

A Review on Bio-based Graphene Derived from Biomass Wastes

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Biomass waste has become a new source for producing graphene due to its carbon-rich structure and renewable nature. In this paper, the research on the conversion of bio-based graphene from different biomass wastes is summarised and discussed. This paper reviews the methods for converting biomass to bio-based graphene. There are two approaches for thermal degradation of biomass: thermal exfoliation and carbon growth. The purpose of the thermal treatment is to increase the carbon content by removing volatile matter from the biomass polymer chain. Pre-treatments that help to break down the complex structure of the biomass are discussed; pre-treatments also remove impurities from the said biomass. Lastly, the characteristics of bio-based graphene produced from different biomass and thermal treatments are summarised.

Keyword: Graphene; Carbon; Bio-based; Biomass; Waste; Agriculture; Thermal treatment; Pyrolysis

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INTRODUCTION

Graphite has been used as the raw material for producing graphene since its first discovery. Structurally, graphite is highly anisotropic, meaning that the in-plane and out-of-plane surfaces have different values in properties (Ke and Wang 2016). A layer of graphite is called graphene. Graphene is produced through layers of graphite compound exfoliated layer by layer until what remains is a 'honeycomb' carbon sheet. In layman's terms, a stack of graphene is graphite. The discovery of graphene started long before the extraction and characterization of graphene (Novoselov *et al.* 2004). Initially, the trend was focused on exploiting a fewer layers of graphite and manipulating the graphite structure using a technique called nanomanipulation (Lu *et al.* 1999). Theoretically, a graphite film with a thickness of an atom is unrestricted in flexibility and manipulation, in the form of bending, rolling, and folding. However, the possibility to extract a compound with these features was impossible until Novoselov *et al.* (2004) extracted graphene. Currently, there are many ways to produce graphene either in single or multiple layers. The two classes of methods for graphene production follow a top-down or bottom-up process (Bhuyan *et al.* 2016). The top-down methods exfoliate or separate graphite (or any graphite derivatives) to form graphene. Bottom-up methods include the growth of small molecular carbon precursors to form graphene. Both methods have their advantages and disadvantages, which have been discussed in the literature (Avouris and Dimitrakopoulos 2012; Bhuyan *et al.* 2016; Dasari *et al.* 2017; Lim *et al.* 2018; Lee *et al.* 2019; Smith *et al.* 2019).

Recently, green alternatives for graphene production have been explored. Biomass is an organic material that normally comes from plants or animals; hence the term renewable is attached to it as both are sustainable materials. In this review, the focus is on biomass waste. Biomass waste will be defined here as any organic materials and wastes generated from agriculture. The discovery of converting biomass wastes into graphene is a successful route for recovering sources. The global annual of biomass wastes is 1×10^{10} MT; this amount will continue to increase based on the projected global cropland area

increment by 2050 (Searle and Malins 2015). Thus, energy debates in most developing countries have included policies to protect the environment (Tursi 2019). In industrialized countries, agricultural wastes have been used as a source for biofuels (Harnesk 2019), biodiesels (Vern *et al.* 2019), biogas (Xue *et al.* 2020), or any other types of bioenergy (Pedroli *et al.* 2013), especially in European countries. Generally, the organic material wastes can be branched out to crop waste, food processing waste, paper industries waste, and many other things that are recyclable. There is potential in producing graphene using biomass wastes as it is sustainable, renewable, and abundant.

Graphene has exceptional properties such as mechanical strength as well as optical and electronic properties. Phenomena related to the properties of graphene have been studied intensively (Wang, 2010; He *et al.* 2014; Zhang *et al.* 2011; Zhang *et al.* 2012; Dasari *et al.* 2017). Graphene is a sp^2 hybridized carbon structure aligned on the same plane with bonds angle of 120° , giving the graphene its honeycomb look, as illustrated in Fig. 1. Each carbon atom is equipped with an unhybridized π -bond giving graphene its high intrinsic mobility and ballistic transport (Bolotin *et al.* 2008). Besides, the combination of the σ -bonds and the π -bonds helps with the toughness of the structure with radical ions presence all around the structure. Hence, graphene can withstand extreme temperatures without damaging its structure. Figure 2 shows graphene derivatives such as graphene oxide (GO) and reduced graphene oxide (RGO), which are the closest versions of graphene in the carbon-based materials family.

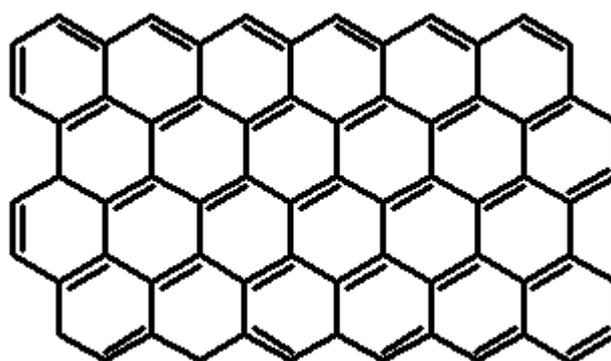


Fig. 1. Graphene honeycomb structure

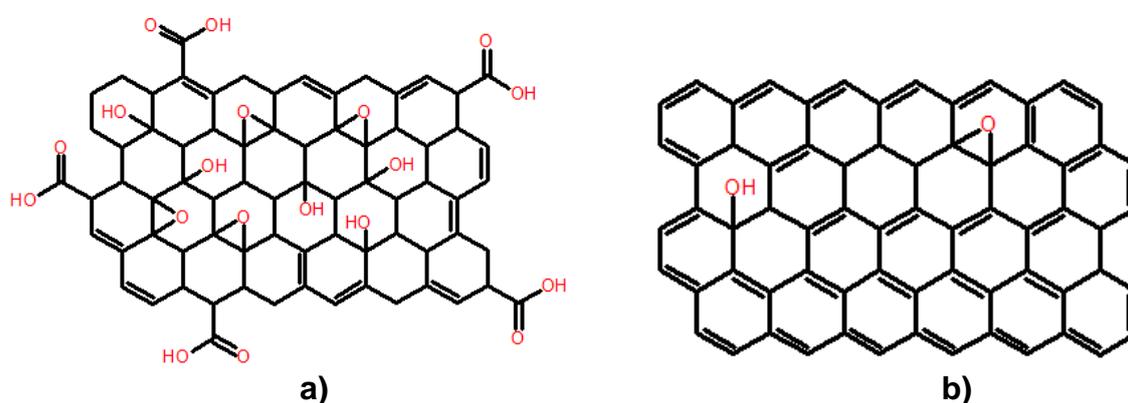


Fig. 2. The structure of a) graphene oxide (GO) and b) reduced graphene oxide (RGO)

GO is an oxidized version of graphene decorated with functional groups, while RGO is produced by removing the oxidized functional groups of GO by reduction. Even though RGO is quite similar to graphene, the severe process of oxidation and reduction introduces defective sites to the RGO sheets. There are the unreacted functional groups that remain attached to the RGO plane. While graphite is abundantly available and cheap,

utilizing biomass as a carbon source is beneficial to the environment because it reduces wastes. Although the GO and RGO produced from biomass are similar to that produced from graphite, property-wise; they are structurally different (Somanathan *et al.*, 2015). This is due to the complexity of the biomass structure, which inherently produces graphene-like structure with impurities. In this review, graphene, GO, and RGO produced from biomass are designated as b-G, b-GO, and b-RGO, respectively.

BIOMASS CONVERSION TO GRAPHENE

Most biomass contains long chains of carbon, hydrogen, and oxygen compounds, with a carbon content as high as 55 wt% (Xie and Goodell 2014). The process of biomass conversion to graphene requires concentrating the carbon content. This process has been used by industries to produce bio-char. Biomass utilization is based on thermal treatment such as gasification, carbonization, liquefaction, and pyrolysis to produce bio-oils (Shahi *et al.* 2020), bio-gas (Abraham *et al.* 2020), biochemicals (Cerazy-Waliszewska *et al.* 2019), or bio-char (Guo *et al.* 2020). The process of increasing the carbon content by removing other elements through thermal treatment is called carbonization; the process of arranging the carbon structures to produce a graphitic-like structure is called graphitization. It should be noted that the carbonization process often produces amorphous carbon rather than a graphite-like structure. Amorphous carbon consists of hard carbon and soft carbon, where the hard carbon is tough to graphitize even at very high temperatures given its chaotic structure (Pan *et al.* 2019). Meanwhile, soft carbon can easily be converted into graphite with thermal treatment. Even though some may consider that the converted carbon structures are not pure graphene due to the number of other carbon materials, the properties that they possessed are somewhat graphene-like. The amount of amorphous carbon correlates to that of exceeding the reaction time and an excessive amount of carbon sample during the thermal treatment (Mamat *et al.* 2018).

