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# Review on Functionalization of Laser-Induced Graphene

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**Abstract:** Owing to carbon materials' diverse functionalization and versatility, the design and synthesis of carbon-based three-dimensional porous structures have become important foundational research topics across various fields. Among the various methods for producing porous carbon structures, laser-induced graphene (LIG) has garnered attention because of its large surface area, controllable structure, excellent electrical conductivity, scalability, and eco-friendly synthesis process. In addition, recent research results have reported more novel functionalities by advancing further from the unique characteristics of LIG through functionalization or compounding of LIG, making it an attractive material for various applications in electronic devices, sensing, catalysis, and energy storage. This review aims to update the research trends in LIG and its functionalization, providing insights to inspire more interesting studies on functional LIG to expand its potential applications ultimately. Starting with the synthesis method and material characteristics of LIG, we introduce the functionalization of LIG, which is classified into surface modification, heteroatom doping, and hybridization based on the interaction mechanism. Finally, we summarize and discuss the prospects of LIG and its functionalization.

**Keywords:** Functionalization, Laser-induced graphene, Surface modification, Heteroatom-doping, Hybridization

## 1. INTRODUCTION

Recently, the importance of nanomaterials has been highlighted because of the need for high-speed electronics and new renewable energy systems in response to industrial advancements. Among various nanomaterials, carbon-based nanomaterials are attractive because of their excellent physical, chemical, and electrical properties. These make them promising candidates for alpha materials that means innovative new materials that represent an era, such as iron and silicon. Carbon nanomaterials have been developed as a broad series of carbon allotropes consisting of zero-dimensional fullerenes and quantum dots, one-dimensional carbon nanotubes (CNTs), two-dimensional graphene, and three-dimensional nanohorns. In particular, graphene, which has a two-dimensional planar structure of carbon atoms arranged in a honeycomb lattice, has attracted considerable research attention owing to its remarkable properties, such as high electron mobility (200,000 $cm^2/V \cdot s$) [1,2], thermal conductivity (~5,000 $W/m \cdot K$) [3], Young's modulus (~1.0 Tpa) [4], and optical transmittance (97.7%) [5]. Currently, the predominant techniques for the mass production of graphene comprise a bottom-up method utilizing chemical vapor deposition (CVD graphene) and a top-down method involving exfoliation flakes from bulk crystals (denoted as flake graphene). CVD graphene offers superior crystallinity [6], scalability [7], and patterning capability [8], but requires complex synthesis processes, including high reaction temperature, long reaction time, and precise regulation of airflow [9]. Flake graphene offers a large specific surface area and ease of production [10], but exhibits limitations in terms

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of poor crystallinity, dispersibility, and uniformity. Recently, laser-induced graphene (LIG) has been proposed as a promising material that can overcome the shortcomings of CVD graphene and flake graphene, while maintaining their advantages. LIG is a material produced through the carbonization and graphitization of a carbon-based polymer thin film using a laser-induced photothermal reaction, enabling synthesis within a few seconds, and has a three-dimensional porous structure [11]. Hence, LIG offers significant advantages over traditional graphene materials, including short synthesis time, vacuum-less process, easy patterning, and high production process efficiency. In addition, it is possible to implement various performances by selectively imparting functional groups only to the necessary areas, thereby inducing differentiated physical properties that existing graphene materials do not have. However, despite its promising properties, there are few studies on the functionalization of LIG compared to other carbon-based materials, which limits the potential of LIG within the field of laboratory research [12-14]. Therefore, it is essential to explore the functionalization possibilities to expand the utility of LIG applications and related research should be actively conducted. This review focuses on the latest advances in LIG functionalization (Fig. 1). Starting with the synthesis method and characteristics of LIG, its functionalization is introduced by classifying it into surface modification, heteroatom doping, and hybridization. Finally, the functionalization of the LIG is briefly discussed.

## 2. SYNTHESIS OF LASER-INDUCED GRAPHENE

Extensive research has been conducted on the synthesis of LIG using a laser with varying wavelengths [11-18]. Among them, the synthesis of LIG using $CO_2$ or UV laser is representative, with slight differences in the synthesis mechanism. The main difference between the LIG synthesis mechanisms of $CO_2$ and UV laser is the primary energy absorption process: $CO_2$ lasers primarily heat up the substrate to high temperatures to induce decomposition and carbon atom release, while UV lasers directly break the C-C bonds in the substrate through rapid lattice vibrations. UV laser, which has

