Review

Comprehensive Review of Graphene Synthesis Techniques: Advancements, Challenges, and Future Directions

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Comprehensive Review of Graphene Synthesis Techniques: Advancements, Challenges, and Future Directions

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Abstract

Graphene, a two-dimensional material with remarkable electrical, thermal, and mechanical properties, has revolutionized the fields of electronics, energy storage, and nanotechnology. This review presents a comprehensive analysis of graphene synthesis techniques, which can be classified into two primary approaches: top-down and bottom-up. Top-down methods, such as mechanical exfoliation, oxidation-reduction, arc discharge, unzipping carbon nanotubes, and liquid-phase exfoliation, are highlighted for their scalability and cost-effectiveness, albeit with challenges in controlling defects and uniformity. In contrast, bottom-up methods, including Chemical Vapor Deposition (CVD) and epitaxial growth on silicon carbide, offer superior structural control and quality but are often constrained by high costs and limited scalability. The interplay between synthesis parameters, material properties, and application requirements is critically examined to provide insights into optimizing graphene production. This review also emphasizes the growing demand for sustainable and environmentally friendly approaches, aligning with the global push for green nanotechnology. By synthesizing current advancements and identifying critical research gaps, this work offers a roadmap for selecting the most suitable synthesis techniques and fostering innovations in scalable and high-quality graphene production. The findings serve as a valuable resource for researchers and industries aiming to harness graphene's full potential in diverse technological applications.

Keywords: graphene synthesis methods; top-down approach; bottom-up approach; chemical vapour deposition; scalable graphene manufacturing

1. Introduction

Graphene is a two-dimensional material consisting of a single layer of carbon atoms arranged in a hexagonal lattice pattern [1], as shown in Figure 1. Each carbon atom in graphene binds to its three neighbors through sp^2 hybridization, forming a sigma bond (σ) in the field, and the pi bond (π) delocalized above and below the field [2]. Since it was first successfully isolated by Andre Geim and Konstantin Novoselov in 2004, graphene has garnered widespread attention among scientists due to

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its remarkable physical properties and potential for diverse applications in various technological fields. This discovery even received international recognition through the award of the Nobel Prize in Physics in 2010 [3].

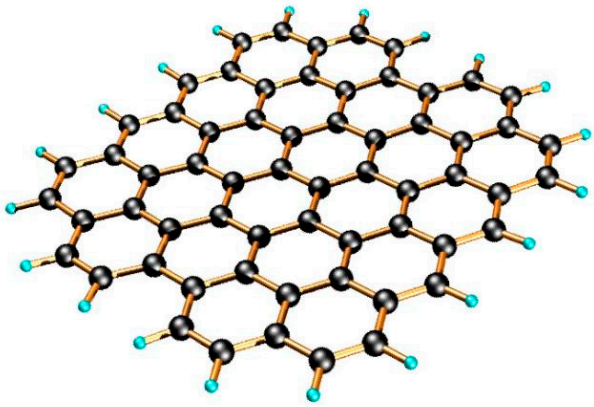

Figure 1. Graphene Structure Pictures [4].

The advantages of graphene lie in its unique combination of properties, including extremely high electrical and thermal conductivity, outstanding mechanical strength, optical transparency, flexibility, and a large specific surface area [5]. These properties are derived from two-dimensional atomic structures and sp^2 bonding systems, which are highly stable and enable the free and efficient movement of electrons in the field. These characteristics make them very promising for application in electronic devices, energy storage, sensors, composite materials, as well as biomedical and environmental technologies [6].

The high prospects of graphene in diverse applications demand efficient and stable synthetic ways to produce quality graphene materials. For its application, graphene has been required to fulfill necessary properties including well-defined layer number, uniform lateral dimension, low defects, and high purity. These characteristics strongly influence graphene's performance in its final application since its electrical, mechanical, and chemical properties are very much dependent on its structure and composition [7]. The choice of raw materials is an important consideration to meet the application requirements of graphene specifications. Graphite is the most popular C precursor due to the fact that it can easily be exfoliated into graphene sheets due to its layered structure [8]. The structural transition from graphite to graphene is depicted in Figure 2. Meanwhile, graphite is known as an abundant material, has a relatively cheap price, and good chemical stability, and thus it can be an excellent precursor source for other graphene synthesis methods [9].

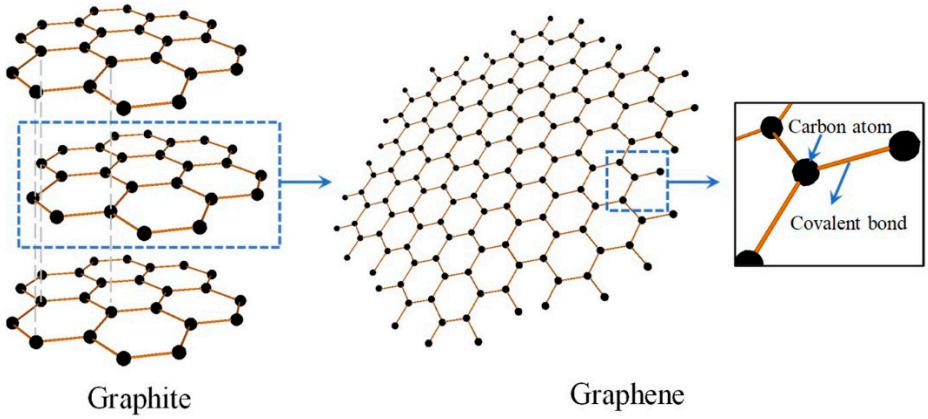

Figure 2. Graphite to Graphene Schematic Diagram [10].

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In addition to the selection of raw materials, the diversity of graphene synthesis approaches that have been developed adds complexity in determining the most effective production strategy. Each of these methods can be used to give a different reaction product, based on reaction conditions, processing stage and added advanced material. Thus, a comprehensive understanding of these methods is necessary for selecting the one that meets most of the application requirements and research questions [11]. However, despite the availability of various methods, the process of graphene synthesis, in general, still faces major challenges that cannot be considered simple. Regardless of the type of raw material and method used, this process requires highly controlled conditions for a layer of graphene to form without damaging its atomic structure [12]. Errors in process control can result in greater structural defects, changes of particle size, or contamination, which reduces the quality and performance of the resultant graphene [13].

As the synthesis of graphene is increasingly researched and developed, it is necessary to prepare a review that would capture all the approaches that have been undertaken as well as highlight the problems encountered and possible frameworks for further development. The purpose of this article is to provide a comprehensive review of different graphite-based graphene synthesis methods, looking into the main characteristics, concentrating on factors determining the quality of the end-product, prospective applications or innovations post-processing, and how these could further advance modern science. In this way, this review will enhance scientific understanding and development of graphene in various technological domains.

2. Graphene Synthesis

The synthesis of graphene is a fundamental stage in the widespread use of this material. Various methods have been developed to obtain graphene from carbon sources, especially graphite, which has a layered structure resembling graphene itself. In the scientific context, these synthesis techniques can generally be classified into two main approaches, namely top-down and bottom-up methods. These two contrasting approaches are illustrated in Figure 3. This classification not only reflects the direction of the synthesis approach but also determines the characteristics of the resulting graphene products [14].

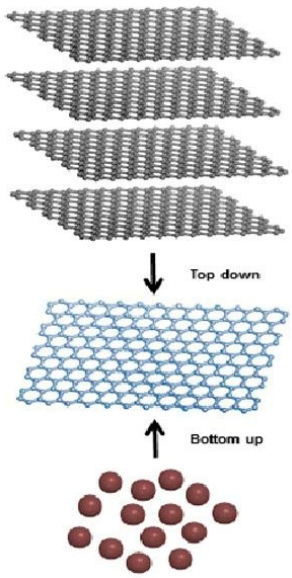

Figure 3. Top Down and Bottom Up Method Schema Diagram [15].