This paper considers two popular methods used to form bio-based graphene, which are exfoliation and carbon growth into producing b-G. Although it might be desirable to focus on the conversion of biomass waste into b-G alone, the exfoliation via an oxidizing technique into products such as b-GO and b-RGO is essential to the achievement of biomass waste utilization. Hence, these two derivatives are included in this review. The exfoliation techniques are the Hummers method or mechanical exfoliation. Carbon growth is similar to the chemical vapor deposition (CVD) method; using carbon gas molecules generated from decomposed biomass that is deposited onto a metal surface. This review will also be focusing on some biomass waste sources as carbon source materials, including agricultural and other wastes such as oil palm waste, rice husk, sugarcane bagasse, wheat straw, lignin, and animal residues. The thermal treatments are summarized in Table 1.

Table 1. Thermal Treatment Conditions for Biomass

Sources	Thermal treatment	Temperature	Atmosphere	Time	Product	Reference
Oil Palm Leaves	Pyrolysis	700 °C	N ₂ gas	3 h	b-GO	(Nasir <i>et al.</i> 2017)
Palm Kernel Shell	Pyrolysis	700 °C	N ₂ gas	3 h	b-GO	(Nasir <i>et al.</i> 2017)
Empty Fruit Brunch	Pyrolysis	700 °C	N ₂ gas	3 h	b-GO	(Nasir <i>et al.</i> 2017)
	Carbonization	800 °C	Air	1 h	b-G	(Hendriansyah <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • Pyrolysis • Graphitization 	<ul style="list-style-type: none"> • 350 °C • 900 °C 	N ₂ gas	2 h	b-G	(Widiatmoko <i>et al.</i> 2019)

Palm Oil Mill Effluent	Carbonization	800 °C	Air	1 h	b-G	(Hendriansyah <i>et al.</i> 2017)
	Carbonization	500 °C	Air	1 h	b-G	(Teow <i>et al.</i> 2019)
Oil Palm Fibres	CVD Pyrolysis	1020 °C	Ar and H ₂ gas	30 min	b-G	(Ayuma <i>et al.</i> 2017)
Palm Oil	DTCVD Pyrolysis	900 °C	Ar gas	10 min	b-GO	(Mamat <i>et al.</i> 2018)
	DTCVD Pyrolysis	450 °C	Ar gas	15 min	b-GO	(Robiah <i>et al.</i> 2017)
	CVD Pyrolysis	800 °C	Ar gas	30 min	b-G	(Rahman <i>et al.</i> 2014)
Sugarcane bagasse	Carbonization	300 °C	Air	10 min	b-G	(Somanthan <i>et al.</i> 2015)
	Carbonization	350 °C	Air	30 min	b-RGO	(Debbarma <i>et al.</i> 2019)
	Carbonization	550 °C	Air	3 h	b-G	(Supriyanto <i>et al.</i> 2018)
	Carbonization	300 °C	Air	20 min	b-GO	(Xiao and Yu 2018)
	Pyrolysis	900 °C	N ₂ gas	1 h	b-G	(Xiao <i>et al.</i> 2017)
	Hydrothermal	500 °C	N ₂ gas	2 h	bG-QDs	(Chai <i>et al.</i> 2019)
	Carbonization	450 °C	Air	5 days	b-G	(Akhavan <i>et al.</i> 2014)
	<ul style="list-style-type: none"> • Pyrolysis • Hydrothermal • Pyrolysis 	<ul style="list-style-type: none"> • 450 °C • 240 °C • 1200 °C 	<ul style="list-style-type: none"> • N₂ gas • Air • N₂ gas 	1 h	b-G	(Tang <i>et al.</i> 2018)
Rice Husks	Pyrolysis	550 °C	Air	3 h	b-G	(Supriyanto <i>et al.</i> 2018)
	Pyrolysis	700 °C	N ₂ gas	2 h	bG-QDs	(Wang <i>et al.</i> 2015)
	Carbonization	900 °C	Air	2 h	b-G	(Singh <i>et al.</i> 2017)
	Carbonization	850 °C	Air	2 h	b-G	(Seitzhanova <i>et al.</i> 2019)
	<ul style="list-style-type: none"> • Carbonization • Chemical activation 	<ul style="list-style-type: none"> • 400 °C • 800 °C 	Air	2h	b-G	(Ismail <i>et al.</i> 2018)
	<ul style="list-style-type: none"> • Carbonization • Chemical activation 	<ul style="list-style-type: none"> • 500 °C • 700 °C 	Air	2 h	b-G	(Sankar <i>et al.</i> 2017)
	Pyrolysis	1300 °C	Ar gas	15 min	b-G	(Fujisawa <i>et al.</i> 2019)
	<ul style="list-style-type: none"> • Pyrolysis • Chemical activation 	<ul style="list-style-type: none"> • 300 °C • 850 °C 	Ar gas	<ul style="list-style-type: none"> • 48 min • 2 h 	b-G	(Azizovna <i>et al.</i> 2018)
Wheat straw	<ul style="list-style-type: none"> • Hydrothermal • Pyrolysis • Pyrolysis 	<ul style="list-style-type: none"> • 150 °C • 800 °C • 2600 °C 	<ul style="list-style-type: none"> • Air • N₂ gas • Ar gas 	<ul style="list-style-type: none"> • 6 h • 3 h • 5 min 	b-G	(Chen <i>et al.</i> 2016)
	CVD	800 °C	N ₂ gas	120 min	b-G	(Fathy 2017)
Lignin biomass	Pyrolysis	1000 °C	Ar gas	90 min	b-G	(Liu <i>et al.</i> 2017)
	Pyrolysis	1100 °C	Ar gas	1 h	b-GO	(Li <i>et al.</i> 2019)
	Hydrothermal	180 °C	Air	12 h	b-G	(Ding <i>et al.</i> 2018)
	Hydrothermal	180 °C	Air	12 h	b-G	(Ding <i>et al.</i> 2020)
Chitosan	<ul style="list-style-type: none"> • Pyrolysis • Chemical activation 	<ul style="list-style-type: none"> 800 °C 900 °C 	N ₂ gas	<ul style="list-style-type: none"> • 3 h • 2 h 	b-G	(Hao <i>et al.</i> 2015)

Dog Faces	CVD	1050 °C	H ₂ /Ar gas	10 min	b-G	(Ruan <i>et al.</i> 2011)
Cockroach	CVD	1050 °C	H ₂ /Ar gas	10 min	b-G	(Ruan <i>et al.</i> 2011)
Mango peel	Pyrolysis	750 °C	Hydrogen and argon gas	15 min	b-G	(Shah <i>et al.</i> 2018)
Camphor leaves	Pyrolysis	1200 °C	Nitrogen gas	4 min	b-G	(Shams <i>et al.</i> 2015)
	<ul style="list-style-type: none"> • Pyrolysis • CVD 	<ul style="list-style-type: none"> • 200 °C • 1020 °C 	H ₂ gas	30 min	b-G	(Kalita <i>et al.</i> 2011)
Soybeans	Pyrolysis	800 °C	Nitrogen gas	2 h	b-G	(Sha <i>et al.</i> 2019)
Spruce bark	<ul style="list-style-type: none"> • Hydrothermal • Pyrolysis 	<ul style="list-style-type: none"> • 180 °C • 1000 °C 	<ul style="list-style-type: none"> • Air • N₂ gas 	<ul style="list-style-type: none"> • 12 h • 2 h 	b-G	(Zhongxin <i>et al.</i> 2018)
Populus wood	Pyrolysis	950 °C	Nitrogen gas	1 h	b-G	(Ekhlasi <i>et al.</i> 2020)
Macadamia nut shell	<ul style="list-style-type: none"> • Hydrothermal • Pyrolysis 	<ul style="list-style-type: none"> • 180 °C • 800 °C 	<ul style="list-style-type: none"> • Air • Argon gas 	<ul style="list-style-type: none"> • 12 h • 2 h 	b-G	(Lu <i>et al.</i> 2019)
Peanut shell	Pyrolysis	800 °C	Argon gas	2 h	b-G	(Purkait <i>et al.</i> 2017)
Bengal gram bean husk	<ul style="list-style-type: none"> • Pyrolysis • Graphitization 	<ul style="list-style-type: none"> • 400 °C • 850 °C 	Nitrogen gas	2 h	b-G	(Gupta <i>et al.</i> 2019)
	<ul style="list-style-type: none"> • Pyrolysis • Graphitization 	<ul style="list-style-type: none"> • 400 °C • 900 °C 	Nitrogen gas	2 h	b-G	(Gupta <i>et al.</i> 2019)
Newspaper	Carbonization	450 °C	Air	5 days	b-G	(Akhavan <i>et al.</i> 2014)

Thermal Treatment

The term carbonization is often used in the context of converting biomass into carbon-related materials. Carbonization is a process in which the subjected material undergoes a heating process to remove light molecular weight compounds. This process increases the carbon content of said material, hence the term. In producing b-G, researchers often have used a low-temperature carbonization process followed by a high-temperature graphitization process (Akhavan *et al.* 2014; Widiatmoko *et al.* 2019). The graphitization process affects the structure of the carbon-material into a graphite-like structure. A thermal carbonization process in the absence of oxygen also known as pyrolysis. It is a well-known process for converting biomass into bio-char, bio-oil, bio-gas, *etc.* The product depends on the parameters set during pyrolysis, including the temperature, heating rate, and holding time. For example, higher temperature results in a more char-like product. A fast heating rate produces more bio-oils, while a slow heating rate promotes re-polymerization of the structure, *i.e.*, more bio-chars. The flexibility in choosing the outcome is one reason why many researchers produce graphene using this route. The other reason is the lack of oxygen during the reaction. A carbonization process surrounding by air can be used to produce oxidized carbon material which can be exfoliated further into b-GO (Gupta *et al.* 2019).

