

A Review on Graphene: Synthesis Methods, Sources, and Applications

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ABSTRACT

The carbon allotrope so-called graphene is gaining popularity in the research and industry sector as it exhibits excellent thermal conductivity, mechanical strength, current density, electron mobility, and surface area. These properties of graphene allow them to be incorporated in many different fields to enhance the features of current materials and to be used for ground-breaking research. This review includes three major categories which discuss several ways to synthesize graphene, its sources, and its application, mainly in the field of battery. As there are many ways to produce graphene, this review would try to evaluate which methods would be best for different applications of graphene to overcome the high production cost of graphene, environmental damages, and low purity of graphene.

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INTRODUCTION

Graphene is a one-atom-thick, two-dimensional (2D) crystal sheet made of sp²-hybridized carbon atoms arranged in six-membered rings [1]. Andre Gaim and Konstantin Novoselov received the 2010 Nobel Prize in Physics for their successful research, which led to the discovery of graphene in 2004 by mechanical stripping [2]. Graphene is considered unique, being the only allotrope of carbon where each carbon atom is covalently and strongly bonded to its neighbors by an electron cloud.

Each monolayer graphene sheets are also connected by the intermolecular force, van der Waals forces which is based on the attraction and repulsion between the atoms, molecules, and surface of the graphene layer [3], [4]. Graphene may be employed in a wide range of applications due to its extraordinary electrical, thermal, and mechanical capabilities, as well as its numerous distinctive characteristics, including a high specific surface area, good flexibility, and chemical and thermal stability [1].

These unique properties made graphene desirable as a potential application in batteries, microbial fuel cells, anti-cancer applications, photovoltaic panel coatings, carbon fiber synthesis, sensor, and biosensor production. However, until now there has not been any breakthrough in producing these products on a mass production. Not to mention that these potential applications will be niche due to the expensive material cost of graphene with only a slight improvement of product usefulness [5], [6].

Most of the applications of graphene demand either high quality single layer or bi-layer graphene. There was some progress in the mass production of graphene by methods such as mechanical and chemical exfoliation, as well as chemical vapor deposition (CVD).

However, these methods do not guarantee high-quality graphene and might produce defects as each method and materials used in the process would determine the morphology and structure of the graphene produced which will affect its potential applications. Thus, achieving high-quality graphene on a large scale is one of the pressing concerns in realizing its wide applications [2].

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METHODOLOGY

There are several ways to synthesize graphene starting from the two-approach top-down and bottom-up method as seen on **Fig. 1** which will be discussed shortly.

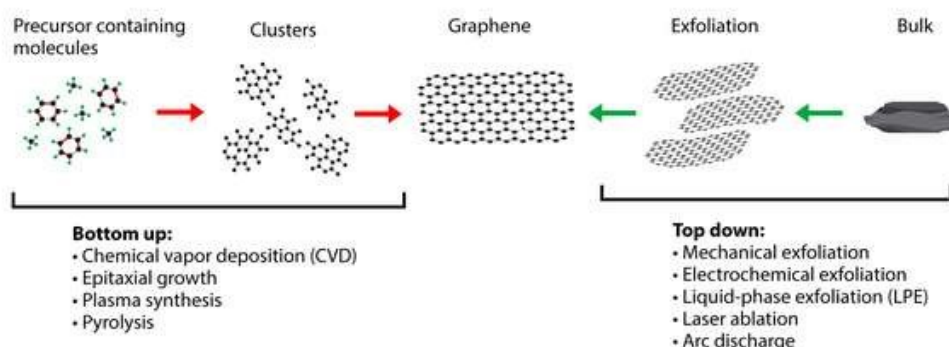


Fig. 1. Method scheme in the synthesis of graphene.

Top-Down

The top-down approach starts with the breakdown of graphite into nano-sized graphene sheets. The benefits of top-down methods include ease of substrate transfer, lower production cost and high accuracy compared with bottom-up approach [7]. The top-down approach are as follows:

Mechanical Exfoliation

Mechanical exfoliation a simple and common technique in producing graphene from natural graphite or other carbon sources. Mechanical exfoliation known as Scotch tape method is the first method to obtain one layer graphene. This method was done by continuously peeling graphite on a substrate using an adhesive tape until a monolayer sheet is formed in the tape [8]. Other mechanical techniques include using a ball mill or other contemporary equipment such as an ultra-sharp single crystal diamond wedge for the peeling of graphite [3].