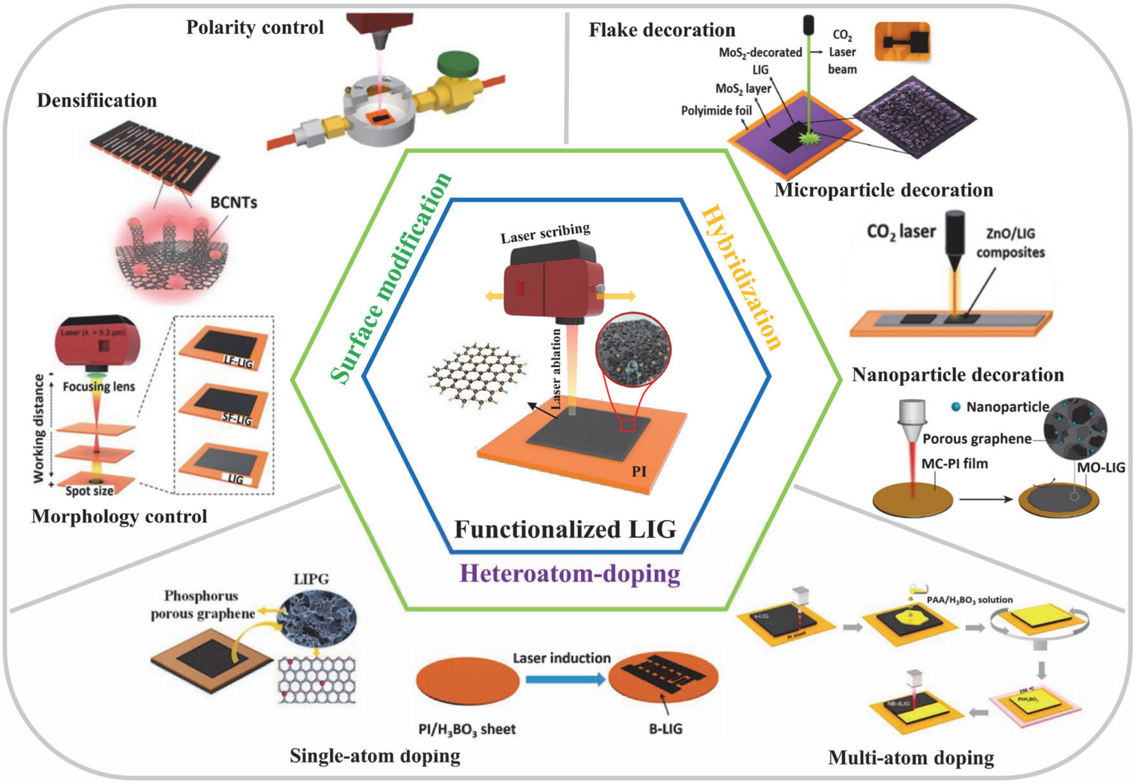

**Fig. 1.** Various approaches to obtain functionalized LIG.

a higher energy level, is absorbed more readily by the carbon atoms in polyimide (PI). The absorbed energy causes rapid lattice vibrations, leading to the release of carbon atoms, such as CO and $CO_2$, as gases from the PI. In contrast, the $CO_2$ laser, with lower photon energy per unit, is less effective in directly breaking C-C bonds in PI. $CO_2$ laser primarily heats up PI to a temperature above 800℃ which causes the decomposition of PI and the release of carbon atoms. Finally, these carbon atoms recombine to form graphene structures due to the high temperature created by the laser [19-24]. From a laser processing perspective, laser intensity, scanning speed, and focal distance are key factors that influence the interaction between laser energy and PI [25]. The laser intensity is a critical factor in determining the amount of delivered energy to PI, an essential parameter for achieving efficient LIG synthesis. Higher laser intensities induce stronger lattice vibrations and promote the emission of carbon atoms, thereby facilitating the formation of graphene structures. Insufficient laser intensity leads to incomplete forms, like amorphous carbon, while excessively high laser intensities can interfere with LIG formation by damaging PI or inducing overheating. The scan speed refers to the duration of laser irradiation on PI, which can significantly affect the synthesis of LIG. The longer interaction times promote the release of more carbon atoms, which leads to the formation of graphene structures. However, excessively long interaction times can cause PI degradation, potentially hindering the formation of LIG. The focal length is another important parameter that affects the laser-PI interaction during LIG synthesis. This determines the spot size of the laser beam on PI, thus affecting the delivered energy

