

# Applications of Laser-Induced Graphene Technology

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Laser-induced graphene (LIG) technology has received a large amount of attention from scholars and has a wide range of applications in supercapacitors, batteries, sensors, air filters, water treatment and so on. A variety of preparation methods for graphene was summarized. The effects of laser processing parameters, laser type, precursor materials, and process atmosphere on the properties of the prepared LIG were focused. Two strategies for large-scale production of LIG were briefly described. The wide applications of LIG in the fields of signal sensing, environmental protection, and energy storage were discussed.

laser-induced graphene

signal sensing

environmental protection

energy storage

## 1. Signal Sensing

The excellent physicochemical properties and 3D porous structure of Laser-induced graphene (LIG) make it an ideal candidate for a variety of sensors. The one-step synthesis of patterned graphene has greatly contributed to the development of smart sensors. Researchers have developed a wide variety of LIG-based sensors or sensor devices with modified LIG materials, such as strain sensors, pressure sensors, temperature sensors, humidity sensors, gas sensors, and biochemical sensors, for monitoring various physical and biochemical signals [\[1\]](#)[\[2\]](#)[\[3\]](#)[\[4\]](#)[\[5\]](#)[\[6\]](#).

A resistive strain sensor is one of the key components for converting strain stimuli into detectable electrical signals and has great potential in healthcare monitoring, human-machine interfaces, and soft robotics and so on. [\[7\]](#)[\[8\]](#)[\[9\]](#). Flexible and stretchable sensors that can be attached to the skin are being developed to monitor the health status of individuals. For example, Luo et al. [\[10\]](#) fabricated a flexible strain sensor with an excellent sensitivity coefficient ( $GF \approx 112$ ) by directly generating porous LIG with different patterns on PI films by direct laser writing (DLW). It could identify and monitor different gesture actions and pulses, and could also use finger gestures to control the robotic arm. Dallinger et al. [\[11\]](#) embedded porous LIG or LIG fibers into 50  $\mu\text{m}$  medical-grade polyurethane, to prepare an LIG-based strain sensor that had excellent stretchability up to 100%. Usually, significant improvements for sensor shape variables are accompanied by a decrease in sensitivity [\[12\]](#)[\[13\]](#), therefore, it is a daunting challenge to maintain the balance between sensitivity and strain range of LIG-based strain sensors. Wang et al. [\[3\]](#) investigated a fingerprint-based resistive strain sensor with balanced sensitivity and strain range. Different process parameters were used to control the high-temperature gradient in the irradiated region and to rationalize the geometry of the LIG lines (e.g., depth, width, and density). The sensitivity was greatly improved without sacrificing too much inherent flexibility (7.4-fold increase in sensitivity at 42–50% strain, while the strain range was only

reduced from 60% to 50%). In addition, this balanced sensitivity and strain sensor could be used to monitor human movement. Take the push-up exercise as an example, a reasonable real-time monitoring of the movement is beneficial for regulating training movements and preventing physical injuries [3]. Zhu et al. [14] prepared a kind of LIG with high conductivity and good mechanical properties on the surface of polydimethylsiloxane (PDMS) films using a diode laser. Then, two identical conductive graphene films were joined with silver paint and copper wire. Finally, they were assembled face-to-face to form a pressure sensor with ultra-high sensitivity ( $\sim 480 \text{ kPa}^{-1}$ ) and good cycling stability ( $>4000$  repetitive cycles). These sensors are considered excellent applications for real-time monitoring of human health.

LIG has high thermal conductivity and low heat capacity, making it an ideal material for thermoacoustic sound sources. Moreover, its porous structure has high sensitivity to weak vibrations and is suitable for sound detection. Therefore, Tao et al. [15] developed a wearable intelligent artificial throat based on LIG, which has both sound generation and detection capabilities. This intelligent artificial throat detected simple laryngeal vibrations of different intensities or frequencies, such as humming, coughing, and screaming of mute people, and converted them into controllable sounds. LIG-based acoustic source devices have been developed by La et al. [16] and Tao et al. [17] by taking advantage of the nanopore structure, good electrical conductivity and low heat capacity per unit area of LIG, which would be likely used in consumer electronics, multimedia systems, ultrasound detection, and imaging.

