basis for subsequent researchers to understand this field quickly.

2. Energy storage mechanisms of porous carbon supercapacitors

According to the different energy storage mechanisms, the current supercapacitors can be roughly divided into three categories: Using electric double-layer capacitors, Faradaic pseudocapacitors that rely on surface redox reactions, and hybrid capacitors that combine the two mechanisms. The porous carbon supercapacitor mainly uses the first principle: placing two non-reactive electrode plates in an electrolyte solution, and then applying voltage at both ends. At this time, under the drive of the external voltage, the positive electrode plate adsorbs anions, the negative electrode plate adsorbs cations, and the potential difference of the two polar plates realizes the storage of energy. Because it only relies on physical adsorption to store energy, this kind of capacitor has a long cycle life. The energy storage is directly proportional to the capacitance of the capacitor. The energy density can be improved by increasing the capacitance. Carbon porous materials have a large specific surface area due to their developed pore structure, so they can adsorb more ions under the same conditions and have greater capacitance than conventional materials. Nevertheless, porous carbon supercapacitors are far inferior to today's commercial lithium-ion batteries in terms of energy density. In order to further reduce this gap, researchers have given multi-angle optimization strategies around the preparation of porous materials: First, use the template method to regulate the pore diameter structure of porous carbon electrodes; second, use the chemical doping of porous carbon electrodes by introducing nitrogen, oxygen, sulfur, and other heteroatoms.

3. Applications of template methods

When the pore diameter of the carbon material is slightly larger than the diameter of the desolvated ion, the ion can undergo desolvation, so as to enter the hole in a denser way, make full use of the huge specific surface area, and improve the capacitance. The idea of the template method is to design the structure of the template in advance, reversely synthesize the material according to its configuration, and then control the shape and size of the finished product. For porous carbon materials, the template method can be used to regulate their size effectively. According to the template category, it can be divided into three types: hard template method, soft template method, and salt template method that has emerged in recent years.

3.1. Hard-template method

The hard template method mainly uses ZnO, MgO, SiO_2, $CaCO_3$, and other rigid inorganic materials as templates, fills the carbon precursor into its voids, calcines and carbonizes at high temperature, removes the template by acid immersion or alkali immersion, and finally obtains carbon porous materials with inverse

replica structure [4].

The traditional hard template method often includes these four steps, and the steps are more complicated and tedious. In response to this problem, the team of Zhejiang Sci-Tech University proposed a more efficient improvement method - mixing tetraethyl orthosilicate as a silicon precursor, resorcinol and formaldehyde as carbon precursors, and mixing with catalysts to complete the template synthesis and material coating in one step, and finally obtain mesoporous hollow carbon nanospheres through hydrofluoric acid etching [5]. After experimental verification, the carbon nanosphere prepared by this method has a specific capacitance of 240F/g at a current density of 1A/g, and the energy density is significantly improved compared with ordinary carbon porous materials; The cyclic stability is also extremely excellent - after 10,000 charges and discharges, the specific capacitance retention is still as high as 93.75%, reflecting the good application potential.

The team's method greatly simplifies the complex and cumbersome preparation process of porous carbon materials. At the same time, the preparation of templates and material coating is carried out. The excellent performance of the preparation of carbon nanospheres also proves the rationality of this method, providing feasible ideas for the low-cost and large-scale preparation of porous carbon materials in the future; However, this method still has its limitations. In the preparation process, it relies on strongly corrosive hydrofluoric acid and fails to find a good green substitute; At the same time, the research has not quantitatively explored the relationship between the template size and the product pore size, and the process stability and the accuracy of pore size control have yet to be verified.

3.2. Soft-template method

The soft template method uses organic molecules such as polymers and surfactants as templates. After they are self-assembled to form a stable structure in the solvent, carbon precursors are added. The two are combined through intermolecular forces such as hydrogen bonds and van der Waals forces. After high-temperature carbonization, the soft template is removed, and finally a carbon porous material with a reverse replication structure is formed [4]. However, the method also faces some existing challenges, where the pore size is difficult to regulate accurately, the self-assembly system is difficult to control fully, and the cost of soft templates is high.

In order to solve these problems, Liang et al. proposed a method of integrating a multi-component collaborative system to synthesize porous carbon spheres [6]. The team used corn straw alkali lignin as the carbon source, CTAB as the soft template, zinc ions as the cross-linking agent, and potassium bicarbonate as the activator. After mixing the four components, hydrothermal synthesis and high-temperature carbonization were carried out, and finally, the finished porous carbon spheres were prepared. Its electrochemical performance is good, and it is tested with a current density of 0.2 A/g. The material has a specific capacitance of 293 F/g, which is also at a high level in biomass derivatives; When the current density increases to 40 A/g, the initial capacity has a retention rate of 70.8%; after 10,000 charging and discharging cycles, the specific capacitance retention rate is 86.7%, demonstrating excellent cycling stability.

