

A Review Study On The Chemical Vapor Deposition Of Graphene

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Abstract— Graphene as a promising nanomaterial with diverse potential applications in electrical, optoelectrical, biomedical, and energy storage devices, has gained wide attention in industrial and research communities. Much research is being carried out to implement the large-scale production of high-quality graphene. Chemical vapor deposition (CVD) has emerged as the most promising method. This review presents the chemistry of graphene and its structure, properties, and applications. Major CVD methods used for graphene growth, include hot-wall, cold-wall, and plasma-enhanced approaches. The role of substrate, reactant gases, and other parameters in the CVD processes for graphene growth are reported. This review also summarizes the limitations of the CVD methods of graphene growth and the advantages of CVD-grown graphene over other techniques.

Keywords— Graphene; CVD; plasma; optoelectronics; nanomaterials

I. INTRODUCTION

Graphene - a two-dimensional crystalline nanomaterial - has gained widespread attention in the research community ever since its first isolation from graphite in 2004 (1). Graphene is an allotrope of carbon comprising of carbon atoms arranged in a hexagonal shape (Figures 1&2). The promising nanomaterial has been discovered to significantly improve the efficiency of photovoltaic devices and reduce their cost of production (2-5). Other areas in which graphene has reportedly been known to have possible applications, include electronics, biomedical, pharmaceuticals, and biosensors (1,6), with more still undergoing experimentation. The increasing advancement in modern technology to satisfy the present demand and meet future needs has led to progressive innovation in semiconductor industries. Although silicon has been regarded as the most prominent semiconductor in recent decades (7), new reports shown by the *International Technology Roadmap for Semiconductors* and *International Roadmap for Devices and Systems* revealed that Moore's law (the number of transistors in an integrated circuit doubles in about every two years) will shortly be exhausted by the limitation imposed by the diameter of the silicon atom.

Because the efficiency of technological device is constantly improved on while the sizes are getting smaller, the need to create a smaller chip becomes inevitable. To avert these future limitations, graphene is being progressively researched as a potential substitute, with its efficiency surpassing silicon. Graphene, with its unique attributes is known to attract electric charges and has been successfully integrated into an electronic circuit. Graphene nanoribbons—short, thin strips of graphene— have been proposed as an option because of their size and semiconductive properties and can be integrated into transistors (8-12). The wide application of graphene has not been entirely commercially successful because of the limitation of its mass production. The three primary graphene production methods include chemical vapor deposition (CVD), chemical or plasma exfoliation from natural graphite, and mechanical cleavage from natural graphite. CVD is the most used technique (13,14).

Multiple layers of graphene, known as epitaxial graphene, have also been developed primarily to achieve a direct layering of graphene on readily available semiconductor materials. However, graphene epitaxy is distinctively different from graphite (15-17). Epitaxial graphene comprises multiple graphene layers, with the electrons of the first layer doped by a built-in electric field while other layers are left undoped. Conversely, graphite is a naturally occurring multiple graphene layer comprising delocalized electrons with high mobility in-between graphene layers (18). *Neeraj et al* (16) gave a brief overview of the recent development of graphene epitaxy grown on silicon carbide by thermal decomposition. The group speculated that this technique would likely bring graphene potentialities into implementation.

Chemical vapor deposition is a process carried out in a vacuum vapor phase to synthesize thin films and nanomaterials on a solid material known as a substrate. Even though the technology requires high temperatures and a high vacuum, it is largely used by semiconductor industries for thin film production. CVD has wide applications in materials technology and has been used for depositing metallic, non-metallic, and semiconductor materials (19). A recent overview of the CVD technique revealed the production of high-quality graphene and the direct growth of graphene on any substrate (20). While continuous effort is being

exerted in the research community and the industry to explore the potentialities of graphene and its applications fully, production techniques need to be examined to ensure the consistent availability of graphene for commercial purposes. This review first summarizes the chemistry of graphene, and its structure, properties, and applications. Secondly, a brief overview of the chemical vapor deposition process, and various CVD methods of graphene growth, applications, and limitations will be discussed. Lastly, the advantages of chemical vapor deposition over other methods of graphene growth will be reviewed.

II. GRAPHENE

Graphene is a 2-dimensional nanostructure material made of layers of carbon atoms covalently bonded in a hexagonal pattern. The atoms are attached by a single or double bond to three surrounding carbon atoms (Figures 1 & 2). It is an allotrope of carbon in which each carbon atom is SP² hybridized with a molecular bond length and thickness of about 0.142 and 0.335 nm, respectively (21). Graphene can be referred to as a single layer of graphite. It is the essential underlying component of carbon allotropes such as graphite, carbon nanotubes, and fullerenes (Figure 1).

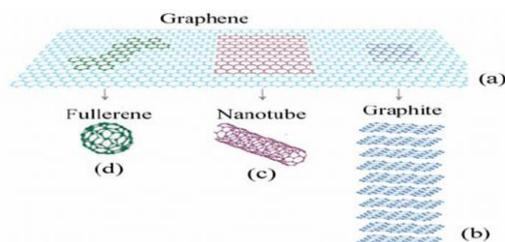


Figure 1: Schematic structure of graphene (a), graphite (b), carbon nanotube (c), and fullerene (d) (22).

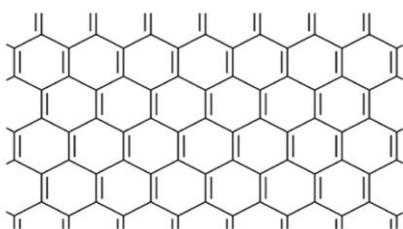


Figure 2: Chemical structure and shape of graphene (23).

Graphene, however, is different from other graphitic forms of carbon, due to its unique properties as an allotrope carbon and a semiconductor material. Furthermore, graphene is known to exhibit a shorter bond length, higher boiling and melting points, greater tensile strength, and higher modulus properties than diamond. In addition, the covalent bond in the lattice structure of graphene makes it stronger and stiffer than diamond (24).

A. Chemical Properties of graphene

Graphene generally demonstrates non-metallic and semiconducting properties. Each carbon atoms possess a free electron in the p-orbital. Also, the lattice structure contains delocalized double bonds, giving room for electrical conductivity in two dimensions. The melting point of graphene was formerly estimated to be 4510 K. In contrast, a recent computational study shows that the bonded atoms of a graphene sheet survive melting at the temperature. In contrast, bond breaking, an indicator of melting, was observed at about 5000 K. However, experimental studies are being anticipated to establish this prediction (25). A single layer of graphene has a surface area of 2630 m²/g; the surface area of the reduced form of graphene has also been investigated (26–28). Graphene can be chemically modified due to the high reactivity of the edge atom i.e, pi-delocalized electrons located on edge (Figure 2). The SP² hybridization of the unsaturated carbon atoms allows graphene reactions with hydrogen, fluorine, oxygen, acids, and free radical atoms. The reactive edge atoms enable further modification of graphene molecules with OH, COOH, CHO, and polymers (29). Recent research has revealed the chemical modification of 2-D graphene to a 3-D honeycomb-like structure, and the electrochemical reactions of graphene have also been investigated (30). Furthermore, different sorts of porosity inside the sheet, which are exceptionally normal, increase its chemical reactivity.

B. Mechanical Properties of Graphene

Graphene is known to possess excellent mechanical properties among other carbon nanomaterials. It is light weighted, flexible, and recognized as the thinnest nanomaterial. Graphene demonstrates ultimate tensile strength (the maximum stress a material can bear before breaking when it is stretched or pulled) of above 1 TPA or 130 GPA (31,32). A theoretical study by Bao et al. shows that a single graphene layer has better mechanical properties than networks of the graphene layer (33). On the other hand, a study by Kumar et al. shows that the increase in the mechanical properties of graphene is directly proportional to the number of layers (34). Recently, computational research was conducted by Gamboa-Suárez et al. to investigate the effects of high temperature on the mechanical properties of graphene. The novel study revealed the resistance of graphene sheets to stiffness degradation at an ultra-high temperature below the melting point (32). The mechanical property of graphene is extraordinary and, as such, can essentially improve the quality of numerous composite materials.