Flexible wearable pressure sensors play an important role in advanced applications, such as electronic skin, real-time physiological signal monitoring, and human-computer interaction. Pressure sensors based on different mechanisms have been widely used in past decades [18][19][20][21]. Inspired by bean sprouts, Tian et al. [19] proposed a flexible self-repairing pressure sensor consisting of polystyrene (PS) microspheres as a microspacer core layer sandwiched between two laser-induced graphene/polyurethane (LIG/PU) films. The porous structure of the LIG provides many cavities for the PS. When subjected to compression, the PS microsphere clusters, which act as spacer layers, modulated the electrical conductance by regulating the degree of physical contact within them. The pressure sensor was highly sensitive, stable, and self-healing. Human arterial pulse monitoring and gait detection were applied, paving the way for scalable production of pressure sensors for human physiological diagnostics and other advanced wearable applications. Li et al. [22] first produced PEEK films with periodic corrugated structures using 3D printing. Porous graphene was then generated on the PEEK films using the LIG technique. Finally, the corrugated LIG (CLIG) films were obtained by transferring them to flexible PDMS films. This corrugated microstructure facilitated the generation of regular cracks during the stretching process and provided initial line contact under normal compression, which effectively improved the sensor performance. Thus, high-performance strain and pressure sensors were successfully prepared based on CLIG films. The CLIG strain sensor had a high resolution of microdeformation ( $1 \mu\text{m}$  or  $0.01\%$ ) and high stability after 15,000 loading cycles. The CLIG pressure sensor had a wide detection range (up to  $500 \text{ kPa}$ ) and high sensitivity ( $678.2 \text{ kPa}^{-1}$ ). It was reported that the CLIG film can measure wrist pulses, swallowing, and even recognize gestures by the subtle differences in muscle contractions.

Temperature sensors are important tools for real-time temperature monitoring in the fields of healthcare and disease diagnosis [23][24]. LIG technology is of great interest because of its low cost, controllability, and scalability.

Due to its high specific surface area, good mechanical stability, electrical and thermal properties, researchers have applied LIG to temperature sensors to improve device performance and reduce preparation costs [25][26]. Kun et al. [27] developed an LIG-based temperature sensor. The sensor was easier to manufacture and operate than conventional thermos-resistance sensors. The accuracy of this sensor was  $\pm 0.15$  °C, which was better than that of the infrared temperature sensor ( $\pm 0.30$  °C). This LIG-based sensor had an accurate and stable temperature response and could accurately measure the surface temperature of the human body. Recently, Chen et al. [28] developed a fast-response, flexible temperature sensor for non-contact human-machine interface using UV laser RGO. Experimental results showed that the temperature sensor had the highest sensitivity ( $0.37\% \text{ } ^\circ\text{C}^{-1}$ ) when the GO concentration was 4 mg/mL and the scan line spacing was 0.12 mm. In addition, this sensor was able to monitor human breathing and contactlessly unlock a combination lock. Han et al. [29] proposed a highly sensitive graphene-based temperature sensor. It consisted of RGO as a temperature-sensitive layer and LIG as an electrode. The sensor exhibited a high sensitivity of  $1.56\% \text{ } ^\circ\text{C}^{-1}$  in the range of 25–45 °C. The sensitivity of the RGO/LIG-based temperature sensor decreased with increasing laser power written to the LIG-based electrode.