The decomposition of the biomass generally starts at about 300 °C, depending on the biomass (Liu *et al.* 2011). The main product is small gas molecules; as during pyrolysis, 70% of the biomass will be decomposed as organic materials (Takeno *et al.* 2006). Debbarma *et al.* 2019 used different pyrolysis temperatures to convert sugarcane bagasse to b-RGO. They used three different temperatures of 250, 350, and 450 °C at different heating rates (1 h, 30 min, and 10 min). The different temperatures and heating rates were important for the degradation of glucose monomers inside the sugarcane bagasse. At 200

°C, glucose monomers degrade into polyaromatic rings, which stack together into a graphitized-like structure (Debbarma *et al.* 2019; Tursi *et al.* 2019). The optimum formation of graphene sheets occurred at 350 °C, as lower temperatures resulted in less aromatic structures, but higher temperatures caused more particle formation rather than sheet formation. Widiatmoko *et al.* (2019) used pyrolysis to convert oil palm empty fruit bunch (EFB) to bio-based graphene using two-stage pyrolysis. The first stage is carbonization at 350 °C for 60 min, and the second stage is graphitization at 900 °C for 90 min under nitrogen atmosphere. The two-stage pyrolysis forces volatile molecular compounds out of the biomass during the first stage. Oil palm EFB is lignocellulosic biomass, and the target temperature for obtaining high char is based on lignin degradation. The second stage for graphitization activates the bio-char to form graphite-like compounds through long pyrolysis. Most importantly, this carbonized/graphitized process enhancing the honeycomb-structure order. Further, metal catalyst is used to diffuse the carbon onto its surface. The same route was used by Ismail *et al.* (2018) to produce graphene from rice husk using two-stage pyrolysis. The first stage was conducted at 400 °C for 2 h. The difference in temperature might reflect the inorganic silica compounds in the rice husk (Uzunov *et al.* 2012). Most pyrolysis of rice husk requires a mixture with potassium hydroxide to release the silica entrapped within the structure. In both cases, the carbonization temperature was set for the decomposition of lignin, as it holds the most carbon linkages of the lignocellulosic components. By concentrating on this portion, one can increase the fixed carbon content, which can be activated later to a graphite-like structure. The high temperature might be the best way to break the biomass structure, but the drawback is the increased overall process time.

Another thermal treatment is hydrothermal carbonization (HTC). The HTC technique is used to overcome the high moisture content in certain biomass. High moisture content makes pyrolysis difficult, as it necessitates high heat for vaporization (Zhao *et al.* 2014). HTC is based on the formation of coal by the combination of pressure and heat. The combination of these forces brings water into its subcritical region. The subcritical water can act as an acid-like catalyst *via* the dissociation of H⁺ ions due to weakened hydrogen bonds. The increase of acidic hydronium ions (H₃O⁺) formation promotes the degradation of biomass (Marcus *et al.* 1999). Thus, HTC is similar to pyrolysis, except for the added pressure, which results in a much lower degradation temperature. Furthermore, the severity of the degradation is controllable with residence time. The reaction time determines the degree of re-polymerization and monomers, as in pyrolysis (Gao *et al.* 2013). By increasing the reaction time, graphite-like condensed aromatic structures are achieved. Unlike pyrolysis, the involvement of water results in high oxygenated functional groups in the structure. Besides, the pre-cursor sample underwent HTC treatment, which resulted in less amorphous carbon formation with more graphite-like structure formation (Barin *et al.* 2014). Huang *et al.* (2018) produced b-GO from fertilizer using HTC. The decomposition of the fertilizer occurred in a stainless steel autoclave at 190 °C for 10 h. The long reaction time was used to increase the formation of sp² carbon content. Chen *et al.* (2016) used a combination of HTC and pyrolysis to synthesize graphene from wheat straw. The HTC was used as carbonization in an autoclave before the biochar was introduced to graphitize by pyrolysis in a furnace. The purpose of the multiple stages of thermal treatment was to unlock the complex structure of wheat straw. Hence, highly concentrated alkali (KOH) was mixed in the autoclave to liquefy the hemicellulose and lignin and to saponify the wax produced from the wheat straw during HTC. Any other thermal treatment can be used as long as the outcome increases the fixed carbon content. Most research on bio-based graphene focuses on increasing the production of graphene; thus, there are no researchers using microwave pyrolysis due to its production limitation.

Exfoliation Method

The exfoliation method with graphitized biomass involves peeling the bulk carbons structure by overcoming the van der Waals force, resulting in graphene sheets. Exfoliation methods used in producing graphene from graphite can be used by substituting it with graphitized biomass. The severity of thermal treatment plays an important role in graphene formation. Chen *et al.* (2016) graphitized the biomass as high as 2600 °C with a combination of multiple thermal treatments, which resulted in the formation of a few layers graphene. They attributed the formation of bio-graphene to the high degree of thermal treatments, which separated crystalline cellulose. The extreme temperature forced the degraded cross-linked polymers into a honeycomb-like structure. Sonication can help with mechanical separation for graphene formation without requiring extreme thermal treatment. Dhand *et al.* (2013) compared the graphene structure with and without sonication. Morphologically, the grain size distribution for the graphene decreases with sonication, which indicates that more exfoliation has occurred. Shams *et al.* (2015) exfoliated bio-char from camphor leaves using sonication. The biomass was treated at 1200 °C, and few graphene layers were suspended after 15 min of sonication. The average I_D/I_G of the graphene was comparable to that of pure graphene.

Another exfoliation technique is oxidizing the bio-char by the Hummers method or any other oxidizing method similar to producing GO using graphite (Hummers and Offerman 1958). The introduction of functional groups forces separation by intercalation between the layers of the bio-char. The Hummers method was derived from Brodie's oxidation method of producing graphite oxide by chemically treated graphite with acids and alkali metals. This process has been the basis for increased GO production. Seitzhanova *et al.* (2019) produced GO from rice husk using the Hummers method. The bio-char was chemically treated with NaNO_3 and H_2SO_4 while being stirred in a round bottom flask. KMnO_4 was introduced slowly after 30 min of reaction. The oxidizing process of bio-char to GO was noticeably faster compared with the Hummers method using graphite powder. A similar result was reported by Li *et al.* (2019) with an oxidation time of 1 h. Morphologically, the GO derived from biomass had a smaller lateral size than the GO derived from graphite. However, a smaller size can be achieved by applying sonication to promote a few layers exfoliation. Another oxidation technique done was based on Marcano's improved oxidation method (Marcano *et al.* 2010). In other words, they managed to remove toxic gas production from the original Hummers method by removing the usage of NaNO_3 . Instead, an excessive amount of KMnO_4 was used as the replacement. Nasir *et al.* (2017) used this oxidation technique to produce GO from oil palm waste. The GO-derived from biomass was similar to GO derived from graphite, albeit with a few defective sites.

Carbon Growth

The long chains of carbon, hydrogen, and oxygen compounds in biomass break down into smaller molecules in the form of gases and condensable vapours during pyrolysis. Using pyrolysis, the process can be customized to produce certain outcomes using the three parameters mentioned above. A CVD-like process can be used for growing carbon onto a metal surface. The carbon-based gases generated from biomass are deposited onto the metal surface, much like in CVD. This process was used by Sun *et al.* (2010); as the growth of graphene was supplied by a polymer instead of gaseous raw material in CVD. Alternative carbon sources are beneficial because the gaseous raw material is expensive, making it difficult to apply at the industrial scale. Using this principle, Mamat *et al.* (2018) pyrolyzed palm oil that generates a gaseous carbon source and deposited the product onto a Ni surface. They established that excessive carbon overwhelms the Ni surface, resulting in a densely compacted graphene structure. The goals of using CVD to produce graphene

are to acquire high-quality graphene or to create a graphene/metal composite. The transferability of graphene is quite problematic, and CVD is a way to overcome this tricky situation (Suk *et al.* 2011; Yang *et al.* 2013; Lavin-Lopez *et al.* 2014). Therefore, a CVD-like process by mixing metal pre-cursor with biomass has been established. The metal catalyst provides surface area for graphene growth and can be dispersed away if needed. Liu *et al.* (2019) mixed iron powder with biomass (kraft lignin) to prepare b-G. The metal melts together with lignin during pyrolysis, allowing the amorphous carbons to encapsulate the melted metal. During this time, the metal is in its nanoparticle form, while the biomass is in metastable amorphous carbon form. At elevated temperature, the process of dissolution and precipitation occurs between the amorphous carbon and the metal surface, resulting in metal encapsulated with graphene as the end-product. Magnetic separation was used to separate the iron particle and graphene structure after washing several times with de-ionized water. For any other metal, an acid wash can be used to separate metal particles from the graphene (Leng *et al.* 2016; Widiatmoko *et al.* 2019).