C. Electronic Properties of Graphene

The fundamental electrical property of graphene is attributed to the zero gaps in its band structure. Carbon, as the elemental constituent of graphene structure, has four valence electrons; three valence electrons are bonded to other carbon atoms in the 2-D plane, leaving one free electron in the 3-D plane available for electronic conduction. These free

electrons, also known as pi electrons, are situated at the top and bottom of the graphene sheet (Figure 1) and foster carbon-to-carbon bonding. These attributes make graphene behaves like a metal, and the difference in the energy level of the valence electron band (bonding) and conduction electron band (antibonding) determines the electronic conductivity of graphene (35).

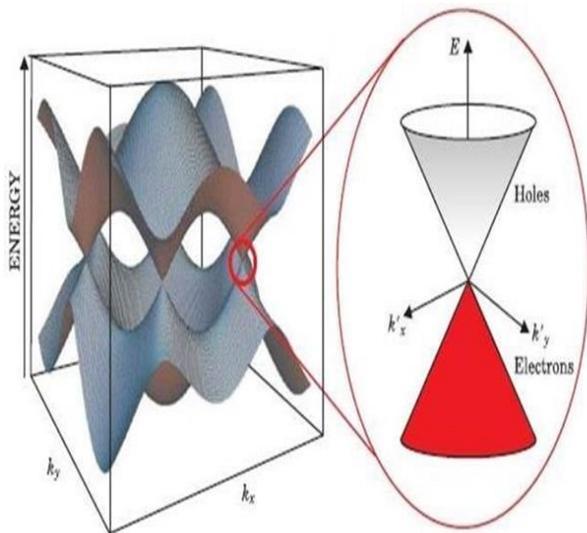


Figure 3: Band structure of a single-layered graphene (36)

The electronic properties of graphene can be modified by doping, i.e., introducing excess electrons and holes into graphene crystals. This approach is being used to create a band gap in graphene. A recent study revealed an improvement in the electronic properties of graphene because of subjection to photochlorination (37). Another study showed that the oxidized form of graphene exhibits poor electronic conductivity; however, graphene oxide can be reduced to improve its electrical conductivity (38,39).

D. Optical Properties of Graphene

One of the unique features of graphene is its ability to absorb electromagnetic radiation over a wide range of wavelengths. Graphene is known to exhibit varying optical responses at each region of the electromagnetic spectrum. This property is attributed to the zero-band gap and how the region interacts with the Dirac fermions of graphene. Also, the graphene monolayer absorbs over 2.3% of the visible light spectrum (40,41). Graphene can also generate some optical transitions when an electric field is applied close to the Dirac point, which causes a shift in the Fermi level. This technique is employed to adjust optical source transmission in electrical circuits. Furthermore, functionalized graphene's linear and non-linear optical properties have been investigated. A study shows that the optical properties of graphene can be impacted by the edge length and SP3 domain of a reduced or oxidized form of graphene (42).

E. Applications of Graphene

Because of its distinctive chemical and exceptional mechanical strength and electronic and optical properties, graphene has gained wide applications in biomedical and electronic devices in solar and energy conversion and as a semiconducting material. Graphene and graphene-based materials are used in photodetectors, light-emitting diodes (LED), and other optoelectronic devices due to their wide spectral ranges and high optical transmissions (42,43). Graphene based materials have reportedly been used as transducing materials in electrochemical biosensors for cancer biomarker detection (44–46). Furthermore, graphene-based materials are also used as active materials in energy storage devices such as supercapacitors, lithium-ion batteries, sodium-ion batteries, fuel cells, and also in thermal energy storage systems such as solar cells (47–50). Environmental applications of graphene involve using functionalized graphene-based materials to remove toxic chemical species in environmental components. In wastewater treatment, graphene-based materials are used to adsorb toxic compounds because of their large surface area. They are also used for electrochemical treatments of wastewater due to their oxidative-reductive abilities (51). A study by *Zafar and Jacob* revealed that graphene is a good material for oil-water separation because of its hydrophobic oil affinity (52). Graphene has also gained wide attention in agricultural management, according to an overview presented by *Chen et al.* The review stated that the oxidized form of graphene (Graphene Oxide) is an ideal material for water conservation due to the water-loving affinity of the graphene's oxygen atoms. Graphene treatment can improve water used by plants, thereby promoting photosynthetic activities in green plants (53). Another review mentioned the use of graphene nanocomposites for plant disease treatment, also as an effective fertilizer coating material that minimizes eutrophication (54). Much research is still ongoing to unveil the more potential applications of graphene and graphene-based materials.

III. CHEMICAL VAPOR DEPOSITION

Chemical vapor deposition is a technique used for depositing thin film materials on a heated solid material. The process employs a thermochemical reaction for making nanomaterials. It is a popular technique adopted in materials technology for semiconductor processing. The CVD process is carried out in a vacuum at a high temperature. Before the main chemical reaction of material deposition on substrates in the reactor, the precursor gases are homogenized in a gas mixer and then transferred into the reactor through the gas inlet system. The gaseous by-products exit the system through the gas outlet (Figure 4).

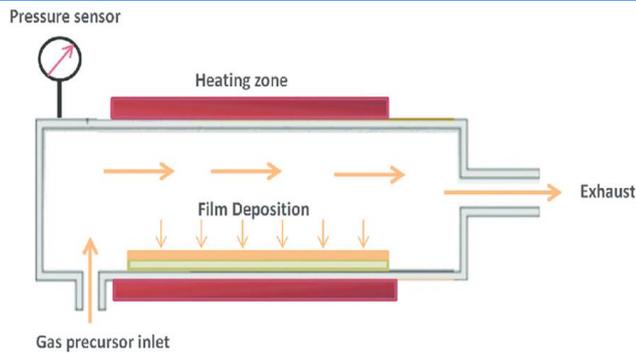


Figure 4: Schematic diagram of a chemical vapor deposition system

The efficiency of a CVD process is determined by the substrate and the process parameters such as temperature, precursor gas ratio, pressure, gas flow rate, and so on (6,56). In the CVD process, the precursor gas serves as the starting material, which undergoes several reaction phases to form the expected product. The purity, ratio, flow rate, and injection mode of the precursor materials can impact the reaction process. Hydrocarbon molecules (C-H) are the major sources of graphene growth, where gaseous hydrocarbons are commonly used as precursors. In addition, liquid hydrocarbons such as benzene, toluene, and xylene and solid hydrocarbons such as polymethylmethacrylate and polystyrene have also been used. Liquid and solid precursors for graphene growth alleviate high-temperature demands. The mechanism also proposes a less sophisticated and inexpensive process for commercial production (57–59). Likewise, alcohol precursors such as 2-phenyl ethanol and ethanol have also been experimented with as carbon sources in the CVD process of graphene growth. The study revealed a successful growth of single and multiple layers of graphene (60). Temperature is another factor that plays a vital role in the CVD process; it supplies the heat energy needed to facilitate material deposition on substrates; however, the temperature range varies at different reaction phases. The temperature gradient of graphene wool synthesized by the CVD process shows a rapid temperature increase at the initial stage, an equilibrium temperature at the intermediate reaction phase, and a gradual temperature decrease at the completing stage (61). Selecting a suitable substrate also determines the performance of CVD growth, substrates do not only provide support for material deposition, but it also has a catalytic effect on CVD growth. Because of the great catalytic abilities of copper (Cu) and nickel (Ni), they have been selected among other transition metals as good fits for graphene growth; however, some difficulties are accompanied by using metal substrates. One of the major setbacks of Ni substrate is the formation of multilayer graphene films. At high temperatures, more carbon atoms become soluble and absorbed by Ni during graphene synthesis, which then isolates to form multiple layers of graphene films upon cooling (62). Cu substrates, on the other hand, produce a uniform