Due to its three-dimensional porous structure and ultra-high specific surface area, LIG provides sufficient surface locations for gas–solid interactions, making LIG also promising for gas-sensitive detection devices and gas sensing [30][31][32][33]. Stanford et al. [34] proposed a gas sensor based on LIG. The high surface area and thermal conductivity of LIG ensured fast response times for all studied gases. When different types of gases were inputted, different degrees of resistivity response occurred, due to the difference in thermal conductivity. Gas sensors were also embedded in the cement to form a refractory composite. These sensors were used to determine the composition of various gas mixtures, such as  $\text{N}_2$  and  $\text{CO}_2$ , which are the most abundant gas species in flue gas. Thus, LIG-based embeddable sensors can be integrated into composite materials, making electronic functional building materials possible. Yang et al. [35] developed a LIG flexible gas sensing platform with a self-heating function. This technology used porous graphene as electrodes and highly sensitive nanomaterials (e.g.,  $\text{MoS}_2$  and  $\text{RGO}/\text{MoS}_2$ ) as gas-sensitive materials to monitor gases, biomolecules, and chemicals. The platform was composed of a fine wire sensing region of LIG and a serpentine connection region of  $\text{Ag}/\text{LIG}$ . This serpentine design increased the tensile properties of the sensor to accommodate different bending variations of the body. Additionally, the platform had good selectivity at slightly higher self-heating temperatures, which allowed the sensor to detect  $\text{NO}_2$  at a concentration of 1.2 ppb. In addition, Zhang et al. [36] coated a solution mixed with  $\text{ZnS}/\text{SnO}_2$  nanoparticles on a PI surface, and  $\text{CO}_2$  laser irradiation was then applied to both sides to convert them into LIG. The LIG, as an electrode, and the semiconductor  $\text{ZnS}/\text{SnO}_2$ , in the middle, formed an ultraviolet photodetector. The lateral electrode structure reduced the total thickness of the device, thus minimizing strain and improving the flexibility of the photodetector. Due to the high flexibility and the ultra-thin characteristics of graphene, the device showed great mechanical flexibility. This simple and cheap manufacturing process is expected to be applied to the field of miniaturized flexible electronics.

With the advantages of masklessness, high resolution, low cost, and high throughput, LIG technology has extended its application to biosensors [37][38]. The conventional screen-printed electrode production process is complicated, time-consuming and expensive, which greatly limits the research progress of electrochemical biosensors [39]. Marques et al. [40] developed a bimolecular system with a double working electrode structure based

on LIG technology for the detection of ascorbic acid (AA) and amoxicillin (AMOX). The combination of electrochemical detection with molecularly imprinted polymer (MIP, a common contaminant in aquaculture) technology could identify specific molecules or compounds, therefore, the two conductive LIG working electrodes modified by MIPs achieved high sensitivity and selectivity for the detection of amoxicillin and ascorbic acid. Cardoso et al. [41] prepared a porous multilayer graphene structure with a resistivity of  $102.4 \pm 7.3 \Omega/\text{square}$  on a PI substrate and used it to design a 3-electrode system. This system was applied to a biosensor using MIPs as biometric elements. Torrente-Rodríguez et al. [42] reported a fully integrated, flexible, and wireless graphene-based sensor for monitoring the correlation between sweating and circulating cortisol. It was demonstrated that changes in sweating cortisol could rapidly be determined under acute stress stimuli, revealing the potential of this sensing system to enable dynamic stress monitoring.

From a practical point of view, these above sensors are single-functional and cannot acquire multiple stimuli at the same time. It has been proposed to integrate multiple single-function sensors onto a single substrate, which can detect multiple stimuli simultaneously [43][44]. This fabrication method is expensive and complex. Therefore, a multiparameter sensor that converts each stimulus into an independent signal can overcome the limitations of the above method. Recently, a two-parameter temperature-strain sensor based on a black phosphorus laser-engraved graphene (BP@LEG) heterostructure suitable for electronic skin was investigated by Chhetry et al. [45]. The introduced polystyrene-block-poly(ethylene-ran-butylene)-block-polystyrene polymer matrix had excellent mechanical strength, high cycle stability, and good contact with human skin. The thermal index of the hybrid sensor was up to 8106 K in the temperature range of 25–50 °C, the strain sensitivity GF was 2765 (>19.2%), and the detection limit was 0.023%. It had excellent durability in 18,400 cycles. According to the research, this hybrid sensor could be applied to body temperature measurement and the full range of human induced deformation [45].

