

Systematic Literature Review on Low Growth Temperature of Single Layer Large Area Graphene Using Chemical Vapor Deposition

Raihana Bahru, A. Atiqah, Maria Abu Bakar, Abdul Rahman Mohmad, Muhammad Aniq Shazni Mohammad Haniff,

Mohd Zulhakimi Ab Razak, Mohd Ambri Mohamed & Azman Jalar*

Institute of Microengineering and Nanoelectronics, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor

*Corresponding author: azmn@ukm.edu.my

Received 8 November 2025, Received in revised form 3 June 2026

Accepted 3 July 2026, Available online 30 September 2026

ABSTRACT

This systematic literature review provides a comprehensive analysis of the current state of the art in fabricating high-quality, large-area, single-layer graphene via Chemical Vapor Deposition (CVD) at low growth temperatures. While graphene's exceptional electrical and mechanical properties are well-documented, the established method for its synthesis, which often requires temperatures exceeding 1,000°C, poses a significant barrier to commercial scalability and integration with temperature-sensitive materials. This study surveyed electronic databases for peer-reviewed studies published between 2009 and 2023, identified and synthesized 73 key studies that meet rigorous inclusion criteria. The analysis delves into the intricate relationship between growth parameters and the resulting material quality, focusing on essential metrics such as defect density, layer uniformity, and structural integrity. A pivotal finding of this research is the successful reduction of synthesis temperatures to a range between 100°C and 300°C, achieved primarily through the implementation of Plasma-Enhanced CVD (PECVD). The method utilized plasma to dissociate carbon precursors, such as benzene, at much lower energy levels than the thermal heating method. By decoupling the carbon radical generation from the substrate temperature, researchers have successfully produced single-layer graphene without compromising scalability. Ultimately, this review highlights promising advancements and emerging trends that pave the way for the direct growth of graphene on flexible substrates and polymers, offering a strategic roadmap for future research directions and the practical application of graphene in next-generation electronics and hybrid nanostructures.

Keywords: Graphene; Chemical Vapor Deposition (CVD); low growth temperature; single layer; large area; systematic review

INTRODUCTION

Over the past two decades, graphene has garnered immense interest owing to its exceptional characteristics, including superior mechanical strength, outstanding electrical conductivity, and remarkable thermal properties. The breakthrough in isolating graphene is by Novoselov et al. (Novoselov et al. 2004) using a simple method known as mechanical exfoliation or the Scotch tape method. Geim and Novoselov won the 2010 Nobel Prize in Physics for their groundbreaking experiments on graphene, a two-dimensional material that has potential for use in nanotechnology (Geim & Novoselov 2007). Until now, many works on graphene including synthesis and applications in various fields shown higher interest globally

and have exponentially increased (estimated 25,000 research publications per year) (Urade et al. 2023). Graphene shares a fundamental structural basis with other carbon allotropes, including charcoal and graphite (Stankovich et al. 2007), fullerenes (Shrestha et al. 2013) and carbon nanotubes (Popov 2004). It has properties that can replace conventional materials in various applications. Examples of excellent properties such as strong mechanical strength (Young modulus of 1TPa, third-order elastic stiffness of $D = -2.0$ TPa, and intrinsic strength of $\sigma_{int} = 130$ GPa) (Lee et al. 2008), high thermal conductivity (5300 W.m⁻¹.K⁻¹) (Balandin et al. 2008; Raja N Othmana 2021), and high electron mobility (200,000 cm².V⁻¹.s⁻¹) (Bolotin et al. 2008; Novoselov et al. 2005). These qualities of graphene represent it as a potential substitute for numerous conventional materials across various applications.

Current applications of graphene include its use in conductive inks for printed electronic circuits (Cortés et al. 2020; Overgaard et al. 2017), incorporation into composites to enhance mechanical and thermal properties (Bokare et al. 2021; Dai et al. 2021), and its role in batteries and supercapacitors to improve energy storage (Cheah et al. 2024). Moreover, graphene is employed in sensors for its high sensitivity and transparent conductive films in touchscreens and flexible electronic devices (Esteghamat & Akhavan 2023; Gaur et al. 2021; Tkachev et al. 2021). Looking ahead, the future applications of graphene are promising, with potential breakthroughs in the semiconductor industry, leading to the development of high-speed transistors and other electronic components. Additionally, graphene may contribute to quantum computing (Alonso Calafell et al. 2019; Wang et al. 2021), thermal management (Ali et al. 2021; Chen et al. 2020; Huang et al. 2021), communication devices (Kuscu et al. 2021; Singh, Andrelloet et al. 2020; Singh, Kumaret al. 2020), and spintronics (He et al. 2022; Khokhriakov et al. 2020; Naganuma et al. 2020), marking it as a versatile material with transformative potential in the world of electronics. It's essential to stay updated on the latest research, as new applications and advancements continue to unfold in this rapidly evolving field.