and high-quality graphene film on a large scale, however, the deposited graphene films need to be transferred which can lead to contamination or damage (63,64). Graphene growth on solid non-metal substrates is considered advantageous as the process offsets the possibilities of contamination caused by graphene transfer. Some of the developed non-metal substrates for CVD-grown graphene, as reviewed by Shi *et al.* include metal oxides such as SiO₂, Al₂O₃, MgO, Ga₂O₃, TiO₂, and ZrO, with the oxides providing the catalytic effect needed for the reaction. In addition, graphene growth on insulated glass materials e.g, quartz, flexible non-metallic substrates such as mica, polyimide, and polydimethylsiloxane (PDMS) were also discussed (65,66). Single and multiple layers of graphene grown on sapphire substrates have also exhibited high stability and are predicted as a good fit for electronic devices (67–69). In an experimental study carried out to investigate the significant process parameters of graphene growth, and quartz substrate was used to grow graphene materials by APCVD. The precursors (CH₄ and H₂) flow rates, temperature, growth time, and cooling rates were controlled to monitor the effects of each parameter on graphene synthesis. At a relatively low temperature, graphene growth was inhibited due to CH₄ inability to decompose to form carbon atoms needed for nucleation. Contrarily, CH₄ decomposition was enhanced at high temperatures leading to more carbon atoms being generated for nucleation to take place. Rapid cooling and high H₂ flow rate were reported to have positively contributed to graphene homogeneity. The CH₄ flow rate was varied from 20 to 200 sccm (represents standard cubic centimeters per minute), and homogenous flake-like graphene was produced at a maximum flow rate of 100 sccm while increasing the flow rate resulted in a non-homogeneity. The study showed that temperature and precursor flow rates play a significant role. At the same time, other process parameters, such as cooling, and growth time, only contribute to the quality of synthesized graphene (65). CVD is a multifaceted and powerful technique with a consistent exploration of its applications. The research communities and industries are constantly working on advancing CVD technology to widen its scope of applications. CVD technology is widely used in the semiconductor industry for thin film synthesis. The technique is preferred over other fabrication processes due to its cost-effectiveness and allows large-scale production. Unlike physical vapor deposition techniques such as sputtering and evaporation, the CVD process generates quality thin film with high purity and density and good coating performance. In addition, CVD techniques require cost-effectiveness; it is widely used in the semiconductor industries for large-scale thin film production (20,56,70).

A. Methods of CVD

CVD technology can be categorized based on the variable process parameters such as substrates heating (cold-wall and hot-wall), temperature (low-temperature, high-temperature, and ultra-high

temperature), precursors (metal-organic and inorganic), pressure (atmospheric pressure, low-pressure and ultra-vacuum), activation techniques (thermal, plasma-enhanced, laser-induced, photo, combustion, and catalyst-assisted). The widely used CVD methods for laboratory and commercial purposes include: 1) hot-wall CVD – the reactor is first heated at a uniform temperature which in turn heats the substrate and also facilitates precursor decomposition, 2) cold-wall - the reactor remains cold while only the substrate is heated up, 3) low-pressure CVD (LPCVD) – the vacuum chamber is operated at a pressure below atmospheric pressure, 4) ultra-high vacuum pressure – the vacuum chamber is operated at relatively high pressure, 5) plasma-enhanced CVD - precursor gases are passed through electrodes and becomes ionized by radio frequency current or electric current form a plasma, the energy generated drives the chemical reaction and enhance material deposition on the substrate at a low temperature (6, 20). A general application of CVD methods will be summarized concerning their principles, features, and applications.

B. Hot-Wall CVD of Graphene

In a hot-wall CVD technology (Figure 5), the substrate is first heated by the radiation from the reactor, after which liquid or gaseous precursors are then introduced into the reaction chamber through the gas delivery system. In the reaction chamber, the temperature is increased to supply the heat energy required to facilitate the chemical reaction. The introduced gas becomes heated up and adsorbed on the substrate surface; a heterogenous chemical reaction takes place between the gaseous materials and the surface. Consequently, the reactive gaseous molecules decompose and become deposited on the substrate while the unreactive molecules exit the system through the gas outlet (21). In a study carried out by *Gausden et al.*, where hot-wall CVD was used to grow graphene on a copper substrate, the copper foil was first preheated in the reactor to a temperature of about 1050 °C at 20 °C/min ramp rate. The precursor gases (CH₄ and H₂) were then introduced at a varied flow rate using nitrogen as the carrier gas (71). Another study on 3D graphene growth using hot wall CVD revealed that the dissociation rate of precursor gases in a hot wall CVD is catalyzed by temperature, thereby increasing the graphene growth rate (72).

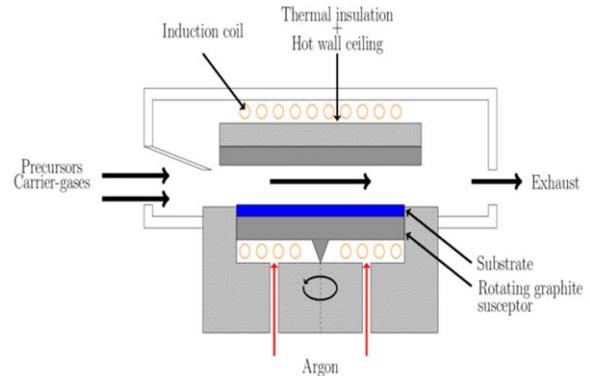


Figure 5: Schematic diagram of hot-wall CVD (73).

In another experiment, a carbon nanotube was grown directly on a silicon substrate using the hot-wall CVD technique. The substrate was pretreated before heating in the reactor; methane was used as a carbon source carried by H₂ gas into the reaction chamber. The reaction was carried out in a large reactor to demonstrate the possibilities of a large-scale deposition. The chemical properties, morphology, and structure of deposited carbon materials were characterized with nano equipment. The study reported that carbon materials produced in a hot-wall CVD, even though on a large scale, are a better fit for microelectronics applications when compared with other CVD methods due to a good mixture of precursor gases (74). Hot-wall CVD has shown to be advantageous as it offers a homogenous coating of materials. It is widely used in depositing materials with thick layers needed for high-voltage supply equipment (75). However, the major drawback is the risk of material deposition on the reactor wall and frequent cleaning of the reactor.

C. Hot-Wall CVD of Graphene

The substrate is only heated in a cold wall CVD system while the reaction chamber is maintained at atmospheric temperature. The substrate is selectively heated up in the reactor chamber by placing it on a hot surface or passing an electric current through it. This method is considered advantageous because it provides fast growth, requires a less complex set-up, and is cost-effective. In addition, particulate contamination is prevented, thereby generating a good-quality graphene film. Likewise, regular cleaning of the reaction chamber is alleviated as there is no film deposition on the reactor wall (21). However, this method is not commonly used for graphene growth due to low throughput caused by the inability to accommodate multiple substrates. *Wasyluk et al.*, efficiently accomplished graphene growth on a cobalt-chromium substrate with the cold wall CVD technique. The result from the study indicated a promising method for coating medical devices as excellent mechanical properties were observed; also, the coated material shows resistance to pro-coagulation (76). A computational analysis was conducted using a fluid dynamics simulation technique to experiment cold wall

CVD method of graphene growth theoretically. The study revealed that low temperature keeps the gas-phase reaction, which improves the quality of graphene produced. Hence, cold wall CVD is a good technique for the large-scale production of high-quality graphene film. The study also proposed further research on the gas-phase reaction of other CVD methods of graphene growth to improve the mass production of an immaculate graphene film (77). Furthermore, the cold wall CVD method is attractive for transferring graphene grown on copper substrates to SiO₂/Si surface. In an investigation by *Soriadi et al.*, graphene monolayers were successfully grown with no crystal defect (78) *Sarno et al.*, had previously demonstrated a successful transfer of graphene grown on copper foil to a polyethylene tetra phthalate (PET) substrate with the possibility of employing the process to develop a transparent electrode and act as a protective device against oxygen degradation in solar cells (79). Likewise, graphene monolayers were successfully grown on Ni substrate by *Faisal et al.* using cold wall CVD. The study revealed that higher-quality graphene monolayers were produced compared to other CVD methods. The improvement in quality was attributed to a better-controlled process of the physical parameters in cold wall CVD and the fast-cooling process (80).

