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Design and Optimization of Graphene Aerogel-Based Electrodes in Supercapacitor

Zhixin Zhang

School of Materials Science and Engineering, Nanjing University of Posts and Telecommunications, Nanjing, China
b23060614@njupt.edu.cn

Abstract. Graphene aerogel, endowed with a unique three-dimensional porous network architecture, an ultra-high specific surface area, outstanding electrical conductivity, and robust mechanical stability, has emerged as one of the most compelling research hotspots in the realm of supercapacitor electrode materials in recent years. This paper undertakes a systematic and in-depth discussion on the regulation mechanisms governing the physical and chemical properties as well as the electrochemical performance of graphene aerogel-based electrode materials. It centers on three core optimization directions, namely intrinsic structure design, composite modification engineering, and multi-level pore structure construction, meticulously elaborating on the underlying action mechanisms, distinctive technical characteristics, and tangible performance improvement effects of each tailored strategy. Furthermore, this paper comprehensively summarizes the synergistic effects generated by the rational combination of different modification approaches and offers a forward-looking perspective on the future application prospects of graphene aerogel supercapacitors in the burgeoning field of high-performance energy storage systems through the integration of multiple advanced technologies.

Keywords: graphene aerogel, supercapacitor, electrode materials

1. Introduction

The rapid development of new energy vehicles, smart grids and portable electronic devices has placed higher demands on the energy density, power density, cycle stability and safety of energy storage devices. Supercapacitors, as a new type of energy storage device between traditional capacitors and secondary batteries, have the advantages of fast charging and discharging, long cycle life, and environmental friendliness. They play an irreplaceable role in scenarios requiring high power output in a short period of time. However, traditional electrode materials have problems such as limited specific surface area, large ion transport resistance, and insufficient capacitance performance, which restrict the further development of supercapacitors. Therefore, developing high-performance electrode materials has become the key to breaking through the bottleneck of energy storage.

Graphene aerogel (GA) is a three-dimensional porous material formed by self-assembly or cross-linking of graphene sheets. Its unique network structure not only gives the material ultra-low

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density, high porosity and high specific surface area, but also builds efficient electron transmission channels, providing sufficient space for the rapid diffusion of ions, making it an ideal choice for supercapacitor electrode materials. However, pure graphene aerogel has defects such as insufficient conductivity, limited pseudo-capacitive contribution, and insufficient mechanical stability, and requires structural regulation and performance optimization to overcome these limitations and expand its application scope.

In recent years, researchers have conducted extensive studies on improving the performance of graphene aerogels. For example, Wu et al, successfully prepared cyanine-C modified nitrogen-doped graphene aerogel (AC-NGA) through the hydrothermal self-assembly method. This material exhibits excellent electrochemical performance under general pH conditions. The symmetric supercapacitor constructed based on it exhibits remarkable energy density, impressive power density and excellent cycling stability in acidic environments, neutral and alkaline electrolytes [1]. Jinwei Zhang et al. successfully fabricated KVPO, F/reduced graphene oxide micro-grid aerogel electrodes using direct writing 3D printing technology. As the positive electrode of a potassium-ion battery, it demonstrates high reversible capacity, excellent cycling stability and good rate performance [2]. This paper will systematically review the regulatory mechanisms of the physicochemical properties and electrochemical performance of graphene aerogel electrode materials, focusing on the three core directions of intrinsic structure design, composite modification design, and three-dimensional morphology design. It will analyze the advantages and disadvantages of each method, explore the collaborative optimization mechanism of different strategies, and look forward to the future development direction and application prospects of graphee aerogel supercapacitors, providing a reference for the research and practical application of high-performance energy storage materials.

2. The regulatory mechanisms of physical and chemical properties as well as electrochemical performance

The core of optimizing the performance of GA lies in the synergy of the three-dimensional porous structure regulation and the multi-component modification strategy. The three-dimensional porous structure constructed through processes such as hydrothermal method and freeze-drying can effectively inhibit the degradation of graphite.The stacking phenomenon of graphene layers caused by π-π interaction and van der Waals forces significantly enhances the specific surface area of the material and optimizes the pore distribution. This not only provides sufficient active sites for charge adsorption, but also promotes the rapid diffusion of electrolyte ions, mitigates the electrode's volume fluctuation in the charge-discharge process, thus boosting its structural stability.