Moreover, it was found that by using highly ordered pyrolytic graphite with mechanical exfoliation method have the ability to synthesize high-quality single layer graphene in the laboratory but not on a large-scale production. This was because the exfoliation technique can lead restacking of graphene layers due to the clinging of graphite sheet [7]. Even if it was possible to produce high purity graphene by mechanical exfoliation, the process is still inconvenient for large-scale manufacture as the process needs to be done gradually making it time consuming[3].

Electrochemical Exfoliation

Electrochemical exfoliation is a substitute for mechanical and oxidation-driven methods for large-scale generation of graphene from graphite. This method uses the mechanism of applying a fixed

voltage to a stack of graphitic material, mainly graphite, in order to weaken the Van der Waals' forces that separate graphene layers by inducing the ions to intercalate into the graphene layer. It uses an anode and a cathode to work in a liquid medium such as organic solvents without the usage of hazardous oxidants [7].

Electrochemical exfoliation does not limit the graphite used as electrodes and can proceed with a variety of morphologic structures such as graphite powders, foils, rods, flakes, or plates. Exfoliated graphene is frequently broken up into little fragments by the introduction of oxygen (O) during graphite exfoliation in liquid electrolytes under atmospheric circumstances. Effective ion intercalations between graphene layers are necessary to increase the interlayer gap during the electrochemical exfoliation of graphite.

Microstructures of graphite electrodes, including particle size, flaws, layer configurations and thickness, composition, and proper pre-treatment, are anticipated to have an impact on ion intercalations [9]. However, as intercalation progresses, the graphite electrode begins to disintegrate, and when graphite fragments are cut off from the electrode, they are unable to continue the intercalation process. Moreover, the breakdown of the graphite rod by electrochemical exfoliation only produces low yields of graphene, and yet requires additional partition of unexfoliated element [10].

Liquid-phase Exfoliation (LPE)

Liquid-phase exfoliation is a method that produces 2D materials like graphene in mass production at a reasonable cost/quality ratio. Typically, the fragmentation and exfoliation mechanisms involved have been associated with

straightforward interaction between solvent molecules with ultrasonic forces. However, not much is known on how dense graphite crystals can separate into thin, tiny graphene flakes [11].

LPE just requires the peeling or exfoliation of natural graphite using a high-shear mixer or sonication to provide a steady scattering of monolayer and few-layer defect-free graphene. Typically, it entails two independent methods of graphite peeling: cavitation in sonication and shear forces in high-shear mixers [12]. Recent research has shown that a microfluidizer may effectively exfoliate graphite in the right aqueous solutions at high shear rates [13]. However, the widespread use of sonication assisted LPE has been avoided because it only yields low concentrations of graphene and requires a lot of energy during the production process, rendering it ineffective.

Choosing the right solvents is one of the most crucial aspects of managing the overall graphene exfoliation yield for LPE. The ideal solvent should successfully overcome the van der Waals interactions between the graphene layers [14]. The low solubility and poor colloidal stability of graphene in the majority of common solvents is one of the challenges in processing graphene utilizing the LPE method [15].

Laser Ablation

Laser ablation is a physical method to produce graphene from graphite. It can take place in a vacuum, argon, or air, however, the one step pulsed laser ablation in liquid (PLAL) mediums offers a number of benefits compared to the other mechanism. The PLAL method produces graphene by exposing graphite target that is submerged in a liquid channel with radiation. The high temperature and pressure required for the creation of nanostructures are provided by the laser – graphite target interaction within the liquid media. This one-step procedure made it possible to accumulate graphene on a foil, triggering nucleation for the creation of graphene quantum dots and exfoliating graphene in a liquid media [12]–[14].

The created nanostructures are discovered in a colloidal solution after a solid graphite target is ablated in a liquid media by laser pulses. It is thought that PLAL is a simple, quick, and easy procedure. Furthermore, because PLAL does not use hazardous chemicals, it is recognized as a green and environmentally beneficial procedure. The flexibility with which different target materials and liquids can be combined to create a variety of nanostructures in the chosen liquids is one of the major benefits of the PLAL approach. Moreover, a significant element effecting the size of the created nanostructures is the amount of time exposed to radiation during

the ablation process or after it is finished [12].