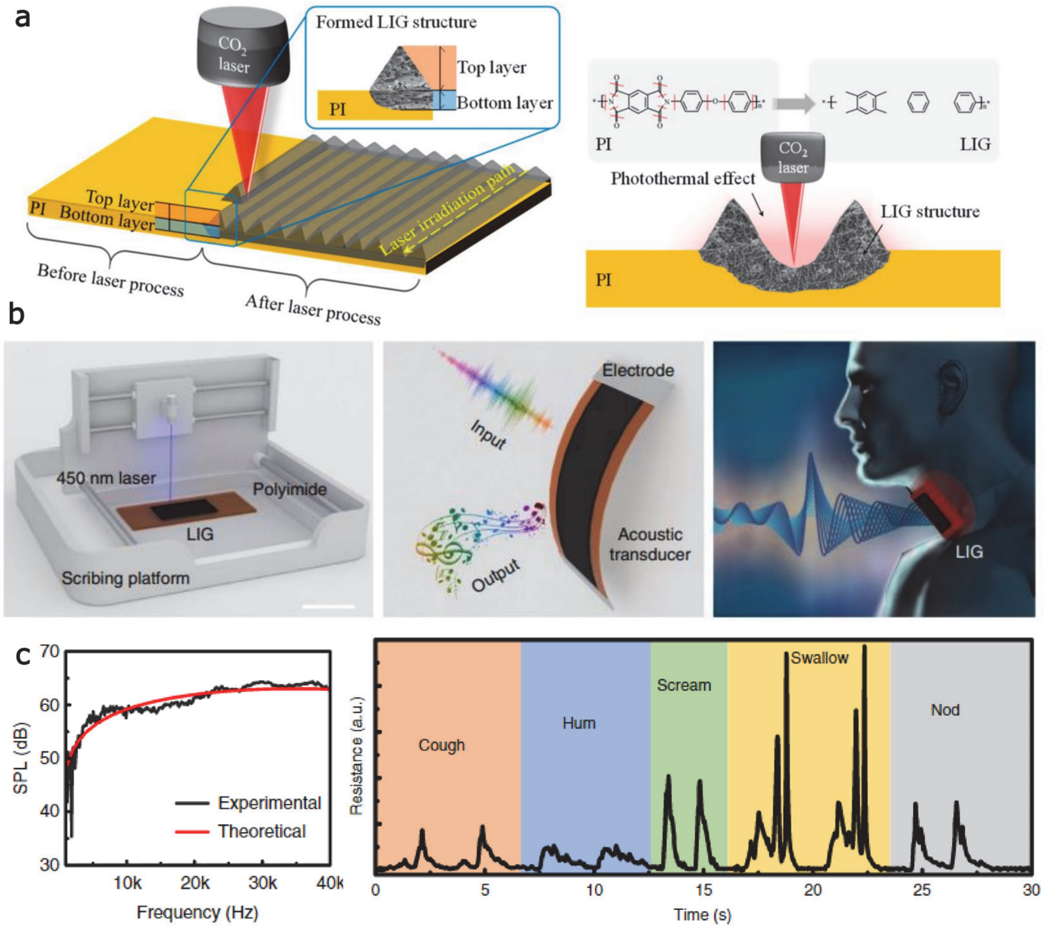

**Fig. 2.** Schematic illustration of (a) fabrication process of LIG with the photothermal effect of laser irradiation on PI film (reprinted with permission from [26] Copyright 2022, Yen, Y. H. et al.), (b) fabrication process of LIG and capabilities of the artificial throat in a single device using LIG, and (c) sound emission performance and responses towards various kinds of throat vibrations of the artificial throat (reprinted with permission from [32] Copyright 2017, Tao, LQ. et al.).

density to PI. A shorter focal length results in smaller spot size and higher energy density, which can promote LIG formation. However, the optimal focal length varies depending on the specific laser parameters and PI. Therefore, careful optimization of these laser process parameters is essential to achieve high-quality LIG. Lin, J. et al. reported a facile and scalable approach for synthesizing and patterning 3D porous graphene on PI films under ambient atmospheric conditions using commercial $CO_2$ lasers [11]. The photothermal effect with localized high temperature and pressure produced by laser irradiation converts a PI film into LIG, as depicted in Fig. 2(a) [26]. The quality of the synthesized LIG was evaluated using the following analytical methods. The Raman spectrum of LIG exhibits three prominent peaks: D (induced by defects or bent $sp^2$ carbon bonds, ~1,350 $cm^{-1}$), G (first-order zone-boundary phonons, ~1,580 $cm^{-1}$), and 2D (second-order zone-boundary phonons, ~2,700 $cm^{-1}$) [11,27]. 2D bands are typically found in graphite composed of randomly stacked graphene layers [28,29]. In addition, the $I_D/I_G$ ratio indicates the degree of disorder or defectiveness in LIG. The X-Ray diffraction analysis (XRD) analysis further confirmed the degree of graphitization through the peak intensity at $2\theta=25.9°$ [11]. Also, the asymmetry observed in the (002) peak can be attributed to the presence of defects distributed in the hexagonal graphene layer. Transmission electron microscopy (TEM) analysis revealed the unusual super-polycrystalline properties of the LIG flakes with disordered grain boundaries [11]. Unlike the traditional honeycomb structure of graphene, the curvature of the LIG layer with a porous structure can be explained by the rich pentagon-heptagon pairs [30]. The important point is that these defects may provide sites for the functionalization of LIG for catalytic applications [31]. Owing to these unique material characteristics and production efficiency of LIG, researchers are conducting extensive research to expand the field of LIG applications [32-40]. As shown in Fig. 2(b and c), Tao, LQ. et al. reported an intelligent artificial throat using LIG that can generate and detect sound in a single device [32]. LIG realizes the functional integration of sound emission and detection owing to its superior thermoacoustic and piezoresistive properties. In conclusion, LIG is an emerging material with considerable attention from researchers. Ongoing research is continuously developing its applications in various fields based on its unique properties and innovative manufacturing methods.