Using metal catalysts, the degradation of the biomass is excessive, which lowers the yield of carbon deposition onto the metal surface. To overcome this problem, Leng *et al.* (2016) performed low-temperature pyrolysis at 300 °C. They controlled the amount of methane gas formed and their deposition onto the metal surface. Additionally, the graphene produced using this method had few defects. However, only a small percentage of graphene was produced, due to the self-limitation of the solid carbon as the carbon source. The biomass can be the carbon source, but due to the complex nature of the structure, it might be difficult for the whole process to be efficient. Yan *et al.* (2018) studied different atmospheric effects on graphene production in pyrolysis. The welding gas atmosphere yielded the most graphene production, as more lignocellulosic components were converted. Under a reducing atmosphere (*e.g.*, H₂), more liquid products were produced due to oxygen conversion in oil fractions, which catalytically transformed it into water. The presence of a reducing agent during pyrolysis can clean up the graphene by removing the oxygen content on the defected graphene structure. This effect was studied by He *et al.* (2019) by using H₂ atmosphere in the pyrolysis of chitosan. The effect of H₂ presence in the pyrolysis was reflected with the low defects of graphene.

Characterization of Bio-based Graphene

For both exfoliation and carbon growth methods, the quality of the graphene is based on the thermal treatment. The thermal treatment affects the product as the conditions during the thermal treatment played an important role in the degradation of the biomass. Biomass is a complex structure, and by using multiple forms of thermal treatment, one might be able to unlock the graphene structure underneath it. However, the cost and practicality of such advanced thermal treatment are questionable. Hence, the use of metal catalysts during pyrolysis is popular. Combining a catalyst with a purging atmosphere during the pyrolysis makes high-quality graphene, similar to that traditionally formed using graphite. The different profile of bio-based graphene is observed by Raman characterization. Table 2 shows the characteristics of bio-based graphene produced from different biomass and thermal treatment. Most of the bio-based graphene have almost flat and broad 2D bands, indicating unstructured multiple layers of graphene (Liu *et al.* 2017). The unstructured layers were due to the complex nature of the biomass; therefore, it is quite difficult to produce high-quality graphene through this process. The oxidation might occur during the pyrolysis as it is impossible to remove the presence of oxygen completely, especially from the biomass itself. High thermal treatment can be the solution (He *et al.* 2019). The defected and multilayers of bio-based graphene might not have the same high quality as graphite-derived graphene, but defective sites of graphene open up the band-gap modulation of graphene suitable for electronic applications (Liu *et al.* 2015; Zaminpayma

et al. 2017). Therefore, the purification process to remove the metal impurities completely is optional. Hence, this method can produce graphene/metal composite as the end-product. The green synthesis route of producing graphene is not entirely a promising alternative quality-wise. However, its success lies in its capability for mass production in large industries. The CVD method can be the answer to high-quality graphene production. By substituting biomass as a carbon source in place of carbon gases, high-quality graphene can be generated. This method sacrifices the quantity of production for higher quality but using biomass as the carbon source might be beneficial in the long term, financially speaking.

Table 2. Graphene Characterization for Different Biomass and Process

Sources	Thermal Treatment	Graphene Properties					Product	Ref.
		I_D/I_G	I_{2D}/I_G	C/O	Layers	Thickness		
Oil Palm Leaves	Hummers method	1.06	-	-	Multi	-	b-GO	(Nasir <i>et al.</i> 2017)
	Hydrolysed with H_2SO_4	4.0	-	-	Multi	-	b-GO	(Fathy <i>et al.</i> 2019)
Palm Kernal Shell	Hummers method	1.14	-	-	Multi	-	b-GO	(Nasir <i>et al.</i> 2017)
Empty Fruit Brunch	Hummers method	1.16	-	-	Multi	-	b-GO	(Nasir <i>et al.</i> 2017)
	Mixing with $FeCl_3$ and $ZnCl_2$	-	-	-	Multi	-	b-G	(Hendriansyah <i>et al.</i> 3017)
	Mixing with $FeCl_3$ and $ZnCl_2$	1.16	0.58	-	4 layer	-	b-G	(Widiatmoko <i>et al.</i> 2019)
Palm Oil Mill Effluent	Mixing with $FeCl_3$	-	-	-	Multi	-	b-G	(Hendriansyah <i>et al.</i> 2017)
	Pre-treatment with H_2SO_4	-	-	3:2	Multi	-	b-G	(Teow <i>et al.</i> 2019)
Oil palm fibres	CVD method	0.16	0.3	70:1	3 layers	-	b-G	(Ayuma <i>et al.</i> 2017)
Palm oil waste	CVD with Nickel substrate	0.04	0.28	4.72	Multi	-	b-GO	(Mamat <i>et al.</i> 2018)
	DT-CVD with Nickel substrate	-	0.43	-	3 layers	-	b-GO	(Robaiah <i>et al.</i> 2017)
	DT-CVD with Nickel substrate	-	1.1	-	3 layers	-	b-G	(Rahman <i>et al.</i> 2014)
Sugarcane bagasse	Mixed with ferrocene	0.76	-	High oxygen	Multi	-	b-G	(Somanathan <i>et al.</i> 2015)
	Carbonize under atm	-	-	-	Multi	-	b-G	(Debbarma <i>et al.</i> 2019)
	Carbonize under atm	0.85	-	-	Multi	-	b-G	(Supriyanto <i>et al.</i> 2018)
	Mixed with Ferrocene	1.16	-	4.27	-	0.34	bRGO	(Li <i>et al.</i> 2018)
	Mixing with $FeCl_3$ and $ZnCl_2$	0.95	-	-	Few layer	-	b-G	(Xiao and Yu 2018)
	Hummers method	-	-	High Carbon	2-11 layer	1.2 nm	b-GO	(Xiao <i>et al.</i> 2017)

	Hydrothermal	0.5	0.55	-	4 layers	3.2 nm	bGQDs	(Akhavan <i>et al.</i> 2014)
	Hydrothermal	-	-	1.05	3 layers	2.26 nm	b-G	(Chai <i>et al.</i> 2014)
	Delignification and hydrothermal	1.23	0.33	10:1	3-5 layers	5 nm	b-G	(Tang <i>et al.</i> 2018)
Rice Husks	Carbonize open air	0.84	-	-	Multi	-	b-G	(Supriyanto <i>et al.</i> 2018)
	Chemical activation with NaOH	0.8	-	-	Multi	20 nm[41]	bGQDs	(Wang <i>et al.</i> 2015)
	Chemical activation with NaOH	0.55	0.67	-	Multi	-	b-G	(Singh <i>et al.</i> 2017)
	Exfoliation with H ₂ SO ₄	0.87	1.56	-	Multi	70µm	b-G	(Seitzhanova <i>et al.</i> 2019)
	Chemical activation with NaOH and sonication	0.93	0.19	-	Multi	-	b-G	(Ismail <i>et al.</i> 2018)
	Chemical activation with NaOH	0.95	0.36	-	5 layers	5 nm	b-G	(Sankar <i>et al.</i> 2017)
	Carbothermic reduction with silicon carbide	1.00	-	-	Multi	-	b-G	(Fujisawa <i>et al.</i> 2019)
	Chemical activation with NaOH and exfoliation with H ₂ O ₂	0.29	2.06	-	Multi	-	b-G	(Azizovna <i>et al.</i> 2018)
Wheat Straws	Hydrothermal and sonication	1.37	0.61	-	Multi	3.5 nm	b-G	(Chen <i>et al.</i> 2016)
	Hydrothermal and CVD	0.75	-	-	-	-	b-G	(Fathy 2017)
Lignin Biomass	Mixing with iron powder	1.16	0.28	-	4 layers	4 nm	b-G	(Liu 2017)
	Iron(III) nickel nonahydrate	1.2	-	-	3 layers	3 nm	B-GO	(Li <i>et al.</i> 2019)
	Hydrothermal	0.94	-	-	Multi	6 nm	bGQDs	(Ding <i>et al.</i> 2018)
	Hydrothermal	0.71	-	-	Multi	2 nm	b-G	(Ding <i>et al.</i> 2020)
Chitosan	Chemical Activation with KOH	0.74	-	-	4 layers	1.5 nm	b-G	(Hao <i>et al.</i> 2015)
Chicken bone	Mixing with FeCl ₃ and ZnCl ₂	0.3	0.65	-	3 layers	3 nm	b-GO	(Akhavan <i>et al.</i> 2014)
Cow dung	Mixing with FeCl ₃ and ZnCl ₂	0.5	0.55	-	3 layers	3 nm	b-GO	(Akhavan <i>et al.</i> 2014)
Dog faces	CVD with Cu Substrate	0.15	1.8	-	Mono layer	-	b-G	(Ruan <i>et al.</i> 2011)
Cockroach leg	CVD with Cu Substrate	-	4.00	-	Mono layer	-	b-G	(Ruan <i>et al.</i> 2011)

