

Preparation Technologies and Applications of Graphene-Based Paper Electronic Devices

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Abstract

Graphene-based paper electronic devices achieve the synergistic unification of high performance and low cost by virtue of graphene's unique advantages of ultra-high carrier mobility and excellent flexibility, and the characteristics of paper-based materials such as wide availability and biodegradability as well as a cost only 1/10,000 that of crystalline silicon, becoming a core research direction in the field of green flexible electronics. This paper systematically reviews mainstream preparation technologies such as paper-based laser-induced graphene (LIG), inkjet printing, and chemical vapor deposition (CVD), focusing on the application progress of devices in sensors, flexible devices, energy, and optoelectronics. Finally, it analyzes current technical bottlenecks and looks forward to future optimization directions, providing references for academic research and industrialization transformation in this field.

Keywords

graphene, paper electronic devices, preparation technologies, flexible electronics, application research

1. Introduction

Traditional silicon-based devices with rigid structure, high preparation cost, and non-degradable nature can no longer meet the demands of emerging scenarios such as wearable devices, portable sensing, and smart terminals against the backdrop of the electronics industry transitioning towards flexibility, greenness, and low cost. As a typical representative of two-dimensional carbon materials, graphene significantly enhances device performance and stability with its ultra-high carrier mobility, excellent mechanical flexibility, and compact two-dimensional structure. Combined with paper-based materials' wide availability and biodegradability, the graphene-based paper electronic device formed by the composite of the two successfully achieve the synergy of high performance and low cost.

In recent years, continuous breakthroughs have been made in preparation technologies in this field. LIG technology can directly generate porous graphene structures on paper substrates, inkjet printing enables patterned preparation through functional inks, and CVD effectively addresses the problem of graphene agglomeration. Applications have expanded to multiple fields including sensors, flexible devices, energy, and

optoelectronics. However, the field still faces key challenges such as graphene agglomeration, insufficient interface bonding stability, and poor consistency in large-scale production.

This paper focuses on the core research directions of graphene-based paper electronic devices, systematically reviews the progress, advantages, and disadvantages of mainstream preparation technologies, deeply analyzes the application status in various fields, and discusses existing bottlenecks and future innovation paths, aiming to provide references for academic innovation and industrial implementation in this field.

2. Mainstream Preparation Technologies of Graphene-Based Paper Electronic Devices

2.1 Laser-Induced Graphene (LIG) Technology

Laser-induced graphene (LIG) technology directly generates graphene structures on polymer substrates through a one-step laser direct writing process, featuring advantages of rapidity, mask-free operation, and low-cost customized preparation. For paper substrates, a two-step method is required for its preparation: First, the paper substrate undergoes laser pre-treatment to prevent carbonization or combustion during subsequent laser irradiation. Common methods include the application of flame retardants, such as phosphate compounds [1]. Subsequently, the paper surface is irradiated with a laser of specific wavelength and power. The laser induces localized pyrolysis and graphitization of carbon within the cellulose, forming a three-dimensional porous graphene structure [2]. LingXiao Wang Team addresses the limitations of conventional transfer methods by femtosecond laser processing, achieving the direct fabrication of multifunctional graphene on heterogeneous substrates. This method exhibits multifunctional properties, including a high water contact angle ($\sim 153.4^\circ$), high absorption rate ($\sim 98.8\%$), low sheet resistance ($\sim 82.0 \Omega/\text{sq}$), and reliable temperature-sensing performance with a sensitivity of approximately ($-0.089\% \text{ }^\circ\text{C}$) [3].

2.2 Inkjet Printing Technology

Inkjet printing technology achieves high-resolution patterning of paper electronic devices by precisely controlling the deposition of conductive inks, can realize micron-scale patterning. The reliability of the printing process is ensured by optimizing the ink viscosity and nanomaterial size, such as achieving a Fromm number < 10 for ethanol-based ink and an average MnO nanoflower size of approximately 600 nm [4]. Layer-by-layer printing of multilayer structures, such as Ag/(CaNaNbO/Ag) or graphene/CaNaNbO/graphene, further improves device performance [5]. Martin-Alex Nalepa used nitrogen-doped carboxylated graphene ink, specifically a water-based and additive-free NGA ink, to produce fully inkjet-printed electrodes, offering the advantage of low cost, while the high carboxyl content of the NGA ink provides opportunities for biomolecule immobilization [6].