## 3. FUNCTIONALIZATION OF LASER-INDUCED GRAPHENE

Functionalization of materials aims to enhance their performance and expand their applications by modifying their chemical, physical, or electronic properties, through the introduction of functional molecules within the surface or bulk structures, as well as by modifying the surface structure. Diverse research on the functionalization of carbon-based materials has been actively conducted [41-47]. Owing to its exceptional electrical conductivity, high specific surface area, and distinctive porous and three-dimensional structure, LIG has become an appealing material for various applications, including energy storage, sensing, and catalysis. However, despite its advantageous properties, LIG also has some limitations that can hinder its practical utilization in certain applications, such as its limited chemical reactivity, hydrophobic surface, and relatively low mechanical strength. By controlling the laser parameters, the structure and properties of the LIG can be dramatically functionalized, providing that the LIG can overcome these limitations and enhance its properties and functionality for various applications. There are various approaches to obtaining functionalized LIG, which involve techniques such as modifying the surface structure, doping with heteroatoms, and hybridizing it with functional materials [48-67]. LIG obtained through these methods can improve the chemical, physical, and electrical properties, enabling increased use of LIG in a variety of fields. Functionalization of LIG has been introduced by classifying it into surface modification, heteroatom doping, and hybridization.

### 3.1 Surface modification

One approach for functionalizing LIG is surface modification [48-53]. Several surface modification techniques have been reported by introducing the results of other carbon materials, such as physical treatments, structure control, and three-dimensional structuring. Physical treatments, such as plasma and UV irradiation, can modify the surface of LIG by introducing oxygen-containing functional groups such as carboxyl, hydroxyl, and carbonyl groups. These can enhance the surface energy and wettability of the material and improve its biocompatibility and chemical reactivity [48]. Structural control involves transforming the structure of LIG into a porous or fibrous form to enhance certain properties needed for applications [49,50]. As another effective method for controlling the structure, coating LIG with functional materials, such as CNTs and polymers, can improve the mechanical and thermal stability of LIG and increase its ability to selectively and sensitively detect target analytes [51,52]. Finally, three-dimensional structuring, using a laser cutter or engraver to cut and shape the LIG material into the desired 3D structure, can be achieved by controlling the laser beam intensity and duration to selectively remove the material in a controlled manner [53]. To expand the range of properties of LIG, Li. et al. demonstrated the fabrication of LIG with different surface morphologies and surface chemistries under selected gas atmospheres, which specifically tuned the hydrophobicity and hydrophilicity of the LIG surfaces, as shown in Fig. 3(a) [48]. The water contact angle of LIG synthesized in a chamber under specific gas atmospheres ranged from 0° (superhydrophilic/under air or $O_2$) to over 150° (superhydrophobic/under Ar, $H_2$, and $SF_6$), while maintaining stability under ambient air conditions. The key parameters needed to form diverse surface morphologies of LIG by tuning the laser irradiation energy enable the fabrication of vertically aligned fibrous LIG forests were studied [49]. To expand this concept further, K. H. Choi. et al. reported the triboelectric properties of structurally controlled LIG to clarify the key factors for improving the energy-harvesting performance [Fig. 3(b)] [50]. Overall, the results showed that LIG with a fibrous structure is the morphology more suitable than a porous structure as the active layer of a triboelectric nanogenerator