Mango Peels	CVD and plasma exposure	0.5	2.76	-	1-2 layer	-	b-G	(Shah <i>et al.</i> 2018)
Camphor	Sonication	0.99	-	-	7 layers	2.37 nm	b-G	(Shams <i>et al.</i> 2015)
	CVD with Cu substrate	0.08	-	-	3 layers	2 nm	b-G	(Kalita <i>et al.</i> 2011)
Soy beans	Dispersed in DMF	1.092	-	9:1	-	-	Nitrogen-doped graphene	(Sha <i>et al.</i> 2019)
Spruce Bark	Hydrothermal	-	-	-	7 layer	10 nm	b-G	
Populus Wood	Pyrolysis	-	-	-	3 layer	50 nm	b-G	(Sun <i>et al.</i> 2018)
Macadamia nut shell	Hydrothermal	1.18	-	-	Multi	-	b-G	(Lu <i>et al.</i> 2019)
Peanut shell	Chemical activation with KOH and sonication with	1.01	--		Multi	-	b-G	(Purkait <i>et al.</i> 2017)
Bengal Gram Bean Husk	Chemical activation with KOH	0.84	-	-	Multi	5 nm	b-G	(Gupta <i>et al.</i> 2019)
	Mixing with FeCl ₃	0.85	-	-	Multi	10 nm	b-G	(Gupta <i>et al.</i> 2019)
Fruit cover plastic	CVD	0.6	0.4	70:1	Multi	-	b-G	(Ayuma <i>et al.</i> 2017)

AGRICULTURAL WASTES

Agriculture wastes are generated yearly. They are the unwanted residues from agricultural activities, including trunks, leaves, husks, or extracted forms like lignin. These agricultural wastes are worth mentioning due to the large quantities that are available annually at predictable locations. Agriculture wastes are lignocellulosic biomass, also known as plant dry matter. The primary components of lignocellulosic are cellulose, hemicellulose, and lignin, as shown in Fig. 3. The percentages of these components differ based on the type of plant. Cellulose is the cell wall of the green plants. It is a chain of glucose units forming a polysaccharide polymer. Hemicellulose is also present in the cell wall of the green plants and is a polysaccharide. The difference between these molecules is in the glucose monomer compositions. Fig. 3(b) shows a partial structure of hemicellulose; one among several common variations of 50 to 200 monomeric units and a few simple sugar residues in a hemi-cellulose structure. Lignin is more complex than cellulose and hemicellulose. As such, lignin possesses toughness and can be found in the cell walls of wood and bark (Isikgor and Becer 2015; Nanda *et al.* 2016). Lignin appears to result from a semi-random polymerization among monolignols that can include those shown in Fig 3(c), where the ratio is different depending on the plant species.

Agriculture wastes are problematic around the world, and policies regarding the recycling of agricultural wastes have been established. However, the generation of agricultural waste far exceeds the capacity for recycling. Therefore, the implementation of biomass in graphene production can be beneficial for all parties. The agriculture wastes need to be processed before conversion into graphene. For example, tree trunks needed to be physically treated by chopping, shredding, and drying before entering the thermal treatment. The process of converting biomass into graphene starts from the pre-treatment before applying the thermal treatment. The complexity of the biomass structure requires

some steps in ensuring a high yield of graphene. The treatment process for bio-based graphene production with different agriculture waste is summarized in Table 3.

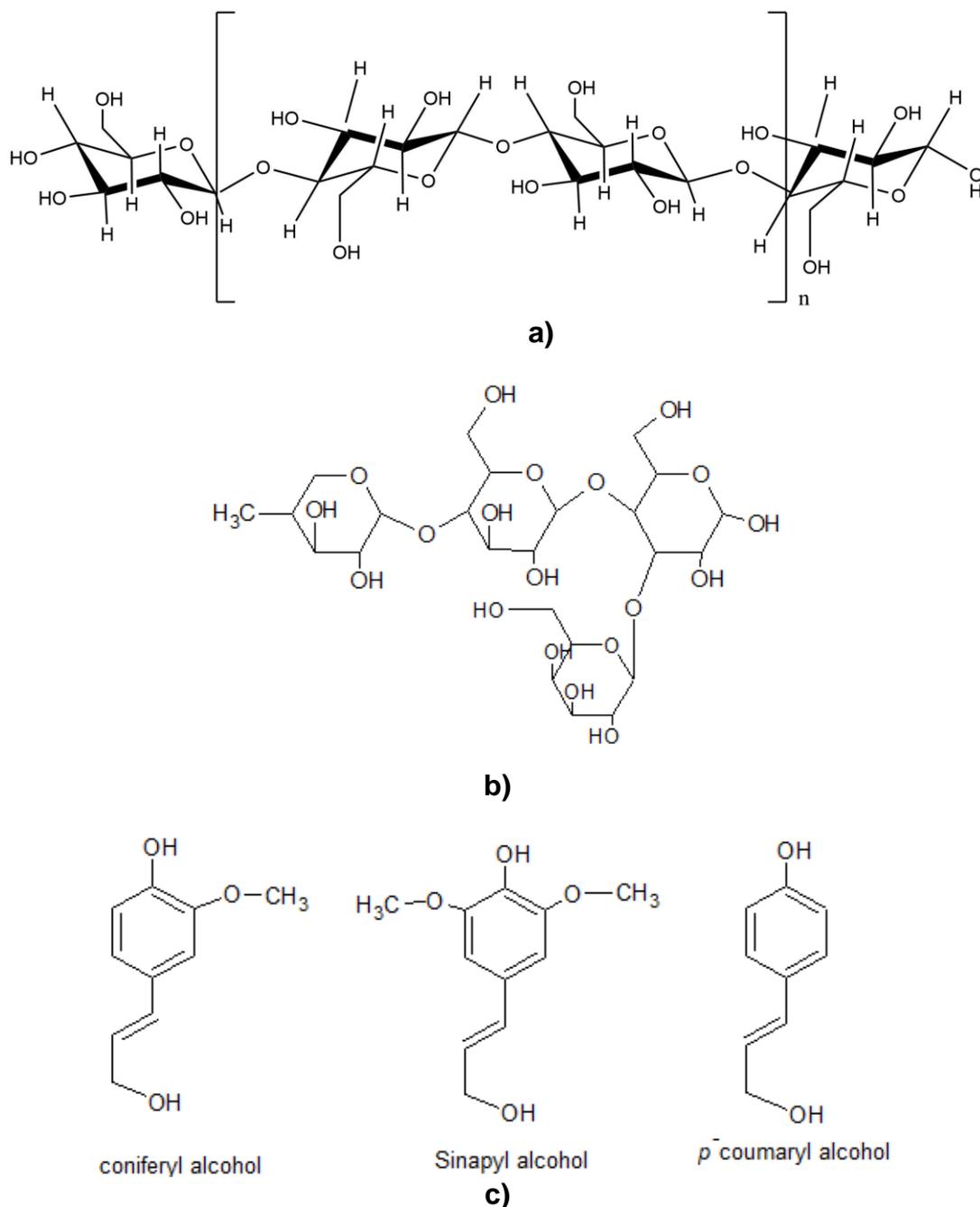


Fig. 3. The structure of a) cellulose, b) hemicellulose, and c) lignin

Table 3. Treatment Process for Agriculture Waste in Producing Graphene

Sources	Experimental detail	Product	Ref
Oil Palm Leaves	<ul style="list-style-type: none"> • Pre-treatment • Pyrolysis • Hummers method 	b-GO	(Nasir <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • Hydrolyzed with H_2SO_4 • Delignification • Catalytic Acid Spray 	b-GO	(Fathy <i>et al.</i> 2019)
Palm Kernal Shell	<ul style="list-style-type: none"> • Pre-treatment • Pyrolysis • Hummers method 	b-GO	(Nasir <i>et al.</i> 2017)