Common techniques of graphene synthesis include exfoliation of graphite, chemical oxidation and reduction, thermal chemical vapor deposition (TCVD), plasma chemical vapor deposition (PCVD), thermal decomposition of silicon carbide (SiC), and unzipping CNTs and other methods. The exfoliation of graphite is done using scotch tape or micromechanical cleavage to repeatedly peel off the graphite until layers of graphene. This method produces low yield, is time-consuming, and is limited to lab-scale which is impractical for mass production (Novoselov et al. 2004). The chemical oxidation and reduction method is a traditional method of chemical reduction of graphene oxide (GO). The approach is an innovation of chemical oxidation and reduction methods to produce the GO via the Hummers method and then reduce using chemical reducing agents such as hydrazine and sodium borohydride. However, the reducing agents are not environmentally safe. Therefore, ascorbic acid is found to be a better-reducing agent due to its being environmentally friendly, cheap, easily available in the market, and practical for mass production (Khan & Shaida 2023).

TCVD consists of the gas decomposition process by gas phase heating (by a hot filament or hot wall), which generates radical species by diffusion, which can be reached and deposited on a suitably placed substrate (Huang et al. 2012). The CVD method is well-known as a high-temperature process (basically more than 1000 °C), able to produce high-quality graphene. The CVD

production of graphene generally involves four steps: (i) synthesizing graphene films on metal foils like copper via CVD, (ii) removing the metal, (iii) transferring the graphene films onto a target substrate, and (iv) doping the graphene film to reduce sheet resistance, if necessary. PCVD is an alternative to growing graphene at lower temperatures. An uncomplicated and easily replicable method is crucial, as it enables the synthesis of flakes without requiring a catalyst on the surface of any substrate capable of withstanding temperatures up to 700 °C (Li et al. 2016; Malesevic et al. 2008). Meanwhile, the thermal decomposition of SiC offers practical graphene growth without transfer mode during synthesis. This method is suitable for fabrication in electronic devices where the Si atoms desorb and leave behind the carbon atoms and form graphitic layers at high temperatures of 1100-1650 °C (Emtsev et al. 2009).

Unzipping CNT is another recent method without chemical methods to produce graphene nanoribbons. The methods involve various zipping techniques such as a mixture of acids, a catalytic cutting method, electric zipping and H-based unzipping (Silva et al. 2018). In principle, the process consists of catalytic hydrogenation of carbon, in which the metal particles cut sp² hybridized carbon atoms along nanotubes resulting in the liberation of hydrocarbon species. Even though this method effectively reduces the width of the graphene nanoribbons, the yield obtained is lower (Eliás et al. 2010). There are various techniques exist for producing and analyzing 2D graphene, with some of these methods currently being scalable, while others are still limited to laboratory-scale applications.

Many companies have started manufacturing graphene materials on a large scale, ranging from ton-level production of flake graphene to hundreds of thousands of square meters for graphene films, with the goal of meeting industrial application demands. The mass production techniques employed must align with industry testing requirements, typically involving kilograms of powder or suspension containing graphene flakes (typically in the micrometre scale) or a thousand pieces for continuous graphene films (typically larger than a millimetre size). Moreover, the cost of production is also important in aligning with a larger scale of mass production. CVD is widely recognized as the primary method for commercial graphene production in industry, alongside other techniques like graphite exfoliation and the chemical oxidation and reduction of graphene oxide (Wang et al. 2018; Zhu, Jiet al. 2018).

The exfoliation of graphite can produce a higher quality of hundreds of tonnes of rGO per year. Recent improvements in graphite exfoliation indicate that keeping temperatures below 10 °C during both the oxidation and quenching stages of the Hummers method helps prevent

carbon loss as CO_2 . This technique yields graphene oxide (GO) with a primarily hexagonal carbon structure, low defect density, and charge-carrier mobility above $1,000 \text{ cm}^2/\text{V.s}$. Moreover, liquid-phase exfoliation is now used for large-scale graphene flake production, though it generates a mix of single-layer and few-layer graphene flakes and can introduce structural damage, including fragmentation or partial oxidation. It is also less cost-effective for scaling up production (for example using microwaves or ovens). Electrochemical exfoliation is the counter approach of low-cost liquid-phase exfoliation with high-quality graphene sheets, but the application is limited to lower charge carrier mobilities (Yang et al. 2016).