2.3 Chemical Vapor Deposition (CVD)

CVD technology deposits solid materials on substrate surfaces through gas-phase chemical reactions. The application of CVD on paper substrates faces challenges related to high-temperature tolerance. Existing studies have shown that graphene deposition can be achieved on paper substrates through low-temperature CVD processes or pre-modified paper surfaces [7]. For example, laser-assisted CVD can generate graphene in local areas, avoiding overall high temperatures that damage the cellulose structure [7]. In addition, transfer technology is crucial for integrating CVD-grown graphene into paper devices: laser etching combined with imprint transfer can reduce and transfer pre-patterned graphene oxide (GO) to polyester substrates, but the roughness of paper substrates requires more precise interface contact control [8].

3. Research Progress on Applications of Graphene-Based Paper Electronic Devices

3.1 Sensor Field

3.1.1 Biosensors and Medical Monitoring

In the fields of biomolecule detection and medical monitoring, laser-induced graphene (LIG) or reduced graphene oxide (rGO) are integrated into cellulose paper-based substrates for detecting ions, small molecules, microRNAs, proteins, and cells, thereby enabling biomolecular sensing. For instance, the Nan Zhao research team directly converted lignocellulosic biomass into porous graphene structures using laser-induced graphene technology, preparing low-cost, large-scale producible flexible electronic devices [9]. The electrochemical sensor demonstrates high sensitivity towards target analytes, such as glucose, while the natural creased structure of the paper enables it to maintain stability under flexible conditions [10], making it suitable for wearable applications. This case validates the sustainability of biomass-derived graphene paper-based devices, such as low cost and environmental friendliness, along with their multifunctional integration capabilities, such as the integration of sensing, heating, and energy storage, providing new insights for disposable medical monitoring devices. However, the mechanical strength of the biopaper may limit its application in complex wearable scenarios, such as long-term friction or humid environments.

3.1.2 Environmental Monitoring Sensors

In the field of pollutant detection, the high specific surface area and electrochemical activity of graphene enable efficient detection of heavy metal ions such as lead and mercury, as well as organic pollutants like benzene series. Paper-based graphene sensors respond to pollutant concentrations through changes in resistance or current, with some designs featuring room-temperature recovery capability, making them suitable for field environmental monitoring [11]. Davide Paolini et al. for the first time integrated laser-induced reduced graphene oxide (rGO) into various cellulose substrates, such as filter paper and cotton paper, directly generating a nanostructured conductive network via a one-step laser ablation technique. The sensor demonstrates high sensitivity (with a detection limit reaching the ppb level) and rapid room-temperature recovery toward various environmental pollutants, including heavy metal ions and volatile organic compounds. For example, its response time to NH_3 is less than 10 seconds, and it maintains good stability in humid environments. However, the non-specific adsorption of gas molecules on graphene results in cross-interference and insufficient selectivity, requiring additional functionalization. Meanwhile, the efficiency of laser processing is limited by substrate uniformity, making it challenging to ensure consistent performance in large-area fabrication [12].

In the field of temperature and humidity sensing, the resistance of graphene varies regularly with temperature changes, enabling it to monitor temperature and humidity values. Recent research has utilized laser-induced graphene (LIG) to directly fabricate 3D porous graphene electrodes on polyimide films, resulting in a novel vertical contact architecture that differs from conventional lateral contact structures. This vertical structure increases the contact area between the electrodes and humidity-sensitive materials, significantly enhancing the exposure of current paths. It addresses the low-response issue in traditional LIG sensors caused by partial structural shielding and subsurface current dominance. However, its production relies on non-fully paper-based polyimide substrates, long-term stability is affected by environmental humidity fluctuations, and laser processing parameters require precise control to maintain the uniformity of the porous structure [13].