Empty Fruit Brunch	<ul style="list-style-type: none"> • Pre-treatment • Pyrolysis • Hummers method 	b-GO	(Nasir <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • Pre-treatment • Mixing with metal catalyst • Pyrolysis • Acid Purification 	b-G	(Hendriansyah <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • Mixing with metal catalyst • Two-stage Pyrolysis • Acid Purification 	b-G	(Widiatmoko <i>et al.</i> 2019)
Palm Oil Mill Effluent	<ul style="list-style-type: none"> • Mixing with metal Catalyst • Pyrolysis 	b-G	(Hendriansyah <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • Pre-treatment with H₂SO₄ • Pyrolysis 	b-G	(Teow <i>et al.</i> 2019)
Oil Palm Fiber	<ul style="list-style-type: none"> • Carbonization • CVD method 	b-G	(Ayuma <i>et al.</i> 2017)
Palm Oil waste	<ul style="list-style-type: none"> • Washed and Filtered • Double thermal CVD Method with Nickel substrate 	b-GO	(Mamat <i>et al.</i> 2018)
	<ul style="list-style-type: none"> • Washed and Filtered • DT-CVD method with Nickel substrate 	b-GO	(Robaiah <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • DT-CVD method with Nickel Substrate • Acid Purification with Nitric acid 	b-G	(Rahman <i>et al.</i> 2014)
Sugarcane bagasse	<ul style="list-style-type: none"> • Mixing with Ferrocene • Carbonization 	b-G	(Somanathan <i>et al.</i> 2015)
	<ul style="list-style-type: none"> • Pre-treatment dried under sunlight • Carbonization at atmosphere • Acid Purification with Nitric Acid 	b-G	(Debbarma <i>et al.</i> 2019)
	<ul style="list-style-type: none"> • Carbonization at atmosphere • Purification with Hydrofuran (HF) 	b-G	(Supriyanto <i>et al.</i> 2018)
	<ul style="list-style-type: none"> • Washed and dried • Mixing with Ferrocene • Carbonization • Convert to rGO by mixed with Copper (II) ions 	b-RGO	(Xiao and Yu 2018)
	<ul style="list-style-type: none"> • Washed and dried • Mixing with metal catalyst • Pyrolysis • Acid Purification with HCl 	b-G	(Xiao <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • Imperfect burning in chimney for 5 days • Mixing with metal catalyst • Hummers method 	b-GO	(Akhavan <i>et al.</i> 2014)
	<ul style="list-style-type: none"> • Washed and dried • Hydrothermal • Pyrolysis • Water purification 	b-GQD	(Chai <i>et al.</i> 2019)
	<ul style="list-style-type: none"> • Delignification • Pyrolysis • Hydrothermal 	b-G	(Tang <i>et al.</i> 2018)
Rice husk	<ul style="list-style-type: none"> • Washed and dried • Carbonization • Purification with Hydrofuran (HF) 	b-G	(Supriyanto <i>et al.</i> 2018)
	<ul style="list-style-type: none"> • Washed, sieved and dried • Pyrolysis 	b-GQDs	(Wang <i>et al.</i> 2015)

	<ul style="list-style-type: none"> • Chemical activation with NaOH at 900 °C atmosphere • Water purification 		
	<ul style="list-style-type: none"> • Washed • Carbonization open air • Chemical activation with KOH • Pyrolysis • Water purification 	b-G	(Singh <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • Preliminary carbonization • Desilication • Chemical Activation with KOH • Exfoliation with H₂SO₄ 	b-G	(Seitzhanova <i>et al.</i> 2019)
	<ul style="list-style-type: none"> • Pre-carbonization open air • Chemical activation with KOH • Centrifuge • Sonication 	b-G	(Ismail <i>et al.</i> 2018)
	<ul style="list-style-type: none"> • Pre-carbonization open air • Chemical activation with KOH • Water purification 	b-G	(Sankar <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • Pyrolysis • Carbothermic reduction with silicon carbide • Sublimation Si and graphene formation 	b-G	(Fujisawa <i>et al.</i> 2019)
	<ul style="list-style-type: none"> • Washed and dried • Pyrolysis • Desilication with NaOH • Chemical Activation With KOH • Exfoliation with H₂O₂ 	b-G	(Azizovna <i>et al.</i> 2018)
Wheat straws	<ul style="list-style-type: none"> • Washed and dried • Hydrothermal • Pyrolysis • Chemical activation with KOH • Ultrasonication • Acid purification with HCl 	b-G	(Chen <i>et al.</i> 2016)
	<ul style="list-style-type: none"> • Washed, milled and dried • Pre-treatment with NaOH • Refluxing in acid medium • Hydrothermal • CVD 	b-G	(Fathy 2017)
Lignin waste	<ul style="list-style-type: none"> • Mixing with Metal Catalyst • Pyrolysis • Acid purification with HCl 	b-G	(Liu <i>et al.</i> 2017)
	<ul style="list-style-type: none"> • Mixing with metal catalyst • Pyrolysis • Hummers method 	b-GO	(Li <i>et al.</i> 2019)
	<ul style="list-style-type: none"> • Pre-treatment with Nitric acid • Ultrasonication • Hydrothermal • Purification 	b-GQDs	(Ding <i>et al.</i> 2018)
	<ul style="list-style-type: none"> • Hydrothermal • Centrifugation • Purification by dialysis • Plasma treatment 	b-G	(Ding <i>et al.</i> 2020)

Oil Palm Waste

The oil palm waste is another type of agricultural waste that is abundantly produced currently. The wastes generated from oil palm industries are up to a million tonnes annually (Abdullah *et al.* 2013). The agronomy of oil palm trees represents major economic expansions for countries such as Malaysia and Indonesia. The main harvest for oil palm is its fruit, which contains edible oils. Approximately 45% of the fruit bunches are used for palm oil, and the rest is thrown away as waste. To harvest the fruit, the trunks, leaves, and fronds are chopped down as wastes. Most oil palm industries utilize these wastes to generate electricity for the plant. However, only 25% can be used for this purpose; the remaining 75% has a high moisture content, which requires additional drying. Most of them are left rotten, unused (Awalludin *et al.* 2015). The waste from oil palm industries consist of wastes produced from the oil pressing mill, *i.e.*, mesocarp fiber (MF), empty fruit bunches (EFB), oil palm shell (OPS), palm oil mill effluent (POME), *etc.*, and wastes produced from the farm, *i.e.*, oil palm trunks (OPT), oil palm fronds (OPF), *etc.* The amount of wastes generated yearly keeps on increasing to the level where the accumulated wastes have overwhelmed the time it took for the waste to degrade naturally; hence the importance of the waste management program. Poor management of this accumulated waste leads to environmental pollution.

Table 4. Lignocellulosic Content for Agriculture Biomass Waste

Biomass	Lignin (%)	Cellulose (%)	Hemicellulose (%)	Ref
Oil Palm:				
EFB	32	37	15	(Sudiyani <i>et al.</i> 2013)
PKS	44	28	22	(Zainal <i>et al.</i> 2016)
MF	21	43	33	(Nordin <i>et al.</i> 2013)
OPT	19	56	16	(Lai and Idris <i>et al.</i> 2013)
OPF	15	33	23	(Zakaria <i>et al.</i> 2014)
Rice Husk	35	25	20	(Hossain <i>et al.</i> 2018)
Sugarcane Bagasse	40	30	20	(Cardona <i>et al.</i> 2010)
Wheat Straw	39	29	25	(Rodriguez-gomez <i>et al.</i> 2012)

The utilization of oil palm biomass starts with cleaning up the raw material. Basic washing with deionized water removes impurities before oven drying (Nasir *et al.* 2017). Many physical treatments can be used to convert biomass into powder form. This step is essential in breaking down the lignocellulosic components (Xia 2017). The most common and easy grinding method is the mortar technique. Ball milling was used by Widiatmoko *et al.* (2019) to produce fine and coarse biomass granules. Fathy and Abdel (2014) pre-treated oil palm leaves with H₂SO₄ to generate monomeric sugars. The high lignin content in EFB makes it a desirable source material for bio-based graphene. Although lignin has a complex structure, it is degraded in a wider temperature range than cellulose and hemicellulose (200 to 500 °C) (Strezov *et al.* 2003). Hence, sustainable carbon sources are available even at low temperatures due to lignin degradation.