CVD is another promising method for producing high-quality, large-area graphene on various substrates under conditions of temperatures above 1000°C and ultra-high vacuum. The market's capacity for large-scale graphene film production has reached around $1,000,000 \text{ m}^2$ annually, exemplified by companies like 2D carbon (Changzhou) Tech. Inc. Ltd, Wuxi Graphene Films Co. Ltd, and Chongqing Graphene Technology Co. Ltd in China; Graphene Square Inc. in Korea; Graphenea Inc. in Spain; BGT Materials Ltd in the UK; and Graphene Laboratories Inc. in the USA (Ren & Cheng 2014; Xiao et al. 2016). However, the challenges faced by CVD methods such as the higher epitaxial growth temperature restricting film sizes, breaking damage of graphene during transfer, sheet resistance on the as-prepared graphene and quality of direct growth on other applications substrate comparable to the copper substrate (Zhu, Jiet al. 2018).

There are many types of graphene such as monolayer graphene (Jin et al. 2011), bilayer graphene (Mccann & Koshino 2013), multilayer graphene, graphene oxide, reduced graphene oxide (Mittra et al. 2023), graphene nanoribbons (Zhang et al. 2023), graphene quantum dots (Ghaffarkhah et al. 2022) and others as illustrated in Figure 1. Each type of graphene has its capabilities to deliver performance. High-quality graphene should be a single layer of carbon atoms with exceptional purity. The previous findings show that many works on sustainable graphene synthesis involve low growth temperatures, established growth catalysts and precursors and a large covered growth area with high-quality film. However, the limitations on synthesis graphene such as low quality of multilayer graphene film, high growth temperature (more than 300°C), high cost of synthesis operations, minimum growth area that is not suitable for commercialization and other challenges. The recent interest in the improvement of synthesis graphene for commercialization are low growth temperatures which range between 100 to 300°C and large growth areas for various applications. The lower growth temperature is significant for the cost-effective synthesis and commercialization of graphene in various applications.

Therefore, with this much information published on the graphene synthesis method, the most suitable method to obtain comprehensive results is using a systematic literature review (SLR). This SLR method will identify and evaluate all relevant literature on a topic to derive conclusions about the question under consideration. Moreover, the SLR employs details and extensive search strategies to locate all relevant studies, whereas traditional reviews may not be as comprehensive. Conventional literature reviews also can lead to a higher risk of bias and a less comprehensive overview of the topic.

In this study, a systematic literature review (SLR) was conducted to evaluate the existing research on the synthesis of graphene in both single-layer and monolayer forms at low growth temperatures (100 to 300°C). The qualitative analysis of all research articles published in this field was conducted and the literature focusing on the current progress and insight of synthesis graphene on a copper substrate using the CVD method was thoroughly reviewed for the first time. The target is to obtain a significant impact on the area and the possibility for improvement in graphene synthesis that will have the potential for mass production.

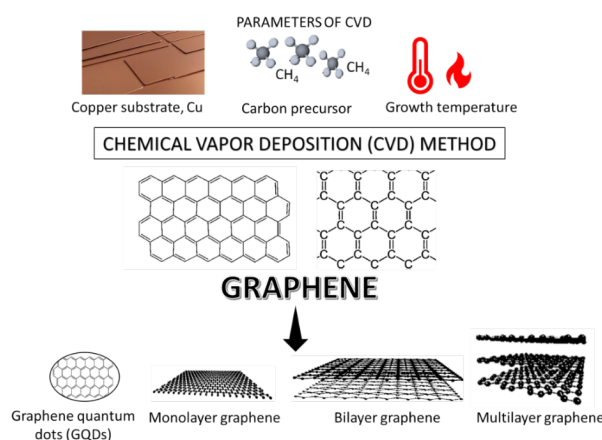


FIGURE 1. Significant parameters involved in the growth of graphene using CVD and types of graphene

METHOD OF ANALYSIS USING SYSTEMATIC LITERATURE REVIEW

For the systematic search, we developed a search strategy to identify relevant literature. This strategy was tailored to two (2) databases which are Scopus and Web of Science (WoS). The keywords search used were the following: “Chemical vapor deposition” AND “Single layer Graphene” OR “monolayer graphene” AND “low temperature” as shown in Figure 2. All searches spanned database inception until 2023 and included journal articles and conference papers that were published in English only.

This paper mostly followed the organized literature review steps recommended by Bokare et al. (Bokare et al. 2021).

From the Preferred Reporting Items for Systematic Reviews and Meta-Analysis (PRISMA) methodology, the identification, screening, eligibility and systematic inclusion of studies were carried out as depicted in Figure

3. The search encompassed scientific articles published between January 2009 and February 2023. A total of 149 articles were initially retrieved, and after eliminating duplicates (45) and manually excluding irrelevant ones (31), the final number was reduced to 73.