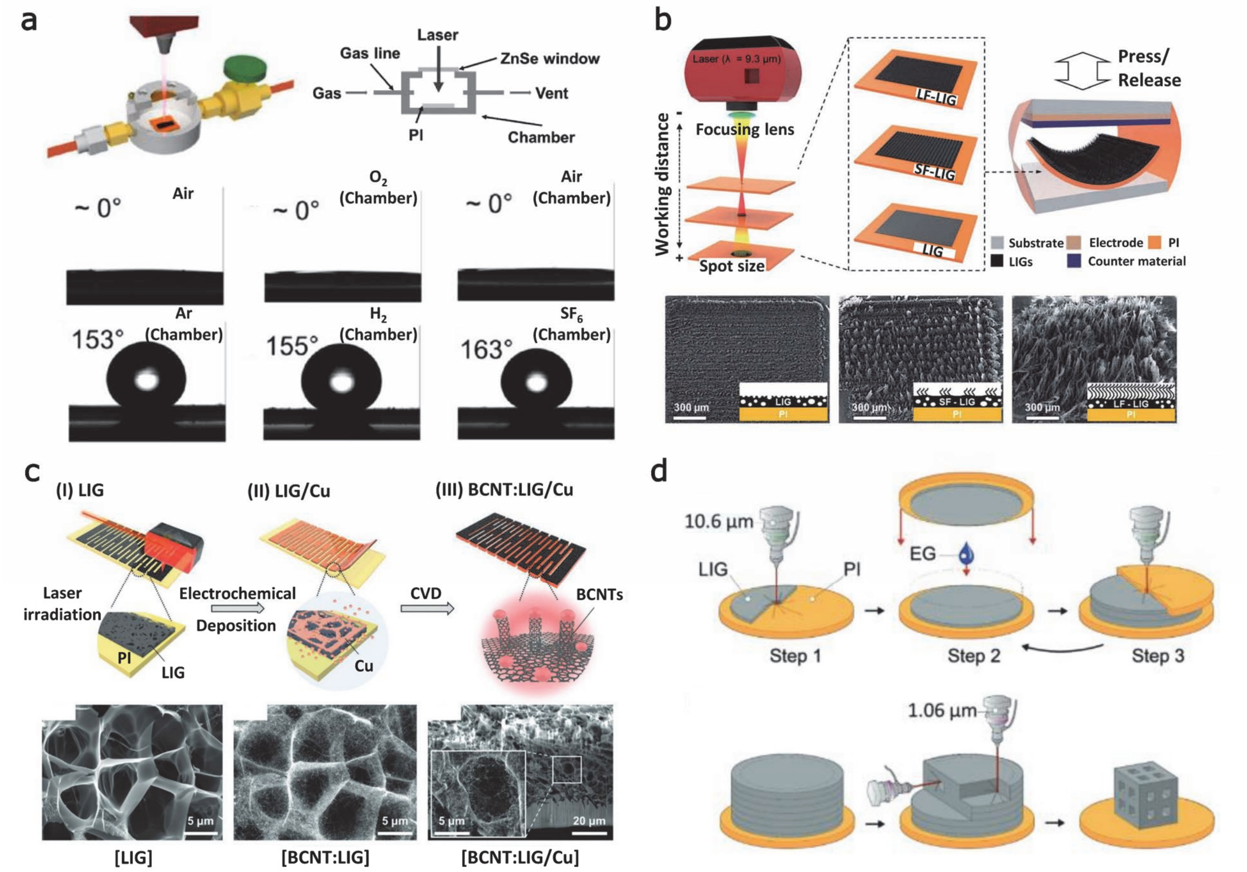

**Fig. 3.** Schematic of (a) synthesis of LIG in a chamber with controlled atmosphere and contact angles of the synthesized LIGs (reprinted with permission from [48]. Copyright 2017, WILEY-VCH), (b) morphology controllable LIG directly synthesized by the working distance modulation method and fabricated press/release-type LIG based TENGs (reprinted with permission from [50]. Copyright 2020, Royal Society of Chemistry), (c) fabrication process of the BCNT:LIG/Cu composite and related scanning electron microscopy (SEM) images (reprinted with permission from [51]. Copyright 2021, WILEY-VCH), and (d) synthesis process of 3D-printed LIG (reprinted with permission from [53]. Copyright 2018, WILEY-VCH).

(TENG). The porous structure of LIG makes it an appealing property for energy storage devices; however, the low packing density resulting from the macroscopic voids inevitably generated during the laser irradiation process hinder the enhancement of device performance. As shown in Fig. 3(c), S. K. Hyeong. et al. reported the fabrication of compacted laser-induced composite electrodes for supercapacitors, which involved filling unused voids with electrochemically active bamboo-like CNTs (BCNTs) to overcome these limitations [51]. As a result, the fabricated composite film (denoted as BCNT:LIG/Cu) provided an energy density approximately 10 times higher than that of LIG-based supercapacitors. In pursuit of high volumetric performance, D. X. Luong. et al. developed a laminated object manufacturing (LOM) technique for the direct fabrication of 3D LIG forms (GFs) and subtractive laser-milling techniques to improve 3D structures [Fig. 3(d)] [53]. The various 3D foam structures of LIG formed by combining these two techniques showed good mechanical strength and electrical conductivity, demonstrating their potential for applications in diverse fields, such as energy storage and flexible electronic sensors.

### 3.2 Heteroatom-doping

Another method of functionalizing LIG is doping heteroatoms or molecules, such as N, B, and S into the LIG lattice during the synthesis process [54-58]. The most widely used method for the formation of heteroatom-doped LIG is the incorporation of a heteroatom-containing precursor during LIG synthesis. Incorporating dopants into a material can modify its electronic characteristics, such as its electrical conductivity or work function, allowing it to be used in electronic applications, such as transistors and sensors. Peng. et al. illustrated the facile synthesis of B-doped LIG under ambient air conditions by the laser irradiation of PI sheets containing boric acid, as depicted in Fig. 4(a) [54]. The B-doped LIG exhibited high-quality and good electrochemical properties, while the cyclability and flexibility of the device