The top-down approach involves separating the graphene layer from the bulk material, such as graphite, through an exfoliation process or chemical reaction, resulting in thin sheets of graphene [16]. Meanwhile, the bottom-up approach forms graphene from simple carbon units through an

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atomic assembly process [8]. Top-down methods are generally better suited for large-scale production at lower costs, although control over the number of layers and structural defects is often a challenge [17]. In contrast, the bottom-up method offers better control over the structure of graphene but with greater complexity and process costs [18]. The following is the schematic of the graphene synthesis method:

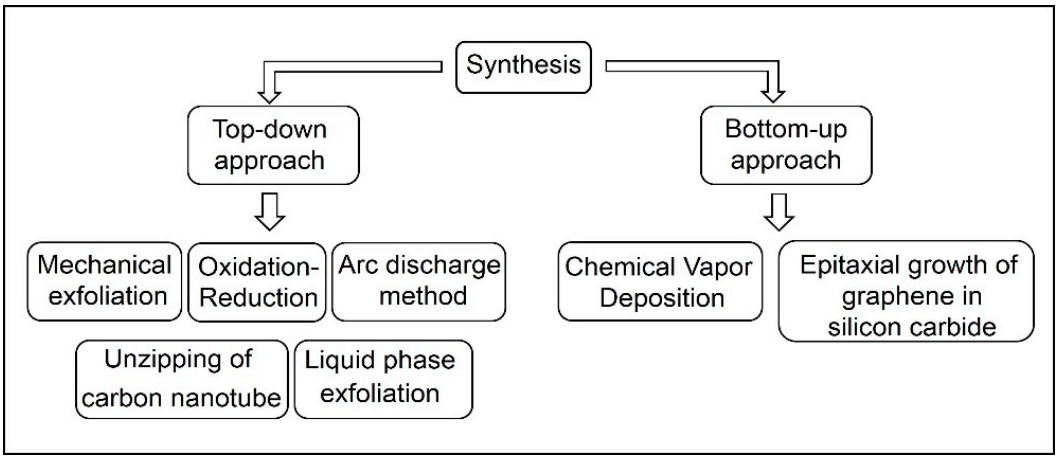

Figure 4. Top Down and Bottom Up Graphene Synthesis Flowchart [19].

Numerous top-down and bottom-up methods for graphene synthesis have been developed. Depending on the advanced treatment and reaction conditions, each method has a different set of working principles, stages, and outcomes. A thorough analysis of these methods is provided in the section that follows, beginning with a top-down strategy and moving on to a bottom-up strategy.

2.1. Top Down

One of the main methods for creating graphene is the top-down approach, which uses physical and chemical procedures to separate the graphene layer from the bulk material, like graphite. Large amounts of graphene are known to be produced using this method, but maintaining product quality is frequently difficult. The following are the top-down graphene synthesis techniques:

2.1.1. Mechanical Exfoliation

Mechanical exfoliation is a top-down method of producing graphene, where layers of graphite are separated using mechanical forces, such as normal or shear forces, to overcome the van der Waals tensile forces between layers. This method avoids major chemical reactions and is comparatively easy and economical by using mechanical energy to exfoliate graphite into single-layer or multilayer graphene [20]. There are several physical methods for performing mechanical exfoliation, and each has unique properties. These are a few methods that are frequently employed in this approach.

Micromechanical Cleavage

Micromechanical cleavage is an exfoliation technique that utilizes the adhesion force of an adhesive (e.g., tape) to physically exfoliate the graphene layer [21]. Geim and Novoselov first introduced this technique, and it was later refined using polymers such as PMMA and PDMS [22]. The use of PMMA is more effective in producing monolayer graphene, while PDMS tends to produce multilayers. This suggests that PMMA is better suited for producing high-quality graphene with a thickness of one [21].

In addition to the exfoliation method using adhesives, a new technique has been developed based on a single diamond tip that is very sharp. This method utilizes a single, very sharp diamond tip for precise exfoliation down to the atomic scale. This approach is capable of producing high-quality monolayer graphene with a thickness of tens of nanometers and a lateral size of about 900 ×

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300 μm. The use of ultrasonic vibrations (33.1 kHz, 2.1 V) was shown to be able to lower the I_D/I_G ratio from ~0.90 to ~0.73 and the crystallite size from ~24 nm to ~17 nm, indicating an improvement in the quality and stability of the graphene layer. TEM analysis showed a more organized structure as well as the emergence of nanohorns, which have the potential for gas storage applications [23].

Sonication

Sonication is a graphene synthesis method that utilizes high-frequency ultrasonic waves to generate mechanical energy through the phenomenon of cavitation. Ultrasonic waves create microscopic bubbles that collapse rapidly, resulting in intense shear forces that can overcome the Van der Waals forces between graphite layers. As a result, graphite flakes efficiently into thin sheets of graphene without the need for aggressive chemical reagents. [24].

The effectiveness of sonication techniques in graphene synthesis is highly dependent on the duration, sonication power, solvent type, and initial treatment of graphite materials. Sonication not only facilitates mechanical exfoliation but can also cause structural defects in graphene sheets, depending on process conditions. Therefore, characterization analysis such as FTIR, SEM, Raman, and XRD is important to evaluate sonication results, ranging from the presence of functional groups and morphology to the degree of defects and crystallinity of graphene [25]. The following is a summary of the characterization data, as presented in Table 1.

Table 1. Summary of characterization data produced by sonication methods.

Methods	FTIR	SEM	Raman	XRD	Ref.
Sonication during oxidation (Hummers method)	Decrease in O–H and C=O functional groups due to strong sonication	Flexible sheet structure with lower oxidation	Increased (many defects)	Intense (002) peak at 2θ = 26.5°, indicating dominant graphite phase	[26]
Sonication for 1–5 hours (with Tween 80)	–	Lateral size decreased from ~5 μm to 317 nm	Ratio increased gradually with longer sonication time	–	[27]
Sonication for 10/20 minutes (30/50 W)	O–H and COOH peak intensities decreased (sample S1 to S4)	Morphology becomes increasingly deformed (S1 to S4)	Increased from 0.84 to 0.95, indicating increased structural disorder	(002) peak shifted from 2θ = 11.34° to 11.14°	[28]
Sonication for 15–45 minutes (electrochemical method)	Appearance of C–O–H vibration, indicating the presence of hydroxyl group	–	–	(002) peak intensity weakened; crystallite size decreased	[29]

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Sonication for 8.5 hours (ethanol: water = 20:80)	Peaks observed O–H, C=C, and C–H	Spherical morphology with particle size ~23–41 nm	I_D/I_G ratio approximately 0.65, suggesting a moderate level of defects	Broad (002) peak at 2θ ~25°; crystallite size ~20 nm	[30]

Sonication effectively exfoliates graphite into graphene, as evidenced by an increase in the I_D/I_G ratio (Raman), a flaky sheet morphology (SEM), and an angular shift of 2θ (XRD), indicating an increase in interlayer spacing. Changes in the intensity of functional groups in FTIR also indicate surface chemical modifications. Overall, sonication is a simple and effective method, although it requires optimization to produce high-quality graphene with minimal defects.

Ball Milling

The ball mill technique is a top-down method for graphene synthesis that involves grinding graphite using crusher balls in a rotating container. The impact that occurs during this process causes the graphite layer to peel off into graphene sheets. This method is known for its simplicity and low cost and is suitable for large-scale production of graphene [31][32].

There are two methods for the ball mill technique: wet methods and dry methods. The dry method is a simpler procedure because it doesn't require solvents. In contrast, the wet method uses solvents like ethanol or water to help create graphene suspensions, lower frictional heat, and speed up the graphite peeling process. However, it requires additional steps for the separation and drying of the final product [33]. The following are the differences in the characteristics of graphene from dry and wet ball milling, as shown in Table 2.

Table 2. Summary of characterization data produced by dry and wet ball milling methods.