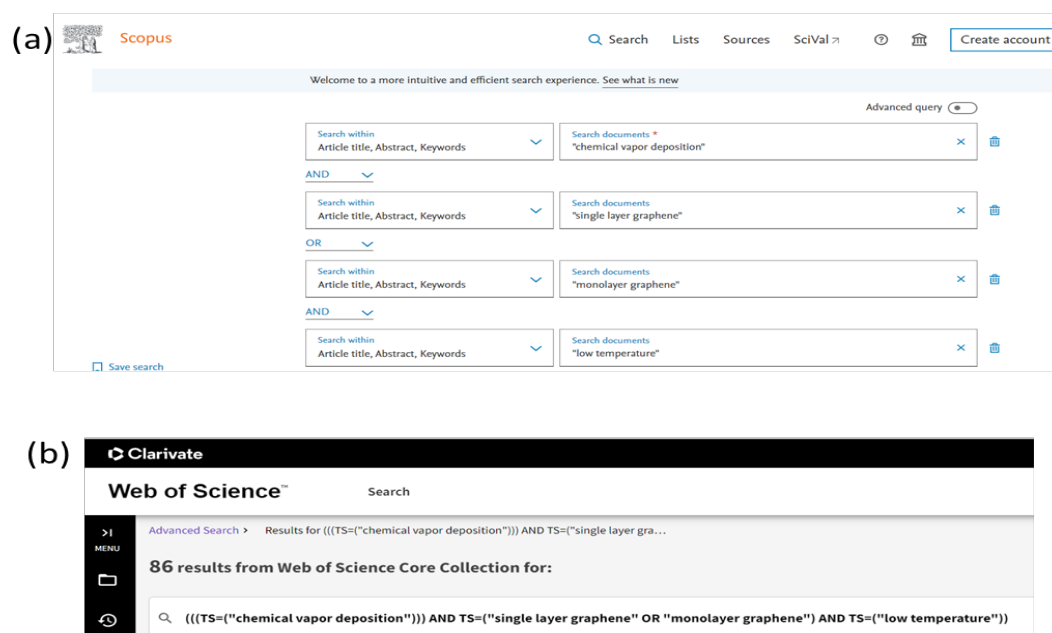


FIGURE 2. Databases search using search engines of (a) Scopus and (b) Web of Science using the following keywords search: "Chemical vapor deposition" AND "Single layer Graphene" OR "monolayer graphene" AND "low temperature".

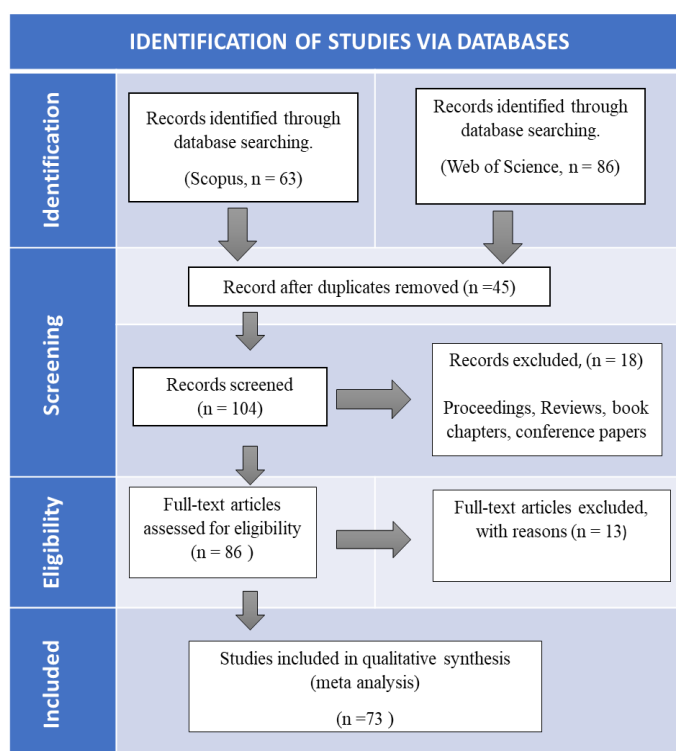


FIGURE 3. PRISMA Framework

RESULT AND INTERPRETATIONS

PUBLICATION YEAR

The publication of the year of study on the synthesis of single layer large area graphene using CVD at a low growth temperature is showing results very differently as shown in Figure 4. The highest number of publications was in

2016. The graphs show that the year 2016 is top-ranked with 16 papers each year. The least publication related to this topic is 2009 with only 1 publication. The articles related to graphene growth through the CVD method are widespread, but the systematic literature reviews on the emphasis on single layer large area graphene using chemical vapor deposition at a low growth temperature have not been published yet.