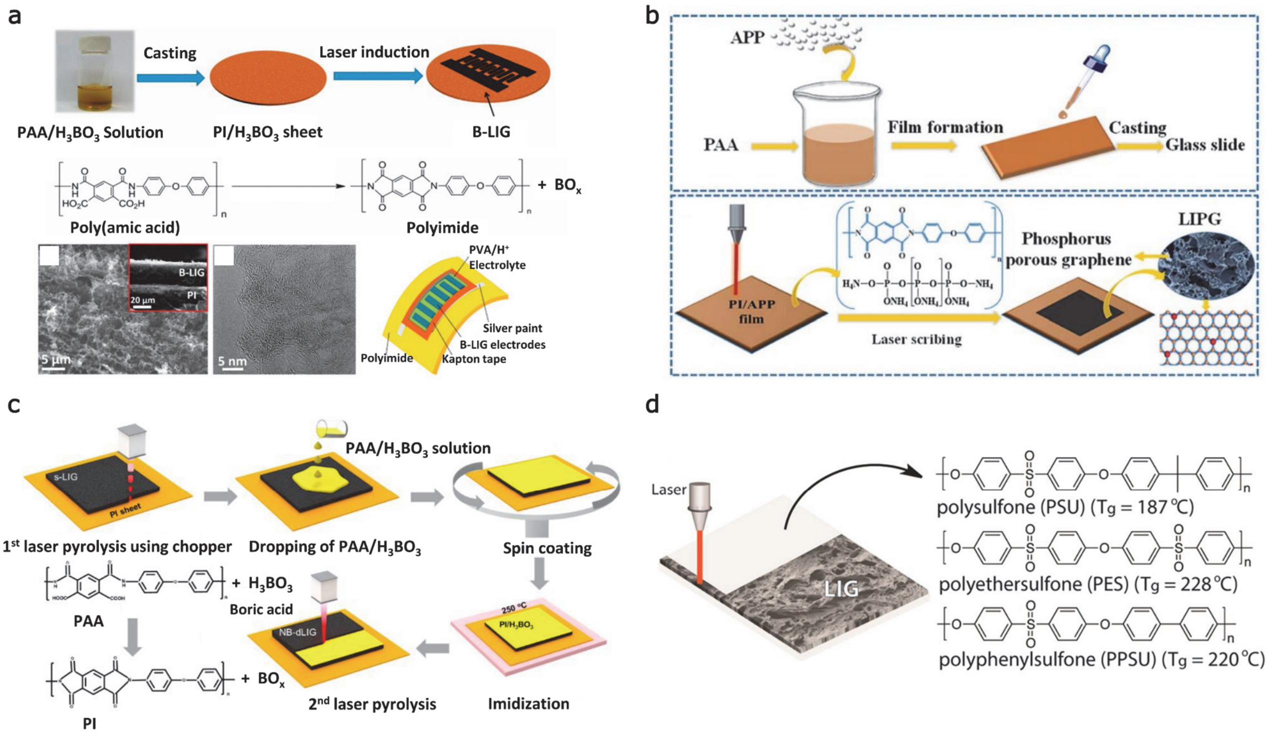

**Fig. 4.** Schematic of (a) synthesis and reaction mechanisms of B-doped LIG and the fabricated MSC device, along with SEM and TEM images (reprinted with permission from [54]. Copyright 2015, American Chemical Society), (b) formation of phosphorous-doped LIG (reprinted with permission from [56]. Copyright 2020, Yang. et al.), (c) synthesis process of N and B co-doped LIG (reprinted with permission from [57]. Copyright 2021, Elsevier), and (d) synthesis of S-doped LIG via laser irradiation on polysulfone-class polymers (reprinted with permission from [58]. Copyright 2018, American Chemical Society).

were well maintained. Moreover, Han. et al. developed a facile and scalable technique for the in situ synthesis of N-doped LIG using a precursor composite approach in conjunction with simple laser irradiation [55]. As shown in Fig. 4(b), Yang. et al. reported a facile and rapid approach for the preparation of P-doped LIG by $CO_2$ laser irradiation of a PI film mixed with ammonium polyphosphate (APP) under ambient air conditions [56]. The P-doped LIG, which exhibited a high specific capacitance, was applied to supercapacitors for further study. To expand the doping range (multiple heteroatom-doping) of LIG, a facile and versatile approach to the fabrication of N and B co-doped and simultaneously densified LIG based on a duplicate laser pyrolysis method was introduced for supercapacitor applications [Fig. 4(c)] [57]. As a result, N and B co-doped LIG, which benefits from the synergistic effect between the two heteroatoms, exhibits significantly enhanced electrochemical performance compared to N-doped LIG and pure LIG without any dopants. Singh, SP. et al. demonstrated a solvent-free and reagent-free method to produce conformal S-doped LIG directly on a polysulfone-polymer class in a single step, which can be seen in Fig. 4(d) [58]. The S-doped LIG was demonstrated to have various potential applications, including its use as a flexible electrode that can generate $H_2O_2$ more efficiently, as well as antifouling surfaces and hybrid membrane-LIG porous filters with antimicrobial properties.