3.1.3 Physical Sensors

In the field of strain and pressure sensing, paper-based graphene sensors are fabricated via laser induction or printing processes. Leveraging the conductive network of graphene and the mechanical flexibility of the paper substrate [14], they can detect minute strains for applications in flexible electronic skin or structural health monitoring. One research team demonstrated the synthesis of laser-induced graphene (LIG) in a single step by directly irradiating common filter paper with a CO_2 laser, eliminating the need for chemical treatment or complex transfer processes [15]. The innovation lies in converting conventional filter paper into a conductive sensing material while retaining the flexibility and wearable properties of paper. The sensor exhibits high sensitivity to bending and strain (gauge factor > 10), fast response time (< 100 ms), and reusability for over 1,000 cycles. Its performance surpasses that of most paper-based sensors fabricated from graphene paste printing, and it requires no additional conductive material modification, making it suitable for

large-scale production. However, the precision of laser processing is affected by the fibrous structure of paper, making it challenging to achieve micron-scale sensing units.

3.2 Flexible Electronic Devices Field

In the field of paper-based field-effect transistors (PFETs), graphene serves as a conductive or semiconductor layer, combined with dielectric materials such as nanocellulose, enabling flexible transistors with low operating voltage (<3 V) and high mobility (>10 cm²/V·s) [16]. Recent research has innovatively integrated graphene and CsPbBr perovskite quantum dots (PQDs) on paper substrates to form hybrid-structured photodetectors (PDs) [17]. This design combines the excellent electrical properties of graphene with the optical characteristics of perovskite quantum dots, enabling high-performance devices directly on paper substrates. It addresses the challenges posed by the roughness and porosity of paper-based materials, exhibiting high sensitivity and efficient photoelectric conversion capabilities. However, the inherently rough and porous nature of paper substrates (e.g., in the absence of polymer coatings) poses significant challenges to device fabrication, which may limit light absorption efficiency and stability. This is particularly evident in flexible applications, highlighting the potential issues of paper substrates in terms of long-term reliability and large-scale production. The team led by Bohdan Kulyk utilized a CO₂ laser to synthesize laser induced graphene (LIG) on conventional filter paper, enabling the direct fabrication of strain and bending sensors. The innovation lies in systematically optimizing process parameters, such as laser power and scanning speed, to achieve in situ conversion of graphene on the paper substrate. This approach eliminates the complex steps of traditional graphitization methods, thereby creating highly sensitive flexible devices [18]. The sensor demonstrates reliable electrical response, capable of accurately detecting strain and bending deformation, making it suitable for applications in bio monitoring and consumer electronics. Studies indicate that optimizing process parameters can significantly improve conversion efficiency and device stability. However, the graphitization process may lead to sensor fragility and performance degradation, which is particularly pronounced on paper substrates [12].

3.3 Energy and Optoelectronics Field

In the field of photodetectors, the zero bandgap and high carrier mobility of graphene make it suitable for flexible optoelectronic devices, but structural designs such as heterojunctions are required to improve the on/off ratio [19]. Recently, a research team utilized laser-induced graphene (LIG) technology to directly pattern graphene electrodes on paper substrates and constructed a heterojunction with MoSSe nanosheets to achieve self-driven (zero-bias) photoresponse [20]. This fabrication method is straightforward, while the MoSSe/graphene heterojunction covers the visible to near-infrared wavelength range. However, non-uniform distribution of MoSSe nanosheets on graphene leads to performance variability in the device, posing challenges in material homogeneity. Additionally, the roughness of paper limits electrode precision, thereby affecting large-scale integration. In the field of energy storage, graphene paper can serve as lightweight electrode material for applications in supercapacitors or batteries [21]. The team led by Qinglu Li developed a freestanding supercapacitor electrode by preparing nitrogen-doped reduced graphene oxide (N-rGO) via a hydrothermal method and combining it with paper-based materials [22]. The incorporation of nitrogen atoms modulates the electronic structure of graphene, significantly enhancing the electrochemical active sites and electrical conductivity of the material. Additionally, the integration of N-rGO with paper-based substrates eliminates the need for binders typically used in conventional electrode fabrication, simplifying the manufacturing process. The resulting electrode demonstrates excellent specific capacitance and cycling stability. However, the hydrothermal method requires precise control over reaction conditions, such as temperature and duration, and the uniformity of nitrogen doping critically affects the performance, which may pose challenges for industrial-scale production.