## 2. Environmental Protection

Due to its large pore structure, large specific surface area, and excellent surface chemistry, LIG has tremendous potential for many environmental applications, such as anti-pollution systems for desalination and water treatment, air filtration, and generation of anti-bacterial/anti-viral surfaces.

Recently, LIG has been considered as a promising candidate material for air filters due to its good microporous structure and strong adsorption of organic molecules [46]. Stanford et al. [47] investigated a filter with bactericidal effect assembled by LIG. The device utilized the microporous structure of the LIG membrane to trap bacteria and contaminants, and the periodic Joule heating mechanism of the graphene area to generate localized high temperature of ultra-high 300 °C. It could be efficiently sterilized by high temperature without the effect of biocides, exhibiting the function of self-sterilization and self-cleaning. It is expected to be used in public health and disease control in the future [47].

LIG also has many applications in water treatment due to its large specific surface area, tunable surface properties, excellent antifouling activity, and high photothermal conversion efficiency. The sulfur-doped porous LIG prepared by Singh et al. [48] on sulfone polymer substrates produced surface electrochemical and electrical effects, resulting

in highly antimicrobial and excellent antifouling effects. This LIG could be applied to membrane aqueous microfiltration. Polyvinyl alcohol (PVA) is characterized by low toxicity, temperature stability, and high film-forming ability [49]. GO is a two-dimensional nanomaterial that can be used to cover porous or nonporous polymeric membrane supports, resulting in separation membranes with enhanced separation surface properties [50]. Therefore, PVA and GO have widely been used in membrane preparation. Thakur et al. [50][51] loaded PVA and GO onto LIG membranes to prepare LIG-PVA composite membranes and LIG-GO ultrafiltration membranes with controlled performance. These LIG membrane composites had high solute selectivity and permeability comparable to polymeric ultrafiltration membranes. The component of the composite (PVA or GO) could change the surface properties and functionality of the composite membrane, thus determining the degree of antifouling effect. For example, when the amount of graphene oxide was increased, the ultrafiltration membrane increased its rejection of bovine serum albumin to 69%, and antimicrobial resistance from 20 to 99.9%. Under non-filtration conditions, these composite membranes showed 83% less biofilm growth than typical polymeric ultrafiltration membranes, exhibiting excellent antimicrobial properties. These composite membranes had excellent antifouling and antimicrobial properties compared with typical polymeric filtration membranes [50]. Due to the presence of the LIG inner layer, these membranes were also electrically conductive and could effectively purify water by mixed culture of bacteria with applied voltage. Moreover, the presence of PVA greatly improved the mechanical strength of LIG and successfully solved the problem of mechanical strength and insufficient separation performance of conventional LIG-coated membranes. PVA can be used to manufacture highly efficient and environmentally-friendly water purification membranes [50][51].

