

Synthesis, Structural Characteristics, and Applications of Vertically Oriented Graphene: A Review

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Abstract. Vertically oriented graphene (VG), few-layer graphene nanosheets that stand upright on the substrate, has attracted significant attention within the carbon nanomaterial family owing to its unusual structural geometry and the resulting electrochemical and physical performance advantages. This review provides a comparative analysis of the two dominant synthesis routes: thermal chemical vapor deposition (Thermal CVD) and plasma-enhanced chemical vapor deposition (PECVD). The plasma sheath electric field is identified as the key mechanism responsible for deterministic vertical growth. The roles of principal process parameters (plasma power density, gas composition, chamber pressure, and substrate bias) in shaping nanosheet morphology and crystallinity are examined. Applications of VG in electrochemical energy storage, high-sensitivity sensing, cold-cathode field emission, and electrocatalysis are surveyed. Outstanding challenges, including large-area uniformity, low-temperature growth, and long-term stability, are discussed alongside emerging trajectories such as machine-learning-driven process optimization and engineered two-dimensional heterostructures. Together, these discussions yield a coherent structure–property–function picture of VG that underpins its push toward practical device deployment.

Keywords: vertically oriented graphene, carbon nanowalls, plasma-enhanced chemical vapor deposition (PECVD), electrochemical energy storage, field emission

1. Introduction

Carbon-based nanomaterials, particularly planar graphene, have reshaped nanotechnology thanks to a remarkable suite of physical and chemical properties [1]. However, a significant limitation persists: horizontal sheets tend to irreversibly restack, resulting in a sharp reduction in the effective specific surface area. Vertically oriented graphene (VG), an architectural variant that sidesteps the problem by self-assembling into an unagglomerated three-dimensional network [2]. Its exposed sharp edges and open nanochannels make it particularly attractive for energy storage and field emission [3]. On the synthesis side, thermal chemical vapor deposition demands a hefty thermal budget. In contrast, plasma-enhanced chemical vapor deposition (PECVD) lets the sheath electric field guide growth into a low-temperature, directionally aligned process [4]. Progress has been rapid, yet the grasp of nucleation mechanisms and morphology control remains incomplete.

This study compares thermal CVD and PECVD to show how growth mechanisms dictate VG morphology and structural quality. It then systematically explores applications in electrochemical energy storage, sensing, field emission, and electrocatalysis, consistently linking functional performance back to these structural features. The study further examines the manufacturing challenges that still limit practical deployment and identifies research directions likely to shape future development.

2. Definition and characteristics of vertically oriented graphene

2.1. Structural definition and spatial configuration

VG, often labeled carbon nanowalls (CNWs) in older papers, belongs to a distinct three-dimensional (3D) branch of the nanocarbon family [2]. What sets it apart from the usual low-dimensional carbon structures is its geometry: its few-layer graphitic sheets rise from the substrate at high inclinations, typically 70° – 90° [2, 5]. Crystallographically, these standing walls are built on highly ordered sp^2 -hybridized carbon networks [2]. The perpendicular orientation, paired with a cross-linked, maze-like connectivity, yields a self-supporting matrix with mechanical robustness to spare; ultra-thin nanosheets that would otherwise collapse under capillary forces or their own weight stay upright inside this framework [5, 6]. Two factors are primarily responsible for this behavior: the high bending stiffness of individual graphene walls and the mechanical interlocking they exhibit within the cross-linked 3D network, which together cancel out the thermodynamic push toward parallel interlayer restacking [2]. Conventional horizontal graphene sheets suffer the opposite fate: strong interplanar van der Waals forces drive them into irreversible aggregation, and the effective specific surface area takes a hit [5]. VG overcomes this limitation: the mutual support between interconnected vertical walls keeps parallel alignment at bay, locking in the high surface accessibility of each layer inside a stable 3D scaffold [2, 5].