### 3.3 Hybridization

The hybridization of LIG with nanoparticles has been widely studied as a technology for forming hybrid materials by combining the advantages of each material [59-64]. Various techniques, such as physical deposition, chemical reduction, and covalent attachment, can be used to create hybrid materials between the LIG and nanoparticles. Physical deposition involves the deposition of nanoparticles on the surface of LIG using physical techniques, such as drop-casting and spin-coating, resulting in the formation of a thin film or layer of nanoparticles on the surface of LIG. Chemical reduction involves reducing metal ions in a solution containing LIG to create metal nanoparticles that attach to the surface of the LIG. This method can be used to produce various metal nanoparticles including Au, Ag, Pt, and Pd. Covalent attachment refers to the modification of LIG with active functional groups that can chemically bond to the surface of the nanoparticles. J. Zhao. et al. reported high-performance flexible planar NiO/NLIG (NiO nanoparticles anchored on N-doped LIG) composite microsupercapacitors (MSCs) fabricated by a facile and rapid laser direct writing method, as presented in Fig. 5(a) [59]. The flexible NiO/NLIG MSCs showed good electrochemical properties with remarkable areal specific capacitance, superior areal energy density at high areal power density, excellent stability, and good cycling performance. As shown in Fig. 5(b), direct laser irradiation of a metal-complex-containing PI film resulted in the in situ formation of nanoparticles embedded in LIG [60]. Nanoparticles synthesized as such were varied, ranging from metal oxides to metal dichalcogenides, and showed high activity in several electrochemical reactions, including oxygen reduction and hydrogen evolution. R. Xu. et al. developed a simple one-step laser irradiation method to design $MnO_2$ nanoparticles uniformly distributed on LIG as shape-controllable electrodes for MSCs ($MnO_2$/LIG MSCs), which can be seen in Fig. 5(c) [61]. The obtained $MnO_2$/LIG MSCs showed remarkable areal specific capacity, high energy density at a high power density, good mechanical flexibility, and modular integration. To develop approaches for the in-situ synthesis of metal oxide/LIG composites for OER or ORR bifunctional catalysts, a facile solid-phase synthesis technique to produce $Co_3O_4$/LIG was studied [Fig. 5(d)] [62]. To simplify the fabrication process of ZnO/carbon composites, J. Rodrigues. et al. demonstrated the synthesis of ZnO-decorated LIG through a direct-laser scribing approach using a $CO_2$ laser under ambient conditions, as depicted in Fig. 5(e) [63]. Electrochemical characterization revealed the presence of charge transfer between LIG and ZnO, and the inclusion of ZnO nanoparticles improved the capacitance of the ZnO/LIG composite compared to that of pristine LIG. In addition, active research has been conducted on new 2D nanomaterials, such as transition-metal dichalcogenides (TMDs), to improve the performances of supercapacitors by increasing their specific capacitance [64-66]. F. Clerici. et al. reported a one-pot synthesis method to produce $MoS_2$-decorated LIG ($MoS_2$-LIG) by directly irradiating a laser onto a PI film [Fig. 5(f)] [67]. The fabricated $MoS_2$-LIG has a 3D arrangement of agglomerates of wrinkled graphene flakes decorated with $MoS_2$ nanosheets. Owing to its good electrical properties and high surface area, the composite is well-suited for use as an

