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Synthesising Graphene with Renewably-sourced Bio-carbon Precursors: A Brief Review

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Synthesising Graphene with Renewably-sourced Bio-carbon Precursors: A Brief Review

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Abstract: Graphene is a 2D material with exceptional properties that surpass those of many other materials in many respects. Conventional methods of graphene synthesis heavily rely on gaseous carbon (C) precursors, primarily hydrocarbons; such as methane or ethylene; that have significantly negative effects on the environment. The global shift towards sustainability and eco-conscious practices has increased the need for graphene production methods that are sustainable. As such, multiple studies have explored alternative sources of C, particularly bio-based materials, as well as waste-to-value processes. Reusing bio-based materials as C-based precursors not only addresses the urgent need for C sources that are sustainable and environmentally friendly but effectively creates a circular economy in the materials science and technology industry. As such, this present study explores the methods of synthesising, applying, and optimising the conversion of bio-based renewable solid carbon (SC) and liquid carbon (LC) precursors derived from a diverse range of C sources; such as lignocellulosic biomass, agricultural residues, and everyday vegetable oils; into high-quality graphene. The findings emphasise the promising role of renewable SC and LC precursors in the pursuit of sustainable and environmentally responsible methods of graphene production.

1. Introduction

The discovery of graphene, via the mechanical exfoliation of pyrolytic graphite using adhesive tape, provided scientists with a deeper understanding of the properties and physical effects of the material. More specifically, Novoselov and Geim (2004) pressed highly-oriented pyrolytic graphite (HOPG) onto a substrate then peeled multiple layers of graphene off of it using commercially available adhesive tape [1]. However, although this method is reliable, it lacks uniformity and reproducibility. As such, more practical and standardised methods of producing high-quality graphene have been developed [2]–[6]. Graphene has since rapidly developed into a family of graphene-based materials; such as single-layered (sLG), bi-layered (bLG), few-layered (fLG), multi-layered (mLG), graphene oxide (GO), reduced graphene oxide (rGO), and nanoplatelets [7]–[10].

Multiple studies have examined the unique properties and applications of graphene as it has been proven to improve the performance of various materials and processes [11]–[13]. However, there is an urgent need for a method of mass-producing graphene to meet ever-growing demands. Several methods of mass-producing graphene have been developed. One such method, chemical vapour deposition (CVD), is the preferred method as it is simple and can produce large-area graphene that is highly crystalline [14], [15]. Chemical vapour deposition (CVD), typically, involves exposing a substrate to a precursor, which reacts and breaks down onto the substrate to produce the desired graphene product. Figure 1 depicts the mechanism of growing graphene in a typical CVD process. Methane and acetylene, both of which are toxic and extremely hazardous, are increasingly used as gaseous carbon (C) precursors as they are stable at elevated temperatures and can control graphene growth [16]–[18] while hydrazine, dimethylhydrazine, and hydroquinone, all of which are highly toxic, are commonly used as reducing agents [19]. As such, precursors and reducing agents that are environmentally friendly and safe must be developed before graphene can be synthesised on a larger scale.

As the synthesis method affects the characteristics of the end product, every method produces graphene that is suitable for different applications. This flexibility facilitates the production of graphene with properties that are tailored for specific applications. For instance, the transparency and flexibility properties of graphene are crucial for electronic applications while its hydrophobicity is vital for coating applications [20], [21]. Similarly, its drawbacks; such as its ability to increase mechanical reactivity and generate magnetism; may not be ideal for applications that require thermal conductivity and mechanical strength [22].

Rapid industrial development has had a profoundly negatively effect on the environment [23]. Furthermore, industrial waste is, almost never, used to its full potential and minimal effort is put into recycle it. It is also difficult to recycle industrial waste due to its composition, contamination, or a lack of established recycling technologies. As such, manufacturers are left with no choice but to rely on virgin materials as appropriate recovery and recycling technologies are non-existent. According to the European Economic Area (EEA), wood products are rarely recycled and manufacturers often recover energy from them [24]. However, as this bio-waste comprises biodegradable substances, it can be converted into valuable materials that enhance its energy-generating potential [25]. Bio-based materials are products that comprise substances derived from naturally occurring or synthesised living matter. This includes products that have been produced via processes that involve biomass. Biomass, typically, contains cellulose, hemicellulose, and lignin [26]. As cellulose is the most abundant component in biomass, multiple studies have examined converting it into new bio-based materials, that are comparable to conventional materials, and integrating it into existing and new products [27]–[31].