Since the outbreak of neocoronavirus pneumonia, masks, as filters for both inhaled and exhaled air, have played a critical role in controlling the spread of the epidemic [52]. However, current surgical masks are not self-sterilizing and, therefore, cannot be reused or recycled for other uses. This causes a large amount of medical waste and results in significant economic and environmental stresses. Zhong et al. [53] investigated a medical mask with excellent self-cleaning and photothermal properties. Since the original medical masks were made of thermoplastics with low melting points (e.g., polypropylene), direct laser transfer of graphene would damage the masks. Therefore, they used a dual-mode laser-induced forward transfer method to deposit a few layers of graphene onto the low-melting-point nonwoven masks. Water droplets were observed to have difficulty remaining on the surface of the treated masks, which had excellent superhydrophobic properties [53]. The hydrophobic LIG was able to effectively inactivate coronaviruses through the synergistic effect of photothermal and hydrophobic properties [54]. After 5 min of sunlight exposure, the surface temperature of the graphene-coated masks rapidly increased to over 80 °C, which is sufficient to extinguish most types of viruses, thus allowing the masks to be reusable after sunlight disinfection. On the other hand, with extension of use, bacteria will accumulate on the mask, and it also becomes a critical issue as to whether the mask can continue to be used. To solve this problem, Huang et al. [55] developed an LIG self-reported antimicrobial mask by modulating laser parameters to regulate the surface properties of the LIG, which were used to feed back the safety information of the mask. The wearer's breathing changes the ambient humidity around the device, leading to an inhomogeneous distribution of protons and generating a detectable potential difference or current. Since the accumulation of environmental substances all have a negative impact on the induced potential, this can reflect the number of bacteria or amount of particulate matter that has accumulated

on the mask, and thus provide valid information on the suitability of the mask for continued uses. In addition, for evaporation of 10 wt% of brine, the graphene-coated masks exhibited better desalination performance compared with PI solar vaporizers using laser scribing. Therefore, the graphene-coated masks can be directly recycled for solar desalination [53].

Desalination is a key technology to solve the global water scarcity problem. Existing desalination technologies have a high economic cost. Desalination driven by solar energy is a sustainable means of obtaining fresh water. It is important to seek a carbon material with high solar vapor generation efficiency and stable buoyancy. Li et al. [56] reported a floating graphene membrane with high efficiency and scalability for evaporation of seawater into freshwater using entirely solar energy. The PI films were completely converted into graphene films by one-step laser scribing. The LIG film had a solar energy conversion efficiency of 90% under one solar illumination, and evaporated water at a rate of  $1.37 \text{ kg m}^{-2} \text{ h}^{-1}$ . This high efficiency was due to the efficient water pumping and high optical absorption of the porous structure. Moreover, the authors also desalinated seawater. The experimental results showed that the water with desalination treatment contained fewer electrolytes than the actual seawater or even domestic water. Thus, graphene membranes can indeed be used for seawater desalination. More importantly, these graphene membranes were self-correcting, and floated firmly at the air-water interface, making the process suitable for practical seawater desalination at the ocean surface [56].

Recently, Luo et al. [57] prepared a porous LIG film by laser processing on PI@MS film. This LIG film achieved an evaporation rate of  $1.31 \text{ kg m}^{-2} \text{ h}^{-1}$  and a photothermal conversion efficiency of 85.4% at 1 solar light intensity. The evaporation performance of the LIG evaporator in high-concentration NaCl solution (10 wt%) could be maintained for up to 12 h, showing excellent stability and salt tolerance. Inspired by the structure of forests that use sunlight efficiently in nature, Peng et al. [58] obtained an LIG with the target structure, named the forest-like LIG, from polybenzoxazine, using a one-step laser etching process. This prepared LIG film had excellent light absorption and photothermal properties (the average absorption was 99.0% under 1 solar irradiation and the equilibrium temperature was about  $90.7 \text{ }^\circ\text{C}$ ). Then, based on this excellent photothermal material, a flexible light-driven driver was designed with short response time and high average speed. Also a salt-resistant bilayer interface solar desalination membrane was designed using the superabsorbent and superhydrophobic properties of the forest-like LIG. Huang et al. [59] used  $\text{CO}_2$  laser irradiation of PI-coated wood to prepare a porous LIG with pore sizes ranging from hundreds of nanometers to tens of microns at its top and bottom. The middle part of the wood was left intact to maintain microchannels for water transport and low thermal conductivity. This structure was used as a small solar water treatment device. The device had a superhydrophobic LIG top layer for solar driven desalination and a superhydrophilic LIG bottom layer for effective repulsion of lipophilic organics and antifouling. The LIG technology retained the low thermal conductivity of the wood, and the LIG was a broadband absorbent material with a high evaporation rate.