Arc Discharge

Arc discharge is another method that can be used to produce bulk graphene nanoplatelets. It has been demonstrated that both a magnetic field and no magnetic field may be used to successfully produce graphene flakes. In comparison to chemical exfoliation, the arc discharge method for producing graphene is a promising method since it creates high purity graphene flakes without the use of harmful chemicals. In order to create an arc discharge, graphite rods of various lengths and diameters that serve as the anode and cathode and are spaced close together in a suitable medium are used.

Here, a spark that vaporizes the electrode material is produced as a result of a very high electric field generated by high voltage conditions. However, the high energy procedure and tight control needed for this technology preclude the synthesis of graphene from scaling up [15]. The graphene produced from arc discharge can be applied for batteries, supercapacitor, sensor, electronic devices, and hydrogen storage materials.

Bottom-Up

Bottom-up approach is a layering method to obtain graphene from the production of small carbon molecule atoms. These procedures frequently include the use of hazardous oxidizers, carboxylic acid, and organic solvents that are damaging to the environment. The use of phytoextracts and micro-organisms can be used as alternatives in this approach due to its eco-friendly nature. The ability to tailor the thickness of the graphene layer generated with the aid of unique surface catalysts and growth conditions is one of the many benefits of the bottom-up method [7]. Bottom-up approaches, on the other hand, have several drawbacks such as high cost of manufacturing, a lower yield, challenging to scale, and difficulty in transferring the finished graphene product to a desired substrate [21].

Chemical Vapor Deposition (CVD)

Chemical vapor deposition is one of the primary methods used for the synthesis of high-quality graphene with a large surface area. Several chemical processes results in the production of graphene on the surface of various transition metals, such as the use of nickel (Ni) and copper (Cu) foils as substrates of carbon source from various vapor classes [7]. The solubility of carbon in metal substrates serves as the basis for the mechanism of CVD. The high-temperature chamber where the gaseous carbon compounds will dissolve into the metal foil substrates is where they are conveyed.

Typically, the CVD method's first step in the production of graphene is described as the generation of carbon-metal solid solutions [16].

Epitaxial Growth

Epitaxial growth is the process by which a crystalline layer forms on a crystalline substrate while adhering to the substrate's structure [23]. A single-crystal substrate and a robust contact between the epilayer and substrate are prerequisites for the formation of single-crystal thin films [13]. The development of epitaxial graphene on SiC substrates by surface Si depletion is a potential method for manufacturing a sizable region of high-quality graphene with uniform thickness. This technology has the advantages of allowing the size of the graphene layer to be as large as the substrate sheet and allowing the production of electrical devices directly on semi-insulation SiC without the need to transfer the graphene layer to another substrate.

The sublimation of Si and C at high temperatures drives the development process of graphene layers. However, the Si and C sides' surface reconstructions and growth kinetics differ, leading to distinct growth rates, structures, and electrical properties. High grade graphene may be produced using this sublimation process for epitaxial growth. It demonstrates intriguing physical properties, including half-eV bandgap structures, high-frequency transistors, a quantum hall resistance standard for metrology, and ballistic transport in nanoribbons [23].

Pyrolysis

Pyrolysis is a method of carbonization to synthesize graphene from various carbon sources such as from plastic wastes, corn stalks, wheats, seaweed, palm leaves, rice husk, and another available biomass. Graphene derived from carbon-like biomass has showed promising performance in areas such as supercapacitors, fuels, catalyst, and additives in composites [18], [19]. It is a key process in the synthesis of carbon nanomaterial, production of bulk carbon, manufacturing of carbon-based technology, generating fuel from organic wastes, and molecule dissolution for analysis using gas chromatography-mass spectroscopy (GC-MS). The foundation of carbon production processes is based on pyrolysis. This process mechanism relies on the heating of certain organic compounds to elevated temperatures in a chemically inactive environment sometimes with the help of a catalyst. This causes the precursor to thermally decompose, releasing non-carbon atoms in a variety of shapes. A portion of carbon is often produced as a by-product in the form of a solid or smoke due to the great thermal stability of carbon in the absence of oxidants.