The team's innovation points in the synthesis process are mainly as follows. First, use low-cost and easily available corn straw as a carbon source to realize the effective use of biomass resources; The second is the introduction of zinc ions to form stable coordination bonds with hydroxyl groups and phenol hydroxyl groups in lignin, thus enhancing the stability of the complex and facilitating the control of the synthetic

self-assembly system; Finally, potassium bicarbonate is added, which will release carbon dioxide during decomposition, which can enrich the pore structure of porous carbon spheres and solve the problem that the conventional soft template method is difficult to regulate the pore size. This work shows that it is completely possible to prepare high-performance porous carbon materials using renewable biomass resources, and also provides new ideas for the rational utilization of biomass resources.

3.3. Salt-template method

The salt template method mainly uses NaCl, KCl, LiCl, and other water-soluble salts or their mixtures as templates. The carbon precursor is fully mixed with salts. After the crystal is precipitated, it is carbonized at high temperature under inert gas conditions. Finally, the salt template is removed with deionized water, and the product can be obtained after drying [2]. Unlike etching agents such as hydrofluoric acid used in the hard template method, the salt template method only needs deionized water to be washed, which is more environmentally friendly. However, the salt template method still has defects such as long washing time and difficulty in completely removing the template.

To address this issue, Feng et al. pioneered a self-sacrificial salt-template method, utilizing ammonium thiocyanate ()—which readily decomposes upon heating, as the self-sacrificial salt [7]. In the process of high-temperature carbonization, ammonium thiocyanate will decompose and evaporate by itself, thus eliminating the subsequent tedious washing steps. At the same time, the decomposition of ammonium thiocyanate also promotes the formation of rich micropores inside the carbon material. The pore diameter is distributed at 0.6–1.1 nm, and the specific surface area of micropores is as high as 770 m^2/g.

This method fundamentally solves the problem of residual template in salt-template-synthesized porous carbons, and this strategy is worth adopting. However, it should also be pointed out that it has limitations; at present, it is only applicable to ammonium thiocyanate, which is easy to decompose, and it is difficult to generalize to conventional salt templates. In addition, the size control of crystal growth has not been solved. Therefore, although the salt template method has significant advantages from the perspective of green synthesis, it still faces great challenges in applying it to industrial production and commercialization.

4. Applications of heteroatom doping

The raw materials doped with heteroatoms contain precursor substances of non-carbon elements such as nitrogen, sulfur, phosphorus, and boron. In the process of high-temperature carbonization, heteroatoms will form a coordinate bond or covalent bond with the carbon skeleton, thus partially replacing the carbon atoms in the carbon lattice, and finally obtaining porous carbon materials with electron-deficient or electron-rich characteristics.

Heteroatom doping can effectively improve the performance of porous carbon electrodes. Take nitrogen as an example - research shows that nitrogen atoms can provide delocalized electrons for the carbon skeleton, thus improving the overall conductivity of the material; nitrogen doping also introduces defects in carbon crystals, effectively increasing its specific surface area, and the more doped functional groups, the greater the energy density [8]. At the same time, nitrogen-containing functional groups (such as amino groups, imino groups, and pyridinic nitrogen, etc.) introduced after doping are often hydrophilic, which can promote electrolyte penetration into the pores through capillary action.

Cui Guangfu's team at Northeast Petroleum University uses longan shell as a carbon source. Under the condition of high-temperature activation and hydrothermal doping, after using urea and nitric acid to dope porous carbon with N and O, the specific capacitance at 0.5 A/g is 463 F/g, and the retention rate of 10,000 cycles under 20 A/g is 89% [9]. The main reasons can be attributed to two points: first, the carbon derivatives prepared from longan shells themselves have a graded porous structure, which can promote the transportation of ions in the pores; the doping of N and O also contributes to part of the pseudo-capacitance, which further improves the surface wettability of carbon materials and is conducive to the full infiltration of the electrolyte solution. The work of the research team provides a green and feasible path for the preparation of high-performance porous carbon, and realizes the value-added utilization of biomass resources.

The doping of other heteroatoms also has a similar modification effect, and there is a certain synergy between them, but the doping mechanism has not been fully exposed. For example, the influence of doping concentration, doping position, doping method, and other factors on the electrochemical properties of materials is still a direction worth exploring in depth.

5. Conclusion

This paper reviews the performance optimization strategy of porous carbon supercapacitors from two aspects: template method and heteroatom doping method. In terms of template method, the hard template method can more accurately regulate the pore size, but the process of removing the template involves the use of strong corrosive substances such as hydrofluoric acid; The soft template method does not need to be etched to remove the template, but it is difficult to regulate the pore size; the salt template method only needs to be washed to remove the template, and even when introducing self-sacrificial salt, it can be exempted from washing, so it is the one with the greenest development potential among the three methods. In terms of the heteroatom doping method, doped heteroatoms can improve the overall electrochemical properties of carbon materials by providing delocalized electrons, improving the wettability of materials, and introducing carbon skeleton defects.

This article summarizes the typical strategies to improve the energy density of porous carbon supercapacitors, which can provide ideas and references for researchers to optimize supercapacitors.

Porous carbon supercapacitors have excellent electrochemical properties, so they are widely used in portable electronic equipment, new energy vehicles, and other fields. Although there is a large difference in energy density compared with commercial lithium-ion batteries, with the advancement of green synthesis technology, a deeper understanding of its energy storage mechanism is likely to become a major trend in future development.

Disclosure statement

The author declares no conflict of interest.

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