The global shift towards a sustainable future has spurred research into green methods of graphene production. Multiple studies have examined synthesising graphene with waste-based solid carbon (SC) precursors via CVD [7], [32], [33], however, examinations into the use of bio-based liquid carbon (LC) precursors is limited [34]. This present study provides a brief overview of the development of graphene synthesis methods, with a focus on pivotal advancements in graphene production, specifically the use of renewably-sourced SC and LC precursors. The primary objectives of this present study were to identify the current challenges and opportunities for the optimisation of bio-based C precursors and their impacts in various applications.

A systematic literature review was conducted to search and collect articles published between 2019 and 2023 on synthesising graphene with bio-based C precursors. Keywords; such as “graphene”, “liquid carbon precursor”, “solid carbon precursor”, “green synthesis”, “renewable”, and “biomass”; as well as Boolean search operators; such as AND, OR, and NOT; were used to search reputable digital libraries; namely, Science Direct, Google Scholar, and Elsevier; for articles based on their titles, abstracts, and methodologies. The gathered references were then sorted according to their synthesis methods and the precursors used to precisely determine the influence of bio-based C precursors on the applications of the produced graphene.

2. Methods of Synthesising Graphene

Most studies examined the mass production of large-area graphene for fundamental applications [35]–[38]. This involves extracting graphene, depending on its purity and the desired end product. The shape and thickness of the synthesised graphene indicates its type and quality which, in turn, affects its properties. As such, many methods of synthesising high-quality graphene with the desired properties have been developed. These can be classified into two groups; namely, "top-down" and "bottom-up" (Figure 2).

The top-down method exfoliates macroscopic precursors; such as bulk graphite; into graphene by breaking the van der Waals bonds between its layers. Although this method does not require substrate transfer and is cost-effective, it is difficult to remove the solvents and reducing agents used in the process and the density of end product is significantly defective [39].

The bottom-up method, on the other hand, arranges graphene layers by aggregating smaller precursor materials. As such, it provides more precise control over the quality of the produced graphene [40]. However, this method is not only costly but the yield is small. Therefore, it cannot be used to mass produce graphene without scaling up [41].

2.1. Chemical vapour deposition (CVD)

Chemical vapour deposition (CVD) is a promising technique that involves heating a substrate and introducing a C-containing gas in a controlled environment to deposit graphene onto the substrate. Carbon (C) can be derived from solid, liquid, or gaseous sources [35]–[37]. Multiple studies have examined using renewably-sourced C precursors to synthesise graphene.

Biomass waste; such as wood waste and peanut seed coats; and other waste; such as plastic and tires; are all examples of renewable C precursors [45]–[48]. As such, these sources of C may be a sustainable method of synthesising graphene while simultaneously overcoming the issue of waste management.

2.2. Hydrothermally

The hydrothermal process involves reacting a solid substance with an alkaline or acidic solution in an autoclave at a low temperature [49], [50]. More specifically, autoclaving a C precursor in an aqueous solution then filtering and drying it. The temperature, pressure, solvent, and C feedstock used during the process determines the type and quality of the end product. Some studies have combine hydrothermal and graphitisation to create nanosized graphene-like materials (GLMs) that possess different properties [51]. Apart from that, hydrothermal-graphitisation has since been proven to produce unique morphologies that recent literature has not discussed.

2.3. Pyrolysis

Pyrolysis is bottom-up method of producing graphene. Pyrolyzing various C sources has now become the standard method of producing graphene on the surfaces of metals [52], [53]. A thermochemical conversion process, it converts biomass into energy without damaging the environment as only high-pressure decomposition is used to change the chemical composition of the biomass [54]. While biochar and bio-oil are common by-products of pyrolyzing organic materials [55]–[57], fast pyrolysis leaves a C residue, typically, graphene [58].

2.4. Chemical exfoliation

Chemical exfoliation, one of the first methods of producing graphene [59], [60], is a top-down method as the graphene is extracted from a highly pure graphite (HPG) powder. This is accomplished by decreasing the interlayer Van der Waals forces and, thereby, increasing the space between the layers, followed by rapidly heating the layers to exfoliate the graphene sheets. However, as HPG is costly and the process uses harmful reducing agents; such as hydrazine and di-methylhydrazine; efforts are afoot to incorporate waste materials into the process. Akhavan et al. is responsible for the recent trend toward optimising environmentally friendly alternatives [61].