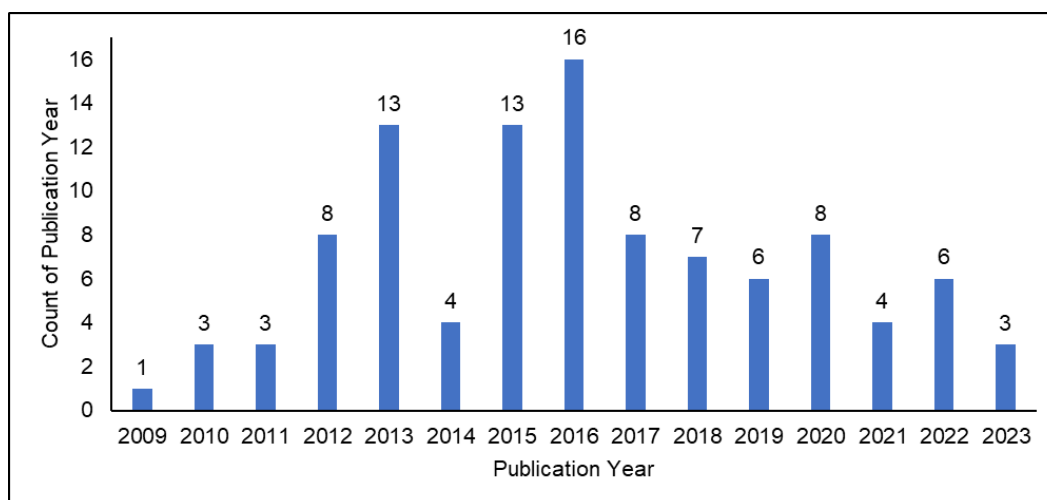


FIGURE 4. Publication per year of Scopus and Web of Science

TYPE OF PUBLICATIONS

The type of published reports on single layer large area graphene using chemical vapor deposition at a low growth temperature shown in Figure 5 consisted of journal articles, book chapters, conference proceedings, review papers, and

conference papers. Journal articles rank the highest, with 88 publications indexed in WoS and Scopus. Review papers are the second most common, totaling 13 publications between 2009 and 2023. Researchers and scientists generally favor publishing research articles to report findings on these topics.

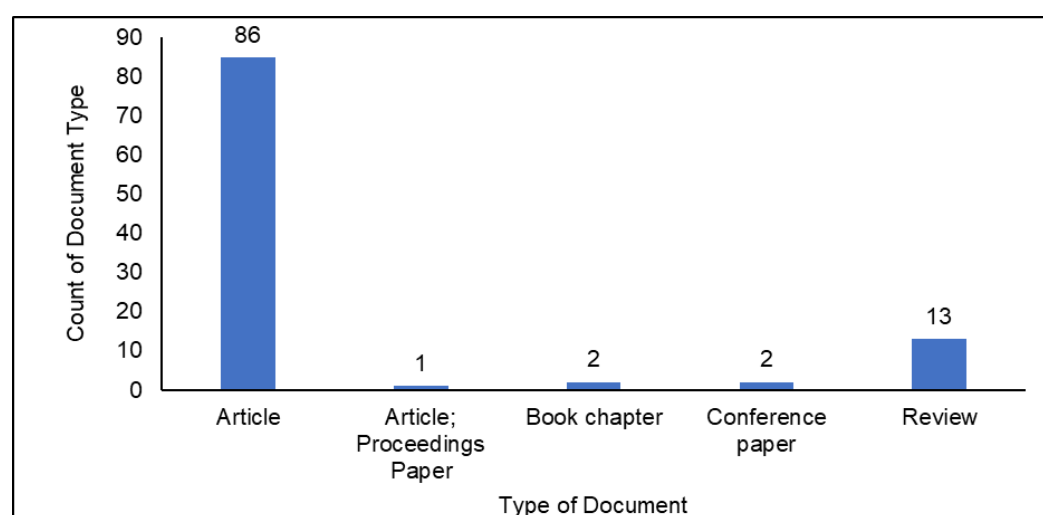


FIGURE 5. Publication categories listed in Web of Science and Scopus

TYPE OF COUNTRY

Figure 6 is a bar chart comparing publication numbers from two sources, Scopus and Web of Science (WoS), across various countries. The chart reveals that China leads in publication counts for both databases. Notably, Scopus shows a considerably higher count than Web of Science for China. The United States follows, with a similar pattern of Scopus having more publications than Web of Science. Other countries like Japan, South Korea, and the United Kingdom also have notable numbers of publications, with Scopus consistently showing more publications than Web of Science for these countries as well. The rest of the countries have fewer publications, with some having very few or no publications in Web of Science, as indicated by the absence or minimal height of the orange bars.

There could be several reasons for the higher publication counts in Scopus compared to Web of Science. Scopus and Web of Science may have different coverage areas, with Scopus potentially including a wider range of publications or sources compared to Web of Science. The databases may have different sources of data, leading to variations in the number of publications included in each database. Scopus and Web of Science may have different indexing policies, leading to discrepancies in the number of publications indexed by each database. Researchers in certain regions or countries may prefer to publish in journals that are more commonly indexed in Scopus than in Web of Science, leading to higher publication counts in Scopus. A time lag in the indexing process between Scopus and Web of Science could lead to differences in the publication counts reported by each database.