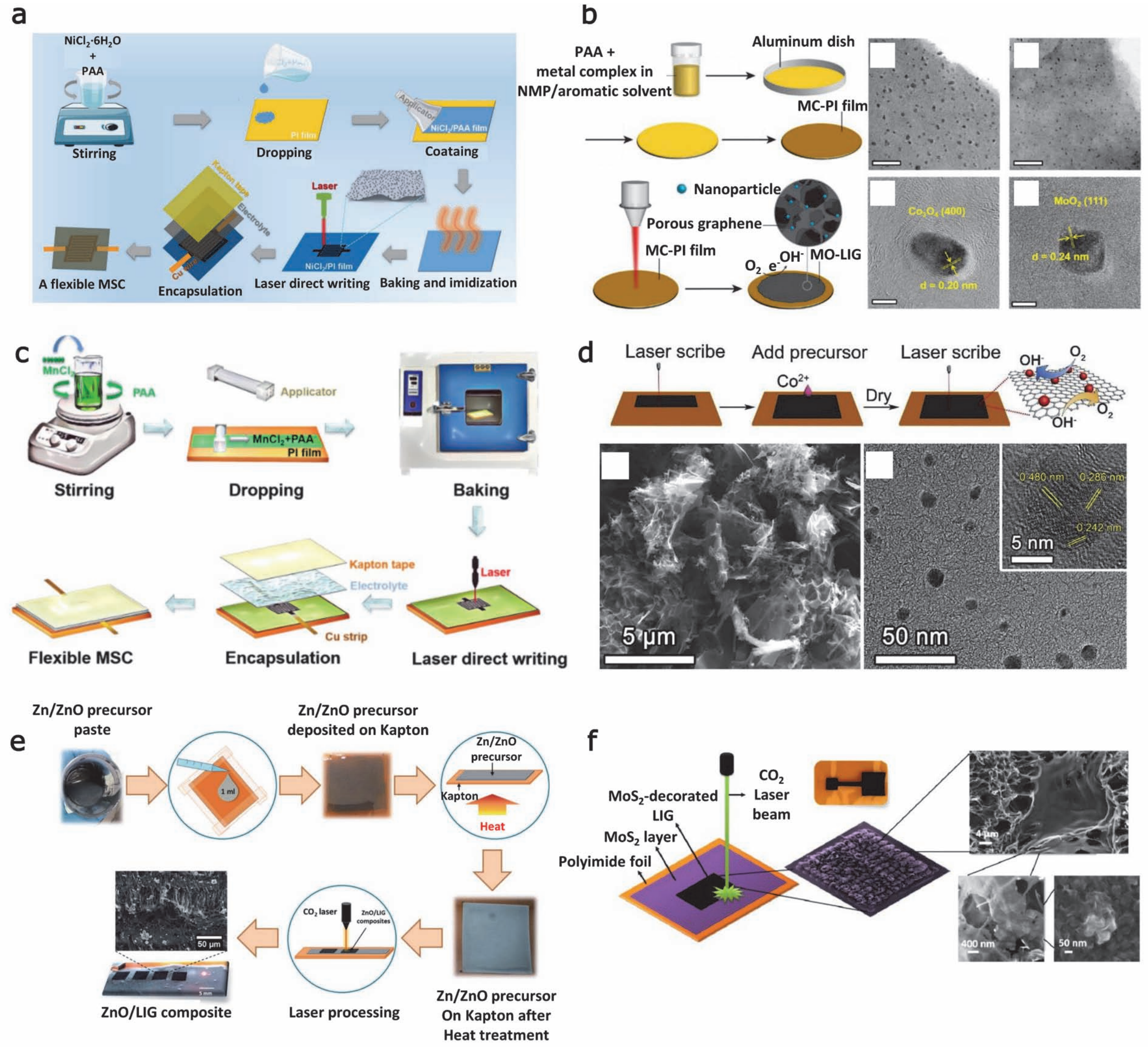

**Fig. 5.** Schematic illustration of (a) fabrication process of flexible planar NiO/NLIG MSCs (reprinted with permission from [59]. Copyright 2022, American Chemical Society), (b) formation of MO-LIG from MC-PI films and related TEM images (reprinted with permission from [60]. Copyright 2015, American Chemical Society), (c) fabrication process of flexible planar $MnO_2$/LIG MSCs (reprinted with permission from [61]. Copyright 2021, Elsevier), (d) preparation process of the $Co_3O_4$/LIG catalyst and SEM and TEM images (reprinted with permission from [62]. Copyright 2018, Elsevier), (e) preparation process of ZnO decorated LIG composites (reprinted with permission from [63]. Copyright 2019, J. Rodrigues. et al.), and (f) laser irradiation on a $MoS_2$-covered PI film to fabricate $MoS_2$-decorated LIG and its SEM images (reprinted with permission from [67]. Copyright 2016, American Chemical Society).

electrode material in supercapacitors, capable of exhibiting both electric double-layer and pseudocapacitance behaviors.

## 4. SUMMARY AND PROSPECTS

This article reviews the synthesis and functionalization of LIG, a porous three-dimensional graphene produced by carbonizing and graphitizing carbon-based polymer thin films using a laser. LIG paves the way for a novel application that relies on distinctive attributes that deviate from those exhibited by conventional CVD and flake graphene. From a technical perspective, functionalization of a material with a laser scribing system offers significant advantages, including short synthesis times, facile patterning, and high processing efficiency under ambient conditions. In particular, this approach distinguishes itself from conventional methods because it allows selective modification of only specific regions on a substrate with functional groups, thereby enabling the realization of various performance characteristics on a single surface. Despite such excellent material and technological potential, research related to the functionalization of LIG still less extensive than for existing carbon nanomaterials. Herein, the functionalization of LIG, which combines these excellent material properties and synthesis technology, is introduced by classifying it into surface modification, heteroatom doping, and hybridization. Functionalization of LIG can be achieved through surface modification techniques such as structural changes, physical treatments, and coating with functional materials. Functionalizing LIG through doping with heteroatoms or molecules such as N, B, and S modifies its electronic characteristics, allowing it to be used in electronic applications such as transistors and sensors. The hybridization of LIG and nanoparticles is a widely studied technology for forming hybrid materials with combined advantages. Various techniques, such as physical deposition, chemical reduction, and covalent attachment, can be used to create hybrid materials which exhibit excellent electrochemical properties and stability, making them suitable for use in supercapacitors. In addition to the research presented in this review, various studies related to LIG functionalization have been conducted. The continuous expansion of research on LIG and its functionalization is expected to drive the emergence of new methods of functionalization and applications, which will ultimately lead to further progress and innovation in the field, particularly considering the vast potential of LIG functionalization in areas such as energy storage, electronics, sensors, and biomedicine.

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