Wet Ball-Mill Method					
Sample	**FTIR**	**Morphology (SEM/TEM)**	**Raman (I_D/I_G)**	**XRD**	**Ref.**
Graphite + DMF	C=O stretching (~1700 cm^{-1})	Thin sheets with folded edges; few-layer structure (0.8–1.8 nm)	Increased, indicating higher disorder due to milling	(002) peak broadened	[34]
Graphite + Water + $KClO_4$	C–O stretching (~1060 cm^{-1})	Small layered nanosheets, graphene oxide (GO) formed	Increased with longer milling time	(002) peak broadened	[31]
Graphite + Water	C=O functional group observed	Large aggregates (BOTTOM60), finer sheets (TOP60); few-layer structure	High value (TOP60), suggesting a small sheet size and more defects	-	[35]
Graphite + Ethanol: Water (7:3)	-	Graphene-encapsulated SiC; few-layer structure	Decreased with increasing speed, indicating improved quality	Graphite peak intensity decreased	[36]

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Graphite + Water	-	More uniform particles; presence of individual sheets	Lowest value at 500 rpm (0.221), indicating high-quality graphene	(002) peak became sharper	[37]
Dry Ball-Mill Method					
Sample	**FTIR**	**Morphology (SEM/TEM)**	**Raman (I_D/I_G)**	**XRD**	**Ref.**
Graphite	-	Nanoparticles with irregular shapes	Increased, suggesting greater defect formation	Crystallinity decreased (weakened graphite peak)	[38]
Graphite	C=O, C–O functional groups	Shaft-like structure with reduced particle size	Increased, reflecting higher structural disorder	Graphite peak intensity decreased	[39]
Graphite	C=O, OH, COOH groups	Thin sheets with open structure; <10 layers	Increased, indicating defect generation during milling	Graphite peaks broadened	[40]
Graphite	OH, C=O groups	Rough surface morphology; reduced particle size	Increased (from 0.21 to 0.97), supporting oxidation process	2θ peaks shifted and broadened	[41]
Graphite	-	Thin and layered flake morphology	-	2θ peaks shifted and broadened	[32]

The wet and dry ball-mill methods both produce thinly coated graphene/GO with increased defects and decreased crystallinity. The wet method results in a more homogeneous structure and higher quality, suitable for electronics, sensors, and energy [34]. The dry method produces graphene with higher oxygen functionalities, suitable for adsorption, catalysts, and composites, and simpler, cheaper, and more environmentally friendly for large-scale production [42].

Fluid Dynamics

Fluid dynamics is a graphene synthesis technique that utilizes the movement of fluids and their interactions with solid surfaces. In graphene synthesis, this principle is used to exfoliate graphite into a thin layer of graphene without the use of harmful chemicals. This process relies on a high-speed flow of fluid that generates mechanical forces, such as friction, turbulence, cavitation, and differential pressure, to overcome the van der Waals forces between graphite layers. This technique is considered efficient, environmentally friendly, and has the potential to be developed on an industrial scale [43].

The three primary fluid dynamics-based methods employed for graphene synthesis are the Vortex Fluidic Device (VFD), Pressure-driven Fluid Dynamics (PFD), and Mixer-driven Fluid Dynamics (MFD). VFD utilizes a fast-rotating tube to produce a thin layer of fluid and applies a gentle shear force to exfoliate graphite. PFD involves the flow of graphite suspension through a narrow channel under high pressure, triggering exfoliation through a combination of shear forces,

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turbulence, and cavitation. Meanwhile, MFDs utilize mixing tools such as rotor-stator mixers or blenders to produce high shear forces evenly, suitable for efficient large-scale graphene production [44]. The following table summarizes the graphene characterization data generated by the three techniques, as shown in Table 3.

Table 3. Summary of characterization data produced by fluid dynamics methods.

Parameters	VFD	PFD	MFD
Thickness	Ranges from < 1 nm to > 20 nm	Up to 79% ≤ 1.5 nm (after 8 hours at 15 MPa)	Average ~1.5 nm; up to 92% ≤ 1.5 nm (after 3 hours)
Number of Layers	1 to > 20 layers	≤ 5 layers: 29% (0.5 h), 63% (4 h), 79% (8 h)	Average < 5 layers; stable across various exfoliation durations
Lateral Size / Area	< 1 μm	Over 85% of flakes < 0.1 μm² (after 8 hours)	Average ~320 nm (AFM); ~0.5 μm (Raman in protein medium)
Thickness Distribution	Uneven; limited data available	Becomes thinner and more uniform over time	Remains stable around 1.5 nm; shifts toward thinner layers
Defect (Raman/XPS)	Minimal defects	Low defect levels, mainly at the edges	Very low defect levels; basal planes largely defect-free

Third, fluid dynamics techniques have great potential to produce quality graphene in a more environmentally friendly manner than chemical methods. VFDs are suitable for products with minimal defects, but production is limited. PFDs are efficient and capable of controlling size, but they require high pressure and a complex design. MFDs offer the most practical and economical solutions for large-scale production. With technological advancements, this approach presents a significant opportunity to support the commercially sustainable production of graphene.

Supercritical Fluids

A supercritical fluid is a substance that exists at temperatures and pressures above its critical point, where the boundary between the liquid and gaseous phases is lost. Under these conditions, the fluid has liquid-like properties with high density while also having high diffusivity, like gases. This combination of properties allows supercritical fluids to penetrate the interlayer gaps of graphite and facilitate exfoliation without the need for harsh chemicals. In addition, when the pressure is lowered, the rapid expansion of the fluid results in an effective separating force between graphite layers [45].

In graphene synthesis, supercritical fluids can be divided into two main types: inert and reactive. Inert fluids such as CO_2 do not react chemically with graphene, making them suitable for producing pure graphene. In contrast, reactive fluids such as supercritical ethanol not only aid in exfoliation but can also modify the surface of graphene through chemical reactions. The selection of this type of fluid is adjusted to the needs of the desired graphene end application [46]. The following table lists the differences in graphene characteristics generated through the two supercritical fluid approaches, as shown in Table 4.

Table 4. Summary of characterization data produced by supercritical fluid methods.

Sample	AFM	Raman	XRD	Ref.
Graphite + SC-CO_2	>10 layers	Weak 2D peak; high I_D/I_G ratio indicating limited exfoliation	Intense (002) peak with slight broadening,	[47]

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			indicating minor delamination	
Graphite + SC-CO_2	Majority <3 layers (yield ~28%)	88% <3 layers; sharp 2D peak confirms few-layer graphene	Clear graphene structure; no signs of oxidation	[48]
Graphite + SC ethanol	~1.0–1.2 nm thickness; 6–10% monolayer content; stable	Low I_D/I_G ratio (~0.17); symmetric 2D peak at 2684 cm^{-1}	(002) peak intensity decreased; increased interlayer spacing	[49]
Graphite + SC ethanol	Few layers (maximum yield ~18.5%)	Slight increase in I_D/I_G ratio; minor defects introduced	Decrease in (002) peak intensity; successful exfoliation	[50]

Exfoliation of graphene with pure SC-CO_2 tends to result in less homogeneous structures and a higher number of defects. In contrast, the use of SC-ethanol is more effective in producing graphene with a more stable thickness distribution, a more regular structure, and a lower defect rate. Thus, SC-ethanol is superior for the synthesis of high-quality graphene.

Detonation Technique

The detonation technique is a graphene synthesis method that utilizes the explosive reaction of carbon compounds, such as acetylene, in the presence of oxygen or oxidizing agents to produce graphene nanosheets [51]. The high temperatures and pressures created during the detonation process break down carbon molecules and facilitate the formation of graphene structures [52]. This method is very fast, does not require a catalyst, and is capable of producing high-quality graphene efficiently, making it suitable for large-scale production [53]. The following table summarizes the graphene characterization data generated by this method, as shown in Table 5.

Table 5. Summary of characterization data produced by detonation methods.

Methods	TEM	Raman (I_D/I_G)	XRD (2θ)	Ref.
C_2H_2 + O_2 gas detonation (ratio 0.4–0.8)	2–3 layers; monolayers observed; lateral size increases with higher O_2 ratio	Decreases from ~1.33 to ~0.28, indicating reduced defects and improved crystallinity	(002) peak at 26.05°, close to graphite (26.6°), indicating preserved graphite structure	[53]
O_2/C_2H_2 detonation (O/C ratio 0.25–0.75)	8–30 layers; turbostratic structure; lateral size 20–200 nm	Decreases when O/C > 0.5, suggesting fewer defects and a more ordered structure	(002) peak shifts from 25.33° to 25.74°, lower than graphite, indicating increased interlayer spacing	[52]
Solid explosive: $CaCO_3$ + Mg + RDX	<10 layers; transparent and crumpled sheets	~0.26, indicating very few defects and high-quality graphene	(002) peak at 26.04°, close to graphite, suggesting good crystallinity	[51]
GO to ER-GO (thermal	Thin, transparent sheets	No numerical value reported; D and G bands	GO: 7.9° (d ≈ 1.09 nm); ER-GO: approximately	[54]

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reduction at 100°C)	are present, indicating moderate defect density	26.3°, indicating partial restoration of graphite-like structure

The detonation technique is capable of producing high-quality graphene with a good crystalline structure, minimal defects (low I_D/I_G), and a varying number of layers. This process is fast, catalyst-free, and suitable for large-scale production, making it an efficient and promising method for graphene synthesis.