2.2. Morphological and microstructural features

VG is readily identified by its distinctive surface morphology: a continuous, maze-like topography woven from interconnected vertical walls [2]. Individual nanowalls are very thin, with cross-sections ranging from a handful of atomic layers to tens of nanometers, placing them squarely in few-layer graphene territory [6]. Lateral extents and vertical heights span hundreds of nanometers to several micrometers, resulting in exceptional aspect ratios [6, 7]. Upon closer examination, a defining trait is revealed: a staggering density of exposed sharp top edges and open boundaries [8]. These terminated edges break sharply from the chemistry of ideal basal planes; they carry unsaturated carbon bonds, non-hexagonal atomic rings, and a variety of localized structural defects, including vacancies and dislocations [6]. The result is a surface with a high density of exposed, unsaturated bonding sites [6]. The nanowall terminations also carry nanoscale tip geometries, with radii of curvature on the nanometer scale, that have become a morphological signature of VG [8].

2.3. Transport phenomena and pore architecture

Aligning the graphitic basal planes perpendicular to the substrate creates sharp anisotropy in both electrical and thermal transport [8]. Travelling along the vertical axis, parallel to individual graphene sheets, charge carriers and phonons sail through continuous, low-resistance pathways with near-ballistic or quasi-ballistic transport [8]. This directional conduction bypasses the grain-boundary

resistance and hopping barriers that impede disordered or horizontally aggregated carbon networks. Moving transversely, perpendicular to the sheets, the behavior changes sharply: interlayer van der Waals gaps and structural discontinuities throttle transport [8]. Complementing this electrical picture, VG's open-pore architecture enhances surface accessibility and accelerates mass transport [5, 9]. Spatial gaps between adjacent vertical walls provide non-tortuous linear nanochannels for electrolytic ions, gases, and liquid species, keeping the geometric surface area accessible and reducing ion diffusion resistance to a level well below that achievable with tortuous porous carbons [3, 5, 9].

3. Comparative analysis of major preparation methods and growth mechanisms of vertically oriented graphene

3.1. Thermal chemical vapor deposition

Thermal chemical vapor deposition (Thermal CVD) was the first route to establish VG. Inside a catalytic CVD reactor, hydrocarbon precursors crack at elevated temperatures, typically 700-1000 °C, on transition-metal substrates such as nickel or copper, releasing carbon radicals that nucleate and self-assemble into graphitic structures. If the catalyst is omitted, pyrolytic decomposition proceeds on its own, producing amorphous or partially graphitized films whose quality depends strongly on temperature [4].

The drawback of Thermal CVD is fundamental: it provides no intrinsic electrostatic mechanism to direct growth perpendicular to the substrate. In the absence of an external orienting field, nascent graphene layers predominantly form parallel to the substrate, and achieving vertical alignment typically requires elaborate post-deposition treatments or specialized substrate engineering. On top of that, the high processing temperatures restrict which substrates can survive the run and inflate fabrication costs, excluding Thermal CVD VG from temperature-sensitive device platforms [5, 6].

3.2. Plasma-enhanced chemical vapor deposition

Plasma-enhanced chemical vapor deposition (PECVD) has become the synthesis route of choice for VG, a shift driven by the orientation problem that hinders Thermal CVD. In a PECVD reactor, radio-frequency or microwave power ionizes the precursor gas mixture, forming a plasma sheath at the substrate surface. Sustained across that sheath is a strong electric field perpendicular to the substrate, which orients the graphene nuclei vertically from the very earliest stages of growth and continuously drives nanosheet elongation along the field direction [7, 8]. Reactive hydrogen species generated within the plasma selectively etch amorphous carbon deposits, leaving only well-ordered graphitic phase behind, and crystallinity and aspect ratio improve without requiring a separate purification step.