2.5. Laser-assisted

The laser-assisted carbonisation of natural sources can be used to mass produce GLMs. The process is quite flexible [62] as modifying the power of the laser and adjusting the exposure time changes the characteristics and scalability of the end product and it can be done at room temperature without the use of hazardous chemicals [63]. Furthermore, the produced GLMs not only have excellent mechanical strength and thermal stability but electrical conductivity as well [64], [65]. Therefore, GLMs can be sustainably produced using natural C resources.

2.6 Liquid-phase exfoliation (LPE)

Liquid-phase exfoliation (LPE) is a versatile technique that isolates bulk graphene or GO into individual graphene layers by combining sonication and shear forces to directly exfoliate the powdered graphite in a liquid [59]. More specifically, the ultrasonic or shear forces separate the graphite layers in graphite or GO into individual sheets or small flakes by overcoming the Van der Waals forces between them [67]. This method is not only scalable but capable of yielding a high amount of graphene as well.

2.7 Laser ablation

Laser ablation is a simple and green method of synthesising graphene by cooling the vapour plumes produced by ablating the graphite onto a target substrate [68]. It is a popular method as it is cost-effective, environmentally friendly, does not use chemicals in the preparation process, or produce any secondary chemical compounds [69]. It is also flexible as the process and laser parameters can easily be altered to suit the manufacturer's needs as well as control of the surface quality and the efficiency of the ablation process [70]. However, although this method produces high-quality graphene, it still lacks a growth method with which to mass produce graphene.

2.8 Chemical reduction

A top-down method that is widely used to synthesise graphene and rGO, chemical reduction involves using chemical agents; such as hydrazine, sodium borohydride, or hydrohalic acids; to convert GO or a precursor into graphene [71]. Nevertheless, multiple studies are examining reducing GO using green reductants [72]–[74]. The characteristics of the produced GO are believed to be influenced by the source and quality of the reducing agent's biomolecules as well as the reaction conditions [75]. However, despite the versatility and cost-effectiveness of this method, careful optimisation is required and the end product does not meet the quality standards nor possess the properties of pristine graphene [76].