Most hydrocarbons undergo bond-breaking and a rearrangement of covalent bonds during pyrolysis, which happens between 300°C and 800°C [20]. Past research demonstrates the poor yield of graphene produced by pyrolyzing biomass. According to past data, the yield of dried products appears to be influenced by the pyrolysis temperature of the first stage. Theoretically, the yield falls as temperature rises because more condensable material is lost. However, since the temperature for thermal degradation of different biomass varies, the fixed temperature to obtain an optimum yield is hard to be determined [21], [22].

Plasma Synthesis

Plasma synthesis does not lead to unwanted defects, morphologies, and contamination as it only uses natural hydrocarbons as its reactant starting material without the use of any harmful chemicals [23]. Advanced synthesis methods that use plasma may create graphene in a matter of seconds without the need of solvents. These methods include atmospheric pressure microwave plasma and plasma-enhanced chemical vapor deposition (PECVD). With the aid of plasma, PECVD, a type of improved CVD method, may reduce the temperature required to produce graphene from its initial high temperature of roughly 1000°C to a lower temperature that lies between 600 to 800°C. Through the assistance of the plasma's massive chemical environment, which creates a variety of molecules, radicals, and ions from hydrocarbon precursors, the production of graphene may be carried out at lower formation temperatures and more quickly compared to thermal CVD [24].

RESULTS AND DISCUSSION

Structure and Properties of Graphene

Graphene is a two-dimensional (2D) atomic-scale hexagonal lattice allotrope of carbon where one atom forms each corner by sp^2 hybridization. The carbon-carbon bond in graphene measures to be 0.142 nm in length with each lattice containing three strong linkages that combine to assemble a sturdy 6-carbon ring structure as seen in **Fig. 2**.

Most of graphene's electrical conductivity is due to the bond, which is positioned vertically to the lattice plane [25]. The tightly packed carbon atoms in graphene with three carbon atomic orbitals – $2s$, $2p_x$, and $2p_y$ – hybridize into sp^2 orbitals that interact chemically with nearby carbon atoms to form covalent bonds. This results in a honeycomb-like hexagonal planar structure, stability, and increased graphene lattice structural toughness [25], [26]. X-Ray diffraction spectrum is an effective equipment for the characterization of crystalline structure and phase purity of solid materials.

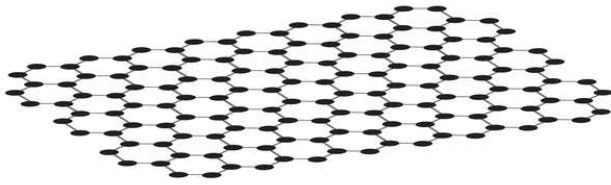


Fig. 2. Structure of graphene sheet.

The XRD spectra measured in a range of 2θ from 5° to 80° (Fig. 3) shows broad diffraction peak at $2\theta = 25.4^\circ$ using X-Ray diffractometer PANalytical Empyrean. The characteristic diffraction peak positioned at around 25.4° corresponds to the reflection of the crystal planes (002). The broad peak for graphene indicates its small crystalline size and disordered structure [10].

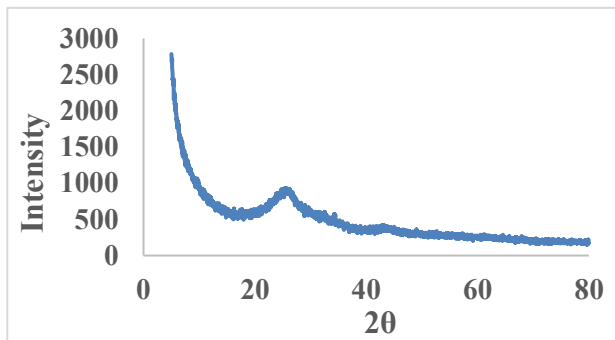


Fig. 3. XRD patterns recorded for commercial graphene.

The use of Raman spectroscopy is to provide chemical and structural data based on the vibrational and rotational modes of a molecule. A disorder (D) band at around 1350 cm^{-1} , a graphitic (G) band at about 1585 cm^{-1} , and a 2D band at about 2700 cm^{-1} all indicate the presence of graphene in Raman spectra. While the 2D band reflects platelet thickness, the intensity ratio of the D and G bands (ID/IG) evaluates the degree of abnormalities [19].