### 3. Energy Storage

Energy storage devices play an important role in energy storage and supply in smart wearable electronics, such as electronic skin and sensors. Due to the excellent electrical conductivity and precise control of the preparation

process of LIG, its application has been extended from supercapacitors and micro-supercapacitors to a wide range of energy storage devices, such as lithium metal batteries and fuel cells [5][57][60][61][62][63].

To meet the needs of portable and wearable electronic devices, and modern microelectronic systems, miniature energy storage devices are receiving increasing attention. Micro-supercapacitors (MSCs) are promising energy storage devices with fast response to electrochemical processes, high power density, and cyclic stability [6][57][58][59]. The electrode material of such devices is one of the key factors determining their performance. Lin et al. [37] designed an LIG-MSC in which the LIG was used as the active electrode and current collector. The LIG-MSCs using LIG electrodes had a high specific surface area capacitance and the cyclic voltammograms were pseudo-rectangular at different laser powers, indicating their good double-layer capacitance. The above study further illustrates that LIG is particularly suitable for energy storage devices with good electrochemical properties. In addition, MSCs fabricated with LIG can be stacked to improve the electrochemical performance for commercial applications. Peng et al. [64] extended an approach based on the previous fabrication of MSCs using LIG. Two flexible solid electrolyte supercapacitors, vertically stacked graphene supercapacitors and in-plane graphene micro-supercapacitors, were fabricated by sandwiching the solid polymeric electrolyte PVA and  $\text{H}_2\text{SO}_4$  between two LIG layers. They both had high electrochemical performance, cyclability, and flexibility. The area capacitance of these devices was up to  $9 \text{ mF/cm}^2$  when the discharge current density was  $0.02 \text{ mA/cm}^2$ , which is more than twice as high as when using aqueous solutions [37]. The performance stability of individual LIG-SCs under mechanical bending were also tested. The results showed that the performance of bent LIG-SCs was almost the same as that of planar LIG-SCs. This indicated that repeated bending had little effect on the electrochemical performance, further illustrating the unique advantages of 2D LIGs for microcapacitor assembly [64]. To further improve the performance of LIG-SCs, two commonly used strategies are heteroatom doping (e.g., B, N, P, and S) [65] and pseudocapacitive material loading (e.g.,  $\text{Co}_3\text{O}_4$  and  $\text{MnS}_2$ ) [12][63]. The above processes are expensive in raw materials and complex in process, which inevitably limits large-scale economic production. It was shown that LIG could convert electrical energy into thermal energy to perform Joule heating. This Joule heating method was able to reduce the density of defect sites in the graphene structure, which resulted in improved physical and chemical properties [66][67][68]. He et al. [69] introduced Joule heating as a key in situ processing strategy combined with laser-induced assembly of graphene paper-based MSCs (LIGP-MSC) to achieve enhanced capacitance.

Although the laser-induced technique has higher fabrication efficiency compared with the conventional method, it is still far from meeting the requirements of rapid mass production and industrialization. Yuan et al. [70] fabricated flexible LIG/ $\text{MnO}_2$  MSCs by the spatially shaped femtosecond laser (SSFL) method. It was reported that the SSFL technique can directly complete the processing of electronic devices in batch. The MSC fabricated by this technology had ultra-high energy density ( $0.23 \text{ Wh cm}^{-3}$ ), ultra-small time constant (0.01 ms), excellent specific capacitance ( $128 \text{ mF cm}^{-2}$  and  $426.7 \text{ F cm}^{-3}$ ), and long-term cycling performance, overcoming the current limitations of low efficiency fabrication and low energy density of micro-supercapacitors. Recently, Le et al. [71] prepared high-quality graphene on fallen leaves using a femtosecond UV laser. The spatial resolution of the laser pattern was improved due to the ultra-short pulse duration that reduced the thermal diffusion in the heat-affected region. The final obtained femtosecond laser-induced graphene (FsLIG) microelectrodes had excellent electrical conductivity with a resistance of up to  $23.3 \text{ } \Omega \text{ sq}^{-1}$ . Finally, flexible FsLIG-MSCs were also prepared using FsLIG.