Rice Husk

Rice cultivation is global, and it is one of the largest food sources, especially in Asia. The demand for rice is rapidly growing every year, and by fulfilling those demands, agriculture waste is also amplified. The waste generated is mostly rice husk; rice husk is recycled for energy as fuel material due to its high calorific value. The by-product of this process is rice husk ash (RHA), which comprises 25% of the raw material (Hossain *et al.* 2018). Traditionally, RHA is disposed of in landfills, which have limitations. Many industries have begun to utilize rice husk as raw material. Rice husk contains high silica and ash contents, which are suitable as raw material for cement, fuel, activation carbon, adsorbent, *etc.* (Liu *et al.* 2012; Franco *et al.* 2016; Prasara-a and Gheewala 2017; Sandhu and Siddique 2017). Table 4 shows the lignocellulosic components of rice husk (Hossain *et al.* 2018). Because rice husk contains almost 20% silica, most biomass utilization requires a pre-treatment with potassium hydroxide or sodium hydroxide. This process releases the entrapped silica impurities inside the rice husk polymeric structure. The silica located in the rigid layer of the structure remains as a by-product with ash after the thermal process. Thus, RHA has been used as raw material for silica production (Abu *et al.* 2016). However, silica is an impurity in graphene and needs to be removed before conversion; pre-treatment with an alkaline solution is necessary. KOH has been used to remove amorphous carbon from rice husk; the product forms a cleaner carbon surface, suitable for making pristine graphene (Muramatsu *et al.* 2014). The KOH induces porosity in carbon materials, making the removal of impurities easier. Rhee *et al.* (2015) found that the amount of KOH influences the morphology of the graphene produced. Because KOH opens the carbon structure, it increases the specific surface area of the graphene. This process is called the activation of the carbon structure. The desilication and activation steps can be done after thermal treatment (Seitzhanova *et al.* 2019). The interaction of KOH and carbon results in the formation of potassium metal, and further purification is needed to eliminate inorganic impurities from graphene. This purification is done by washing with DI water and hydrogen peroxide (Rhee *et al.* 2015). The pre-treatment that is required to enable rice husk as a suitable carbon source for graphene is quite challenging. However, it should be noted that the pre-treatment with KOH has resulted in a high surface area of the b-G. Furthermore, increasing the amount of KOH does increase the amount of graphene component and reduces the content of amorphous carbon (Seitzhanova *et al.* 2019).

Sugarcane Bagasse

Sugarcane is cultivated in tropical and subtropical climates. It is mainly grown for sugar production. Harvesting sugarcane includes cutting the cane, washing, and grinding to extract the juice. The residue from this process is sugarcane bagasse. Almost 28% of sugarcane is retrieved as bagasse after the extraction process (Arni 2018). As lignocellulosic biomass, sugarcane bagasse consists of 20% lignin, 40% cellulose, and 30% hemi-cellulose (Cardona *et al.* 2010). Moreover, sugarcane bagasse consists of a large amount of carbon and oxygen with low ash content. Currently, bagasse is considered a promising feedstock for biofuel production, as it can be processed into bioethanol. The conversion of sugarcane bagasse to bioethanol requires a pre-treatment to increase its degradation. This pre-treatment process can be done chemically, biologically, or physically as long as it increases the accessibility of cellulose, hemi-cellulose, and lignin (Niju and Swathika 2019). The pre-treatment process can be applied for converting it to graphene. The pyrolysis of sugarcane bagasse has been studied extensively. During the thermal treatment of sugarcane bagasse, the decomposition of lignocellulosic components releases glucose and sucrose. The release of these reducing sugars also discharges acids (Wongsiriwan *et al.* 2010). The acids released can reduce GO (Gan *et al.* 2018). Moreover, the loose structure of the bagasse is suitable for HTC. Chai *et al.* (2019) used HTC to

convert sugarcane bagasse into graphene. The purpose of using HTC was to produce fermentation sugar and graphene as the by-product. In this study, the cellulose and hemicellulose high reducing sugar content were converted to sugars, while graphene was derived from the dissolved lignin during HTC. However, most pyrolysis of sugarcane bagasse for graphene production has focussed on cellulose decomposition. Debbarma *et al.* (2019) found that the highest carbon content was achieved with pyrolysis at 350 °C because the cellulose and glucose monomers degrade heavily at this temperature. The glucose monomer in sugarcane bagasse will help the formation of the aromatic structure of graphene.

Wheat Straw

Wheat straw is the by-product of wheat grain cultivation. It is the stalk left behind when the wheat grains are harvested. Similar to other crops, the biomass waste generated from wheat straw agriculture reaches a million tonnes annually. By tradition, these straws are burned, but with recent environmental awareness, it has been tested as a raw material for the production of bioethanol and other bio-products. Wheat straw is composed of 39% cellulose, 20% hemicellulose, and 25% lignin, while the rest is protein and ash (Rodriguez-gomez *et al.* 2012). Wheat straw and rice husk are both from the grain group, which means that they have almost identical traits. However, wheat straw contains a lower percentage of silica than rice husk. Similar to rice husk, pre-treatment with KOH removes silica impurities while chemically activating the wheat straw structure. Wheat straw has high calcium and potassium contents, which require high thermal temperature for processing. Potassium is released from 1000 to 1500 °C (Trubetskaya *et al.* 2016). However, the complicated formation of silicate matrix trapped some of the potassium, which has the consequence that only around 70 % of them are released. Therefore, it is necessary to pre-treat the wheat straw to open up the structure. Another method to loosen the wheat straw structure is by removing the lignin and hemicellulose. Almost 40% of wheat straw is cellulose. Chen *et al.* (2016) used HTC and KOH to dissolve hemicellulose and lignin and converted the wheat straw to cellulose fibres. These cellulose fibres then underwent multiple thermal treatments before they were converted into graphene. Converting wheat grass into cellulose first can be a good method to overcome the complex structure of the biomass. There are many pre-treatments available to increase the percentage of cellulose in the straw biomass, such as acid pre-treatment with sulfuric acid (Chen *et al.* 2011), organic solvent pre-treatment with glycerol (Fuelbiol *et al.* 2015), alkaline pretreatment, *etc.* Like other lignocellulosic materials, cellulose can be converted into graphene or other carbon materials.

Lignin Waste

Lignin is one of the most abundant natural polymers, having high carbon content and high aromaticity (Rinaldi *et al.* 2016). Compared with other lignocellulosic components, the structure of lignin is much more complex. This is evidenced by the wide range of thermal decomposition of the structure, ranging from 200 to 500 °C. The rigidity of lignin is important because it gives mechanical support for the plant wall cell (Bonawitz and Chapple 2010). Lignin is a combination of methoxy, phenolic hydroxyl, and aldehyde groups. Lignin is considered to be a waste material because it is the by-product of bioethanol production. The bioethanol process uses feedstock as raw material and converts cellulose and hemicellulose using hydrolysis. Hydrolysis separates the lignin, which remains as waste. Most lignin produced is recycled as burn material to power factories. Notably, the pulping industry generates tonnes of lignin as it de-lignifies biomass to produce paper. In this case, kraft lignin is the by-product of the pulping process in a liquid form known as black liquor (Gustafsson *et al.* 2017). Similar to bioethanol production,

most of the kraft lignin is utilized as a fuel source within the factory where it has been separated from the cellulose and hemicellulose. Additional steps can be taken to separate kraft lignin from the black liquor, and most solvents used in the pulping process can be recycled from the remaining black liquor by extraction.

Generally, lignin can be used to derive carbon-based material due to its rich phenolic carbon structures (Stojanovska *et al.* 2018). Furthermore, lignin is decomposed within a wider range of temperatures, enabling it to be utilized as a carbon source for both low or high pyrolysis temperature (Leng *et al.* 2016; Liu *et al.* 2017). This also highlights that lignin conversion to b-G has multiple routes; for example via the carbon growth route. The wider temperature decomposition range gives a consistent supply of carbon sources throughout the process. It should be noted that a metal catalyst mixed with lignin also can serve as a catalytic surface that provides a place for carbon to deposit while helping to break down the lignin complex structure resulting in more stable aromatic rings compound with high carbon content (Cui *et al.* 2019). Others have used a combination of metal and solvent such as THF to further infuse the metal onto the lignin structure. Previously it was mentioned that the lignin differs depending on the type of the plant source. In the pulping industry, both softwood and hardwood are widely used, and the proportions of them to be employed depends on the type of paper products. Softwood fibres are lengthier than the fibres contain in hardwood. In addition, a larger amount of lignin is typically present in the softwood compared to the of hardwood (Chatterjee and Saito 2015). From the authors' knowledge, there has been no study comparing the quality of b-G produced from softwood and hardwood lignin.

ANIMAL RESIDUES

Animal residues are generally described as the by-products of livestock farming, in the form of solid materials or carcass. However, in this review animal residue cover anything related to the waste generated from animals including aquaculture. If these by-products are not being well-utilized, it will lead to problems with the environment and health. Most livestock industries are well aware of the consequences of unchecked livestock residues and are taking steps to recycle them. The residue generated from this industry increases yearly; thus any utilization of the residue is valuable. Animal residue has been used as raw material for carbon-based compounds such as activated carbon (Li *et al.* 2018), bio-chars (Qiu *et al.* 2014), and porous carbon material (Cheng *et al.* 2019). Chicken bone and cow dung can be used for GO production (Akhavan *et al.* 2014). Due to the complexity of the structure for some of these animal residues, pre-treatment with metal catalysts is the preferred method. In most cases, animal residues are not presented in powder form. Moreover, it is hard to purify most of the materials considering the presented matrix. In this case, dehydration and drying steps are used to remove moisture and harden the materials before they are used.