At the same time, through modification methods such as doping with sulfur, nitrogen and other heteroatoms, or introducing metal nanoparticles, the intrinsic conductivity of graphene can be improved. The composite of graphene with conductive polymers and metal oxides, combined with the enhancement effect of the three-dimensional porous structure on the efficiency of charge transmission, can collaboratively optimize the specific capacitance and energy density that balance the double-layer capacitance and Faraday capacitance, laying a key performance foundation for the application of this material in electrochemical energy storage.

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3. Intrinsic structure optimization

3.1. Construction of three-dimensional porous networks

The three-dimensional porous structure of GA is regulated through various methods, with the core objective being to optimize the pore size distribution, porosity and pore structure, in order to enhance the physical and chemical properties of the material.

The template method demonstrates unique advantages in the preparation of GA, enabling flexible control over the pore size and microstructure of the material. The core mechanism involves the reduction self-assembly process of graphene oxide (GO) or reduced graphene oxide (rGO) on the surface of a three-dimensional scaffold template. After the assembly is completed, the template is removed, and a porous GA with a cross-linked structure can be obtained. Researchers used foam nickel as the growth template and employed chemical vapor deposition (CVD) technology to decompose CH_4 at 1000℃ to deposit graphene films. After PMMA support, etching the template with hydrochloric acid or ferric chloride, and removing the support with acetone, they fabricated three-dimensional foam graphene (GFs) with a high specific surface area of 850 m^2/g and a pore volume of up to 99.7% [3].

The self-assembly method uses GO sheets as the core structural unit. After uniformly dispersing them in a liquid phase system, the GO sheets undergo self-assembly through an inducing effect to form graphene hydrogel (GH). Subsequently, through subsequent treatments such as critical drying or freeze-drying, GA is finally prepared. This preparation path endows the material with unique characteristics such as high specific surface area, extremely low density, and superior thermoelectric performance, enabling it to demonstrate significant application potential in supercapacitors.

The chemical cross-linking method utilizes the carboxyl and hydroxyl groups on the surface of graphene and its derivatives to undergo coupling reactions with specific functional groups in the hydrogel matrix, thereby constructing a highly cross-linked three-dimensional porous network structure. GO with good water solubility and dispersion was prepared using the Hummers method. GO was dispersed in water to prepare a uniform GO solution ranging from 1.0 g/L to 3 mg/mL; cross-linking agents such as EDA and L-cysteine (Lcy) were added to the GO solution in proportion and ultrasonically dispersed; the cross-linking conditions were adjusted to trigger cross-linking: first, 0.1 mol/L sulfuric acid was used to wash away the by-products, then the solution was repeatedly washed with ultrapure water to remove impurities, and finally, it was obtained through freeze-drying or critical point drying as GA [3,4].

3.2. Defect engineering

Defect engineering is a method that alters the electronic structure and physical and chemical properties of materials by introducing or controlling defects within them.

During the reduction process of GO, some carbon atoms are oxidized, forming oxygen-containing groups (such as hydroxyl groups and carboxyl groups). Subsequently, these groups are removed through chemical or thermal reduction, leaving vacancies and defects. This significantly increases the specific surface area and active sites of GA, enhancing its specific capacitance and energy density.

By introducing oxygen-containing groups or nitrogen-containing groups (such as amino groups) to modify the chemical activity of graphene, the adsorption capacity of it for target gas molecules can be increased. The chemical activity and adsorption capacity of GA are enhanced [5].

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By bombarding GA with ion beams, vacancies or topological defects are formed, which can regulate the thermal conductivity and mechanical properties of GA. It is possible to precisely control the density of defect types in GA and optimize its mechanical stability [6].