2.2. Oxidation-Reduction

The oxidation-reduction method is a graphene synthesis technique that involves oxidizing graphite to produce graphene oxide (GO), followed by exfoliation and chemical reduction to form reduced graphene oxide (rGO) [55]. This process begins with the oxidation of graphite using strong oxidizers, such as sulfuric acid, nitric acid, or potassium permanganate, which introduces oxygen functional groups, including hydroxyl, epoxy, and carboxyl, into the graphite layer [56]. This function group increases the distance between layers, allowing exfoliation into GO sheets through sonication in solvents such as water [57]. The resulting GO is hydrophilic and easily dispersed; however, it lacks the conductive properties of graphene due to the disruption of the sp^2 structure caused by oxidation [58].

The reduction stage is performed to remove the oxygen group and restore the sp^2 carbon structure, using reducing agents such as hydrazine, sodium borohydride, or thermal methods to produce reduced graphene oxide (rGO) [59]. Although this method enables the production of large quantities of graphene at a low cost, rGO often exhibits structural defects and oxygen group residues that degrade its electronic properties compared to pure graphene [60]. The quality of graphene can be improved by optimizing the reduction conditions, such as using environmentally friendly reducing agents or electrochemical reduction methods [61]. The oxidation-reduction method is very popular due to its scalability and ease of process; however, the main challenge is minimizing defects for applications that require high-quality graphene [62].

The different processes for oxidizing and reducing graphene will be covered in this section, along with how they affect the final product's quality and characteristics. The primary focus is on how these techniques affect graphene's conductivity, defects, and structure for the best possible applications.

2.2.1. Oxidation Method

The properties and applications of graphene can be greatly impacted by the classification of oxidation methods used in graphene synthesis, such as chemical, thermal, and electrochemical oxidation. It is essential to comprehend this method in order to optimize graphene for a variety of applications.

2.2.2. Chemical Oxidation

Chemical Oxidation is the initial stage in the synthesis of rGO, where graphite is converted to graphene oxide (GO) using a strong oxidizing agent in an acidic solution. The most common method is the Hummers method or its modification, which uses a combination of potassium permanganate ($KMnO_4$) and sulfuric acid (H_2SO_4), often with the addition of sodium nitrate ($NaNO_3$) [63]. In some variations of these methods, oxidizing agents such as nitric acid (HNO_3), peroxide (H_2O_2), or a mixture of phosphoric acid (H_3PO_4) and potassium permanganate ($KMnO_4$) are also used [64]. This reaction introduces various oxygen groups such as hydroxyl, epoxy, and carboxyl into the graphite structure, making GO water-soluble and ready for the reduction process to rGO [65].

Thermal Oxidation

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In order to introduce oxygen groups at high temperatures, thermal oxidation entails heating graphite with oxygen or other oxidizing gases. To increase the effectiveness of functional group recognition, this technique is frequently used in conjunction with chemical oxidation [66]. Compared to chemical oxidation, thermal oxidation produces graphene with fewer defects because it allows for controlled oxidation at high temperatures. In addition, this method is often used in conjunction with chemical reduction to produce rGO that has better conductivity [67].

Electrochemical Oxidation

This method utilizes an electric current to oxidize graphite in an electrolyte solution, which can be a neutral salt solution, such as sodium sulfate (Na_2SO_4), or an environmentally friendly electrolyte solution that is less harsh than a strong chemical agent [68]. This process avoids the use of harmful chemical oxidizers such as $KMnO_4$ and strong acids, making it safer and more environmentally friendly [69].

Reduction Method

The following is the reduction of graphene oxide (GO): The structure and electrochemistry of rGO are impacted by techniques like chemical, thermal, and electrochemical treatments, which in turn affect the properties and uses of materials.

Chemical Reduction

The cost-effectiveness and scalability of chemical reduction make it a popular choice. In order to eliminate the oxygen group from graphene oxide (GO) and partially restore the graphitic structure, reducing agents such as sodium borohydride, hydrazine, metals, and L-ascorbic acid are used in this process [70]. Reduction capacity, toxicity, and environmental impact can all be impacted by changing the choice of reducing agents [60].

Thermal Reduction

Thermal Reduction is achieved by heating graphene oxide (GO) in an inert atmosphere, such as argon or nitrogen, to remove oxygen groups as gases (e.g., CO and CO_2) [71]. This method does not require any additional chemicals and can produce rGO with high conductivity. However, the process requires good temperature control to prevent damage to the carbon structure [72].

Electrochemical Reduction

Electrochemical Reduction involves the use of an electric current to reduce graphene oxide (GO) inside an electrochemical cell [73]. GO is used as an electrode, and when it is given a negative voltage, the oxygen groups on its surface are removed through electron transfer. This method is fast, clean, and allows for good control over the reduction results, making it widely considered for sustainable applications [74]. The following table summarises the graphene characterization data obtained via the oxidation-reduction method, as shown in Table 6.

Table 6. Summary of characterization data produced by oxidation-reduction methods.

Oxidation	Reduction	FTIR	Raman (I_D/I_G)	XRD	Ref.
Thermal	Thermal	High intensity of –OH, C=O, and C–O functional groups	Decreases with increasing temperature and time	(002) peak at 26.5°, interlayer spacing d ≈ 3.36 Å	[75]

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Chemical (Hummers)	Chemical ($NaBH_4$)	High –OH and C=O intensity; epoxy group is reduced	0.98	Interlayer spacing d = 0.388 nm; average number of layers ≈ 1.4; crystallite size ≈ 22 nm	[70]
Electrochemical	Chemical (hydrazine)	Presence of –OH, C–O–C, C–C, and C=O groups	0.849	2θ = 26.52°; grain size ≈ 23 nm	[76]
Chemical (Hummers)	Electrochemical	Decrease in –OH, C=O, and C–O functional groups	1.24	Interlayer spacing d = 0.3554 nm	[77]
Chemical (Hummers)	Chemical (Ascorbic acid)	Decrease in C=O, C–OH, and C–O–C; partial restoration of sp^2 structure	Decreases from 0.805 to 0.788 with increasing temperature	GO peak at ~11.9°; rGO peaks between 24.8° and 25.2°; d-spacing ≈ 3.55 Å	[78]
Chemical (Hummers)	Chemical (hydrazine)	C=O and C–O functional groups are reduced	2.23	GO: ~10.9° (001); rGO: ~26.4° (002); interlayer spacing decreases	[79]
Chemical (Hummers)	Thermal	C=O and C–OH groups are reduced	-	GO: ~10.5°; rGO: 24.7° to 26.2°; d-spacing for GO ≈ 0.84 nm; for rGO ≈ 0.34–0.36 nm	[80]
Chemical (Hummers)	Chemical (Zn metal)	Decrease in C–O–C, C–OH, and C=O groups; oxygen-containing groups are weakened	1.01	GO (002) peak at ~26°; additional ZnO peak with wurtzite structure observed	[81]

The synthesis of rGO begins with the oxidation of graphite using thermal, chemical (Hummers), or electrochemical methods to introduce oxygen groups that facilitate exfoliation. Furthermore, reductions are carried out chemically (using $NaBH_4$, hydrazine, or ascorbic acid), thermally, or

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electrochemically to remove these oxygen groups, improve the sp^2 structure, and decrease the defects, which is reflected in the decrease in the Raman I_D/I_G ratio and changes in the XRD pattern.

2.2.3. Arc Discharge Method

The arc discharge method is a graphene synthesis technique that uses the discharge of electricity between graphite electrodes in a gaseous environment to form a carbon plasma, which condenses into graphene, usually few-layer graphene (FLG) [82]. High-quality graphene with a good crystalline structure is produced by this process, which evaporates graphite at high temperatures. To guarantee homogeneity and purity, however, exact control of variables like gas pressure and current is necessary [83]. Large-scale production is possible with this straightforward process, but minimizing contamination and flaws is a challenge [84]. According to the research that has been done, the following factors influence the quality of graphene using the arc discharge method:

Effect of Buffer Gas Type

Table 7 summarizes how the type of buffer gas greatly affects the properties and quality of graphene produced by arc discharge.

Table 7. Summary of the effect of gas type on the quality of graphene via arc discharge methods.

