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Macroscopic Configurations of Graphene Electrodes for Flexible Supercapacitors: A Comparative Review

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Abstract: Currently, flexible wearable electronic devices are developing rapidly, so there is a need for lightweight, high-efficiency flexible energy storage devices. Graphene has a unique two-dimensional structure and excellent physicochemical properties, so it is a prominent candidate material for flexible supercapacitors. This paper presents a summary of the research advancements in one-dimensional fibers, two-dimensional films, and three-dimensional porous graphene macroscopic configurations, while comparing them based on structural attributes, mechanical flexibility, and electrochemical performance. According to this comparison, this paper discusses the selection strategy for suitable configurations in graphene-based flexible supercapacitor development, offering a guide for electrode design.

Keywords: Graphene; Flexible supercapacitors; Macroscopic configurations; Electrode design.

1. Introduction

The flexible wearable electronic devices, like smart bracelets and electronic skin, pose diverse requirements on their on their energy-storage components [1]. Although supercapacitors are very valued in flexible electronics because they have high power density and long cycle life, electrode materials with a single configuration usually don't meet the varied demands of different application scenarios [2]. To fully exploit the advantages of graphene and meet the structural needs of various flexible devices, researchers have assembled graphene into macroscopic architectures, including one-dimensional (1D) fibers, two-dimensional (2D) films, and three-dimensional (3D) porous graphene [3]. Each of these macro configurations has its own advantages and disadvantages. But existing research has mostly addressed them individually rather than holistically. The absence of a thorough comparison across the three architectures leaves researchers without well-defined criteria for configuration selection in response to particular application needs. In this review, the mechanical and electrochemical properties of the three configurations are compared, and strategies for matching configuration to application scenario are discussed, in order to provide a reference for the selection of electrode configuration of graphene-based flexible supercapacitors for different application requirements.

2. Classification of Graphene Macroscopic Assemblies

Graphene macroscopic assemblies represent macroscale materials constructed by assembling graphene sheets. According to Liu et al. [3], graphene can be assembled into different structural dimensions, namely,1D fiber,2D film, and 3D porous structures—depending on the assembly approach.

2.1. One-dimensional graphene fiber

1D graphene fiber is a macroscale linear material composed of graphene nanosheets aligned along the fiber axis.

At present, the main preparation methods of 1D graphene fibers are wet spinning, confined hydrothermal method and film shrinkage method [4]. Wet spinning is a relatively mature mainstream method. In this process, graphene oxide is dispersed in water or an organic solvent to form a spinning solution, which is then injected into a coagulation bath and solidified into precursor fibers. The precursor fibers are subsequently converted into graphene fibers via chemical reduction or high-temperature thermal reduction[3,5].The confined hydrothermal method seals the graphene oxide (GO) solution in a tubular reaction vessel, and graphene fibers are obtained through subsequent hydrothermal treatment[3,6].The film shrinkage method immerses the graphene film grown by chemical vapor deposition (CVD) in an organic solvent to induce crimping and shrinkage, and then extracted it from the solvent to form graphene fibers[5].

1D graphene fibers exhibit excellent flexibility, allowing them to be bent, knotted, or even woven into textiles. But they generally suffer from a relatively low specific capacitance, and their internal resistance increases with fiber length, which limits the energy storage efficiency of long-fiber devices [2,5].

2.2. Two-dimensional graphene film

2D graphene film is a macroscopic planar material formed by assembling and stacking graphene nanosheets [3].

There are two commonly used preparation methods for 2D graphene film: vacuum filtration and CVD. The vacuum filtration method deposits the graphene colloidal suspension onto a microporous membrane by suction filtration, and after drying and peeling off the membrane, a free-standing graphene film is obtained [3,7]. The CVD method catalyzes the growth of large-area graphene films on a metal substrate, which can then be transferred onto a flexible substrate [2,8,9].

Although 2D graphene films possess excellent in-plane conductivity, their sluggish ion transport across the stacked layers results in limited rate performance [2].

2.3. Three-dimensional porous graphene

3D porous graphene is a framework material with interconnected pore networks assembled from graphene nanosheets [10].

3D porous graphene can be prepared by the CVD template method, the chemical reduction self-assembly method, and

the 3D printing method. In the CVD template method, graphene is grown on the surface of metal foam by high-temperature deposition, and 3D porous graphene is obtained after removal of the metallic substrate [3]. In the chemical reduction self-assembly method, the oxygen-containing functional groups on GO lamellae are removed via reduction, which restores their hydrophobicity and interlayer interactions, thereby promoting cross-linking and assembly of the lamellae into a hydrogel. Subsequent freeze-drying of this hydrogel yields 3D porous graphene [10]. The 3D printing process uses GO as the ink to construct periodic macroscopic structures through programmed control [3,12,13].