Several interdependent process parameters ultimately dictate the geometry and quality of the resulting VG. Plasma power density governs both reactive species density and ion bombardment energy; excessively high power degrades edge crystallinity. The CH₄/H₂ ratio balances carbon supply against etching: hydrogen-rich conditions selectively remove disordered carbon, improving the I_{2D}/I_G Raman ratio. Chamber pressure shifts mean free path and plasma uniformity, while substrate temperature drives surface diffusion and graphitization kinetics. Applying a negative substrate bias increases ion energy at the growth front, sharpens edges, and increases the field enhancement factor relevant to field emission [4, 10].

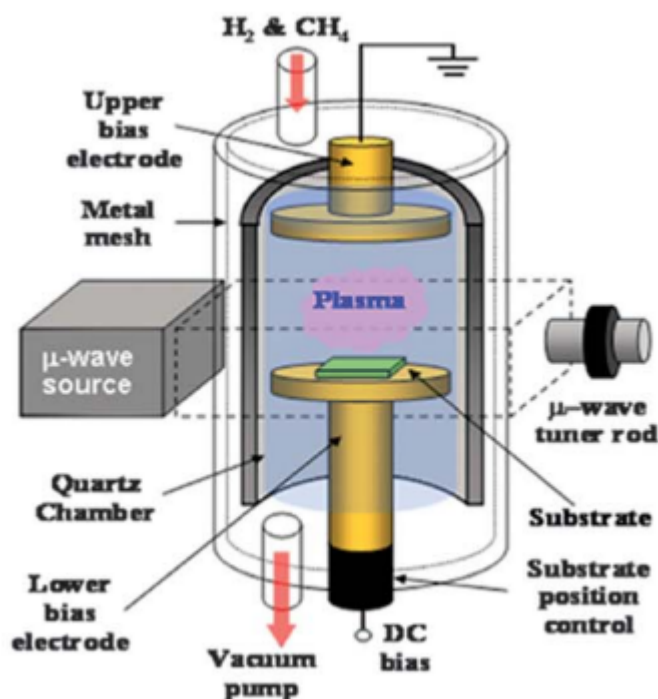


Figure 1. Schematic of the PECVD growth mechanism of VG, illustrating the plasma sheath electric field perpendicular to the substrate as the directional force behind nanosheet nucleation and vertical elongation, and contrasting it with the substrate-parallel growth of thermal CVD [7]

3.3. Comparative assessment

What separates the two methods fundamentally is how they handle orientation. Thermal CVD has only thermodynamic driving forces to lean on, with no lateral electrostatic control, which is why reproducible vertical alignment proves so elusive. PECVD introduces a directing field via the plasma sheath, enabling deterministic vertical growth across a broad substrate range and at much gentler thermal budgets, on certain substrates as low as 300–400 °C [4]. Raman spectra reported for PECVD-grown VG typically show relatively low ID/IG ratios and well-resolved 2D bands, consistent with fewer in-plane defects and longer coherence lengths [10]. Films deposited by Thermal CVD are separately reported to carry higher disorder under comparable carbon-supply pressures [11]. Because these findings come from independent studies with differing reactor geometries and precursor chemistries, they cannot be compared quantitatively without dedicated side-by-side experiments.

In terms of scalability, Thermal CVD draws on mature reactor infrastructure and is compatible with large-area roll-to-roll processing [11]. PECVD equipment demands a higher capital investment but affords independent control over ion bombardment, plasma chemistry, and substrate temperature, capabilities that individual studies have each linked to tunable nanosheet morphology and broader substrate compatibility [4, 6].