Here, the D band originates from out-of-plane vibrations linked to the existence of structural flaws, whereas the G band is caused by in-plane vibrations of sp^2 -bonded carbon atoms. It can be said that the quality of the graphene sample increases with a decrease in the ID/IG ratio value. Fig. 4 shows that the D-band was found to be around $1300\text{--}1350\text{ cm}^{-1}$, G-band approximately 1600 cm^{-1} , and lastly, the 2D-band around 2700 cm^{-1} . From the spectra, the ID/IG ratio was estimated to be 0.188 which shows low defect ratio.

In a graphene layer, the sp^2 hybridization of carbon atoms can be identified as out-of-plane or in-plane. An out-of-plane π bond is composed of $2p_z$ orbitals that are perpendicular or crosswise to the planar structure. Meanwhile, an in-plane σ bond is made up of three sp^2 ($2s$, $2p_x$, and $2p_y$) hybridized orbitals. The monolayer graphene has extraordinary mechanical capabilities from the short covalent σ bond of interatomic length 0.142 nm , which makes it even stronger than the sp^3 hybridized carbon-carbon bonds in diamonds [27].

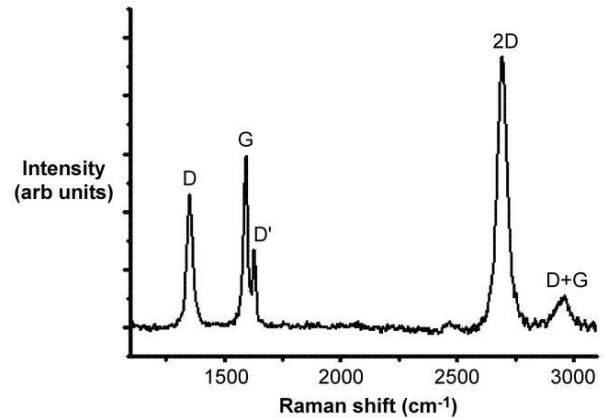


Fig. 4. Raman spectra of N-doped single-layer graphene [38].

Due to the strong covalent bonds between the carbon-carbon atoms, graphene has a high Young's modulus, an extremely high intrinsic charge mobility, a high specific surface area, good thermal conductivity, and a high optical transmittance due to its one atom thickness [25], [26]. It has a 97.7% optical transmittance which means that it only absorbs 2.3% of visible light. It also means that between layers of graphene and substrate, there is only 2.3% transparency difference.

By increasing the graphene layers, the amount of white light absorbed increases around the same amount 2.3%. As a result, simulations employing the non-interacting Dirac fermion theory show that the transparency of graphene can effectively indicate the number of graphene layers [25]. Saturable absorption starts to happen whenever the optical intensity crosses a particular threshold. Whole band mode locking has been accomplished due to the wavelength-insensitive ultrafast saturable absorption characteristics of graphene [28].

The inherent strength of graphene is another one of its exceptional qualities. Graphene is one of the strongest substances ever known, with an ultimate tensile strength of 1.3×10^{11} Pascals (or 130 gigapascals), due to the strength of its carbon bonds. Moreover, it is very small, weighing only 0.77 milligrams per square meter. In addition to having elastic qualities, graphene can return to its original size even after being elongated. According to research, graphene sheets with a thickness of 2-8 Nm had spring constants ranging between 1-5 N/m and a Young's modulus of 0.5 TPa, which differs from that of parent three-dimensional (3D) graphite.

Graphene differs from other condensed matter being a semi-metal with zero-gap semiconductors and four electronic properties. Graphene has a very high electrical and heat conductivity, mechanical durability, and has a zero-overlap semimetal with both holes and electrons acting as charge carriers which makes them favorable [29]. Graphene, being the toughest crystal structure ever found, has a strength limit of 42 N/m with tensile strengths and elastic moduli of 125 GPa and 1.1 TPa. To give an

example, it can be said that the strength of graphene is around 100x greater than that of steel at the same thickness; 1 m² of graphene can support up to 4 kg of weight [25].

Sources of Graphene

Graphene and its derivatives are frequently synthesized from graphite as a basic material. It is abundant in nature and simple to obtain. Additional carbon sources, including any type of waste, can be obtained from our environment, and used as primary ingredients in the manufacturing of graphene. The use of waste such as biomass can be viewed as an effective way to create new and useful products while also solving the environmental challenge. This is considered as a green synthesis method which uses biomass for the production of graphene [30].

