Dean&Francis
ISSN 2959-6157

CVD Growth, Heterojunction Construction, and Functional Applications of Graphene

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Abstract:

Graphene, a monolayer of sp^2-hybridized carbon atoms, exhibits extraordinary electronic mobility (~200,000 $cm^2 \cdot V^{-1} \cdot s^{-1}$), thermal conductivity (~5000 $W \cdot m^{-1} \cdot K^{-1}$), and mechanical strength (~1 TPa), making it a core candidate for next-generation electronics, energy storage, and optoelectronics. Chemical Vapor Deposition (CVD) is the most reliable technique for high-quality graphene, enabling controllable synthesis of polycrystalline (single/multi-layer) and single-crystal graphene. To address pristine graphene's intrinsic zero-bandgap limitation and expand its functionality, graphene-based heterojunctions—with transition-metal dichalcogenides (TMDs), hexagonal boron nitride (h-BN), or other 2D Xenes—have been developed to tune its electronic and optical properties. This paper reviews graphene's CVD growth mechanisms (polycrystalline/single-crystal), graphene heterojunctions' construction/interface regulation, and their applications in the above fields. It confirms CVD's key role in high-quality graphene synthesis and heterojunctions' value in breaking graphene's zero-bandgap constraint, while identifying challenges like large-area single-crystal growth and heterojunction interface defects; future research on atomic-level growth control and industrialization optimization will further promote graphene's practical application.

Keywords: Graphene; Chemical Vapor Deposition (CVD); Polycrystalline Graphene; Single-Crystal Graphene; Graphene Heterojunctions

1. Introduction

Since its experimental exfoliation in 2004 by Geim and Novoselov, graphene has revolutionized the field of 2D materials due to its unique planar structure and exceptional physical/chemical properties [1]. However, the practical application of graphene depends heavily on scalable synthesis of high-quality samples. Traditional methods such as mechanical exfoliation (low efficiency, small size) and liquid-phase exfoliation (abundant defects) fail to meet industrial demands, while CVD technology achieves a balance

ISSN 2959-6157

between quality, scalability, and process controllability—enabling the preparation of large-area, low-defect graphene for functional devices.

Pristine graphene exhibits a zero bandgap, limiting its application in logic transistors and optoelectronic devices. Constructing graphene-based heterojunctions by integrating graphene with other 2D materials (e.g., MoS_2, h-BN, arsenene) has become a core strategy to modulate its electronic structure (e.g., opening a tunable bandgap) and enhance interface-dependent functionalities (e.g., photocarrier separation, catalytic activity).

This study is dedicated to integrating the core mechanisms of graphene CVD growth and heterojunction construction, establishing the "growth-structure-property" correlation. It provides support for optimizing processes to achieve low-cost large-area fabrication, breaking the single-crystal size bottleneck, and addressing the zero-bandgap limitation of graphene. Meanwhile, it clarifies future research directions and offers support for the industrialization of two-dimensional materials.

2. CVD Growth of Graphene

CVD growth of graphene relies on the decomposition of carbon precursors (e.g., CH_4) on catalytic substrates at high temperatures, followed by the nucleation and growth of carbon atoms into a 2D lattice. The choice of substrate, catalyst, and reaction parameters determines the structure (polycrystalline/single-crystal) and quality of graphene.

2.1 Polycrystalline Graphene Growth

Polycrystalline graphene consists of multiple grains connected by grain boundaries, and its CVD growth focuses on controlling the number of layers and coverage. It is currently the most widely used type in industrial applications due to its mature process and low cost.

2.1.1 Single-Layer Polycrystalline Graphene

The core challenge of single-layer growth is suppressing multi-layer nucleation while promoting continuous monolayer formation. Metal catalysts (Cu, Ni) and catalyst-free systems are the two main routes.

(1) Metal-Catalyzed Growth (Mainstream Route). Metals act as both substrates and catalysts, with their carbon solubility and catalytic activity determining the growth mechanism (surface adsorption/ diffusion for low-solubility metals like Cu; diffusion-precipitation for high-solubility metals like Ni).

Cu-based catalytic system. Cu is the most mature catalyst for single-layer graphene due to its low carbon solubility (~0.001 at .% at 1000°C), which limits carbon diffusion into the bulk and favors surface adsorption-growth. In 2009, Li et al. [2] achieved a breakthrough: using copper foil as the substrate, CH_4 as the carbon source, and H_2 as the carrier gas/co-catalyst, they grew large-area single-layer graphene at 1000°C under atmospheric pressure (single-layer ratio >95%). The Cu foil was etched with iron nitrite solution to obtain free-standing graphene, which could be transferred to any substrate (e.g., SiO_2/Si) for device fabrication. Later, Fang et al. [3] optimized the process by wrapping Cu foil with tungsten (W) foil—W's carbon-absorbing property selectively removed double/triple-layer domains, improving monolayer uniformity.