After 50,000 cycles, the capacitance of the device retained about 99% of the initial capacitance with good electrochemical stability. The maximum surface energy density of the device was  $1.204 \mu\text{Wh cm}^{-2}$  and the power density was  $324.39 \mu\text{W cm}^{-2}$ , which is comparable to other advanced SCs and MSCs [72][73][74].

With the growth of demand for Li-ion batteries, there is a need to continuously improve the coulomb efficiency, lifetime, stability, and safety factor of batteries. The high porosity, high electrochemical stability, and excellent electronic conductivity of LIG could meet the basic requirements for an ideal collector. A large specific surface area and a good electronic conductivity would reduce the local current density during lithium deposition and slow down the high-volume change of lithium during cycling. Yi et al. [75] laser irradiated PI film on copper foil to obtain an array structure consisting of copper foil, PI column, and LIG. Among them, the copper foil acted as a conductive channel for lateral electrons, the PI pillar relieved the stress generated by Li deposition, and the LIG film acted as a nucleation site for Li. Moreover, it was concluded that the large number of defects and heteroatoms in the LIG lowered the nucleation potential barrier of Li, thus promoting stable cycling of the Li-metal anode. As a result, this approach allowed the Li-metal anode to achieve a high coulombic efficiency of 99% at  $1 \text{ mA cm}^{-2}$  and a high cycle life of 400 h. Yang et al. [76] proposed a novel CuS/Cu<sub>2</sub>S aqueous solution cell. They used a composite of LIG mixed with sulfur as the cathode of this cell. The large specific surface area of LIG sheets (up to  $294.69 \text{ m}^2 \text{ g}^{-1}$ ) provided a large number of surface-active sites. The characterization revealed that the LIG was multilayered, multi-doped and defective, which facilitated the sulfur loading. Due to the synergistic effect of the new redox couple and LIG, when the current density was  $0.8 \text{ A g}^{-1}$ , the discharge capacity of the battery reached  $1654.7 \text{ mAh g}^{-1}$  at the initial cycle. Moreover, 91.2% of the reversible capacity was retained after 328 cycles.

Proton exchange membrane fuel cells have received the most attention as fuel cells in recent decades. However, production cost and lifetime limit their commercialization applications [77][78]. Tiliakos et al. [79] transferred non-sacrificial Pt-coated LIG directly onto Nafion membranes as a microporous layer in proton exchange membrane fuel cells by an easily scalable and inexpensive low-temperature decal method. Due to the more favorable porosity, conductivity, hydrophobicity, and material transport characteristics of LIG, the electrochemical active area of the LIG-based membrane-electrode assembly (MEA) was relatively high at the optimum test conditions of  $80 \text{ }^\circ\text{C}$  and 80% RH. The LIG-based fuel cell effectively improved the power performance by 20%, compared with the reference MEA with the same catalyst loading [79]. Recently, Kong et al. [80] developed an integrated flexible enzyme biofuel cell (EBFC) based on nitrogen-doped graphene obtained by laser scribing method. EBFC is a new type of green energy source and is considered as an alternative power source for wearable devices. Importantly, due to the large specific surface area and good electrical conductivity of laser-scribed N-doped graphene electrodes, electron transfer can be achieved directly without electron mediators. Furthermore, after 20 days of storage, the open-circuit voltage of EBFC still maintained 78% of the initial value, and there was little change after 100 times of bending, indicating that the fuel cell had good stability and mechanical robustness.

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