D. Plasma-Enhanced CVD of Graphene

Plasma is considered the fourth state of matter; it is generated when a gaseous molecule becomes ionized when it is subjected to extreme heat or intense electromagnetic radiation. This plasma-enhanced CVD (PECVD) method of graphene growth provides an advantage of a faster deposition rate at a low temperature. In a PECVD set-up (Figure 6), the precursor gases pass in between a grounded and an energized electrode; the gaseous molecules become ionized by the electrode to form a plasma, which generates a reaction with the product being deposited on the substrate. The energized electrode introduces radio frequency, microwave, or a direct current to excite the molecules (21).

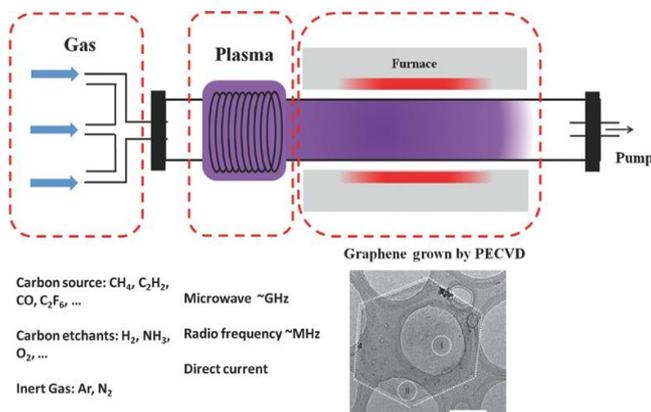


Figure 6: Schematic diagram of plasma enhanced CVD (81)

Lu et al., demonstrated graphene growth on copper ink and copper substrate using a single-step PECVD method with a microwave as the plasma generator. In

the study, CH₄ and H₂ were used as the precursor gases, while nitrogen served as the carrier gas. The partial pressure of the precursor gases was monitored before the plasma formation. Graphene growth on the copper ink and substrates was confirmed using Raman spectroscopy after the reaction, while surface topography and roughness were characterized before and after the process. Some of the advantages the PECVD technique provides from the study include an improvement in the quality of copper ink, elimination of graphene transfer after growth, elimination of graphene annealing, and formation of graphene flakes on copper substrates (82). *Duan et al.*, in their study on PECVD graphene growth on a copper substrate using radio frequency as a plasma generator, highlighted the drawback of low substrate temperature in the PECVD process. Their study demonstrated a solution to this shortcoming by keeping the reactant gases at a low temperature and atmospheric pressure during the reaction while the copper substrate was being heated up at the same time. In addition, a mixture of CH₄ and H₂ was compared to pure CH₄ to examine the anti-corrosive properties of the synthesized graphene. At a lower temperature (<200 °C) and longer time (>900 min), both tests showed an equivalent anti-corrosiveness. At the same time, pure CH₄ exhibits a superior anti-corrosive property between 200 °C – 300 °C (83). Similarly, *Yen et al.*, had previously revealed in their study that H₂ gas restricts the nucleation and growth mechanism of graphene in the PECVD technique with a radio frequency energizer (84). Furthermore, *Hong et al.*, displayed in their study on graphene growth using inductively coupled plasma CVD that the etching effect resulted from the increase in the partial pressure of hydrogen radicals caused by an increase in the plasma generator power, and a low-power power can alleviate the effect. From the study, the overall growth rate was observed to increase with plasma power. However, high-quality, uniform graphene film was produced at a lower plasma power of about 50 W (85). Even though PECVD is efficient in graphene growth rate, the technique also compromises the quality of graphene film produced. Graphene film grown by PECVD exhibits higher nucleation density, thereby impairing the electronic application. *Li et al.*, demonstrated in their study that the nucleation density of graphene film synthesized with the PECVD technique could be reduced by using a Faraday cage (86). In the experiment, the copper substrate was pretreated and placed in a Faraday cage- before the in-flow of the precursor gases. The essence of the cage is to protect the electric field and control the reactant's flux without taking part in the reaction. This procedure reduces the nucleation density and deposition rate of graphene film; also, long graphene grains were produced with high electronic conductivity.

E. Limitations of CVD

Although CVD is regarded as a good technique for large-scale synthesis of graphene film, the promising techniques also exhibit some drawbacks. One of the

major problems is the generation of toxic by-products. During the CVD process, the reaction between the reacting gases and substrate generates some secondary gaseous species which do not take part in the reaction. These by-products are removed through an exhaust system (81). The thermal CVD technique requires high temperature for the operation, which is energy and cost-consuming. In addition, graphene grown using this method exhibit minimal applications in electronic and optoelectronic devices as the process requires a low temperature (87). Another drawback of this CVD method is the inefficiency in depositing graphene film on a thin-metal substrate. Because the process requires heating the reaction chamber and the substrate at a high temperature, some metal films become vaporized during the heating process (88). On the other hand, graphene is grown using the CVD at a low temperature, and atmospheric pressure compromises the film quality. For instance, Hong et al. compare the thermal CVD and PECVD methods of graphene growth using CH₄ as the carbon source. From the investigation, CH₄ remains undissociated in the gas phase at the commonly used temperature for graphene growth in thermal CVD. In PECVD, even though the process is carried out at a low temperature, the generated plasma provides activation energy, and more than 80% of CH₄ dissociated to form H, H₂, CH₃, CH₂, and C₂H₂.

Meanwhile, the increasing hydrogen molecules increase the nucleation density, causing etching on the graphene, thereby reducing the uniformity and quality of the graphene film generated (89). To improve the quality of graphene produced using PECVD, *Li et al.*, proposed using a Faraday cage, which significantly decreased the nucleation density of synthesized graphene, as demonstrated in their study.⁸⁶ Aside from the enormous cost involved; further research is still needed to provide more information on the effectiveness and feasibility of synthesizing high-quality graphene with PECVD.

F. Advantages of CVD Graphene Growth Over Other Methods

Among the three popular methods of producing graphene – chemical vapor deposition, chemical or plasma exfoliation from natural graphite, and mechanical cleavage from natural graphite- CVD is the most viable method of large-scale production of graphene film. Mechanical exfoliation involves the cleavage of graphene layers from natural graphite. It requires using scotch tape to peel off graphene layers from graphite. Similarly, chemical exfoliation consists of the intercalation of graphite in an acidic solution to enhance the easy peel-off of graphene layers from graphite. It is the simplest method for graphene growth; however, the process is time-consuming and produces thick graphene with no uniform quality (90,91). Likewise, these methods' industrial application of graphene has not been ascertained. Another method of graphene growth is epitaxial growth on silicon carbide. In this method, SiC is heated up at a very high temperature, Si molecules

sublime while the carbon atoms condensed to form graphene layers. The major disadvantage of this technique is the difficulty in transferring the synthesized graphene film from one substrate to another (92). CVD method is widely recognized as a large-scale synthesis of high-quality graphene film. Graphene grown using the CVD technique is transferrable from one substrate to another. It is cost-efficient with wide industrial applications, especially for film deposition in semiconductor industries. In addition, CVD methods generate high-quality graphene film with excellent uniformity. The films are used in biomedical, energy storage devices, electrochemical sensors, electronics, and optoelectronics devices.

IV. SUMMARY

With its diverse potential applications, graphene needs to be produced on a large scale for industrial purposes. Chemical vapor deposition has emerged as a promising technique for the commercial production of quality graphene film. The chemical, mechanical, electrical, and optical properties of graphene were discussed. Various CVD methods of growing graphene were also considered in this review. Hot-wall CVD requires heating the reactor chamber and substrate at a high temperature and generates uniform graphene coating on metal substrates. Cold wall CVD requires heating the substrates only while maintaining the reaction chamber at a low temperature; it requires a less complicated setup and a fast deposition rate. The PECVD method is facilitated by plasma generated from electromagnetic radiation while the reaction is maintained at atmospheric pressure and a low temperature. Generally, CVD methods is widely recognized as techniques for large-scale synthesis of high-quality graphene film. However, the viable approach still has some shortcomings. Further research on minimizing the by-product generated during the CVD process is recommended. In addition, more investigation should be carried out on efficient graphene transfer from substrate to another, and high-quality graphene synthesis using PECVD method.