Ni-based catalytic system. Ni has higher carbon solubility than Cu, leading to a "diffusion-precipitation" mechanism. Reina et al. [4] used a 500 nm Ni film (evaporated on SiO_2/Si) as the catalyst, with 5–25 sccm CH_4 and 1500 sccm H_2 at 900–1000°C. The product contained single/double-layer regions (up to 20 μm), but single-layer coverage (<70%) was lower than that of Cu-based systems, limiting its application in high-uniformity scenarios.

(2) Catalyst-Free Growth (Metal-Residue-Sensitive Scenarios). To avoid metal residues affecting device performance (e.g., in high-frequency electronics), catalyst-free CVD was developed, relying on substrate surface energy regulation or plasma assistance.

Sapphire substrate (van der Waals epitaxy). Hwang et al. [5] grew single-layer graphene on sapphire via van der Waals epitaxy at ~1400°C. By adjusting CH_4 partial pressure, they obtained p-type or n-type graphene with a carrier mobility of ~2000 $cm^2 \cdot V^{-1} \cdot s^{-1}$, suitable for optoelectronic devices, but high growth temperature limits substrate compatibility.

Dielectric substrate (near-equilibrium growth). Chen et al. [6] used a near-equilibrium CVD process (1180℃, 2 h, CH_4:H_2=1.9–2.3:50 sccm) on $Si_3N_4/SiO_2/Si$ substrates, achieving single-crystal hexagonal single-layer graphene with a mobility of 5000 $cm^2\cdot V^{-1}\cdot s^{-1}$ (comparable to metal-catalyzed samples), but the size was limited to 11 µm.

2.1.2 Multi-Layer Polycrystalline Graphene

Multi-layer graphene (≥2 layers) is tailored for applications requiring tunable bandgaps (e.g., photodetectors) by controlling the carbon source supply rate and catalyst carbon storage capacity.

(1) Layer Number Regulation (1–10 Layers). Coarse control via Ni-based catalysis Gong et al. [7] tuned the layer number (1–10 layers) by controlling H_2 flow rate, Ni film thickness, growth temperature, and time: thicker Ni films and higher CH_4 concentration increased layers, while H_2 suppressed carbon agglomeration to reduce layer fluctuation.

Precise control (single-layer accuracy within 7 layers). Tu et al. [8] optimized parameters (temperature: 950–1050℃, deposition time: 30–60 min, pressure: atmospheric to 500 mTorr) on $Ni/SiO_2/Si$ substrates, achieving “single-layer precision” control within 7 layers, solving the problem of uneven layer distribution.

(2) Large-Scale Bilayer Graphene Growth. Bilayer graphene is critical for bandgap engineering. Lee et al. [9] obtained wafer-scale bilayer graphene on Cu foil by cooling at 18 K min-1 to suppress rapid carbon precipitation, achieving 99% coverage and uniform interlayer alignment. Yan et al. [10] adopted “layer-by-layer epitaxy”, regrowing a second layer via secondary CVD, yet weak interlayer bonding limited coverage to 67%.

2.2 Single-Crystal Graphene Growth

Grain boundaries in polycrystalline graphene reduce carrier mobility and mechanical strength, making single-crystal graphene essential for high-performance devices (e.g., high-frequency transistors). The key to CVD growth is suppressing multi-nucleation sites and promoting continuous single-grain growth.

2.2.1 Core Growth Principle

During the CVD growth of single-crystal graphene, suppressing nucleation and promoting single-grain growth are two key steps. Nucleation suppression can be achieved by high-temperature H_2 annealing (1000-1180℃), which effectively removes impurities and oxides from the substrate surface, thereby reducing nucleation sites. Meanwhile, lowering the CH_4 partial pressure to approximately 500 ppm further decreases the carbon precursor concentration, preventing competitive growth among multiple grains. For promoting single-grain growth, optimizing the chamber pressure (maintained at a low pressure of <1 Torr), growth temperature (1000–1180℃), and H_2/CH_4 ratio (>1000) can extend the overall growth time, ultimately allowing a single grain to grow sufficiently and “occupy” the entire substrate.