Types of Gas	Result	Ref.
Ar	~12% graphene sheets comprising 1–10 layers; interlayer spacing of 0.34–0.39 nm	[85]
Ar	High-quality, very pure 4-layer graphene	[86]
H_2	2–4-layer graphene, free of nanotube contaminants	[87]
H_2-N_2	Up to 5 layers of graphene, low defect density, suitable for mass production	[88]

By using various gases, the arc release method creates graphene with unique properties: argon for high-quality multilayers, hydrogen for pure graphene with little contamination, nitrogen-hydrogen mixtures for scalability and quality balance, and helium for monolayers. The particular requirements of the application are taken into consideration when choosing the gases.

Effect of Current Type

The structure, purity, and general quality of the synthesized graphene are greatly influenced by the type of electric current used in the arc discharge, whether it be direct current (DC) or alternating current (AC). The relative impact of the two existing types on different facets of graphene production is compiled in Table 8.

Table 8. Summary of the effect current type on the quality of graphene via arc discharge methods.

Aspects	Air conditioning	DC	Ref.
Structure	Nanohorns, carbon onions, 1–5 layers of graphene	Carbon nanotubes, 2–4 layers of graphene	[89],[88],[86],[87]
Purity	Very pure; minimal carbon contamination	Less pure; mixed with non-graphitic carbon	[89], [86]

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Process & Control	Flexible, frequency-controlled process	Less flexible; dependent on gas, metal catalyst & pressure	[89],[90],[91]
Graphene Quality	Low defects, optimal with N_2/H_2 mixture	Minimal defects, optimal with argon gas	[88],[86]
Scalability	Large-scale, suitable for industry	Suitable for small–to medium-scale and specialized applications	[88],[91]
Efficiency & Results	High efficiency when optimizing gas composition & frequency	Generally lower efficiency than the air-conditioning method	[88],[90]

Although the DC and AC arc release methods have their advantages, their selection depends on specific needs in graphene synthesis, such as the desired quality, structure, and production scale. The DC method is generally preferred to produce high-quality graphene with double or more layers and minimal defects, while the AC method is superior in large-scale production as it offers more flexible control over the structural characteristics of graphene.

Effects of Pressure

Because it directly affects the final graphene's morphology, purity, and structural quality, pressure is a crucial parameter in arc discharge techniques. The effects of various pressure ranges on graphene properties are compiled in Table 9.

Table 9. Summarizes the impact of pressure using arch discharge techniques on graphene quality.

Pressure	Number of Layers	Purity	Ref.
Low	Formation of nanohorns and nanospheres, no coated graphene layers	Low purity, a hybrid of various carbon nanostructures	[92]
Moderate	Approximately 4-layer graphene, thermally and structurally stable	High purity, well-ordered structure without toxic intercalates	[86]
High	Graphene with 2–10 layers, pronounced condensation observed	High purity, uniform, defect-free graphene structure	[92]

The trade-off between pressure regulation and the desired graphene properties must be taken into account, even though the arc release method works well for graphene synthesis. While medium and high pressures are more beneficial for creating graphene with fewer layers and higher purity, low pressures are less suitable for creating high-purity graphene. The pressure choice needs to be customized to meet the demands of the final graphene's particular application.

Effect of Reaction Temperature

Reaction temperature plays a crucial role in determining the number of layers, growth rate, and purity of graphene synthesized via arc discharge. The kinetic energy available at different temperatures influences the atomic mobility, plasma behavior, and overall structural integrity of the resulting material. Table 10 summarizes the effects of various temperature ranges on graphene quality.

Table 10. Summary of reaction temperature against quality graphene via arch discharge methods.

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Temperature	Number of Layers	Purity	Ref.
Low	Single-layer graphene grown at low energy	High purity, low-carbon atomic mobility, minimal defects	[93]
Moderate	2–4 layer graphene with balanced growth energy	High purity, stable plasma, minimal fouling	[86], [87]
High	> 4-layer graphene; rapid growth due to high energy	Lower purity, increased mobility lead to more defects and contamination	[93], [94]

The arc release method is effective in producing graphene, but the temperature must be carefully controlled to optimize the number of layers and purity. High temperatures accelerate growth but can also contribute to defects, whereas low temperatures yield single-layer graphene with fewer defects. Medium temperatures provide balance, resulting in multi-layer graphene with high purity, which is crucial for tailoring it to specific applications.

Effect of Reaction Time

The number of layers, growth rate, and purity of graphene produced by arc discharge are all significantly influenced by the reaction temperature. Atomic mobility, plasma behavior, and the final material's overall structural integrity are all influenced by the kinetic energy accessible at various temperatures. Table 11 summarizes the relationship between reaction time and graphene quality.

Table 11. Summary of reaction time against quality graphene via arc discharge methods.

Duration	Number of Layers	Purity	Ref.
Short	2–4 layer graphene	High purity; slightly distorted structure	[87], [95]
Moderate	4–6 layer graphene	High purity; some layer non-uniformity and minor structural flaws	[82], [94]
Long	Up to ~20 layers graphene	Decreased purity; more defects, mitigable with buffer gas	[83]

The arc removal method has the ability to control the number of layers and the purity of graphene, which is very important for adapting its properties to a specific application. Rapid synthesis is particularly suitable for applications that require high purity and fewer layers, whereas longer synthesis can produce graphene with more layers, making it suitable for applications that require thicker structures. However, the balance between the number of layers and purity must be carefully managed to optimize material performance according to the application's specific needs.

Effect of Chamber Type

The configuration of the reaction chamber influences the quality, purity, and layer number of graphene. Table 12 summarizes the effects of different chamber types on graphene produced via arc discharge.

Table 12. Summary of reaction chamber type against quality graphene via arch discharge methods.

Chamber Type	Number of Layers	Purity	Ref.
Closed chamber	Approximately 4 layers of graphene	High purity, minimal defects	[82],[86]
Semi-open chamber	Moderate, depending on parameters	Moderate purity, possible contamination	[82]

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Open chamber	Multilayer graphene (many layers)	Low purity, high defect density	[96]

Although enclosed spaces typically produce graphene with high purity and fewer layers, this type may be less suitable for applications that demand large yields. In contrast, open and semi-open spaces, while at risk of producing more defects, are more suitable for mass production. Therefore, the selection of the type of space should be tailored to the needs of the application, taking into account the balance between the number of layers, purity, and yield.

2.2.4. Unzipping of Carbon Nanotube

The unzipping of Carbon Nanotubes (CNT) is a *top-down* method for the synthesis of graphene, specifically graphene nanoribbons (GNR), by splitting the cylindrical structure of the CNT into flat graphene tapes. This process utilizes a CNT wall composed of rolled graphene sheets, resulting in GNR with width and length that depends on the dimensions of the CNT [97][98]. While effective for producing graphene with controlled edges, this method can cause structural damage [99]. Various methods as follows, can unzip carbon nanotubes (CNT) into graphene nanoribbon (GNR):

Oxidative Zipper Retraction

This method utilizes strong oxidizers to break the carbon bonds in carbon nanotubes (CNTs), resulting in graphene nanoribbons (GNRs) [100]. This process is general and flexible, allowing the regulation of GNR properties through oxidation rates [101]. Nonetheless, harsh chemical conditions often damage the structure of CNT crystals and degrade their conductivity [102].

Catalytic Zipper Opening

This technique uses microwaves and metal nanoparticles, like palladium, to efficiently accelerate the unzipping of carbon nanotubes. Although layered GNR can be produced using this process, the primary obstacles are the possibility of metal contamination and the requirement for stringent reaction control [103].

Electrochemical Zipper Opening

The CNT zipper can be opened with a high degree of precision and little damage thanks to this method, which uses an electric field in the electrolyte medium [104]. Although it necessitates specialized equipment and rigorous electrochemical condition control, this method works well for nitrogen-doped CNTs [101].

Sonochemical Zipper Opening

This process utilizes ultrasonic waves to open the CNT mechanically. This method is relatively inexpensive and simple, and can produce GNR with smooth edges. However, the quality of the final product may vary due to limited control over the process [105].

A comparison of the characteristics of graphene nanoribbon (GNR) generated through various carbon nanotube (CNT) unzipping methods is presented in the following table to highlight the advantages and limitations of each approach, as shown in Table 13.

Table 13. Summary of characteristics of GNR against various methods for unzipping CNT.