4. Composite modification design

4.1. Introducing pseudo-capacitance

When GA is combined with pseudo-capacitive materials such as metal oxides (RuO_2, MnO_2, etc.), hydroxides ($Ni(OH)_2$, etc.), and MXenes, it exhibits excellent comprehensive performance through the synergistic effect. The pseudo-capacitive materials enhance the specific capacitance and energy density through rapid redox reactions, while the high specific surface area and conductive properties of GA provide support and an efficient conductive network for it, significantly optimizing the electrochemical performance of the composite material. It complements the high energy storage characteristics of the pseudo-capacitive materials, possessing outstanding multi-functional adaptability [7].

Select different types of pseudocapacitive materials based on application requirements: metal oxides, hydroxides or MXenes. By optimizing the composite ratio, the shortcomings of a single material can be compensated, and the synergistic effect can be maximized.

By adopting the strategy of uniformly loading nanoparticles, the graphene sheet layers are prevented from aggregating and the utilization rate of active sites is improved; at the same time, a three-dimensional porous structure is constructed to provide convenient channels for ion transmission, further enhancing the specific surface area and electrochemical activity.

Nanoscale composite of pseudocapacitive materials and GA was achieved through the sol-gel method. Nanoscale structures (nanorods/pebbles) were orientedly grown on the surface of GA by the hydrothermal method, or the electrochemical deposition method was used to ensure the uniform distribution of particles and high loading capacity, precisely regulating the material structure and performance [8].

Compared with pure double-layer capacitive materials, the introduction of pseudocapacitance through the addition of redox components or the optimization of doping strategies has significantly enhanced the energy density and power density of the device, effectively improved the cycle durability of the electrode, optimized the ionic transport efficiency and electronic conductivity of the electrode, and broadened its application prospects in energy storage fields such as supercapacitors and batteries.

4.2. Conductive material composites

The three-dimensional porous network structure of GA can effectively prevent the stacking of graphene layers, thereby maintaining its large specific surface area and outstanding electrical conductivity. It provides a direct electron transmission path for conductive fillers and reduces the resistance of electron transmission. At the same time, the π-π interactions and chemical cross-linking between graphene layers further enhance the stability of the conductive network.

The vacuum impregnation method uses GA as the framework. Through vacuum impregnation, polymers such as epoxy resin and polyurethane are immersed into the pores of the aerogel, and then cured to form a composite material. This ensures that the polymer matrix is uniformly penetrated into the pores of GA, forming a dense composite structure while retaining the three-dimensional network structure of GA. Wang Mu et al. prepared a graphene-pyrrole aerogel/epoxy resin

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composite system using a vacuum-assisted impregnation process. The resulting material has both lightweight porous characteristics and a complete three-dimensional graphene network structure. This structure can provide a fast path for electron migration, thereby significantly enhancing the conductivity of the material [9].

The freeze-drying method is used to prepare GA through freeze-drying technology, and then it is combined with polymer matrices (such as polyvinyl alcohol, polyurethane) to form conductive composite materials. It can retain the three-dimensional porous structure of GA, and by introducing crosslinking agents (such as polyvinyl alcohol), the mechanical strength and conductivity of the aerogel can be further enhanced. Wu/Zou et al. successfully constructed an aerogel with a three-dimensional cross-linked network structure by mixing GO, aramid nanofibers (ANF), and polyaniline nanotubes (PANIT) in the DMSO system, followed by 90 ℃ heat treatment, freeze-drying, and 300 ℃ low-temperature annealing, significantly improving the mechanical strength and electrical conductivity of the material [10].

The in-situ reduction method prepares GA through a chemical reduction process, and then it is combined with polymer matrix such as phenolic resin to form a conductive composite material. This method enables the in-situ generation and composite of GA, simplifying the preparation process while retaining the excellent electrical conductivity of graphene. Zhen Qi et al. ingeniously completed the reduction of GO, in-situ polymerization of PPy, and the composite of MXene/Ca nanofibers in one step through the pyrolo-assisted hydrothermal method, constructing a three-dimensional porous conductive network composed of two-dimensional nanosheets (MXene/rGO), one-dimensional nanofibers (CA), and conductive polymer (PPy) through multiple chemical bonds [11].