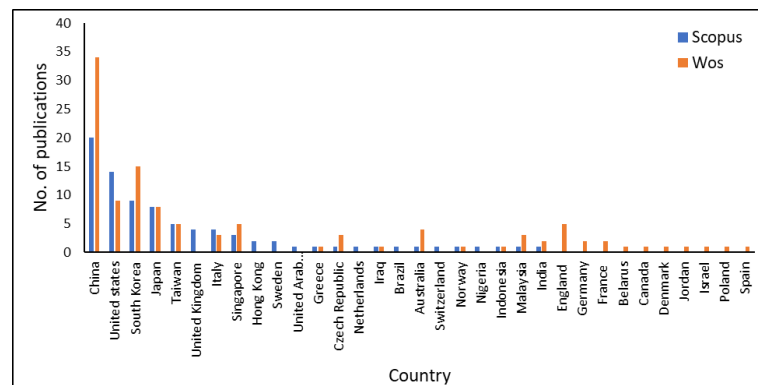


Figure 6. Number of publications of Scopus and Web of Science from country around the world.

TYPE OF LANGUAGE

Figure 7 depicts a bar graph with the type of language publications of Scopus and Web of Science which compares the frequency of three types of languages: Chinese, English, and Korean. The vertical axis represents the total of language occurrences, and the horizontal axis lists the types

of languages. Chinese language is represented by a single bar with a total publication of 2. English has a significantly higher total number of publications, represented by a bar reaching up to 100. Korean is represented by a bar with only one publication. The graph visually illustrates that English is the most frequent language among the three, with Chinese and Korean having much lower counts.

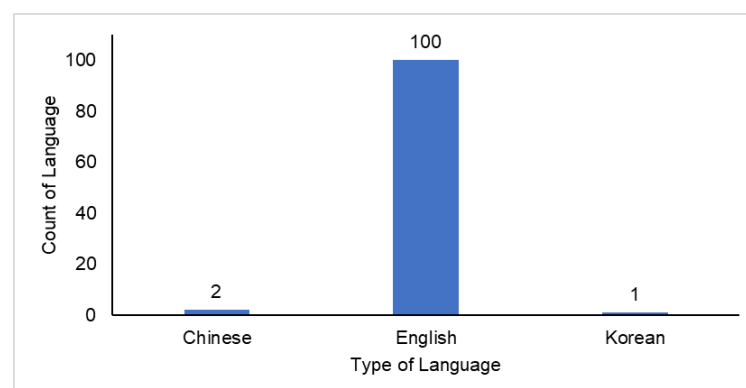


FIGURE 7. Type of language publications of Scopus and Web of Science

CLASSIFICATION

After analysis on an Excel sheet, the studies are categorized based on the process. The study is then segmented into four (4) distinct categories to comprehend the research work conducted. Each category is thoroughly examined to

extract relevant information from previous research. Table 1 represents the summary of included references using keywords: “Chemical vapor deposition” AND “Single layer Graphene” OR “monolayer graphene” AND “low temperature”.

TABLE 1. Summary of included references using keywords: “Chemical vapor deposition” AND “Single layer Graphene” OR “monolayer graphene” AND “low temperature”.