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REFERENCES

- [1] Novoselov, K. S.; Geim, A. K.; Morozov, S. v.; Jiang, D.; Zhang, Y.; Dubonos, S. v.; Grigorieva, I. v.; Firsov, A. A. Electric Field Effect in Atomically Thin Carbon Films. *Kluwer*. **2000**, Vol. 404.
- [2] Zhang, J.; Fan, J.; Cheng, B.; Yu, J.; Ho, W. Graphene-Based Materials in Planar Perovskite Solar

Cells. Sol. RRL. John Wiley and Sons Inc. 2020. <https://doi.org/10.1002/solr.202000502>.

[3] Czerniak-Reczulska, M.; Niedzielska, A.; Jędrzejczak, A. Graphene as a Material for Solar Cells Applications. *Adv. Mat. Sci.* 2015, 15 (4), 67–81. <https://doi.org/10.1515/adms-2015-0024>.

[4] Wang, J. T. W.; Ball, J. M.; Barea, E. M.; Abate, A.; Alexander-Webber, J. A.; Huang, J.; Saliba, M.; Mora-Sero, I.; Bisquert, J.; Snaith, H. J.; Nicholas, R. J. Low-Temperature Processed Electron Collection Layers of Graphene/TiO₂ Nanocomposites in Thin Film Perovskite Solar Cells. *Nano Lett.* 2014, 14 (2), 724–730. <https://doi.org/10.1021/nl403997a>.

[5] Cai, L.; Yu, G. Recent Advances in Growth and Modification of Graphene-Based Energy Materials: From Chemical Vapor Deposition to Reduction of Graphene Oxide. *Small Methods. John Wiley and Sons Inc.* 2019. <https://doi.org/10.1002/smtd.201900071>.

[6] Saeed, M.; Alshammari, Y.; Majeed, S. A.; Al-Nasrallah, E. Chemical Vapor Deposition of Graphene—Synthesis, Characterization, and Applications: A Review. *Molecules MDPI.* 2020. <https://doi.org/10.3390/molecules25173856>.

[7] Yeh, C. H.; Liang, Z. Y.; Lin, Y. C.; Chen, H. C.; Fan, T.; Ma, C. H.; Chu, Y. H.; Suenaga, K.; Chiu, P. W. Graphene-Transition Metal Dichalcogenide Heterojunctions for Scalable and Low-Power Complementary Integrated Circuits. *ACS Nano.* 2020, 14 (1), 985–992. <https://doi.org/10.1021/acsnano.9b08288>.

[8] Feng, C.; Xiang, J.; Liu, P.; Wang, X.; Wang, J.; Hu, G.; Huang, M.; Wang, Z.; Zhang, Z.; Liu, Y.; Lu, Y.; Xiang, B. Magnetic Logic Inverter from Crossed Structures of Defect-Free Graphene with Large Unsaturated Room Temperature Negative Magnetoresistance. *Nano Res.* 2019, 12 (10), 2485–2489. <https://doi.org/10.1007/s12274-019-2472-y>.

[9] Wang, K.; Li, L.; Lan, Y.; Dong, P.; Xia, G. Application Research of Chaotic Carrier Frequency Modulation Technology in Two-Stage Matrix Converter. *Math. Probl. Eng.* 2019. <https://doi.org/10.1155/2019/2614327>.

[10] Kim, W.; Li, C.; Chekurov, N.; Arpiainen, S.; Akinwande, D.; Lipsanen, H.; Riikonen, J. All-Graphene Three-Terminal-Junction Field-Effect Devices as Rectifiers and Inverters. *ACS Nano* 2015, 9 (6), 5666–5674. <https://doi.org/10.1021/nn507199n>.

[11] Kurzmann, A.; Overweg, H.; Eich, M.; Pally, A.; Rickhaus, P.; Pisoni, R.; Lee, Y.; Watanabe, K.; Taniguchi, T.; Ihn, T.; Ensslin, K. Charge Detection in Gate-Defined Bilayer Graphene Quantum Dots. *Nano Lett.* 2019, 19 (8), 5216–5221. <https://doi.org/10.1021/acs.nanolett.9b01617>.

[12] Qiu, H.; Liu, H.; Jia, X.; Liu, X.; Li, Y.; Jiang, T.; Xiong, B.; Yang, Y.; Ren, T. Ink-Injected Dual-Band Antennas Based on Graphene Flakes, Carbon

Nanotubes and Silver Nanowires. *RSC Adv.* 2018, 8 (65), 37534–37539. <https://doi.org/10.1039/c8ra08018f>.

[13] Bhuyan, Md. S. A.; Uddin, Md. N.; Islam, Md. M.; Bipasha, F. A.; Hossain, S. S. Synthesis of Graphene. *Int. Nano Lett.* 2016, 6 (2), 65–83. <https://doi.org/10.1007/s40089-015-0176-1>.

[14] Sun, B.; Pang, J.; Cheng, Q.; Zhang, S.; Li, Y.; Zhang, C.; Sun, D.; Ibarlucea, B.; Li, Y.; Chen, D.; Fan, H.; Han, Q.; Chao, M.; Liu, H.; Wang, J.; Cuniberti, G.; Han, L.; Zhou, W. Synthesis of Wafer-Scale Graphene with Chemical Vapor Deposition for Electronic Device Applications. *Adv. Mater. Technol.* 2021. <https://doi.org/10.1002/admt.202000744>.

[15] Beshkova, M.; Hultman, L.; Yakimova, R. Device Applications of Epitaxial Graphene on Silicon Carbide. *Vacuum.* 2016, 128 pp 186–197. <https://doi.org/10.1016/j.vacuum.2016.03.027>.

[16] Mishra, N.; Boeckl, J.; Motta, N.; Iacopi, F. Graphene Growth on Silicon Carbide: A Review. *Pss (RRL).* 2016, pp 2277–2289. <https://doi.org/10.1002/pssa.201600091>.

[17] Yazdi, G. R.; Iakimov, T.; Yakimova, R. Epitaxial Graphene on SiC: A Review of Growth and Characterization. *Crystals.* 2016. <https://doi.org/10.3390/cryst6050053>.

[18] de Heer, W. A.; Berger, C.; Wu, X.; First, P. N.; Conrad, E. H.; Li, X.; Li, T.; Sprinkle, M.; Hass, J.; Sadowski, M. L.; Potemski, M.; Martinez, G. Epitaxial Graphene. *Solid State Comm.* 2007, 143 (1–2), 92–100. <https://doi.org/10.1016/j.ssc.2007.04.023>.

[19] Pierson, H. O. Principles, Technology, and Applications. *Handbook of Chemical Vapor Deposition: Noyes Publications* 1999. pp 29. ISBN: 0-8155-1432-8.

[20] Sun, L.; Yuan, G.; Gao, L.; Yang, J.; Chhowalla, M.; Gharahcheshmeh, M. H.; Gleason, K. K.; Choi, Y. S.; Hong, B. H.; Liu, Z. Chemical Vapor Deposition. *Nat Rev Methods Primers.* 2021, 1 (1), 5. <https://doi.org/10.1038/s43586-020-00005-y>.

[21] Saeed, M.; Alshammari, Y.; Majeed, S. A.; Al-Nasrallah, E. Chemical Vapor Deposition of Graphene—Synthesis, Characterization, and Applications: A Review. *Molecules MDPI.* 2020. <https://doi.org/10.3390/molecules25173856>.

[22] Sergey Mikhailov. Electronic Properties of Graphene Probed at the Nanoscale. *Adv. Mater. Technol. John Wiley and Sons Inc.* 2020, pp 353. 978-953-51-4496-0

[23] Özkan, S. A.; Dedeoğlu, A.; Karadaş Bakirhan, N.; Özkan, Y. Nanocarriers Used Most in Drug Delivery and Drug Release: Nanohydrogel, Chitosan, Graphene, and Solid Lipid. *Turk J Pharm Sci.* 2019, pp 481–492. <https://doi.org/10.4274/tjps.galenos.2019.48751>.

[24] Sun, B.; Pang, J.; Cheng, Q.; Zhang, S.; Li, Y.; Zhang, C.; Sun, D.; Ibarlucea, B.; Li, Y.; Chen, D.;

Fan, H.; Han, Q.; Chao, M.; Liu, H.; Wang, J.; Cuniberti, G.; Han, L.; Zhou, W. Synthesis of Wafer-Scale Graphene with Chemical Vapor Deposition for Electronic Device Applications. *Adv. Mater. Technol. John Wiley and Sons Inc.* **2021**. <https://doi.org/10.1002/admt.202000744>.