4. Multifunctional applications of vertically oriented graphene

4.1. Electrochemical energy storage

The open, non-stacked vertical channels of VG provide straight, unobstructed ion-transport pathways, keeping the entire graphitic surface electrochemically accessible even at high scan rates. Miller et al. demonstrated a near-ideal capacitive phase angle at 120 Hz and a characteristic frequency in the kHz range for VG-based supercapacitors [3]. Shiji et al. confirmed that PECVD-grown VG films retain a much larger share of their low-rate capacitance during fast charge–discharge than planar graphene does, an advantage they attribute directly to the vertical channel geometry [10]. The binder-free monolithic structure offers an additional benefit: it eliminates the parasitic contact resistance introduced by polymer binders in conventional composite electrodes, reducing equivalent series resistance and enhancing power delivery. In batteries, VG scaffolds accommodate the ~300% volumetric expansion of silicon anodes during lithiation, maintain continuous electron transport through the graphene framework, and, when nitrogen-doped—chemically anchor lithium polysulfide intermediates in lithium-sulfur (Li-S) cathodes to mitigate the shuttle effect [8, 12].

4.2. Sensing applications

VG electrodes exhibit exceptional electrochemical biosensing performance, and the dense population of unsaturated edge carbon atoms plays the dominant role, they serve as efficient electron-transfer mediators. Those edge sites carry an elevated local density of states, which speeds up heterogeneous electron transfer for the oxidation of glucose, dopamine, and other bioanalytes; representative VG-based sensors achieve detection limits as low as 0.1 μM for dopamine and ~2 μM for glucose with sensitivities one to two orders of magnitude higher than bare glassy carbon electrodes (GCE), alongside linear ranges extending to several millimolar [12, 13]. The same three-dimensional open network excels in gas sensing: a high accessible surface area maximizes analyte adsorption, and the vertical channel geometry eliminates lateral diffusion constraints, yielding response times of 30–60 s and sub-ppm detection thresholds for NO_2 and NH_3 [13]. Selectivity can be turned by heteroatom doping or metal-oxide nanoparticle decoration, widening the applicability envelope from industrial safety monitoring to exhaled-breath diagnostics.

4.3. Field emission

Atomically sharp edges and high aspect ratios enable VG nanosheets concentrate the local electric field so intensely, courtesy of the classical tip-enhancement effect; field enhancement factors β are on the order of 10^3 . Substituting these parameters into Fowler–Nordheim theory yields an exponential rise in the emission current density with β , enabling substantial cold-cathode emission at macroscopic fields well below those required by conventional emitters [2, 3]. The turn-on field, however, is sensitive to nanosheet packing density: when the array is packed too tightly, inter-tip electrostatic screening occurs, paradoxically raising the turn-on threshold. Zhu et al. showed that density-optimized VG arrays achieve turn-on fields below 1 $\text{V } \mu\text{m}^{-1}$ and current densities above 10 mA cm^{-2} at 5 $\text{V } \mu\text{m}^{-1}$, providing sufficient evidence that VG belongs in the next-generation cold-cathode emitters for electron microscopy [6].

4.4. Electrocatalysis and water treatment

VG carries measurable HER activity in its intrinsic edge defects, and in situ nitrogen or phosphorus doping during PECVD growth amplifies that activity substantially: heteroatom incorporation shifts the free energy of H^* adsorption toward the thermoneutral region ($\Delta GH^* \approx 0$), reducing overpotential to levels that are comparable to benchmark non-precious catalysts [11]. In alkaline-medium oxygen reduction reaction (ORR), pyridinic and graphitic nitrogen sites unlock an efficient four-electron reduction pathway with onset potentials closing in on commercial Pt/C, plus superior methanol tolerance [7]. Decorating the three-dimensional VG scaffold with transition-metal nanoparticles builds synergistic metal–carbon active sites, while the scaffold itself stops nanoparticle sintering and keeps electron-transport resistance low [12]. In capacitive deionization, the high surface area paired with VG's vertical channel structure delivers greater salt adsorption capacity and faster ion uptake than activated-carbon electrodes; the material's chemical stability under aggressive electrolytes enables removal of heavy-metal ions, chiefly Pb^{2+} and Cr^{6+} among them, from industrial wastewater [10, 13].