In aquaculture, chitosan is one of the most used materials as animal waste. Chitosan can be used to treat diseases such as cholesterol, obesity, and Crohn's disease (Mahapatro and Singh *et al.* 2011). Chitosan is a polysaccharide compound produced from chitin, and it is the second most abundant polymer after cellulose (Pandiselvi and Thambidurai 2014). Chitosan is extracted from chitin using an alkaline solution. The decomposition of chitosan has been studied extensively; it involves the breaking of its long-chain structure and releasing volatile aromatic compounds such as pyrazines, pyridines, pyrroles, and furans (Zeng *et al.* 2011). Besides, there are amino groups in the chitosan structure, which give a possibility of nitrogen-infused to the graphene as a doped compound as shown by studies

conducted by Hao *et al.* (2015). KOH is used to open up the structure for easier graphitization. The list for graphene produced from animal residue is shown in Table 5.

Table 5. Treatment Process for Animal Residue in Producing Graphene

Sources	Experimental details	Product	Ref.
Chitosan	Freeze drying Carbonization Chemical activation with KOH Pyrolysis Acid purification with HCL	b-G	(Hao <i>et al.</i> 2015)
Chicken bone	Imperfect burning in chimney for 5 days Mixing with metal catalyst Hammers method	b-GO	(Akhavan <i>et al.</i> 2014)
Cow dung	Imperfect burning in chimney for 5 days Mixing with metal catalyst Hammers method	b-GO	(Akhavan <i>et al.</i> 2014)
Dog feces	Dehydrated in vacuum oven CVD method with Cu substrate Purification with HCl	b-G	(Ruan <i>et al.</i> 2011)
Cockroach leg	CVD method with Cu Substrate Purification with HCl	b-G	(Ruan <i>et al.</i> 2011)

OTHERS

Many other biomass wastes are not commonly produced as agriculture and livestock waste, such as nut shell, soya beans, mango peels, and camphor leaves. The nut shell is considered as biomass waste, as consumers and the industries both dispose of the shell. Nut shells can be converted into bio-products through pyrolysis (Mgaya *et al.* 2019). The carbon and fixed carbon content of nut shells are high enough for producing bio-char, which can be converted to graphene. Lu *et al.* (2019) used macadamia nut shell to produce graphene using a combination of HTC and pyrolysis. The rigidity of the nut shell can be loosened by KOH activation. In most cases, high and multiple thermal treatments are needed to organize the graphene structure. Acetone can be used to clean up the material. Oven drying and grinding, which are normally used for biomass pre-treatment, are applied for purification. Shams *et al.* (2015) used D-tyrosine for better separation of the graphene sheets after pyrolysis, as it acts as a stabilizer. D-tyrosine was chosen because it is removed by washing with strong acids or bases. Other biomass wastes used to produce bio-based graphene are shown in Table 6.

Table 6. Treatment Process for Other Biomass Wastes in Producing Graphene

Sources	Experimental detail	Product	Ref
Mango Peel	<ul style="list-style-type: none"> Washed and dried CVD method with CU metal substrate Plasma exposure 	b-G	(Ruan <i>et al.</i> 2011)
Camphor leaves	<ul style="list-style-type: none"> Washed and dried Pyrolysis Ice bath sonication with D-tyrosine and trichloromethane solvent Centrifugation 	b-G	(Shams <i>et al.</i> 2015)
	<ul style="list-style-type: none"> Washed and dried CVD method with Cu substrate Purification with chlorobenzene 	b-G	(Kalita <i>et al.</i> 2011)
Soybeans	<ul style="list-style-type: none"> Washed and dried Pyrolysis Dispersed in DMF 	Nitrogen-doped graphene	(Sha <i>et al.</i> 2019)
Spruce bark fibres	<ul style="list-style-type: none"> Washed and dried Hydrothermal Chemical activation with KOH Acid purification 	b-G	(Sun <i>et al.</i> 2018)
Populus wood biomass	<ul style="list-style-type: none"> Washed, crunched and dried Pyrolysis Acid purification 	b-G	(Ekhlas <i>et al.</i> 2020)
Macadamia nutshell	<ul style="list-style-type: none"> Ground, washed and dried Chemical activation with KOH Hydrothermal Pyrolysis Acid purification with Nitric acid 	b-G	(Lu <i>et al.</i> 2019)
Peanut shell	<ul style="list-style-type: none"> Washed and dried Pre-treatment under sunlight Pyrolysis Chemical activation with KOH Sonication by dissolved in H₂SO₄ 	b-G	(Purkait <i>et al.</i> 2017)
Bengal Gram Bean husk	<ul style="list-style-type: none"> Pre-carbonization Chemical activation with KOH Pyrolysis 	b-G	(Gupta <i>et al.</i> 2019)
	<ul style="list-style-type: none"> Pre-carbonization Mixing with metal catalyst Pyrolysis 	b-G	(Gupta <i>et al.</i> 2019)

APPLICATION OF BIO-BASED GRAPHENE

Apart from certain chemical vapour deposition (CVD) processes, most b-G is multi-layered. Even so, the mass production of graphene also subjected to a multi-layer structure due to the complication in mass-producing monolayer graphene. As of now, there are many applications conducted using b-G that are not solely from biomass waste material. The porous structure of the b-G with a high specific surface area suitable for fast ion-transport applications such as supercapacitors. Carbon materials have been used as positive and negative electrodes for supercapacitors for many years. Porous material can provide a well-balanced pore distribution for high energy density and power density (Ke *et al.* 2016). Purkait *et al.* (2017) used b-G derived from peanut shells to fabricate a binder-free supercapacitor. The electrode had ample pore volume to achieve high energy density and power density of 58.125 W h Kg⁻¹ and 37.5 W Kg⁻¹, respectively. Similarly, Jung *et al.* (2018) produced a porous b-G electrode that was physically and chemically activated. The b-G generated a specific surface area of 3,657 m² g⁻¹, which translated to high energy

density and power density of 74 W h Kg^{-1} and 408 W h Kg^{-1} , respectively. This result is comparable to that of other graphene electrodes.

The application of graphene in fuel cells is already well documented (Tennyson *et al.* 2017; Leeuwner *et al.* 2019). As in the case of a supercapacitor, a large surface area is one of the requirements for a good fuel cell electrode. Most fuel cells have utilized multi-layered graphene as opposed to monolayer graphene due to the surface area it provides (Shen *et al.* 2015). The high surface area provides great electrocatalytic activity for more efficient oxygen reduction reaction (ORR) performance. Liu *et al.* (2014) prepared a graphene-like fuel cell material with a high surface area of $1510 \text{ m}^2 \text{ g}^{-1}$. The material was doped with nitrogen for better electrocatalytic performance as opposed to Pt-doped versions used by other carbon-related fuel cell materials. Meanwhile, Yan *et al.* (2019) utilized the large active sites of b-G in a lithium-ion battery. The numerous pores in the structure provided improvement in the permeability of electrolytes, which promoted free passage to the lithium ions and charges. The b-G posed specific capacitance and discharge capacity of 271.7 F g^{-1} and 310 mA h g^{-1} with a good rate performance and cycle stability. The future for bio-based graphene is bright, as there are plenty of applications that require high surface area materials. B-G can be used as an adsorbent or as a catalyst that can utilize the porous structure of b-G.

CONCLUSIONS

Graphene and its derivatives are novel materials that have yet to reach their peak in technology fields. Although there has been much discussion related to this structure, there are still barriers that need to be overcome to reach the goal of easily available material. Graphene technology is limited by graphene production. Thus, graphene production is expensive, even though it is produced from cheap and abundantly available material, graphite. The utilization of biomass waste can alleviate this problem while reducing the associated pollution. Graphene can be produced from biomass waste by removing volatile compounds and increasing the carbon content in the structure. This mechanism can be achieved by using a thermal treatment, which is already used for bio-materials production. High-temperature pyrolysis incorporating metal pre-cursors mixed with the biomass has been chosen by many researchers. The high temperature ensures the decomposition of the biomass structure, while the metal catalyst provides a surface for volatile carbon materials released from the biomass to deposit. Moving forward, low-temperature thermal treatment will be necessary, as the time required for heating and cooling is added to total reaction time. Bio-based graphene might not surpass the quality one can get through the graphite route. However, the development of graphene using the green synthesis route can be considered as one step forward in graphene technology. One thing to take from this route is the possibility of producing a massive amount of multi-layer graphene, GO and RGO. Even though quality-wise, bio-based graphene is lacking, it is still usable for most current graphene-based applications.

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