3D porous graphene exhibits excellent specific capacitance and rate performance, but its compressive cycling stability is relatively poor, and its performance tends to degrade [10].

3. Comparative Study of Graphene Macrostructures

The mechanical and electrochemical properties of the three configurations are fundamentally governed by their respective structural disparities.

3.1. Structural characteristics

The mechanistic differences among the three configurations arise from their distinct assembly methods for graphene sheets: in 1D graphene fibers, the sheets are aligned along the axial direction; in 2D film graphene, they are stacked in layers; and in 3D porous graphene, they are crosslinked to form an interconnected network.

In 1D graphene fibers, graphene sheets are aligned along the fiber axis, which confers good axial conductivity and tensile strength. But the dense radial accumulation and scarce porosity hinder the penetration of electrolyte ions in the radial direction. Therefore, ions enter the interior mainly through the fiber end face, resulting in low utilization of the effective electrochemical surface area [5,6].

In 2D graphene films, the graphene sheets are arranged in a layer-by-layer manner, forming an in-plane tiling pattern. In the in-plane direction, the π-π conjugated structure provides continuous electron transport pathways, enabling the film to exhibit exceptional conductivity. But interlayer spacing is extremely narrow, ions have to migrate through the confined channels between layers, which makes diffusion paths long and transport resistance high. This contradiction between high in-plane conductivity and sluggish interlayer ion diffusion is the reason for the limited rate capability of 2D graphene films [2,11].

In 3D porous graphene, sheets are crosslinked through face-to-face, side-to-side, and side-to-side configurations to form a multilevel interconnected porous network. This structure allows free ion diffusion in three dimensions and provides a continuous electron conduction path, thus avoiding significant interlayer resistance problems [3,10].

3.2. Mechanical properties

The mechanical properties of the three configurations are mainly determined by their deformation ability and structural stability.

1D graphene fibers are the most flexible of the three configurations because they have an elongated morphology and the lamellae is axially oriented. Zhang et al. [5] reported that graphene fibers exhibit excellent mechanical flexibility and can be easily integrated into fabrics by weaving. When the fiber bends, the outer layer can slide relative to the inner layer to accommodate the bending stress. But this interlaminar slip also means that irreversible structural rearrangement is prone to occur during axial tensile process.

2D graphene films exhibit excellent bending flexibility, allowing repeated bending and folding without structural degradation, while maintaining stable performance over multiple bending cycles. Li et al. [2] reported that graphene film-based flexible supercapacitors exhibit negligible performance degradation when bent to arbitrary angles. This flexibility stems from the fact that its thickness is much smaller than the in-plane size, resulting in negligible stress near the neutral plane when bending. At the same time, the relative sliding between adjacent thin plates can effectively relieve the bending stress.

3D porous graphene is compressible, but irreversible structural damage occurs during compression cycles. Li et al. [2] reported that the network structure of 3D porous graphene synthesized by CVD is bound together by covalent bonds, which endows it with excellent mechanical properties. During the compression process, the hole wall can absorb energy through bending deformation. But the lack of interlayer sliding in covalently bonded networks makes the hole wall connections prone to fracture under repeated compression. The intrinsic brittleness of the carbon skeleton further exacerbates the structural degradation during compression-rebound [10]. 3D porous graphene is not as mechanically flexible as its fiber and film counterparts, and its compressive stability is relatively poor.

3.3. Electrochemical properties

The differences in ion transport pathways and structural stability among the three configurations collectively affect their specific capacitance, rate capability, and cyclability.

3.3.1. Specific capacitance and rate capability

Among the three configurations,3D porous graphene generally exhibits the highest specific capacitance. Its interconnected porous structure offers ample diffusion space and shortened pathways for ion transport, thereby enabling full utilization of the graphene sheet surfaces. Wang et al. [10] reported a specific capacitance of 150 F/g and a charge transfer resistance as low as 1.3Ω at a current density of 0.1A/g. Its rate capability is also the highest among the three configurations. The meso-macroporous network is interconnected, which allows for rapid ion transport, so the electrode can maintain a high capacitive output even at high current densities.

The specific capacitance of the two-dimensional film is the second highest among the three structures. While the in-surface orientation provides a continuous electron conduction pathway, the migration of ions across the layers is greatly restricted. As a result, only the graphene layers on or near the surface can store charges, while the inner active surface is largely unused. As a result, the actual specific capacitance is considerably lower than the theoretical value. Its rate capability is also limited because the compact stacking of layers requires ions to pass through narrow interlayer channels with long diffusion paths and high electrical resistance, which leads to significant capacitance decay at high rates [2].