The preceding sections reveal a consistent set of structure–property relationships across all application domains. Table 1 maps these four structural features onto their derivative functional advantages and the application fields in which they have been demonstrated.

Table 1. Mapping of VG structural characteristics onto their derivative functional advantages and the application domains where they show up

Structural Feature of VG	Derived Physical/Chemical Advantage	Representative Application Fields
Highly oriented, non-stacked vertical channels	Unobstructed linear diffusion of electrolyte ions and gas molecules, minimizing mass-transport resistance	Electrochemical energy storage requiring high-rate charge/discharge kinetics; resistive gas sensing requiring rapid analyte adsorption
Binder-free monolithic electrode formed by in situ growth on conducting substrates	Elimination of parasitic interfacial resistance and inactive binder mass; seamless covalent electron pathway to current collector	Li-ion/Li-S battery electrodes; electrocatalytic electrodes for HER and ORR where minimizing ohmic drop is critical
Atomically sharp graphene lattice edges with nanometer-scale tip radii and high aspect ratio	Intense local electric-field concentration at nanosheet tips ($\beta \sim 10^3$); substantially reduced effective work function	Cold-cathode field emission sources for electron microscopy, flat-panel displays, and X-ray generation
High-density unsaturated edge carbon atoms and structural lattice defects	Abundant electrochemically active sites with elevated local density of states; facile surface functionalization	Electrochemical biosensors for biomolecule detection; non-precious HER and ORR electrocatalysts

5. Challenges and prospects

5.1. Bottlenecks in controllable synthesis

Even though PECVD has matured into the preferred VG synthesis route, a handful of engineering hurdles still stand between laboratory-scale demonstrations and dependable industrial production.

5.1.1. Spatial non-uniformity in large-area growth

The most pressing issue is spatial non-uniformity: large-area reactors and roll-to-roll configurations inherently produce non-uniform plasma sheath electric fields. The resulting gradients in ion energy and radical flux cause pronounced variations in nanosheet height, density, and crystallinity across the substrate. Tackling this means turning to advanced multi-anode or magnetically confined plasma sources and pairing them with computational fluid dynamics (CFD) optimized gas flow patterns [8, 9].

5.1.2. Low-temperature growth and structural quality

A second tension sits between low-temperature growth and structural quality. Dropping substrate temperature below roughly 400 °C is essential for compatibility with flexible polymer substrates and back-end semiconductor processes, yet reducing thermal energy impairs precursor decomposition efficiency and slows graphitization, leaving behind higher defect densities and lower aspect ratios. Developing highly reactive radical precursors, or pulsed plasma excitation schemes that decouple thermal and plasma contributions, looks promising on paper but is technically demanding in practice [4, 14].

5.1.3. Reactor cost and feedstock sustainability

One more obstacle: the high capital cost of radio-frequency and microwave plasma reactors, combined with continued reliance on fossil-derived hydrocarbon feedstocks, blocks cost-competitive scale-up. Active investigations into biomass-derived carbon sources and atmospheric-pressure plasma alternatives are aiming squarely at that bottleneck [12, 13].

5.2. Application-level challenges

5.2.1. Electrochemical and mechanical degradation

Translating VG from the laboratory to durable device operation presents its own set of challenges. In electrochemical storage and catalysis, repeated ion intercalation and harsh electrolyte exposure gradually induce edge bending, nanosheet aggregation, and root delamination from the substrate, each of which erodes the open-channel architecture on which performance depends. Mechanically, individual vertical nanosheets lack lateral cross-linking, leaving them vulnerable to collapse under compressive stress; one proposed remedy is to build three-dimensionally interconnected, honeycomb-like VG architectures with deliberately integrated cross-link junctions [6, 10].