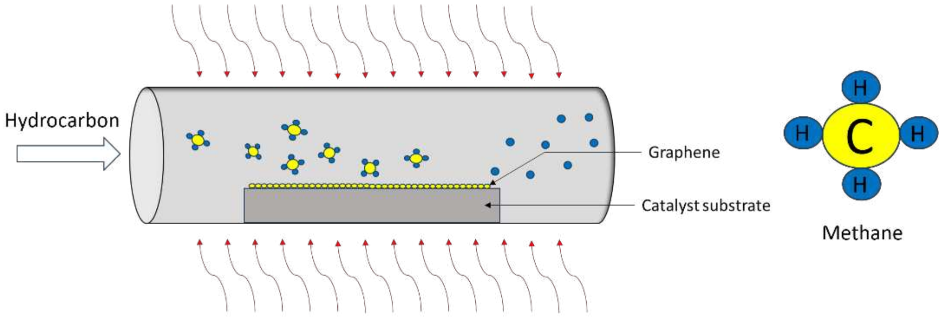

Figure 1 The graphene growth mechanism in the CVD method

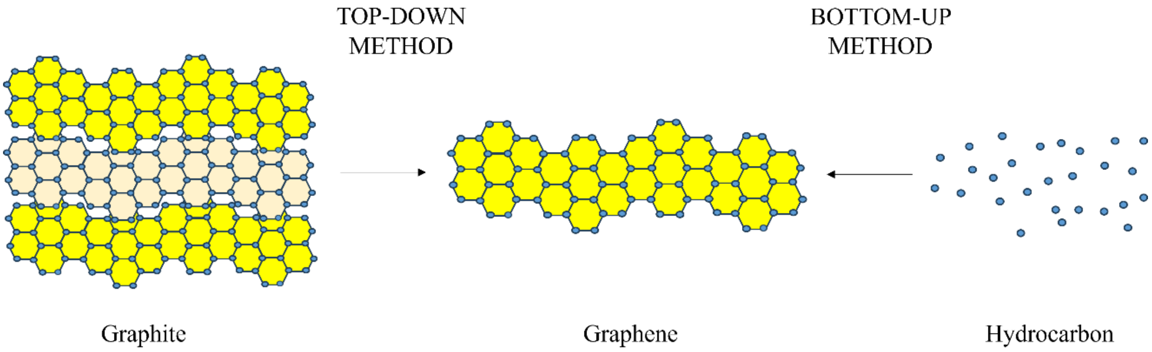

Figure 2 The top-down and bottom-up graphene synthesis methods

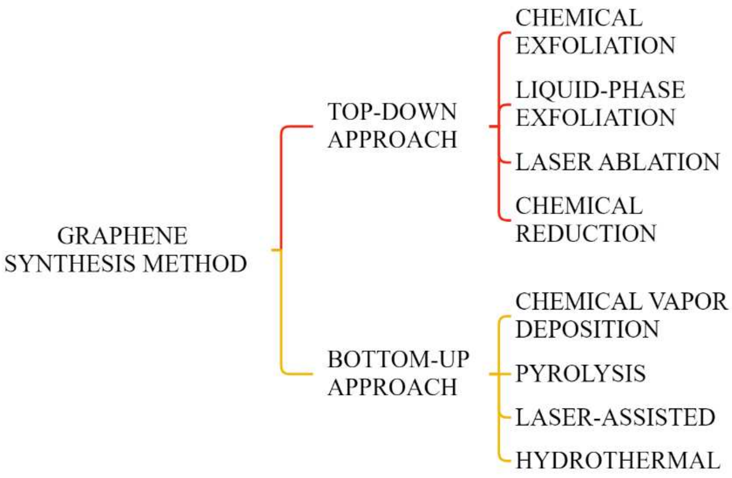

Figure 3 The many methods of synthesising graphene

3. Methods of Synthesising Liquid Carbon (LC) Precursors

Jalani et al. synthesised mixed mLG and bLG on a nickel (Ni) substrate via CVD with a refined cooking palm oil (rCPO) precursor [77]. When combined with a rapid cooling technique, the high concentration of hydrogen produced during the growth process not only improved uniformity but prevented defects from forming in the graphene layers. It is believed that rapidly cooling a substrate halts the production of C post-heating, which decreases the likelihood of excessive precipitation forming on the substrate [78]. Likewise, rapidly cooling a copper (Cu) substrate during sLG synthesis using rCPO decreased the number of defects in the graphene layer [79].

Seo et al., similarly, found that slow cooling promotes excessive C aggregation on the Ni bulk [78]. The amount of precursor used was also found to affect the quality of the produced graphene film. More specifically, an excessive amount of precursor caused the Ni bulk to be oversaturated with C while an insufficient amount caused oxygen species; namely, C-O amorphous C; to form.

Graphene synthesised with waste chicken fat (wCF) had the lowest heating temperature (400°C), and its low molecular-weight C molecules were found to facilitate graphene growth [80]. Copper (Cu) annealing was also found to expand the grain size and smoothened the surface, resulting in only a minor lattice mismatch in the graphene. Therefore, wCF can be used to synthesise sLG that are uniform.

Azam et al. synthesised graphene nanoparticles (GNP) via catalytic CVD with a waste cooking oil (wCO) precursor [81]. As wCO is rich in methylene, it formed stacked bLG that had superlattice structures and insignificant wrinkles or crumpling, which differs from commercial GNPs. Robaiah et al. synthesised mLG via double thermal CVD with a wCPO precursor [82]. It was critical to maintain an optimum deposition temperature as a lower temperature formed imperfect mLG with numerous pores and cracks while a high temperature formed graphene layers of different shapes and densities.

Nurfazianawatie et al. synthesised mLG via double thermal chemical vapor deposition (DT-CVD) with a wCO precursor. [83]. The temperature of the precursor was found to play a role in obtaining optimal mLG. A Raman analysis revealed that low precursor temperatures caused no graphene peaks due to low vaporisation energy. As the C-H bonds in wCO are weaker, it enables graphene to synthesise at lower temperatures. As such, an optimum temperature (350°C) easily breaks the C-H chain and forms mLG.

Salifairus et al. synthesised high quality mLG via CVD with a palm oil precursor [84]. Although a deposition temperature of 900°C was found to produce the best results, it yielded the highest surface roughness (378 nm) due to the poor uniformity of the deposition as the graphene was only located at the edges of the Ni substrate.

Renewable sources of LC have been used to synthesise graphene for some applications (Table 1). As seen, these precursors produce graphene products with fewer defects, however, they, along with their applications, have rarely been examined. Therefore, more research on the performance of graphene that has been synthesised with LC precursors and its specific applications is required to better understand the characteristics and applications of LC precursor as they may open new and sustainable innovation opportunities.