Method	**Number of Layers**	**Purity**	**Ref.**
Oxidative	Single to multi-layer graphene	Multiple defects; presence of oxygen-containing functional groups	[100],[106],[107]

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Catalytic	4 – 8 layers graphene	Slight defects, traces of residual metal catalyst	[103]
Electrochemical	Single to multi-layer graphene	High purity, minimal defects	[108]
Sonochemistry	Predominantly bilayer graphene, with some monolayer	Smooth edges, low interference	[109]

Each CNT *unzipping method* produces GNR with its advantages and limitations. The oxidative method is easy to do but poses many defects. Catalytic and electrochemical methods produce purer GNR with a more intact structure, although the process is more complex. Sonochemistry is relatively simple, yielding smooth-edged results, but it is not easy to control. The selection of methods depends on the target application and practical considerations such as purity, efficiency, and environmental impact.

2.2.5. Liquid Phase Exfoliation

Liquid-phase exfoliation (LPE) is a top-down method of graphene synthesis that utilizes solvents or chemicals to separate the graphene layers from the graphite. Unlike mechanical exfoliation, which is physical in nature, this method utilizes chemical interactions or reactions to weaken the forces between layers [110]. Some of the techniques included in chemical exfoliation include:

Graphite Intercalation Compounds (GIC)

GIC is a compound that results from the insertion of chemicals (such as metals or acid molecules) into a graphite layer to widen the distance between layers to facilitate the process of exfoliating graphene through heating or ultrasonication [111]. Intercalation with alkali metals, such as potassium, can increase the distance between graphite layers from 0.34 nm to approximately 0.53 nm. This increase weakens the inter-layer van der Waals force, making it easier to exfoliate into graphene [112].

Intercalation with acid molecules, such as $HClO_4$, can produce graphene with high exfoliation efficiency. This intercalation widens the distance between graphite layers, as evidenced by the XRD peak at $2\theta = 23.1°$. After sonication, the separate layers reform into graphene. The Raman spectrum showed an increase in the peaks of D and G, signalling an increase in disorder and successful exfoliation [110].

Chemical Exfoliation with Organic Solvents

Chemical exfoliation with organic solvents is a method of separating the graphene layer from graphite by utilizing a solvent with a surface tension close to that of graphene (30–40 mJ/m^2) [112]. Solvents such as N-methyl-2-pyrrolidone (NMP), dimethylformamide (DMF), and dimethyl sulfoxide (DMSO) are commonly used because they are able to stabilize graphene in suspension and improve exfoliation efficiency [113].

Method Chemical Exfoliation with pure organic solvents produces low-concentration graphene (<0.1 mg/mL). The addition of organic salts such as sodium citrate, sodium tartrate, potassium sodium tartrate, and EDTA disodium has been shown to significantly improve exfoliation efficiency, resulting in high-quality graphene (1–3 layers, oxide-free and defective) with concentrations of up to ~1 mg/mL in 2 hours of sonication [114].

Chemical Exfoliation with Ionic Liquid Exfoliation

Liquid phase exfoliation with ionic liquid is an environmentally friendly top-down method to produce graphene from graphite. The ionic liquid acts as a stable solvent with a corresponding

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surface tension, thus allowing the separation of the graphene layer without damaging its structure [115], as shown in Table 14.

Table 14. Summary of ionic liquids type against characteristic graphene via LPE.

Ionic Liquids	SEG (mg/mL)	Number of Layers
$[C_4C_1im][Ntf_2]$	~1.8	≤5
$[Pyrr_{4,1}][Ntf_2]$	~1.8	≤5
$[N_{4,1,1,1}][Ntf_2]$	~1.8	≤5
$[C_{10}C_1im][Ntf_2]$	<1.8	≤5
$[BnzmC_1im][Ntf_2]$	<1.8	≤5
$[C_4C_1im][C(CN)_3]$	<0.5	>30
$[C_2C_1im][N(CN)_2]$	<0.5	≤5
$[C_4C_1im][C_1SO_4]$	<0.5	≤5
$[C_2C_1im][OTF]$	<0.5	<5

$[Ntf_2]$ based ionic liquids, specifically $[C_4C_1im][Ntf_2]$ and $[Pyrr_{4,1}][Ntf_2]$, produce graphene with high concentrations, ≤5 layer counts, and the best purity. In contrast, ionic liquids with other anions, such as $[C(CN)_3]$ and [Otf], show low efficiency and high oxidation rates, signalling the crucial role of ionic structures in exfoliation quality [116]. Ionic liquids have been shown to produce high-concentration graphene dispersions without chemical modifications, but their use is still limited due to their high price and high viscosity, which affect the efficiency of exfoliation [115].

Chemical Exfoliation with Surfactants

Liquid exfoliation with surfactants is a method to separate graphite into graphene sheets in solution using ultrasonication, where surfactants function to stabilize graphene sheets so that they do not clump together. This method is effective, simple, and suitable for large-scale graphene production [117], as shown in Table 15.

Table 15. Summary of Surfactants on Characteristics of Graphene via LPE.

Surfactant	CG Max (mg/mL)	Optimal Csur (mg/mL)	CMC (mg/mL)
SDOC	~0.10	~1.0	5.0
SDBS	~0.11	~0.7	0.7
SDS	~0.09	~2.0	2.3
HTAB	~0.12	~0.3	0.33
Tween 80	~0.08	~0.015	0.0157
Triton X-100	~0.29	~1.0	0.343

From Table 15, the six types of surfaces studied, exfoliation with Triton X-100 resulted in the highest concentration of graphene (~0.29 mg/mL) in (~1 mg/mL) surfactant. TEM and AFM show thin sheets with a thickness of 1–3 nm and <5 layers. The average flake size of 46.83 μm^2 obtained from SDOC samples indicates a complete and quality graphene morphology. Raman and XPS in the Triton X-100 sample showed low defects and dominance of sp^2 structures. The best dispersion stability is also demonstrated by Triton X-100 and Tween 80, with more than 80% of the graphene remaining dispersed after 700 hours [118].

Exfoliate with Low Boiling Point Solvent

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Exfoliation with a low-boiling-point solvent is a method of separating a layer of graphite into graphene using a volatile solvent. High-boiling point solvents were previously widely used because they were effective, but they were difficult to vaporize and could cause clumping during drying. The use of low-boiling-point solvents is a solution to overcome this problem [119], as shown in Table 16.

Table 16. Summary of Solvent type against effectiveness via LPE.

Solvent	Boiling Point (°C)	Concentration (mg/mL)	Result
Acetone	56	~0.08	Low concentration, suitable for low-boiling dispersion.
Chloroform	61	~0.5	Stable (≥75 % remains suspended after 100 h), produces medium-sized flakes.
Isopropanol	82	~0.5	Highly stable (>90% fixed suspended >200 hours); produces good quality flakes
Cyclohexa-none	156	~1.0	Effectively exfoliates graphene; high boiling point makes solvent removal challenging.
NMP	204	~1.0	Effectively exfoliates graphene; high boiling point makes solvent removal challenging.
DMF	153	~1.0	Effectively exfoliates graphene; high boiling point makes solvent removal challenging.

Exfoliation with volatile solvents yields good quality graphene, with a thickness of <5 layers and a low defect rate based on TEM and Raman analyses. Solvents such as isopropanol and chloroform are not only easy to evaporate but are also able to maintain the stability of graphene dispersion. In contrast, high-boiling point solvents produce higher concentrations, but they tend to cause aggregation during drying, which can degrade the quality of graphene [120].

Exfoliation with Electrochemistry

Electrochemical exfoliation is a fast and environmentally friendly method of producing graphene oxide from graphite using an electric current in an electrolyte solution [121]. This process does not require strong oxidizers, is suitable for large-scale production, and is capable of producing <10-layer nanosheets with a lateral size of >1 μm [122].

The shape of graphite has a significant effect on the results of electrochemical exfoliation. Compressed graphite powder produces graphene with a lateral size of >30 μm and a yield of 65% [123]. Natural powder produces GO with an oxygen content of 25.3 at.%, an I_D/I_G ratio of 0.85, and ±9 layers [124]. Graphite foil produces ~1.0 nm thick GO, consisting of 1–3 layers, with a yield of up to 96%, while rods break easily and flake exfoliate quickly but with a lower yield (<40%) [121].