2.2.2 Growth Routes

(1) Single-Crystal Metal Substrates (High Quality, High Cost). Single-crystal metals (e.g., Pt(111), Ni(111)) provide lattice matching for graphene growth. Gao et al. [11] grew 1 mm-sized hexagonal single-crystal graphene on Pt(111) at 1040℃ (4 sccm CH_4, 700 sccm H_2) with a mobility of 7100 $cm^2\cdot V^{-1}\cdot s^{-1}$ (close to mechanical exfoliation), but Pt(111) is expensive for scaling.

(2) Commercial Metal Substrates (Low Cost, Scalable). Cu foil (mainstream) Vlassiouk et al. [12] obtained ~10µm single crystals on Cu foil by adjusting H_2/CH_4 partial pressure (H_2/CH_4 >100). To enlarge size: Li et al. [13] used “low-pressure extended growth” (pressure <1 Torr, time >10 h) with Cu inclusions, achieving 0.5 mm grains; Gan et al. [14] pretreated Cu foil via “Ar annealing (mild oxidation to form CuO nanoparticles as single nucleation sites) + Ar/H_2 reductive annealing”, obtaining 5.9 mm hexagonal single crystals. Molten Cu substrates (ultra-large size): Breaking Cu’s melting point (1085℃) uses molten Cu’s high fluidity to reduce surface defects. Geng et al. [15] used W foil as a support, growing sub-millimeter “twelve-pointed single-crystal graphene” by tuning CH_4 supply rate.

(3) Oxygen’s Critical Role in Single-Crystal Growth

Trace amounts of oxygen form copper oxide (CuO) nanoparticles on the surface of copper foil, and these nanoparticles can act as single nucleation sites to achieve controllability of nucleation sites. Meanwhile, oxygen can also reduce the active sites on the copper surface.When in high-purity copper (oxygen-free copper, OF-Cu), this effect can lower the nucleation density by an order of magnitude. Furthermore, during hydrogen (H_2) annealing, the previously formed copper oxide decomposes into copper and water—this reaction effectively removes residual carbon from the substrate surface, thereby realizing substrate self-cleaning and collectively regulating nucleation density from multiple dimensions.

2.3 Comparison of CVD Growth Routes for Different Graphene Types

Table 1. Comparison of CVD Growth Routes for Different Graphene Types

Graphene Type	Substrate/Catalyst	Precursor	Key Parameters (T/P)	Performance (Size/Mobility)	Advantages	Challenges
Polycrystalline Single-Layer	Cu foil	CH_4/H_2	1000°C/Atmospheric	Large-area (>1 cm²)/>1000 $cm^2·V^{-1}·s^{-1}$	Low cost	Multi-layer domains
Polycrystalline Multi-Layer	Ni foil/Cu foil	CH_4/H_2	950–1050°C/Atmospheric-Low	1–10 layers/500–2000 $cm^2·V^{-1}·s^{-1}$	Layer controllable	Interlayer uniformity
Single-Crystal (Small)	Cu foil	$CH_4/H_2/Ar$	1000–1050°C/Atmospheric	10–20 μm/5000–8000 $cm^2·V^{-1}·s^{-1}$	Low cost	Low cost Size limitation
Single-Crystal (Large)	Molten Cu/Ge/Si	CH_4/H_2	1075–1180°C/Low	0.5–5.9 mm/7000–15000 $cm^2·V^{-1}·s^{-1}$	High quality	Long growth time

Table 1 systematically contrasts the CVD growth routes of four key graphene types, clarifying the correlation between growth conditions and material performance: polycrystalline single-layer graphene uses Cu foil as the substrate, and achieves large-area fabrication (>1 cm²) at 1000°C under atmospheric pressure with low cost, though it faces issues of multi-layer domains. Polycrystalline multi-layer graphene relies on Ni/Cu foils, enables controllable layer numbers (1-10 layers) at 950-1050°C but struggles with interlayer uniformity. Small-sized single-crystal graphene adopts Cu foil as the substrate, features low cost and high mobility (5000-8000 $cm^2·V^{-1}·s^{-1}$) yet is limited by small size (10-20μm). Large-sized single-crystal graphene uses molten Cu/Ge/Si substrates, delivers high quality (mobility 7000-15000 $cm^2·V^{-1}·s^{-1}$) but requires long growth times. Overall, the table highlights the trade-offs between cost, scalability, and performance in CVD growth, providing a clear reference for selecting appropriate routes based on application demands (e.g., low-cost industrial devices for polycrystalline types, high-performance electronics for single-crystal types).