[25] Ganz, E.; Ganz, A. B.; Yang, L. M.; Dornfeld, M. The Initial Stages of Melting of Graphene between 4000 K and 6000 K. *Phys. Chem. Chem. Phys.* **2017**, *19* (5), 3756–3762. <https://doi.org/10.1039/c6cp06940a>.

[26] Montes-Navajas, P.; Asenjo, N. G.; Santamaría, R.; Menéndez, R.; Corma, A.; García, H. Surface Area Measurement of Graphene Oxide in Aqueous Solutions. *Langmuir.* **2013**, *29* (44), 13443–13448. <https://doi.org/10.1021/la4029904>.

[27] Zhang, S.; Wang, H.; Liu, J.; Bao, C. Measuring the Specific Surface Area of Monolayer Graphene Oxide in Water. *Mater. Lett.* **2020**, *261*, 127098. <https://doi.org/10.1016/j.matlet.2019.127098>.

[28] Abdelaal, S.; Elmaghraby, E. K.; Abdelhady, A. M.; Youssf, M.; Rashad, A. M.; Bashter, I. I.; Helal, A. I. The Physical Structure and Surface Reactivity of Graphene Oxide. *Diam Relat Mater.* **2020**, *101*. <https://doi.org/10.1016/j.diamond.2019.107613>.

[29] Wang, X.; Shi, G. An Introduction to the Chemistry of Graphene. *Phys. Chem. Chem. Phys.* **2015**, *17* (43), 28484–28504. <https://doi.org/10.1039/c5cp05212b>.

[30] Kamedulski, P.; Skorupska, M.; Binkowski, P.; Arendarska, W.; Ilnicka, A.; Lukaszewicz, J. P. High Surface Area Micro-Mesoporous Graphene for Electrochemical Applications. *Sci. Rep.* **2021**, *11* (1). <https://doi.org/10.1038/s41598-021-01154-0>.

[31] Su, L. S.; Tsai, J. L. Characterizing the Mechanical Properties of Nanocomposites with Aligned Graphene. *Polym. Compos.* **2021**, *42* (8), 4005–4014. <https://doi.org/10.1002/pc.26112>.

[32] Gamboa-Suárez, A.; Seuret-Hernández, H. Y.; Leyssale, J.-M. Mechanical Properties of Pristine and Nanocrystalline Graphene up to Ultra-High Temperatures. *Carbon Trends* **2022**, *9*, 100197. <https://doi.org/10.1016/j.cartre.2022.100197>.

[33] Bao, Q.; Yang, Z.; Lu, Z.; He, X. Effects of Graphene Thickness and Length Distribution on the Mechanical Properties of Graphene Networks: A Coarse-Grained Molecular Dynamics Simulation. *Appl. Surf. Sci.* **2021**, *570*. <https://doi.org/10.1016/j.apsusc.2021.151023>.

[34] Kumar, Y.; Sahoo, S.; Chakraborty, A. K. Mechanical Properties of Graphene, Defective Graphene, Multilayer Graphene and SiC-Graphene Composites: A Molecular Dynamics Study. *Phys. Rev. B Condens. Matter* **2021**, *620*. <https://doi.org/10.1016/j.physb.2021.413250>.

[35] Malik, R.; Tomer, V. K.; Chaudhary, V. Hybridized Graphene for Chemical Sensing. *Adv. Mater. Technol. John Wiley and Sons Inc.* **2018**, pp 323–338. <https://doi.org/10.1016/B978-0-12-814548-7.00016-7>.

[36] Kumar, K.; Yadav, B. C. An Overview on the Importance of Chemical Vapour Deposition Technique for Graphene Synthesis. *Adv. Sci. Eng. Med.* **2018**, *10* (7), 760–763. <https://doi.org/10.1166/ asem.2018.2241>.

[37] Mesquita, F.; Copetti, G.; Tumelero, M. A.; Gusmão, M. A.; Radtke, C.; Pureur, P. Electrical Magnetotransport Properties of Chlorinated CVD Graphene. *Phys. Rev. B Condens. Matter* **2021**, *609*. <https://doi.org/10.1016/j.physb.2021.412893>.

[38] Sharma, N.; Tomar, S.; Shakir, M.; Kant Choubey, R.; Singh, A. Study of Optical and Electrical Properties of Graphene Oxide. *Mater. Today: Proc.; Elsevier* **2019**, Vol. 36, pp 730–735. <https://doi.org/10.1016/j.matpr.2020.04.861>.

[39] Chan, K. Y.; Baktash, A.; Demir, B.; Mayes, E. L. H.; Yang, D.; Pham, D. Q.; Lin, K. te; Mouritz, A. P.; Ang, A. S. M.; Fox, B.; Zhu, B.; Lin, H.; Jia, B.; Lau, K. T. Tailoring Mechanical and Electrical Properties of Graphene Oxide Film for Structural Dielectric Capacitors. *J Power Sources* **2021**, *482*. <https://doi.org/10.1016/j.jpowsour.2020.229020>.

[40] Wang, F.; Shi, S. Optical Properties of Graphene. In *2D Materials: Properties and Devices; Cambridge University Press* **2017**, pp 38–51. <https://doi.org/10.1017/9781316681619.004>.

[41] Lin, H.; Lin, K. Te; Yang, T.; Jia, B. Graphene Multilayer Photonic Metamaterials: Fundamentals and Applications. *Adv. Mater. Technol. John Wiley and Sons Inc* **2021**. <https://doi.org/10.1002/admt.202000963>

[42] Afrozeh, A. Dependence of Linear and Non-Linear Optical Properties to Sp³ Domains Level and Edges Length in Graphene-Based Nanomaterials. *Optik (Stuttg)* **2021**, *226*. <https://doi.org/10.1016/j.ijleo.2020.165903>.

[43] Sang, M.; Shin, J.; Kim, K.; Yu, K. J. Electronic and Thermal Properties of Graphene and Recent Advances in Graphene-Based Electronics Applications. *J. Nanomater; MDPI AG* **2019**. <https://doi.org/10.3390/nano9030374>.

[44] Kansara, V.; Shukla, R.; Flora, S. J. S.; Bahadur, P.; Tiwari, S. Graphene Quantum Dots: Synthesis, Optical Properties and Navigational Applications against Cancer. *Mater. Today Commun; Elsevier* **2022**. <https://doi.org/10.1016/j.mtcomm.2022.103359>.

[45] Lee, J. H.; Park, S. J.; Choi, J. W. Electrical Property of Graphene, and Its Application to Electrochemical Biosensing. *J. Nanomater; MDPI AG* **2019**. <https://doi.org/10.3390/nano9020297>.

[46] Kamedulski, P.; Skorupska, M.; Binkowski, P.; Arendarska, W.; Ilnicka, A.; Lukaszewicz, J. P. High

Surface Area Micro-Mesoporous Graphene for Electrochemical Applications. *Sci. Rep.* **2021**, 11 (1). <https://doi.org/10.1038/s41598-021-01154-0>.

[47] Liu, Z.; Navik, R.; Tan, H.; Xiang, Q.; Wahyudiono; Goto, M.; Ibarra, R. M.; Zhao, Y. Graphene-Based Materials Prepared by Supercritical Fluid Technology, and Its Application in Energy Storage. *J. Supercrit. Fluids; Elsevier* **2022**. <https://doi.org/10.1016/j.supflu.2022.105672>.

[48] Li, Z.; Kan, A.; Wang, K.; He, Y.; Zheng, N.; Yu, W. Optical Properties and Photothermal Conversion Performances of Graphene-Based Nanofluids. *Appl. Therm. Eng.* **2022**, 203. <https://doi.org/10.1016/j.applthermaleng.2021.117948>.

[49] Nauman Javed, R. M.; Al-Othman, A.; Tawalbeh, M.; Olabi, A. G. Recent Developments in Graphene and Graphene Oxide Materials for Polymer Electrolyte Membrane Fuel Cells Applications. *Renewable Sustainable Energy Rev.; Elsevier* **2022**. <https://doi.org/10.1016/j.rser.2022.112836>.