In addition, the type of electrolyte also determines the quality of the graphene formed. H_2SO_4 (0.5 M) electrolytes produce I_D/I_G of 0.35 with rapid blistering, Li_2SO_4 (0.5 M) produce I_D/I_G of 0.25 with moderate exfoliation, and $NaClO_4$ (1 M) produce I_D/I_G of 0.17 with non-destructive intercalation [125]. Electrolytes such as $HClO_4$ and HNO_3 form epoxy and alkoxy groups, $(NH_4)_2SO_4$ allow doping of N and S, while ozone produces 1–3 layers of GO with 16.37 at.% oxygen and 1.21 I_D/I_G [121]. This data confirms that the combination of graphite shape and the right type of electrolyte greatly determines the quality and efficiency of GO production.

2.3. Bottom up

The *bottom-up* approach is a graphene synthesis strategy that is carried out by gradually forming the structure of graphene from small molecular units such as carbon atoms, usually through chemical

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processes or deposition. This method generally produces high-quality graphene with a good level of structural control, although scalability and production costs are major challenges. The following techniques are used in the synthesis of graphene by *the bottom-up* method:

2.3.1. Chemical Vapor Deposition (CVD)

Chemical Vapor Deposition (CVD) is a vacuum deposition method to produce high-quality graphene through the chemical reaction of a gas precursor on a hot substrate, such as copper or nickel [126]. This process produces the carbon atoms that make up graphene by breaking down gases like methane [127]. For electronics and sensor applications, CVD makes it possible to create graphene with a large surface area and a consistent crystal structure [128]. To reduce flaws in the graphene layer, this procedure necessitates rigorous regulation of temperature, pressure, and gas flow [129].

Temperature

The temperature of synthesis has a major impact on the quality of graphene generated using the CVD process. The number of layers, purity, degree of defects, and interaction with the substrate are all influenced by temperature [130]. High-quality graphene is characterized by uniform coatings, slight defects, high purity, and good substrate interactions, all depending on temperature settings during the process [131]. The following sections discuss the impact of various low, high, and ultra-high temperature ranges on these aspects, as shown in Table 17.

Table 17. Summary of temperature reactor conditions against graphene characterisation via CVD.

CVD Temperature	Layers	Defects	Substrate Interactions	Ref.
Low	Slow film growth, non-uniform coverage	High defects, insufficient energy	Weak interactions, delamination potential.	[132]
High	Fast carbon diffusion, uniform and continuous graphene film	Reduced defects, more perfect structure	Strong interaction, adhesion and stability are better.	[133], [134]
Ultra-High	Rapid growth, increased film thickness	Further reduced defects, though new defects may emerge	Potential substrate etching and adverse interactions	[135],[136]

By increasing layer uniformity, decreasing defects, and enhancing substrate interactions, high temperatures enhance graphene quality; however, they can also increase energy consumption and cause substrate damage. Although they use less energy, low temperatures can cause quality degradation. For some applications, extremely high temperatures are advantageous, but they must be carefully managed to avoid aggravating flaws or damaging the substrate. These factors must be balanced in order to optimize the CVD process and produce graphene of the appropriate quality.

Pressure

The quality of graphene produced by CVD is greatly influenced by process pressures, whether atmospheric, low, or ultra-vacuum. This pressure affects the number of layers, defects, purity, and interaction with the substrate, with each condition offering its own advantages and challenges [137], as shown in Table 18.

Table 18. Summary of Pressure reactor conditions against graphene characterization via CVD.

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CVD Pressure	Coating & Uniformity	Defects	Substrate Interactions	Ref.
APCVD	Large area, fairly uniform, limited control	High defect, decreased purity	Strong substrate interaction, difficult to transfer	[138],[133]
LPCVD	Uniform, high-quality monolayer	Low defects, high purity	Weak interaction, easy to transfer	[139]
Ultra-Vacuum	Highly uniform, high precision	Very low defects, optimal purity	Minimal interaction, ideal for transfer	[140]

Each pressure condition has its own advantages depending on the needs of the application. Ultra-vacuum is suitable for high-precision applications because it produces the purest graphene, while APCVD is more efficient and suitable for large-scale production despite higher defects.

Wall/Substrate

The quality of graphene synthesized with CVD is affected by the configuration of the walls/substrates, both cold walls and hot walls [141]. Here are the differences in graphene results obtained from cold wall and hot wall configurations in the CVD method, as shown in Table 19.

Table 19. Summary of cold wall and hot wall configurations against graphene characterization via CVD.

Aspects	Cold Wall CVD	Hot Wall CVD	Ref.
Number of Layers	Uniform and thin layer formation	Thicker layers, less uniform	[142],[143]
Defects	Low defects, high purity	Higher defect density, reduced purity	[142],[144]
Substrate Influence	Better process control, high-quality graphene	Greater substrate influence; increased defect formation	[145],[133]

The hot-wall method is still useful for applications that value cost and scalability, but the cold-wall CVD method produces graphene with superior quality. By optimizing both approaches, research and technological advancements can lessen the disparity in quality between them.

Deposition Time

The quality of graphene produced by CVD is greatly influenced by deposition times, including continuous, disconnected, and pulsed methods. Each approach has a different impact on the number of layers, defects, and purity, so it is important to choose the right timing strategy to optimize the structural and functional properties of graphene [135], as shown in Table 20.

Table 20. Summary of deposition times against graphene characterization via CVD.

CVD Method	Coating & Uniformity	Defects	Ref.
Continuous	Uniform, suitable for large areas	Low defect density, high material purity	[146],[147]
Diconected	Less uniform	Higher defect density, lower purity	[148]

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Pulsed	Controlled and tunable deposition	Low defect density, high material purity	[135],[136]

The continuous, intermittent, and pulsed CVD methods have their own advantages depending on the application. Continuous CVD produces the most uniform graphene and minimal defects; intermittent CVD is suitable for property modification, while pulsed CVD offers high control for customization.

Gas Flow State

The quality of graphene produced through CVD is affected by the gas flow method, which is an open and closed CVD system. Open systems use a continuous flow of gases, while closed systems maintain a static gas environment. These two methods affect the number of layers, the defect density, and the purity of graphene in different ways [149], as shown in Table 21.

Table 21. Summary of gas flow configurations against graphene characterization via CVD.

CVD System	Gas Flow & Dynamics	Defects & Purity	Scalability	Ref.
Open	Continuous flow (methane, H_2), stable reaction	Slight defects, high purity	Suitable for large-scale, even gas distribution	[138]
Closed	Static gas, layer growth control	High crystallinity & purity, sensitive to reaction conditions	Less suitable for large-scale use without process optimization	[150]

The choice between open and closed CVD systems depends on the specific quality needs of the graphene application. Open systems are better suited for large-scale production with fewer uniformity and defects, while closed systems offer better control over the purity and thickness of the coating, ideal for applications that require high quality.

Activated Manner

The synthesis of graphene by various CVD methods, such as thermal, plasma, and laser, has a major effect on the number of layers, defect density, and purity. Each method has advantages and disadvantages that affect the final quality of graphene [151], as shown in Table 22.

Table 22. Summary of CVD techniques against the quality and efficiency of graphene synthesis.

Method	Defects & Purity	Layer	Efficiency	Ref.
PECVD	Low defects, high purity	SLG–FLG (dependent on parameters)	Fast, low temperature, suitable for large-scale	[152],[153], [154],[155]
TCVD	Higher defect density; RT-CVD offers improvement	Generally monolayer	Slow, high temperature, RT-CVD is more efficient	[156]
Laser-CVD	Low defects with precise control	Multi-layer (dependent on	Fast, ideal for specific patterns	[157]

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	laser parameters)

The selection of the CVD method depends on the application requirements and the desired properties of graphene. PECVD is suitable for pure graphene with low defects; lasers are ideal for fast patterns, and superior thermal methods for large areas, albeit slower. Efficiency and quality are largely determined by the process parameters and the end goal of using graphene.

2.3.2. Epitaxial Growth of Graphene Silicon Carbide

Epitaxial growth of graphene on silicon carbide (SiC) substrates is a synthesis method in which a layer of graphene is formed directly on the surface of the SiC crystal through a process of thermal decomposition at high temperatures, generally above 1200 °C [158]. At this temperature, the silicon atoms from the surface of SiC evaporate, leaving behind carbon atoms that then compose themselves into a graphene structure [159]. This process is often carried out in the atmosphere of inert gases such as argon to control the sublimation rate and improve the uniformity of the graphene layer [160]. The uniqueness of this method is that it does not require the process of transferring graphene to other substrates, thus reducing the risk of contamination and mechanical damage [161].