3. Graphene-Based Heterojunctions: Construction and Performance Regulation

Pristine graphene's zero bandgap limits its application in logic devices. Graphene-based heterojunctions—integrating graphene with other 2D materials—enable bandgap opening, enhanced light-matter interaction, and multi-functional integration.

3.1 Classification and Construction Strategies

Graphene heterojunctions are classified by the type of companion material, with common systems including graphene/TMDs, graphene/h-BN, and graphene/Xenes (e.g., arsenene, antimonene).

3.1.1 Construction Methods

One method for constructing graphene heterojunctions involves van der Waals stacking, where exfoliated 2D materials (such as MoS_2 and h-BN) are manually stacked onto graphene through van der Waals forces. This approach effectively avoids chemical damage to the materials. For instance, graphene/h-BN heterojunctions prepared via this method exhibit enhanced stability, thanks to the insulating property and chemical inertness of h-BN [16]. Another commonly used construction strategy is CVD direct growth, which leverages Chemical Vapor Deposition (CVD) technology to grow graphene directly on 2D material substrates (e.g., MoS_2/SiO_2), resulting in the formation of strong interfacial bonding. Lee et al. [17] applied this method to grow graphene on epitaxial Ge films deposited on Si wafers, successfully fabricating 2-inch wafer-scale graphene/Ge heterojunctions and laying the groundwork for the integration of Si-based devices.

3.1.2 Interface Interaction Mechanisms

In the interfacial interaction of graphene heterojunctions, van der Waals forces play a dominant role in systems like graphene/hexagonal boron nitride (h-BN) and graphene/molybdenum disulfide (MoS_2), and this force ensures the intact crystal structures of both components in the heterojunctions. For instance, graphene/MoS_2 heterojunctions not only maintain the direct bandgap of MoS_2 (approximately 1.8 eV) but also enhance electron transfer efficiency [18]. Meanwhile, covalent bonding is typically formed through post-treatment methods such as plasma activation, and this type of bonding helps improve the interfacial charge transfer of heterojunctions. Specifically, Yan et al. [10] successfully achieved covalent bonding between graphene and antimonene via nitrogen (N_2) plasma treatment, which not only increased interfacial adhesion but also improved carrier mobility by 30%.

3.2 Performance Regulation via Heterojunctions

Graphene heterojunctions enable effective regulation of graphene's key properties to address its intrinsic limitations. Specifically, they can induce a tunable bandgap in graphene: graphene/h-BN heterojunctions generate a bandgap of approximately 50-100 meV, which is attributed to lattice mismatch and dielectric screening effects [16], while graphene/MoS_2 heterojunctions open a bandgap of around 200 meV through charge transfer processes. Beyond bandgap engineering, these heterojunctions also enhance optoelectronic performance; for example, graphene/MoS_2 heterojunctions exhibit a photoresponsivity of $\sim 10^3$ $A \cdot W^{-1}$—100 times higher than that of pristine graphene—thanks to efficient separation of photogenerated carriers at the heterojunction interface. Additionally, they improve the material's stability: graphene/h-BN heterojunctions prevent graphene from oxidizing in air, allowing the material to retain 90% of its initial mobility even after 6 months of exposure [17].

4. Applications of Graphene and Graphene-Based Heterojunctions

Graphene and its heterojunctions have been widely applied in electronics, optoelectronics, energy, and biomedicine, leveraging their tunable properties.

4.1 Electronic Devices

4.1.1 Field-Effect Transistors (FETs)

Graphene field-effect transistors (FETs) based on copper-catalyzed Chemical Vapor Deposition (CVD) exhibit a carrier mobility of approximately 10^4 $cm^2 \cdot V^{-1} \cdot s^{-1}$ at room temperature [5], and this excellent performance allows them to meet the requirements of high-frequency devices with a cutoff frequency exceeding 100 GHz. On the other hand, graphene/molybdenum disulfide (MoS_2) heterojunction FETs achieve an on/off ratio of about 10^5—a value 10^3 times higher than that of pristine graphene FETs—thanks to the bandgap property of MoS_2 [11], which is sufficient to satisfy the core performance requirements of logic devices.

4.1.2 Sensors

Gas Sensors: Graphene-based NO_2 sensors show a detection limit of ~25 ppb at room temperature, with response/recovery times of 26/290s [18]. Graphene/antimonene heterojunctions enhance sensitivity to NH_3 by 5 times via synergistic adsorption [7].

Biosensors: Graphene field-effect biosensors detect DNA with a limit of $\sim 10^{-18}$ M [19], leveraging graphene's high surface sensitivity to biomolecule adsorption [20].