4. Methods of Synthesising Solid Carbon (SC) Precursors

Multiple studies have examined synthesising graphene with SC precursors derived from biomass. Solid carbon (SC) precursors are preferable to gas precursors as using biomass to synthesise graphene manages waste and decreases greenhouse gas emissions. Multiple ongoing studies are examining a variety of waste-based precursors with which to synthesise GLMs as it would transform waste into value-added products as well as provide a method of producing graphene that is both environmentally friendly and economical.

Ruan et al. is believed to have pioneered using low-valued feedstocks to grow high-quality sLG via CVD [85]. This was accomplished by combining purification with a growth method. The SC source on the top side of the Cu foil decomposed, indicating that purification had occurred as unwanted elemental residues had been left behind, and high-quality pristine graphene with few defects was deposited on the underside.

Akhavan et al. synthesised multi-layered graphene oxide (mGO) sheets via chemical exfoliation of GO and rGO using Hummer's method with carbonaceous waste materials as a precursor; namely, wood, leaf, bagasse, and fruit waste [61]. Although the chemical states of the synthesised GO were the same, they all contained different residual elemental impurities from the carbonaceous materials. The lowest residual impurity accomplished was $< 0.7\%$. Chen et al. synthesised pristine wheat straw-derived graphene sheets via a combination of hydrothermal and graphitisation methods [86]. The hydrothermal method separated the cellulose-rich fibres of the wheat straw, which was then graphitised to form fLG.

Zhang et al. synthesised wheat straw-derived three-dimensional graphene foam (3DGF), that only had a few defects, by combining rapid pyrolysis and atmospheric-pressure CVD (AP-CVD) [87]. Waste pyrolysis gases were used as an alternative to conventional carrier gases, thereby, decreasing the environmental impact of the method.

Chyan et al. synthesised GLMs via a laser-assisted method with conventional fossil-based polymers and organic C precursors [88]. Apart from that, a grape molasses precursor was found to yield GLMs that have a turbostratic phase and ultra-low sheet resistance ($10\ \Omega\ sq^{-1}$) [89]. Higher laser fluence is believed to have increased energy deposition on the target materials, resulting in more intense heating, which can potentially cause changes in the surface morphology.

Apart from agricultural waste, other sources of biomass have been used to produce value-added products. For instance, nutshells can be pyrolyzed to produce bio-oils [90]. For instance, Lu et al. synthesised GLMs from macadamia nutshells for supercapacitors via activated hydrothermal treatment [91]. The physical structure and characteristics of a bio-C material is believed to affect the morphology of the produced graphene. Activated hydrothermal treatment was found to optimise the morphology of macadamia nuts and yield a GLM that had a porous and fluffy structure, which is desirable for supercapacitor applications.

The conventional method of synthesising graphene on metal surfaces from different C sources or activated C has limitations in terms of yield. Few-layered graphene (fLG) was synthesised via one-step pyrolysis of dead camphor leaves (DCL) without a metal catalyst [92]. Although the high porous morphology of DCL facilitates quick heat transfer during pyrolysis, the Raman peaks of the synthesised fLG were weak, which could be due to the presence of residual chemicals and the multi-layer structural defects that formed. This method also yielded very little fLG; specifically, only 0.8mg of graphene from 0.1g of DCL. Therefore, although the results demonstrate that biomass precursors can be used sans a catalyst or chemical treatment, further optimisation is necessary if it is to be used to mass produce graphene.

The abundance of agricultural waste produced is a pressing matter that warrants immediate redress. As such, Mat Tahir et al. [93] synthesised mLG with oil palm fibre (OPF) and fruit cover plastic waste (FCPW). The best synthesised graphene sample; which was the graphene-coated substrate; exhibited impressive tribological performance when tested in a micro ball-on-disc tribometer. More specifically, it exhibited better wear resistance and lower coefficient of friction (COF) than an uncoated surface at an applied load of 3N. Therefore, it would prolong the lifespan of a component which would, in turn, improve the overall performance of a system. However, these results do not fully indicate the maximum tribological performance of the synthesised graphene coating as the micro ball-on-disc tribometer had a limited range of applied loads.