Plasma-assisted method		
Parameters	Remarks	Ref.
SLG, 100-150 °C, Ti-buffered substrate, methane	In this study, a two-step plasma-assisted thermal chemical vapor deposition (PATCVD) technique was developed, allowing the in-situ formation of a titanium (Ti) buffer layer on the target substrate, followed by the low-temperature deposition of monolayer graphene (mGr) on the Ti layer. The Ti buffer layer plays a key role in controlling the mGr growth mechanism.	(Han et al. 2021)
MLG, 100 °C, Benzene/ammonia/argon system, polycrystalline nickel (Ni)	This study demonstrated the ability to produce nitrogen-doped nanocrystalline graphene films with enhanced nitrogen incorporation at temperatures as low as 100 °C, utilizing a cold-wall plasma-assisted CVD system. The resulting films are characterized by highly defective, nitrogen-rich polycrystalline nanodomains.	(Zainal Ariffin et al. 2021)
Hydrogen activates surface-bound carbon, 850°C, Ar and H ₂ gas, Copper foil substrate	Graphene is synthesized uniformly as a continuous, high-quality monolayer film at a power level of 50 W. However, as plasma power increases beyond 50 W, the grain size decreases, which is attributed to the etching effect of hydrogen during the initial growth phase. Higher plasma power seems to not only accelerate growth rates but also leads to excessive layer formation.	(Hong et al. 2019)
1000 °C, H ₂ (30 sccm) and CH ₄ (50 sccm), Copper foil	Low-temperature oxygen plasma treatment enhances the surface roughness of single-layer graphene (SLG) and introduces C-OH and C=O functional groups into the graphene, resulting in weakened sp ² hybridization and increased disorder. After oxygen plasma treatment, both the defect density and grain refinement of the sample increase.	(Zhu, Denget al. 2018)
Vertical Gr flakes, 400-600 °C, not mentioned various types of glasses methane	Adjustment of the growth time achieved a high density of vertical graphene flakes on various types of glasses. The perpendicular electric field induced by the plasma contributes significantly to the vertical growth of graphene flakes.”	(Sun et al. 2015)
SLG, 600 °C, Copper, Methane	This study determined that the optimal distance between the initial plasma stage and the sample is 30 cm. Additionally, it was found that the optimal plasma power is 170 W. The intensity of the 2D peak increased as the plasma power was raised from 0 W to 170 W. This increase was due to the cocatalytic activity of hydrogen that was formed by the decomposition of methane plasma. The hydrogen increased the size of the synthesized graphene’s grains and prevented disorder in its growth.	(Chan et al. 2015)
SLG, 600C, H ₂ , Cu foil, Methane	The findings of the study suggest that maintaining a certain distance between the initial plasma stage and the deposition stage is crucial for the diffusion of plasma to the substrate. Additionally, the research reveals that a higher concentration of hydrogen facilitates the synthesis of a high-quality graphene film, which is indicated by Raman spectra.	(Chan et al. 2013)
SLG, 500 °C, H ₂ , Copper, CH ₄	The growth of monolayer graphene is affected by the catalytic nature of Cu at high temperatures, while radicals generated in the plasma dominate the growth of multilayer graphene and growth at low temperatures.	(Terasawa & Saiki 2012)
Precursors		
Mono to few layers, 500 °C, C ₂ H ₂ and H ₂ , Ni/Cu thin film, Ni/Cu alloy catalysts	This paper presents a graphene synthesis technique that utilizes in situ manipulation of carbon precursors to produce continuous graphene films at lower temperatures on thinner catalyst layers.	(Zietz et al. 2020)

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SLG, 800-1020 °C (high temperatures), Ar, H ₂ , Cu imidazole and melamine	There are three chemical configurations where nitrogen atoms bond to carbon atoms in graphene: (1) by substituting one carbon atom with nitrogen. To synthesize nitrogen-doped graphene, a mixture of imidazole and melamine is used as nitrogen-containing precursors. This mixture enhances both overall nitrogen content and graphitic nitrogen content in the layers of graphene.	(Vishwakarma et al. 2016)
SLG, 300 -400 °C, H ₂ , Copper, PMMA, polystyrene, benzene	At temperatures as low as 400 °C, solid precursors such as PMMA and polystyrene are employed to synthesize monolayer graphene films that can extend over several centimeters. When benzene is used as the hydrocarbon source, high-quality monolayer graphene flakes can be produced at an even lower growth temperature of 300 °C.	(Li et al. 2011)
Catalyst		
Ni and Ni alloy catalysts, 500°C, C ₂ H ₂ precursor	With the addition of 1 wt% Au and 2 wt% Cu, a noticeable reduction in multilayer formation occurs, although some multilayer islands persist. Finally, a Ni alloy catalyst with 1 wt% Au and 1 wt% Cu was used, indicating that most of the graphene film consists of monolayers, though some multilayer regions are still observed.	(Zhan et al. 2020)
Polycrystalline bimetal Ni-Au catalyst, 450 °C, Ar and H ₂ , SiO ₂ /Si wafer, C ₂ H ₂	Ni-Au catalysts were created by remotely pre-alloying the catalyst during electron beam evaporation, resulting in a catalyst that is immediately ready for graphene growth at 450 °C.	(Tracy et al. 2019)
Substrate		
SLG, 300 C, Flexible Polyimide (PI) substrate, methane	Metal electrodes serve as an adhesion layer to facilitate the bonding between the substrate and the copper (Cu) foil. The Cu foil was attached to the polyimide (PI) substrate, enabling the successful in situ growth of a large-area, high-quality monolayer graphene film on the polyimide.	(Hu et al. 2023)
SLG, 500 °C, ethylene, nickel foams	The graphene only deposited on half of the probed surface and interacts with the foam as strongly as on the flat ordered Ni substrate whereas the remaining surface appears nearly free-standing.	(Petrone et al. 2022)
Redistilled Isopropanol vapour, 750, 800 °C and 850 °C, (4, 8, 10, 12, and 20 min), gas Ar and H ₂ , Ni electroplated Cu foil	The absence of a defect intensity (D-band) peak indicates that the spectra exhibiting a 2D-band peak at low substrate temperatures correspond to single-layer graphene, while the intensity of the G-band peak is associated with few-layer graphene.	(Arikpo & Onuu 2020)
Methanol, hydrogen flow, 800°C, Si ₃ N ₄ /SiO ₂ /Si substrates	Liquid Ga was dropped on the substrate as a catalyst and flowed away after growth, achieving graphene on Si ₃ N ₄ (100nm)/ SiO ₂ (100nm)/Si and recovering most of Ga.	(Yang et al. 2019)
1050°C for 30min, Ar/H ₂ flow, Cu substrate hexamethylenetetramine ((CH ₂) ₆ N ₄)	Large, hexagonal-shaped monolayer NG single crystals were grown on copper substrates using HMTA as a solid precursor via CVD.	(Lin et al. 2018)
550 °C -650 °C, H ₂ /C ₂ H ₂ , high resistivity (ρ=3000 Ω·cm, p doped) silicon wafers (111) with native oxide were used as substrates.	At a lower growth temperature of 550 °C, monolayer graphene films display high crystallinity and typically have a grain size of over 300 nm.	(Muñoz et al. 2018)
SLG, 800°C, Treated Ni(111) substrate by Ar ion radiation and annealing, C ₂ H ₄	A monolayer of graphene was grown on a clean Ni (111) substrate using previously published procedures.	(Liu et al. 2016)
SLG, 1600°C, Ar, SiC	A large-scale highly uniform monolayer graphene film was synthesized on the SiC substrate.	(Ohno et al. 2016)
SLG, 1050 °C (high temperatures), H ₂ , Cu, methane	Growth of Graphene (Gr) was carried out on Copper (Cu) at a temperature of 1050 °C. The Gr was then transferred onto a Silicon (Si) substrate, and finally, Gallium Nitride (GaN) nanorods were grown on the Gr at a temperature of 870 °C. The resulting n-GaN NR-graphene hybrid structure showed an improvement in photocurrent, sensitivity, and photoresponsivity. These enhancements were attributed to the photoconductive channel's extremely low carrier trap density.	(Kang et al. 2015)