[50] Ajravat, K.; Rajput, S.; Brar, L. K. Microwave-Assisted Hydrothermal Synthesis of N Doped Graphene for Supercapacitor Applications. *Diam Relat Mater.* **2022**, 129, 109373. <https://doi.org/10.1016/j.diamond.2022.109373>.

[51] Saravanan, A.; Kumar, P. S.; Srinivasan, S.; Jeevanantham, S.; Vishnu, M.; Amith, K. V.; Sruthi, R.; Saravanan, R.; Vo, D. V. N. Insights on Synthesis and Applications of Graphene-Based Materials in Wastewater Treatment: A Review. *Chemosphere* **2022**, 298. <https://doi.org/10.1016/j.chemosphere.2022.134284>.

[52] Zafar, M. A.; Jacob, M. v. Synthesis of Free-Standing Graphene in Atmospheric Pressure Microwave Plasma for the Oil-Water Separation Application. *Appl. Surf. Sci.* **2022**, 11. <https://doi.org/10.1016/j.apsadv.2022.100312>.

[53] Chen, Z.; Zhao, J.; Cao, J.; Zhao, Y.; Huang, J.; Zheng, Z.; Li, W.; Jiang, S.; Qiao, J.; Xing, B.; Zhang, J. Opportunities for Graphene, Single-Walled and Multi-Walled Carbon Nanotube Applications in Agriculture: A Review. *Crop Design* **2022**, 1 (1), 100006. <https://doi.org/10.1016/j.croprd.2022.100006>.

[54] Singh, S.; Hasan, M. R.; Sharma, P.; Narang, J. Graphene Nanomaterials: The Wondering Material from Synthesis to Applications. *Sensors International. KeAi Communications* **2022**. <https://doi.org/10.1016/j.sintl.2022.100190>.

[55] Zhang, Q.; Sando, D.; Nagarajan, V. Chemical Route Derived Bismuth Ferrite Thin Films and Nanomaterials. *RSC J. Mater. Chem.* **2016**, pp 4092–4124. <https://doi.org/10.1039/c6tc00243a>.

[56] Miao, C.; Zheng, C.; Liang, O.; Xie, Y.-H. Chemical Vapor Deposition of Graphene. *Adv. Mater. Technol. John Wiley and Sons Inc.* **2016**. <https://doi.org/10.5772/15543>.

[57] Zietz, O.; Olson, S.; Coyne, B.; Liu, Y.; Jiao, J. Characterization and Manipulation of Carbon Precursor Species during Plasma Enhanced Chemical Vapor Deposition of Graphene. *J. Nanomater.* **2020**, 10 (11), 1–11. <https://doi.org/10.3390/nano10112235>.

[58] Hansen, Clayton. Synthesis of Graphene by Liquid Precursors at Lower Temperatures. *Physics CapstoneProjects*. **2020**. https://digitalcommons.usu.edu/phys_capstoneproject/83

[59] Li, Z.; Wu, P.; Wang, C.; Fan, X.; Zhang, W.; Zhai, X.; Zeng, C.; Li, Z.; Yang, J.; Hou, J. Low-Temperature Growth of Graphene by Chemical Vapor Deposition Using Solid and Liquid Carbon Sources. *ACS Nano.* **2011**, 5 (4), 3385–3390. <https://doi.org/10.1021/nn200854p>.

[60] Campos-Delgado, J.; Botello-Méndez, A. R.; Algara-Siller, G.; Hackens, B.; Pardo, T.; Kaiser, U.; Dresselhaus, M. S.; Charlier, J. C.; Raskin, J. P. CVD Synthesis of Mono- and Few-Layer Graphene Using Alcohols at Low Hydrogen Concentration and Atmospheric Pressure. *Chem. Phys. Lett.* **2013**, 584, 142–146. <https://doi.org/10.1016/j.cplett.2013.08.031>

[61] Schoonraad, G. L.; Madito, M. J.; Manyala, N.; Forbes, P. Synthesis and Optimisation of a Novel Graphene Wool Material by Atmospheric Pressure Chemical Vapor Deposition. *J. Mater. Sci.* **2020**, 55 (2), 545–564. <https://doi.org/10.1007/s10853-019-03948-0>.

[62] Dong, Y.; Guo, S.; Mao, H.; Xu, C.; Xie, Y.; Cheng, C.; Mao, X.; Deng, J.; Pan, G.; Sun, J. The Growth of Graphene on Ni–Cu Alloy Thin Films at a Low Temperature and Its Carbon Diffusion Mechanism. *J. Nanomater.* **2019**, 9 (11). <https://doi.org/10.3390/nano9111633>.

[63] Hu, B.; Wei, Z.; Ago, H.; Jin, Y.; Xia, M.; Luo, Z.; Pan, Q.; Liu, Y. Effects of Substrate and Transfer on CVD-Grown Graphene over Sapphire-Induced Cu Films. *Sci. China Chem.* **2014**, 57 (6), 895–901. <https://doi.org/10.1007/s11426-014-5073-3>.

[64] Wood, J. D.; Schmucker, S. W.; Lyons, A. S.; Pop, E.; Lyding, J. W. Effects of Polycrystalline Cu Substrate on Graphene Growth by Chemical Vapor Deposition. *Nano Lett.* **2011**, 11 (11), 4547–4554. <https://doi.org/10.1021/nl201566c>.

[65] Shi, Q.; Tokarska, K.; Ta, H. Q.; Yang, X.; Liu, Y.; Ullah, S.; Liu, L.; Trzebicka, B.; Bachmatiuk, A.; Sun, J.; Fu, L.; Liu, Z.; Rummeli, M. H. Substrate Developments for the Chemical Vapor Deposition Synthesis of Graphene. *Adv. Mater. Interfaces; Wiley-VCH Verl.* **2020**. <https://doi.org/10.1002/admi.201902024>.

[66] Chen, Z.; Qi, Y.; Chen, X.; Zhang, Y.; Liu, Z. Direct CVD Growth of Graphene on Traditional Glass: Methods and Mechanisms. *J. Adv. Mater; Wiley-VCH Verlag* **2019**. <https://doi.org/10.1002/adma.201803639>.