Table 2 provides a summary of graphene products that have been synthesised with renewably-sourced SC precursors. As SC precursors have been more extensively studied, they are known to produce graphene with desirable properties. Furthermore, the multitude of methods with which to synthesise graphene with SC precursors indicates their adaptability and customisability to specific applications. Many types of renewable waste can be used as C precursors to synthesise graphene. Furthermore, as technology and research advance, more renewably-sourced SC precursors may be discovered with which to produce high-quality graphene.

The use of renewably-sourced SC precursors for graphene synthesis offers a range of advantages and limitations as the precursor and synthesis method affect the produced graphene. The use of bio-based materials increases the environmental sustainability and decreases the C footprint of graphene production. Zhang et al. examined the life-cycle of their 3DGF and found that it had a smaller environmental footprint than those produced using conventional CVD [87].

Furthermore, using renewable sources to produce graphene aligns with the 2030 Sustainable Development Goals (SDGs) of the United Nations, especially Goal 12; namely, responsible consumption and production. The waste-to-value approach is a comprehensive strategy with which to decrease waste, add value, use resources sparingly, and effectively conserve resources [94] as it uses materials that would, otherwise, be discarded or underutilised, thereby, decreasing the waste of resources and environmental impacts. In short, using bio-based materials as C precursors to synthesise graphene creates a more sustainable and circular economy.

However, it is proving challenging to identify the optimal parameters with which to grow graphene products with bio-based sources. This is primarily because the synthesis method must be able to control the size, quality, and morphology of the synthesised graphene [95]. Apart from that, not many studies have examined the commercial method of synthesising graphene with bio-based precursors, which is important for the sustainable mass production of graphene. The impact of bio-based C precursors on the environment should also be examined more thoroughly.

Table 1 A summary of the synthesis methods, properties, and applications of graphene produced with renewably-sourced liquid carbon (LC) precursors

C Source	End Product	Synthesis Method	Graphene Properties		Applications	Reference
			I_G/I_D	I_{2D}/I_G		
wCF	sLG	LP-CVD	<0.1	>3.0	-	[80]
wCO	Graphene nanoplatelets	Catalytic CVD	-	0.93-0.99	Energy storage	[81]
wCPO	mLG	DT-CVD	-	0.36	-	[82]
Commercial palm oil	mLG	Thermal CVD	0.27	1.19	-	[84]
Soybean oil	Graphene film	Thermal CVD	0.15-0.25	0.95-1.50	Biosensing electrodes	[78]
wCO	mLG	CVD	-	0.34-0.38	-	[83]
rCPO	mLG and bLG	Thermal CVD	-	0.62-3.01	-	[77]

Table 2 A summary of the synthesis methods, properties, and applications of graphene produced with renewably-sourced liquid carbon (LC) precursors

C Source	End Product	Synthesis Method	Graphene Properties		Applications	Reference
			I_G/I_D	I_{2D}/I_G		
Food, waste, and insects	sLG	CVD	<0.1	>1.8	-	[85]
Vegetation waste, animal waste, semi-industrial waste, and industrial waste	GO and rGO sheets	Chemical Exfoliation	-	>0.3	Supercapacitors	[61]
Wheat straw	fLG	Hydrothermal + Graphitization	1.37	0.61	Anode for lithium-ion batteries	[86]
Cellulose lignin, wheat straw, and sawdust	3DGF	CVD	-	-	Energy storage, supercapacitor electrodes, and adsorption of organic liquids or oils	[87]
Grape molasses	GLM	Laser	~ 0.2	0.9-1.0	Supercapacitors	[89]
Camphor leaves	fLG	Pyrolysis + Dispersive Agent	0.99	-	-	[92]
OPF and FCPW	mLG	CVD	-	0.89-1.09	Tribology	[93]
Macadamia nut shells	GLM	Hydrothermal Treatment	1.18	-	Supercapacitor	[91]

5. Applications of Graphene

As graphene has exceptional qualities, it is used in various fields. Furthermore, the ground-breaking inventions that have been made with it have significantly advanced technology as well as enhanced our way of life. Therefore, this remarkable material is anticipated to revolutionise other industries and pave the way for more innovations.

Graphene has excellent tribological properties due to its self-lubricating and wear resistance capabilities [96]–[98]. Furthermore, the weak van der Waals forces between its layers enables them to be easily sheared, which results in low COF and wear rate [99], [100]. For instance, mLG synthesised from OPF performs better tribologically than FCPW as its COF and wear rate are significantly lower [101]. Furthermore, when used as a lubricant, graphene nanoflakes enhance the tribological properties of Jatropha oil as the tribo-layer that it forms impedes metal-to-metal contact, thereby, decreasing the COF and wear rate once again [102].