The specific capacitance of 1D fiber is the lowest among the three configurations. Its dense radial structure hinders ion penetration into the interior, making charge storage mainly dependent on the fiber surface, while the inner layer remains

largely underutilized. In addition, the fiber length leads to internal resistance, which further limits the specific capacitance. Its rate capability is equally poor. Under the condition of high-rate charging and discharging, ions are required to migrate in a very short time. But the dense radial structure makes the diffusion dynamics a rate limiting factor, resulting in a rapid decline in the effective capacitance [5].

3.3.2. Cycling stability

Both 1D fibers and 2D films exhibit high-capacity rates after multiple charge/discharge cycles, thanks to all the volumetric changes in their layered structures during cycles and the good structural stability of the active materials [2, 5]. In contrast, 3D porous graphene exhibits significant capacity decay during cycling. Wang et al. [10] reported that three-dimensional porous graphene exhibits a high initial specific capacitance, but the capacitance gradually decreases at cycling. This is attributed to structural fatigue of the 3D interconnect network during repetitive charge/discharge cycles. Ion repetition/extraction leads to gradual degradation of pore wall structure and active material utilization, and ultimately to capacity decay under long-term cycles.

Based on the above comparison, each of the three configurations provides obvious advantages: 1D fibers exhibit flexibility but have low specific capacitance and poor rate capability; 2D films provide good in-plane conductivity but show limited rate capability; Specific capacitance and rate capability provided by 3D porous graphene, although with poor mechanical flexibility compared to fiber and film counterparts. It's clear that no one configuration excels on all performance metrics.

4. Configurations for Different Application Scenarios

The differing emphases of the three configurations on mechanical flexibility and electrochemical performance lead to their divergent application scenarios.

4.1. Requirements for flexible electrodes in different application scenarios

Flexible wearable electronic devices come in many forms—from smart bracelets and electronic skin to smart textiles—and different devices place different demands on energy storage electrodes. Wearable energy fabrics and smart textiles demand electrodes that combine excellent flexibility and weaveability, the ability to tolerate repeated bending and stretching without structural damage, and full compatibility with textile processing technologies [5]. For planar micro-devices and electronic skin, electrodes must be thin and lightweight, integrate seamlessly with planar fabrication processes, deliver outstanding in-plane conductivity, and be suitable for on-chip integrated energy storage. Compressible high-output power supplies are required to deliver stable performance under frequent compression or high-current charge/discharge cycling, imposing stringent requirements on both rate capability and compression resistance.

4.2. Recommendations on configuration selection

According to the comparative analysis, the guiding principles for selecting configurations in different application scenarios are as follows: The flexibility shown by 1D fibers, which can be integrated into fabrics, is ideal for applications involving repeated bending and deformation, such as art textiles and wearable energy fabrics [5]. But they have low specific capacitance and poor rate capability, so they aren't suitable for high-power-density applications.2D films have a flat structure, is highly compatible with planar processing, and has superior in-plane conductivity, so they are suitable for planar micro-supercapacitors, on-chip integrated energy storage, flexible displays, and renders electronic skin [2]. Nevertheless, their limited rate capability makes them insufficient for high-power-density applications. 3D porous graphene has the best specific capacitance and rate capability, which is perfect for compressible energy storage and high-frequency power supplies with stringent power and energy density requirements [10]. But it has weak mechanical flexibility, so in applications where there are frequent, large-scale bending, we need to carefully select.

5. Conclusion

The choice between these three configurations depends largely on the target application, as each configuration prioritizes different performance aspects. Optical fiber (1D) electrodes typically provide surface capacitance in the range of tens to hundreds of mF/cm ^{2}. They have excellent mechanical flexibility but tend to have a low energy density. 2D films provide similar values of area capacitance and excel in in-plane conductivity; but their rate capabilities are limited. Three-dimensional porous structures can reach specific capacitance of over 150 F/g and show faster ion transport as well as excellent rate performance, although their mechanical compliance remains inadequate. Obviously, no one-dimensional electrode configuration can be used for all requirements at the same time.

Cross-dimensional hybridization combined with structural optimization has been shown to break through these limitations. For example, weaving 1D fibers into 2D fabrics expands the electrode area while providing flexibility, and integrating 3D porous skeletons with 2D films allows one to take advantage of the ion transport advantages of 3D networks as well as the high in-plane conductivity of 2D layers. Despite these advances, significant gaps remain. The trade-off between mechanical flexibility and electrochemical performance has not been described with quantitative models, which seriously hinders application-oriented electrode design. In addition, standardized testing protocols for flexible devices are still missing, which means that the impact of bending period and curvature on performance is not consistently evaluated. This situation complicates tailoring collaborative strategies for specific applications and developing uniform evaluation criteria for flexible systems. Closing these gaps is critical to accelerating the transition of graphene-based flexible supercapacitors from the laboratory to practical applications.

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