4. Challenges and Future Outlook

Although laser-induced graphene (LIG) technology offers the advantage of single-step processing, its application on paper substrates still faces challenges such as unstable electrical conductivity due to insufficient process optimization, and the cost-effectiveness of large-scale production remains unclear. During repeated laser ablation, variations in the surface morphology and pore structure of the paper substrate lead to significant batch-to-batch fluctuations in the conductivity of graphene films, with resistance variations exceeding 30%,

making it difficult to meet device uniformity requirements [23]. Inkjet printing technology relies on post-treatment annealing to enhance electrical conductivity, yet high-temperature treatment tends to damage the paper substrate, causing degradation and deformation, which increases equipment and energy costs [24]. The rheological properties of graphene inks, such as viscosity and surface tension, significantly affect printing precision. Aggregation of nanosheets can easily clog nozzles, resulting in higher circuit breakage rates; for line widths greater than 20 μm , the yield is typically less than 80% [25]. Chemical vapor deposition (CVD) can produce high-quality graphene, but it involves high-temperature reactions and noble-metal catalysts, requiring substantial equipment investment and lacking compatibility with low-cost paper-based materials. When transferred onto paper substrates, interfacial defects such as wrinkles and cracks can reduce carrier mobility by more than 50% [26]. Meanwhile, graphene synthesis relies on strong oxidants or high-temperature reduction, with waste treatment accounting for 63% of the total production cost, while residual metal catalysts (e.g., Ni, Cu) pose environmental risks [27]. Its long-term stability also presents significant challenges. Looking ahead, the development of large-scale, low-cost production methods for graphene-based paper electronic devices, such as inkjet printing and laser-induced graphene technology, will enable mass production of high-quality graphene films. At the same time, efforts will be made to advance multifunctional system integration, for example, by achieving fully integrated devices on paper substrates that incorporate logic gates, memory units, and sensor arrays, enhancing flexibility and low-power performance. In terms of materials, hybrid composites combining graphene with other sustainable materials such as nanocellulose or MXene can be created to optimize electrical conductivity, mechanical strength, and biocompatibility, supporting high-performance applications [28]. The application scope will further expand to wearable devices, Internet of Things (IoT) solutions, biosensors, and energy storage systems, addressing demands for environmental sustainability and ultra-low power consumption. Moreover, the adoption of green manufacturing processes and biodegradable designs will improve overall sustainability and reduce electronic waste. Together, these directions will drive the commercialization of graphene-based paper electronics, unlocking their tremendous potential in flexible electronics [29].

5. Conclusions

Significant breakthroughs have been achieved in the core fabrication technologies of graphene-based paper electronics. The laser-induced graphene (LIG) technique enables the facile preparation of flexible graphene circuits on polyimide or paper substrates through a one-step laser direct-writing process, eliminating the complex post-processing required in conventional ink printing and demonstrating excellent mechanical flexibility and environmental stability. Inkjet printing technology, by optimizing graphene ink formulations—such as through the addition of ethyl cellulose—or by developing high-resolution indirect mask lithography (IML) processes, has realized precise circuit patterning with line widths as low as 20 μm , providing a new pathway for low-cost, high-throughput manufacturing. Advances in low-temperature chemical vapor deposition (CVD) processes ($< 200\text{ }^\circ\text{C}$) have enabled the direct growth of high-quality graphene on plastic substrates, facilitating the integration of transparent flexible devices.

Graphene-based paper electronics have brought transformative impact to the field of flexible electronics. In terms of device performance, the excellent electrical conductivity (2385 S/cm) and thermal conductivity (1324 W/m·K) of graphene paper provide an efficient thermal management solution for high-power flexible electronics, while its bendable nature ensures reliable device operation in dynamic environments. From the perspective of application expansion, this technology has enabled fully integrated paper-based systems: LIG-based paper biosensors have achieved low-cost disposable detection; inkjet-printed graphene circuits drive soft actuators, enabling controllable deformation at low voltages as low as 1V; and multifunctional electronic systems constructed with composite biopaper have demonstrated the feasibility of self-powered wearable platforms. At the industrial ecology level, the integration of scalable manufacturing technologies and green materials has significantly reduced production costs, promoting the large-scale application of paper electronics in the Internet of Things and wearable devices. Nevertheless, long-term environmental stability, manufacturing consistency, and biocompatibility standards remain critical bottlenecks that require further breakthroughs.

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Conflicts of Interest

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