The sugar molecule glucose, having the chemical formula C₆H₁₂O₆, is a plentiful and renewable source of carbon. Glucose can be converted from waste cellulose such as sugarcane bagasse, milk cartons, newspapers, and corrugated

fiberboard by hydrolysis using acids and enzymatic processes known as saccharification. With this technique, a high glucose yield of between 70% and 89% can be produced. Since glucose is carbon based, a variety of ways have been experimented to use glucose as a natural resource for the synthesis of graphene. A variety of graphene structures and purities could be obtained using different methods and materials [31].

Graphene can also be synthesized from a variety of sources depending on which method of production was used as shown in **Fig. 5**. For instance, plants can be used as raw material through the pyrolysis method, which involves thermal decomposition at high temperatures in an inert atmosphere. The pyrolysis procedure produces graphene-sand composites if a mixture of sugar and sand or asphalt is used as the precursors. Meanwhile, a range of foods, polymers, and wastes can all be used to make graphene using CVD process.

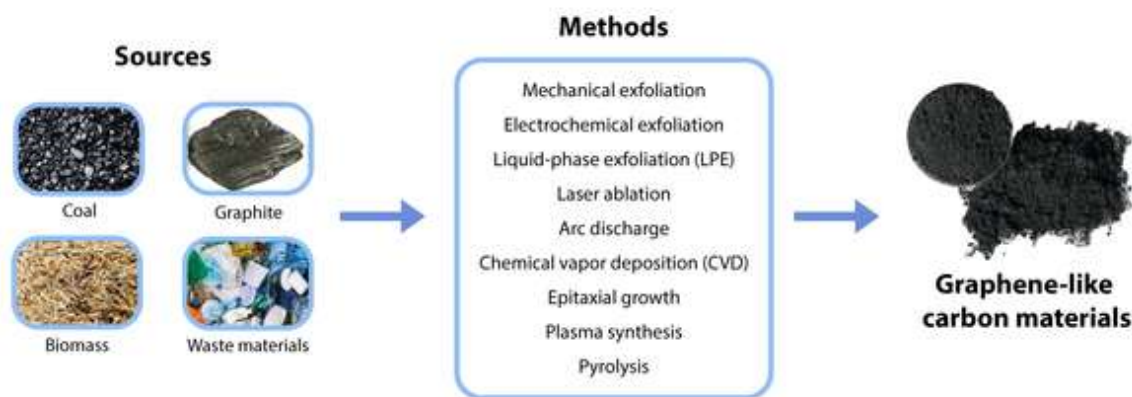


Fig. 5. Synthesis techniques of graphene from various sources.

In this method, graphene is created by the breakdown of the source into gases that subsequently interact with a metal catalyst substrate at elevated temperatures and in a reductive gas environment. Reactive plasma technology, on the other hand, enables the manufacture of customized graphene structures, such as vertical graphene sheets and metal-doped graphene, from a variety of natural precursors in a matter of minutes without the assistance of a catalyst and at a low temperature [32].

Applications

The extraordinary quality of graphene gives enormous potential that compelled scientists to continue their studies. Graphene could be used to replace many present existing materials and can be incredibly helpful in a variety of applications. It can be said that the applications of graphene were divided into four categories: energy, environmental, biomedical, and sensor/biosensor.

Some examples of the applications of graphene include batteries, solar cells, electrochemical devices, wastewater purification, tissue engineering, and could be utilized for other purposes [26], [33].

materials, has a high conductivity which was thought to be perfect for high-speed electronics and battery storage. However, graphene's commercial applications are still in its early stages. Since graphene has a zero-band gap, more research is necessary before it can be used in industrial applications. In order to develop high-speed graphene transistors for consumer electronics, graphene research is currently being expanded. Furthermore, batteries, supercapacitors, fuel cells, solar cells, and fuel cells are all examples of energy storage technologies. These energy storage materials have currently been used with the addition of graphene and thoroughly researched.

Based on recent research, the use of graphene in data storage in replace of indium tin oxide electrodes was said to have 10 times more power which can increase the capacity levels of storage devices [34], [35].

Research also shows that graphene can be used to overcome the poor conductivity and ion diffusion problem in lithium-ion battery by increasing the electronic conductivity of the composite. Recently, graphene nanosheets were added in the LiFePO_4 cathode of li-ion batteries as a new carbon conducting additive and it was proven to enhance the rate capability effectively [30], [36].