Apart from that, as the porous structure of graphene has a high specific area, it is ideal for use in applications that require rapid ion transfer; such as supercapacitors [103], [104]. As such, high-performance supercapacitors that have been made from various renewably-sourced graphene have consistently outperformed C-based supercapacitors [105], [106]. For instance, a porous C-based GLM synthesised with macadamia nut shells yielded significant capacitance (251 F g^{-1}) at the current density of 1 A g^{-1} and had a capacitance of 96.4% after 1000 cycles [91]. Athanasiou et al. also found that C-based GLMs have superior supercapacitor properties with little sheet resistance [89].

As graphene efficiently transports electrons, it is ideal for use in lithium-ion batteries as it decreases internal resistance and increases output power [107]–[110]. Its superior mechanical properties may also increase the stability of electrode materials, which would increase the latter's rate capabilities and cyclic stability [111], [112]. A wheat straw-derived fLG was found to possess the unique physical and chemical properties that anode materials require [86]. Meanwhile, Zhang et al. found that 3DGF has stable electronic transport properties at only 27°C [87]. Additionally, soybean oil-derived graphene films are promising biosensing electrodes. More specifically, they have highly sensitive and selective bio-detection capabilities and facilitate the detection of both miRNAs, based on the efficient immobilisation of probes, and non-complimentary miRNAs [78].

Multiple studies on C-based nanomaterials have proven their efficacy as an adsorbent of several chemical contaminants [113]. Nanomaterials that have a large surface area, high chemical reactivity, and good mechanical properties are suitable for wastewater treatment [114]. Wastewater treatment involves removing contaminants and pollutants from effluents, rendering it safe for disposal or reuse. Graphene-based materials have shown promising potential in wastewater treatment applications. For instance, GOs are good adsorbents of dyes, heavy metals, and pharmaceuticals [52], [115], [116] as their negatively charged sites have a strong affinity for cationic dyes which, when combined with C materials, develop positively charged sites, thereby, enabling the GOs to effectively remove cationic and anionic dyes [103]–[105]. Furthermore, the large surface area and porous structure of graphene-based materials provide ample adsorption sites with which to efficiently remove cationic dyes; such as methylene blue [120]; from wastewater. Apart from that, the large surface area (3000 m^2 g^{-1}) of 3D graphene sponges synthesised with wood waste enables it to rapidly and extremely efficiently adsorb o- and p-nitrophenols, even when the pollutant concentration was doubled [45]. However, it is difficult to synthesise graphene-based materials that have adsorption selectivity, thereby, making it difficult to selectively remove only specific contaminants from wastewater [121] and leave behind essential ions or compounds. As most studies have only examined the adsorption capabilities of graphene on one or two pollutants, its adsorption capabilities and selectivity warrant further investigation.

The use of bio-based materials may decrease the environmental impact of the graphene production process. Furthermore, the practice of using renewable materials promotes sustainable practices as well as decreases reliance on petroleum-based precursors and toxic fumes [122]. The use of graphene in wastewater treatment applications also contributes to environmental protection and sustainability by decreasing the amount of harmful pollutants in water bodies and protecting aquatic ecosystems and human health [123].