ISSN 2959-6157

4.2 Optoelectronic Devices

4.2.1 Photodetectors

Graphene/MoS_2 heterojunction photodetectors achieve a broadband response (400-1100 nm) and a detectivity of ~10^{13} Jones [11], suitable for visible-NIR imaging. Graphene/h-BN heterojunctions further improve detectivity to ~10^{14} Jones via reduced dark current [17].

4.2.2 Solar Cells

Graphene-based heterojunction solar cells: Graphene/P3HT:PCBM solar cells exhibit a power conversion efficiency (PCE) of ~8% [21], while graphene/$MAPbI_3$ perovskite solar cells achieve a PCE of ~20% by enhancing light absorption and charge transfer [7].

4.3 Energy Storage and Conversion

4.3.1 Lithium-Ion Batteries (LIBs)

Graphene anodes: CVD graphene anodes exhibit a reversible capacity of ~1050 $mAh \cdot g^{-1}$ (3 times higher than graphite) due to their layered structure and high Li^+ diffusion rate [22].

Heterojunction anodes: Graphene/Sb heterojunctions achieve a capacity of ~660 $mAh \cdot g^{-1}$ with 90% retention after 500 cycles [23], solving Sb's volume expansion issue.

4.3.2 Catalysis

Hydrogen Evolution Reaction (HER): Graphene/Ni heterojunctions exhibit an overpotential of ~50 mV at 10 $mA \cdot cm^{-2}$(close to Pt/C) due to Ni's catalytic activity and graphene's electron transfer.

CO_2 Reduction: Graphene/Cu heterojunctions convert CO_2 to CH_4 with a Faradaic efficiency of ~70% [22], leveraging graphene's high surface area and Cu's selectivity.

4.4 Biomedical Applications

Photothermal Therapy (PTT): Graphene quantum dots (GQDs) exhibit a photothermal conversion efficiency of ~42%, achieving tumor ablation in mice under 808 nm laser irradiation [23].

Bioimaging: Graphene/Fe_3O_4 heterojunctions enable magnetic resonance imaging (MRI) and photoacoustic imaging (PAI) dual-modal imaging, with a PA signal-to-noise ratio of ~30 dB [24].

5. Challenges and Future Prospects

5.1 Current Challenges

Large-Area Single-Crystal Growth: The size of single-crystal graphene is still limited to the ~cm scale; achieving inch-scale single crystals with atomic-level uniformity remains difficult.

Heterojunction Interface Compatibility: Lattice mismatch between graphene and companion materials (e.g., ~10% between graphene and MoS_2) causes interface defects, reducing device performance.

Long-Term Stability: Graphene oxidizes in air, and heterojunctions degrade due to interfacial water adsorption, limiting practical application.

5.2 Future Prospects

Atomic-Level Growth Control: Develop "substrate atomic polishing + precise oxygen regulation" to achieve cm-scale single-crystal graphene with <1 defect per μm^2.

Heterojunction Functional Integration: Construct multi-component heterojunctions (e.g., graphene/MoS_2/h-BN) for multi-functional devices (e.g., self-powered photodetectors).

Industrialization Optimization: Develop roll-to-roll CVD systems for large-area graphene production, reducing cost to <$10 per cm^2.

6. Conclusion

This paper systematically reviews graphene's Chemical Vapor Deposition (CVD) synthesis, graphene-based heterojunction construction and performance modulation, and their applications in electronics, energy, and biomedicine which maps its progress from lab research to potential industrial use. CVD is pivotal for high-quality graphene, outperforming traditional methods like Cu-based systems lead polycrystalline graphene production for mass-produced devices, while molten Cu/Ge substrates show promise for single-crystal graphene (critical for high-performance electronics) but face scaling and long-growth-time challenges. Graphene heterojunctions solve its intrinsic zero-bandgap issue. Integrates with 2D materials (e.g., h-BN, MoS_2) tunes bandgaps, enhances optoelectronic properties (e.g., 10^3 $A\cdot W^{-1}$ photoresponsivity in graphene/MoS_2), and improves stability (90% mobility retention in graphene/h-BN after 6 months). Key applications include high-mobility graphene FETs, high-capacity graphene anodes, and graphene quantum dot (GQD)-based photothermal therapy, yet hurdles remain—limited single-crystal size, heterojunction interface defects, and oxidation. Future work focusing on atomic-level growth control, multi-component heterojunctions, and roll-to-roll CVD will drive industrialization, with CVD and heterojunctions as dual pillars to bridge graphene's lab discoveries to real-world impact.

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