The use of graphene in environmental applications ranges from photocatalyst and desalination membrane to pollutant adsorption, detection, and monitoring. Graphene-based sensors are viewed as a potential technology because of their high sensitivity to the detection of impurities and their ability to scale down to nanoscale size. However, the interaction of the side chains with other molecules is problematic when using graphene-based sensors since they are sensitive to environmental variables including temperature, pH, and ionic strength, which causes many false positives [37].

It has been claimed that graphene is an excellent sorbent that can be recycled. It has been demonstrated that graphene can absorb liquids 600 times its weight. Graphene also gives an outstanding performance in the adsorption of gasoline, organic pollutants like ethanol, olive oil, nitrobenzene, acetone, and dimethyl sulfoxide as well as heavy metals like arsenic, mercury, cadmium, chromium, and lead are other compounds that graphene may be able to adsorb. Fast electron transport in graphene and graphene-based materials with sp^2 hybridized carbon atoms enables excellent charge separation during photocatalysis to remove pollutants from wastewater. Due to its smooth, frictionless surface and quick water flow, graphene was also examined for desalination technology, a method to address the issue of fresh water scarcity in the environment [34], [37].

In the field of nanomedicine, graphene, and some of its derivatives, specifically graphene oxide (GO), have drawn much interest as bacterial inhibitors, imaging contrast agents, and drug/gene delivery systems because of their suitable broadened surface area, biocompatibility, and simplicity of surface functionalization. Additionally, the near-infrared (NIR) light absorption properties of graphene nanostructures enable their usage as photo-thermal therapy agents.

As a result, these graphene-based nanomaterials (GBNs) combine the potentials of transporting medicines and improved photo-heat conversion, making them an excellent choice for usage as heat-responsive nano-sized drug carriers in therapeutic applications like cancer [42].

The biosensor is a device that can detect biological chemicals by sensing their concentration and converting it to electrical impulses. Biosensors offer unprecedented technical support for the early examination of diseases and the discovery of particular elements in contemporary medicine. It was discovered that graphene materials may often give improved sensitivity and reduce reaction time for existing biological detection methods due to their strong adsorption capacity and superior conductivity when considering the implementation of graphene in biosensors in the last 10 years. Therefore, it is possible to say that graphene's function in sensor materials was to increase the sensitivity of the sensor, decrease the reaction time, and cover a larger area [38].

The benefits of using graphene in sensors can be summed up as the following: First, compared to silicon, graphene-based sensors are more responsive due to the vast specific surface area and atomic thickness of the graphene layers that allow carbon atoms to come in direct contact with analytes. Additionally, by using graphene-based sensors, a conformal close contact with organs of interest may be formed due to the mechanical flexibility and thin layer of graphene. It can be said that graphene-based sensors are essential for obtaining high-quality signals free of disturbance. Moreover, graphene is the ideal material for seeing a clear image of bio-tissue with no visual defects and disruption due to its excellent optical transparency and electrical conductivity.

Furthermore, high electrical conductivity-based conformal integration and efficient signal transmission may be employed to acquire electrophysiological data with a high signal-to-noise ratio (SNR). Last but not least, due to graphene's exceptional performance in biosensors, which includes its high electron transfer rate, broad potential window, and large specific surface area, receptors such as enzymes, antibodies, and deoxyribonucleic acid (DNA) may be successfully mounted on its surface [41].

CONCLUSION

This review has shown that there are many advantages of graphene which include high electrical conductivity, exceptional electrochemical, electronic, thermal, and mechanical properties that cause them to be desirable in many applications. This paper reviews recent research in the production

and synthesis of graphene whether it may be for industrial purposes or laboratory uses. Many procedures and approaches have been developed in producing high-quality graphene. These methods include various types of exfoliations, laser ablation, pyrolysis, epitaxial growth, and other processes mentioned in the previous discussion. It could be summarized that future research in graphene would allow applications in the field of energy, environmental, biomedical, and sensors. These future opportunities would give immense advantages and improve daily activities such as in energy storage, LCD screens, nanomedicine, and biosensors. Thus, further study on graphene and its application would be necessary to overcome the best way to synthesize graphene for commercial uses.

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AUTHOR CONTRIBUTION

E. Kartini, T. A. Setiadi and M. Fakhrudin contributed equally as the main contributors of this paper. All authors read and approved the final version of the paper.

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