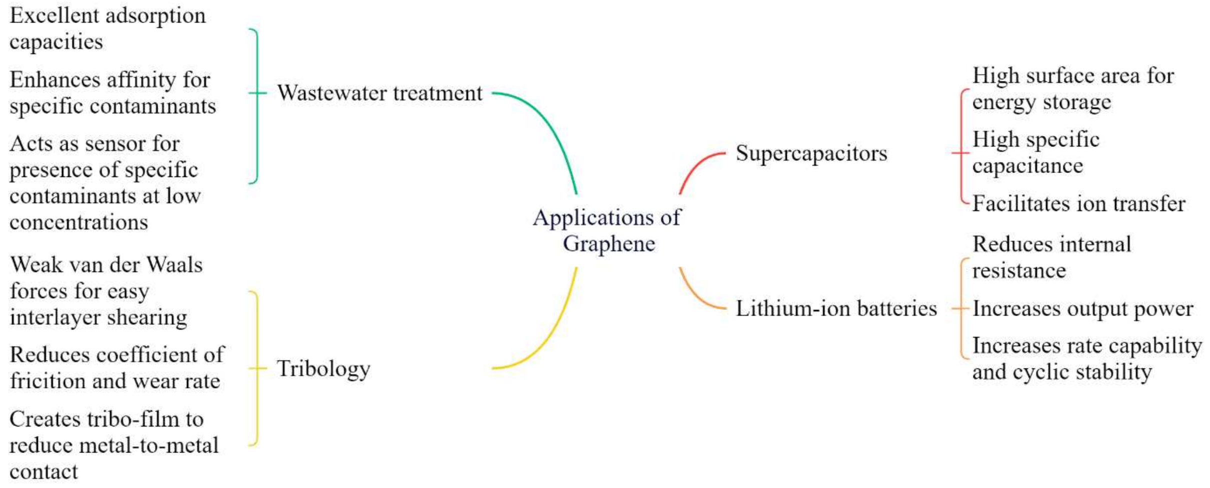

Figure 4 The applications of graphene

6. Conclusions and Future Perspectives

As the demand for graphene continues to rise, there are growing concerns over its synthesis methods and sources. Although multiple studies have examined graphene and its derivatives, it has yet to reach a peak in the technology industry. The efficacy of graphene for biomedical, wastewater treatment, and supercapacitor applications is well documented. However, not many studies have examined the tribological properties of renewably-sourced graphene. As such, more research is needed to investigate the tribological properties of graphene as it could benefit various industries, from aerospace to automotive engineering. Furthermore, most studies have examined the properties and quality of renewably-sourced graphene but only a handful have examined its quality for its intended applications. As such, the unique properties of graphene produced using each type of precursor must be further examined before it can effectively be used for specific applications.

This present study provided a brief overview of synthesising graphene with renewably-sourced bio-based C precursors. Several renewably-sourced LC and SC precursors produce graphene with properties that surpass that of traditionally grown graphene. However, the environmental effects of mass-producing graphene with bio-based precursors are still being investigated. Furthermore, although these precursors contribute towards sustainable graphene production, there are still obstacles in terms of scalability and cost-effectiveness. It is believed that more research and development could lead to the discovery of more innovative applications for graphene. Therefore, the use of renewably-sourced C-based precursors for graphene synthesis must be further investigated to discover their full potential.

Declaration Statement

The manuscript has not been previously published, is not currently submitted for review to any other journal and will not be submitted elsewhere before a decision is made by this journal. All the data shown in this manuscript originally obtained from our experimental results. All authors are aware of this manuscript and agree with and have approved its content and its submission to the Journal of the Brazilian Society of Mechanical Sciences and Engineering.

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Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Nomenclature

CVD	Chemical vapour deposition
LP-CVD	Low-pressure chemical vapour deposition
AP-CVD	Atmospheric pressure chemical vapour deposition
DT-CVD	Double thermal chemical vapour deposition
LPE	Liquid-phase exfoliation
HPG	Highly pure graphite
GO	Graphene oxide
rGO	Reduced graphene oxide
FLG	Few-layer graphene
GNP	Graphene nanoplatelets
3DGFs	3D graphene foams
FWHM	Full width at half maximum
OPF	Oil palm fibre
FCPW	Fruit cover plastic waste
WCF	Waste chicken fat
WCO	Waste cooking oil
WCPO	Waste cooking palm oil
miRNA	MicroRNA
RMS	Root mean square
COF	Coefficient of friction
FESEM	Field emission scanning electron microscopy
TEM	Transmission electron microscopy
XRD	X-ray diffusion
Ni	Nickel
Cu	Copper
H_2	Hydrogen
I_{2D}	Peak intensity of peak 2G
I_G	Peak intensity of peak G

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RESPONSE TO REVIEWERS

Reviewer #2

1. No sufficient supporting information in the introduction part of the manuscript.
- Thank you for the comment. The author does not entirely understand which statement the reviewer is referring to, but a few statements and an additional paragraph have been added into the introduction part. Please refer to the turquoise highlighted part in the introduction section.

2. I feel the authors have missed some of the important references in this field.
- Please refer to response 1.

3. Clarity of figures need to be improved.
- All figures' clarity has been improved. Please refer to the turquoise highlighted figures in page 4 and 9.

4. Language editing of the manuscript is required.
- The revised manuscript has been sent for proofreading. The certificate of proofreading as in the supplementary file.

5. Applications part need to be explained with pictures for readers clarity.
- Thank you for your comment. A figure on the applications of graphene has been added in page 9 as well as an additional paragraph on tribology. Please refer to the turquoise highlighted part in the application section.

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