5. Multi-level pore structure design

The design of a multi-level pore structure for GA electrode materials is a key method for optimizing their electrochemical performance by regulating the micro-pore distribution. The multi-level pore structure can effectively improve ion transport, increase specific surface area and active sites, thereby significantly enhancing the capacitance performance and cycling stability of the electrode materials.

5.1. Sol-gel method

The liquid crystal properties of GO were regulated by the sol-gel method. Surface active agents (such as Nafion) were introduced to reduce the agglomeration of GO, forming a multi-level pore size structure; or low-temperature pre-freezing combined with room-temperature drying was used to prepare three-dimensional GA with stable pore size and low density. Wang Yong et al. chemically modified the surface of GO with Nafion and prepared graphene oxide hydrogel by reducing with ethylenediamine. Finally, GA was formed through freeze-drying. The prepared GA had pore sizes much smaller than traditional GA (20-100 nm), and exhibited high specific surface area. The high porosity leads to an approximately 40% improvement in the electrochemical capacitance performance compared to traditional aerogels [12].

5.2. Electrostatic self-assembly method

The electrostatic self-assembly method utilizes the electrostatic attraction between molecules or nanoparticles with opposite charges, through electrostatic adsorption and combination, to form an

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initial micro-nano scale composite structure. Subsequently, when the solvent is removed through freeze-drying or supercritical drying, the voids left by the solvent evaporation will form large pore structures, and ultimately, a multi-level pore system of micropores, mesopores and macropores will be coherently constructed. Wang Yankun et al. successfully prepared a three-dimensional layered porous structure of TiO_2/reduced oxidized GA (TGAs) using negatively charged graphene oxide (GO) colloids and positively charged $Ti(OH)_4$ colloids. Its specific capacitance reached 268.3 F/g at 5 mA/g, and still maintained approximately 93% of the initial specific capacitance after 3000 cycles [13].

5.3. Ice template method

The ice templating method involves freezing a suspension containing GO or other precursor materials to enable the directional growth of ice crystals to form a template. During the freezing process, the ice crystals repel solutes, forming a pore framework; subsequently, by drying, the ice crystals are removed, leaving a porous structure. Deng Bo-wen et al. constructed a mixed layer of porous graphene oxide (HGO) and $Ni(OH)_2$ active materials on a nickel foam framework through a one-pot hydrothermal reaction, and used the ice templating method to prepare oriented oxidized graphene aerogel as a filling material, forming an asymmetric solid-state supercapacitor (ASC) device. The specific capacitance reached 479.8 mF/cm^2, and the energy density and power density reached 1.69 Wh/m^2 and 9 $W/m^{2,}$ respectively, demonstrating excellent energy storage capabilities [14].

6. Conclusion

This paper conducts a systematic study on the performance optimization of GA as a supercapacitor electrode material. The core content includes: clarifying the synergistic mechanism of the regulation of GA's physical and chemical properties and electrochemical performance; focusing on analyzing the specific strategies, implementation paths, and performance improvement effects of the three core directions: intrinsic structure optimization, composite modification design, and multi-level pore structure design. In response to the performance bottlenecks of traditional supercapacitor electrode materials and the application limitations of Pure GA, the paper systematically analyzes the performance regulation rules of GA electrode materials, integrates the collaborative optimization ideas of structure design and composite modification, fills the gap in the comparative analysis and collaborative mechanism research of different regulation strategies, and provides a basis in support of the development of high-performance and high-quality products.

The research on stable energy storage materials provides theoretical support and technical references. In the future, we will further explore the synergistic optimization path of multiple modification strategies, precisely control the microstructure and interface characteristics of GA, and achieve the simultaneous improvement of electrochemical performance and mechanical stability; at the same time, we will promote a breakthrough in the low-cost large-scale preparation.

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