5.2.2. Field emission and substrate adhesion

In field emission, high-voltage operation produces localized Joule heating at nanosheet tips, which can ablate them and gradually degrade their morphology. VG adheres to non-metallic substrates (glass, flexible polymers, and the like), mostly through weak van der Waals interactions, which leaves contact resistance elevated and peel strength wanting; plasma pre-activation of the substrate surface to introduce covalent anchoring sites, or deposition of thin transition-metal interlayers, both offer paths to mitigation [6, 8].

5.2.3. Characterization standards and reproducibility

A broader systemic issue is also worth noting: the field has yet to converge on internationally harmonized characterization standards for VG. Different research groups quantify nanosheet density, accessible surface area, and active-site concentration using inconsistent protocols, making cross-laboratory data difficult to validate and hindering progress toward reliable structure, property relationships.

5.3. Future directions

Three emerging directions are poised to shape the next chapter of VG research. The first is machine-learning-assisted process optimization, which offers a tractable way through the sprawling, high-dimensional PECVD parameter space, power, pressure, gas ratios, temperature, and bias voltage, en route to on-demand nanosheet geometry with sub-nanometer precision; early demonstrations have already appeared in analogous thin-film deposition systems [11, 12].

Second, multifunctional integrated device architectures that leverage VG's overlapping electrochemical, sensing, and thermal-management capabilities are expected to displace single-function implementations, especially in smart wearables and automotive electronics, where mass, volume, and energy budgets are tightly squeezed.

Third is atomic-scale heterostructure engineering: pairing VG with complementary two-dimensional materials such as MXenes, black phosphorus, or metal–organic framework-derived carbons, an approach expected to unlock synergistic properties beyond what either component delivers alone, enabling quantum electronic devices and catalysis under extreme conditions [7, 13].

Table 2 presents a structured overview of the current manufacturing bottlenecks, their underlying root causes, and proposed research strategies to address them.

Table 2. Summary of current manufacturing bottlenecks, root causes, and proposed remedies for VG materials

Current Limitation	Scientific/Technical Root Cause	Proposed Future Solution
Non-uniform large-area growth	Plasma sheath field distortion and radial gradients in radical flux across large substrates	Multi-anode / magnetically confined plasma sources; computational fluid dynamics-guided reactor design
Thermal damage to flexible substrates	Insufficient precursor decomposition and graphitization at low temperatures	Highly reactive radical precursor chemistries; pulsed plasma excitation to decouple thermal and plasma contributions
Nanosheet collapse during service	Lack of lateral cross-linking in isolated vertical structures	Three-dimensional honeycomb-interconnected VG architectures with deliberate cross-link junctions
Root delamination from non-metallic substrates	Weak van der Waals bonding between carbon and glass/polymer surfaces	Plasma pre-activation of substrate surface; deposition of thin covalent-bonding transition-metal interlayers

6. Conclusion

This review has systematically presented the preparation, growth mechanisms, and multifunctional applications of VG. Placing Thermal CVD and PECVD side by side shows that the plasma sheath electric field in PECVD is the decisive ingredient behind deterministic perpendicular growth, getting around the fundamental orientation limitation built into thermally driven synthesis. Tuning plasma power density, gas composition, chamber pressure, and substrate bias translates directly into control

over nanosheet height, edge sharpness, aspect ratio, and crystalline quality, structural variables that, in turn, govern performance across every application domain examined here.

The structure-to-function mapping running through this review demonstrates: the open vertical channel architecture drives ion-transport kinetics in energy storage and mass-transport kinetics in gas sensing; the atomically sharp nanosheet edges generate the field enhancement factors that cold-cathode field emission demands; the high-density edge defect sites supply the electrochemically active surface that biosensing, HER, and ORR catalysis cannot depend on. For all electrochemical applications, the binder-free monolithic electrode eliminates interfacial resistance. Addressing the remaining obstacles of large-area uniformity, low-temperature substrate compatibility, and long-term stability will require machine-learning-guided PECVD optimization, cross-linked three-dimensional nanoarchitectures, and heterostructure integration with emerging two-dimensional materials.

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