In the process of epitaxial growth of graphene in silicon carbide, there are several important factors that significantly affect the quality and characteristics of the graphene layer produced. Factors that affect the quality of graphene with this method include as following:

Surface

SiC substrates have two surface sides, namely the silicon side (Si-face) and the carbon side (C-face), which significantly affect the growth outcome of graphene. In Si-face, growth usually results in a uniform monolayer graphene layer and bonds to the substrate via a buffer layer, which can modify the electronic properties of graphene [162]. In contrast, the growth on the C-face produces a multilayer of graphene with a random orientation, resulting in electronic properties that resemble substrate-free graphene [163]. Due to its advantages in uniformity, stability, and ability to produce graphene layers over large areas, this epitaxial method holds great promise for high-speed electronics applications and quantum metrology standards [159]. The following are the results of graphene analysis with the difference between Si-face and C-face, as shown in Table 23.

Table 23. Summary of the effect of substrates on graphene characteristics via SiC methods.

Parameters	Si-face	C-Face
Number of layers	1–2 layers (monolayer)	Up to ~30 layers (multilayer)
Raman Spectrum	$I_D/I_G < 0.02$ (sharp G and 2D peaks)	$I_D/I_G < 0.05$, broader peaks with creases
Electron mobility	Relatively low	Relatively high

Graphene from Si-face has a regular epitactic structure, smooth surface, and controllable monolayer thickness, making it suitable for precision electronics applications such as field effect transistors (FETs) and sensors. In contrast, graphene from the C-face tends to grow in multilayer forms with random orientations and corrugated surfaces, yet offers higher electron mobility and weak interactions with substrates, making it more suitable for applications such as capacitors, batteries, or transparent conductors [164].

Temperature

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Growth temperature plays an important role in the quality of graphene synthesized through the epitaxial growth method on silicon carbide (SiC) because it directly affects the evaporation rate of silicon (Si) from the SiC surface and the formation of graphene layers [165]. At high temperatures, Si atoms evaporate from the surface, leaving behind carbon atoms that are then composed into graphene structures [166]. Temperatures that are too low can lead to incomplete or irregular graphene formation [167], while too high a temperature can lead to too rapid evaporation of Si, resulting in a thick, non-uniform, or deformed layer of graphene [168]. The following are the results of graphene analysis with temperature differences, as shown in Table 24.

Table 24. Summary of Temperature growth against graphene characteristics via SiC methods.

Temperature	Number of Layers	Electron Mobility	Description
Low	0 – 0.6 layers	Very low (up to 81 cm^2/Vs)	Graphene is not fully formed; significant exposure of SiC surface remains
Optimal	~1.2 – 1.4 layers	Highest (~370 cm^2/Vs)	Nearly monolayer graphene, maximum mobility, uniform surface morphology
High	>1.6 layers	Decreasing (up to 77 cm^2/Vs)	Multilayer graphene, presence of grain boundaries and "giraffe stripe" patterns

Optimal growth temperatures produce graphene monolayers with the highest electron mobility, while temperatures that are too low or too high produce low-quality graphene due to inadequate thickness or structural defects [169].

Pressure

Pressure greatly affects the evaporation rate of silicon during the epitaxial growth of graphene from SiC. At low pressures, silicon evaporates faster, which can lead to uncontrolled growth of graphene and rough surfaces [170]. In contrast, the use of an inert atmosphere, such as argon at high pressure, can slow the evaporation of silicon, allowing the carbon to be composed more stably into a finer and more uniform layer of graphene [171]. The following are the results of graphene analysis with pressure differences, as shown in Table 25.

Table 25. Summary of pressure effects against graphene characteristics via SiC methods.

Pressure	Raman	Surface Morphology	Graphene Quality
Low Pressure (~10^{-7} mbar)	Weak G and 2D bands, dominant D band	Rough surface with many defects	Low-quality, non-uniform graphene
Inert Atmosphere (~10^{-3} mbar)	Strong G and 2D bands, D band nearly absent	Smooth surface with uniform morphology	High-quality, thin, and uniform graphene layers

The pressure during the epitaxial growth of graphene greatly affects its quality. At low pressures, silicon evaporates too quickly, so graphene is heavily deformed and morphologically rough. Whereas at inert atmospheric pressure, the sublimation rate is inhibited, resulting in a smoother, uniform, and high-quality layer of graphene [167].

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Catalyst

By aiding in the breakdown of silicon from the SiC surface, catalysts in the epitaxial growth of graphene in silicon carbide contribute to the acceleration and direction of the graphene layer formation process. Graphene can grow more regularly and with fewer defects if a catalyst is present to control the silicon sublimation rate. Because they make it easier for carbon atoms to come together to form graphene structures, metal-based catalysts like nickel and copper are frequently employed.

However, the use of metal catalysts must also be considered so as not to cause contamination that can degrade the quality of graphene, especially in its electronic properties. Additionally, catalysts can influence the morphology and size of graphene domains, enabling growth with larger crystals and more uniform layers [172]. The following are the results of graphene analysis without a catalyst and with a catalyst, as shown in Table 26.

Table 26. Summary of Catalyst effects against graphene characteristics via SiC methods.

Method	Number of Layers	Raman	Surface Morphology	Graphene Quality
No Catalyst	6–7 layers	G and 2D wideband, I_D/I_G > 0.4	Rough surface, many defects, non-uniform	Low-quality, multilayer graphene with small crystallite
With Ni–Cu Catalyst	Monolayer	G and 2D sharp band, I_D/I_G ~0.24, 2D > G	Smooth surface, uniform morphology	High quality, uniform monolayer graphene with large crystallites (35–60 nm)

The use of Ni–Cu catalysts on graphene epitaxial growth in 3C–SiC/Si significantly improved the quality of graphene, resulting in a monolayer structure with a low I_D/I_G ratio (~0.24), smooth surface, and uniform morphology. Without catalysts, the graphene produced is multilayer, high defects, and has a rough surface[172].

3. Conclusions

Graphene is a two-dimensional material that is stronger, more conductive, and more heat-resistant than other materials. Scientists have come up with several techniques to produce graphene, employing both top-down and bottom-up methods. Top-down processes like mechanical exfoliation, oxidation-reduction, arc discharge, unzipping carbon nanotubes, and liquid phase exfoliation are all examples of procedures that are usually easy to use and may be employed to produce things on a large scale. But this method usually creates graphene that has structural problems and sizes that aren't always the same.

On the other hand, bottom-up technologies like Chemical Vapour Deposition (CVD) and Epitaxial Growth on Carbide may generate high-quality graphene with a nice crystal structure and a number of layers that can be controlled. However, this method does need intricate, costly, and inefficient equipment for making things in large quantities. Because of this, it is better for high-tech uses that need very high purity and accuracy.

There is still no one-size-fits-all approach to manufacturing graphene that works for all purposes. So, the way that graphene is made should alter depending on the final goal, which could be to generate the best graphene possible or to speed up the process. We need to work on building graphene synthesis procedures that are better for the environment and use green chemistry to help make production more sustainable in the future.

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Abbreviations

The following abbreviations are used in this manuscript:

PMMA	Polymethyl methacrylate
PDMS	Polydimethylsiloxane
FTIR	Fourier Transform Infrared
SEM	Scanning Electron Microscopy
XRD	X-ray Diffraction
AFM	Atomic Force Microscopy
TEM	Transmission Electron Microscopy
DMF	Dimethylformamide
NMP	N-Methyl-2-pyrrolidone
SC	Supercritical
RDX	Research Department Explosive
ER-GO	Explosively Reduced Graphene Oxide
SEG	Solvent-Exfoliated Graphene
SDOC	Sodium Deoxycholate
SDBS	Sodium Dodecylbenzenesulfonate
SDS	Sodium Dodecyl Sulfate
HTAB	Hexadecyltrimethylammonium Bromide
CG	Concentration of Graphene
Csur	Concentration of Surfactant
CMC	Critical Micelle Concentration
APCVD	Atmospheric Pressure Chemical Vapor Deposition
LPCVD	Low Pressure Chemical Vapor Deposition
PECVD	Plasma Enhanced Chemical Vapor Deposition
TCVD	Thermal Chemical Vapor Deposition

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