- [67] Ueda, Y.; Maruyama, T.; Naritsuka, S. Effect of Growth Pressure on Graphene Direct-Growth on an A-Plane Sapphire Substrate: Implications for Graphene-Based Electronic Devices. *ACS Appl. Nano Mater.* **2021**, *4* (1), 343–351. <https://doi.org/10.1021/acsanm.0c02634>.
- [68] Shan, J.; Sun, J.; Liu, Z. Chemical Vapor Deposition Synthesis of Graphene over Sapphire Substrates. *Chem Nano Mat; John Wiley and Sons Inc* **2021**, pp 515–525. <https://doi.org/10.1002/cnma.202100079>.
- [69] Chang, C. J.; Tsai, P. C.; Su, W. Y.; Huang, C. Y.; Lee, P. T.; Lin, S. Y. Layered Graphene Growth Directly on Sapphire Substrates for Applications. *ACS Omega* **2022**, *7* (15), 13128–13133. <https://doi.org/10.1021/acsomega.2c00554>.
- [70] Goto, T., Katsui, H. Chemical Vapor Deposition of Ca–P–O Film Coating. In: Sasaki, K., Suzuki, O., Takahashi, N. (eds) *Interface Oral Health Science*. Springer **2015**. https://doi.org/10.1007/978-4-431-55192-8_9
- [71] Gausden, J.; Siris, R.; Stimpel-Lindner, T.; McEvoy, N.; Duesberg, G. S.; Hallam, T. Nitrogen as a Suitable Replacement for Argon within Methane-Based Hot-Wall Graphene Chemical Vapor Deposition. *Phys. Status Solidi B Basic Res.* **2019**, *256* (12). <https://doi.org/10.1002/pssb.201900240>.
- [72] Zeng, J.; Ji, X.; Ma, Y.; Zhang, Z.; Wang, S.; Ren, Z.; Zhi, C.; Yu, J. 3D Graphene Fibers Grown by Thermal Chemical Vapor Deposition. *J. Adv. Mater.* **2018**, *30* (12). <https://doi.org/10.1002/adma.201705380>.
- [73] Kordina, O.; Henry, A.; and Janzen, V. SiC Epitaxial Growth on large Area Substrates: History and Evolution. *Silicon Carbide Epitaxy* **2012** pp 1-25. ISBN: 978-81-308-000-9.
- [74] Kudarenko, I. P.; Malykhin, S. A.; Orekhov, A. S.; Puzyr, A. P.; Kleshch, V. I.; Ismagilov, R. R.; Obraztsov, A. N. Detonation Nanodiamond-Assisted Carbon Nanotube Growth by Hot Filament Chemical Vapor Deposition. *Phys. Status Solidi B Basic Res.* **2018**, *255* (1). <https://doi.org/10.1002/pssb.201700286>.
- [75] Janzén, E.; Bergman, P.; Danielsson, Ö.; Forsberg, U.; Hallin, C.; ul Hassan, J.; Henry, A.; Ivanov, I. G.; Kakanakova-Georgieva, A.; Persson, P. O. Å.; Wahab, Q. SiC and III-Nitride Growth in Hot-Wall CVD Reactor. *Materials Science Forum* **2005**, *483–485*, 61–66. <https://doi.org/10.4028/www.scientific.net/msf.483-485.61>.
- [76] Wasyluk, Ł.; Boiko, V.; Markowska, M.; Hasiak, M.; Saladino, M. L.; Hreniak, D.; Amati, M.; Gregoratti, L.; Zeller, P.; Biały, D.; Borkowski, J.; Wawrzyńska, M. Graphene Coating Obtained in a Cold-Wall CVD Process on the Co-Cr Alloy (L-605) for Medical Applications. *Int. J. Mol. Sci.* **2021**, *22* (6), 1–22. <https://doi.org/10.3390/ijms22062917>.
- [77] Jia, K.; Ci, H.; Zhang, J.; Sun, Z.; Ma, Z.; Zhu, Y.; Liu, S.; Liu, J.; Sun, L.; Liu, X.; Sun, J.; Yin, W.; Peng, H.; Lin, L.; Liu, Z. Super clean Growth of Graphene Using a Cold-Wall Chemical Vapor Deposition Approach. *Angew Chem.* **2020**, *59* (39), 17214–17218. <https://doi.org/10.1002/anie.202005406>.
- [78] Soriadi, N.; Abdullah, M. F.; Md Yakin, F. S.; Mohamad Badaruddin, S. A.; Syono, M. I. Effect of Cu Thickness and Temperature on Growth of Graphene on 8-Inch Cu/SiO₂/Si Wafer Using Cold-Wall CVD Reactor. *Mater. Today Commun; Elsevier* **2021**, Vol. *42*, pp 2948–2952. <https://doi.org/10.1016/j.matpr.2020.12.800>.
- [79] Sarno, M.; Rossi, G.; Cirillo, C.; Incarnato, L. Cold Wall Chemical Vapor Deposition Graphene-Based Conductive Tunable Film Barrier. *Ind. Eng. Chem. Res.* **2018**, *57* (14), 4895–4906. <https://doi.org/10.1021/acs.iecr.7b05281>.
- [80] Faisal, M.; Bhattacharyya, S.; Jha, P.; Agarwal, A.; Chaudhury, P. K.; Islam, S. S.; Husain, M. Cold Wall CVD (CWCVD) in the Synthesis of Few Layered Graphene on Ni. **2017**. <https://doi.org/10.20944/preprints201705.0217.v1>.
- [81] Saeed, M.; Alshammari, Y.; Majeed, S. A.; Al-Nasrallah, E. Chemical Vapor Deposition of Graphene—Synthesis, Characterization, and Applications: A Review. *J. Molecules. MDPI* **2020**. <https://doi.org/10.3390/molecules25173856>.
- [82] Lu, C.-H.; Leu, C.-M.; Lee, T.-M.; Yeh, N.-C. Single-Step Plasma-Enhanced Chemical Vapor Deposition of Graphene on Cu Ink and Sputtered Cu Thin Films. *J Soc Inf Disp.* **2020**, Vol *51* pp 1565-1568. <https://sid.onlinelibrary.wiley.com/doi/epdf/10.1002/sdtp.14190>.
- [83] Duan, J.; Guo, Z.; Liu, Y.; Chen, Y. Coat Multi-Layered Graphene on Cu with Radiofrequency Plasmas for Anti-Oxidization in Hot Air. *Mater. Chem. Phys.* **2021**, *274*. <https://doi.org/10.1016/j.matchemphys.2021.125155>.
- [84] Yen, C. C.; Chang, Y. C.; Tsai, H. C.; Woon, W. Y. Nucleation and Growth Dynamics of Graphene Grown through Low Power Capacitive Coupled Radio Frequency Plasma Enhanced Chemical Vapor Deposition. *Carbon N Y* **2019**, *154*, 420–427. <https://doi.org/10.1016/j.carbon.2019.08.042>.
- [85] Hong, H. K.; Kim, N. Y.; Yoon, A.; Lee, S. W.; Park, J.; Yoo, J. W.; Lee, Z. Synthesis of High-Quality Monolayer Graphene by Low-Power Plasma. *Curr. Appl. Phys.* **2019**, *19* (1), 44–49. <https://doi.org/10.1016/j.cap.2018.11.003>.
- [86] Li, N.; Zhen, Z.; Xu, Z.; Zhang, R.; Mu, R.; He, L. The Growth of Large-Sized Graphene Domains by Faraday Cage-Assisted Plasma Enhanced Chemical

Vapor Deposition. *Appl. Surf. Sci.* **2021**, *6*.
<https://doi.org/10.1016/j.apsadv.2021.100154>.

[87] Wang, J. bin; Ren, Z.; Hou, Y.; Yan, X. L.; Liu, P. Z.; Zhang, H.; Zhang, H. X.; Guo, J. J. A Review of Graphene Synthesis at Low Temperatures by CVD Methods. *Xinxing Tan Cailiao/New Carbon Mater.* **2020**, pp 193–208. [https://doi.org/10.1016/S1872-5805\(20\)60484-X](https://doi.org/10.1016/S1872-5805(20)60484-X).

[88] Xu, S.; Zhang, L.; Wang, B.; Ruoff, R. S. Chemical Vapor Deposition of Graphene on Thin-Metal Films. *Cell Rep. Cell Press* **2021**.
<https://doi.org/10.1016/j.xcrp.2021.100372>.

[89] Hong, H. K.; Kim, N. Y.; Yoon, A.; Lee, S. W.; Park, J.; Yoo, J. W.; Lee, Z. Synthesis of High-Quality Monolayer Graphene by Low-Power Plasma. *Curr. Appl. Phys.* **2019**, *19* (1), 44–49.
<https://doi.org/10.1016/j.cap.2018.11.003>.

[90] Kumar, D.; Tripathi, C. C.; Singh, R.; Kumar, D.; Tripathi, C. C. Graphene: Potential Material for Nanoelectronics Applications. *Indian J. Pure Appl. Phys.* **2015**, *53*(8) 501-513.

[91] Sun, B.; Pang, J.; Cheng, Q.; Zhang, S.; Li, Y.; Zhang, C.; Sun, D.; Ibarlucea, B.; Li, Y.; Chen, D.; Fan, H.; Han, Q.; Chao, M.; Liu, H.; Wang, J.; Cuniberti, G.; Han, L.; Zhou, W. Synthesis of Wafer-Scale Graphene with Chemical Vapor Deposition for Electronic Device Applications. *Adv. Mater. Technol; John Wiley and Sons Inc* **2021**.
<https://doi.org/10.1002/admt.202000744>.

[92] Kataria, S.; Wagner, S.; Ruhkopf, J.; Gahoi, A.; Pandey, H.; Bornemann, R.; Vaziri, S.; Smith, A. D.; Ostling, M.; Lemme, M. C. Chemical Vapor Deposited Graphene-From Synthesis to Applications. *Phys. Status Solidi (A)*. **2014**, *211*(11) 2439-2449.
<https://doi.org/10.1002/pssa.201400049>.