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SLG, 1000 °C, H ₂ , Cu, CH ₄	The process involves placing a single layer of graphene between two layers of La ₂ /3Sr1/3MnO ₃ (LSMO). The graphene film, grown on a copper foil, is then transferred onto an STO substrate that has an LSMO electrode, using a wet transfer method.	(Li et al. 2014)
SLG, 600 °C, Ar + C ₂ H ₂ gas mixture, (RF power of 100 W), Si, (CuNi, AuNi), Cu and Au on SiO ₂ substrate PMMA	Based on DFT calculations, the incorporation of nickel (Ni) atoms at surface sites can increase the catalytic activity of typically inactive copper (Cu) and gold (Au), enabling graphene growth at lower temperatures compared to pure Cu or Au.	(Woo et al. 2013)
SLG, suspended monolayer graphene (SMG), SF ₆ /C ₄ F ₈ , Cu foils	The differential conductance versus voltage (dI/dV) spectrum of single-layer graphene (SMG) displays a nonlinear local density of states (LDOS) near the Fermi energy (EF), which is in contrast to the V-shaped spectrum of multilayer graphene (MG) on a copper foil. This unexpected behavior is attributed to the electron-electron interaction in SMG when there is no dielectric substrate present.	(Zhao et al. 2013)
SLG, 950 – 1000 °C, H ₂ , Copper, CH ₄	This technique can be readily applied to other metallic substrates, as it relies on the dissipation of heat generated by laser irradiation through the substrate. Consequently, the primary requirement for the substrate is a high thermal conductivity.	(Piazzi et al. 2012)
SLG, 1020 °C, Ar/H ₂ , Copper (Cu) foil	This work highlights an innovative approach for producing highly conductive graphene sheets, utilizing camphor as a natural precursor, followed by nitrogen doping (N-doped graphene) through a low-temperature post-annealing process.	(Vemuri et al. 2022)
SLG, ~350 °C, Tellurium (Te) vapor, H ₂ , Polymethylmethacrylate (PMMA)	This study employed Te-assisted, space-confined graphene growth using the CVD method within a custom face-to-face SiO ₂ reaction chamber. This straightforward and versatile growth strategy has potential for scaling up multilayer graphene production and for the direct integration of graphene-based functional electronics in the future.	(Liu et al. 2021)
SLG, Twisted bilayer graphene (tBLG), 1020 °C H ₂ , methane, Cu foil (tBLG)	This paper is about fabricating a SLG-tBLG junction device.	(Rui et al. 2020)
Pyridine as the sole source of carbon and nitrogen, 1000 °C, gas Ar and H ₂ , Quartz, rock crystal, and sapphire pyridine	Single graphitic-N doping leads to more prominent n-type transport behaviors, owing to the absence of p-type pyrrolic-N and pyridinic-N.	(Yang et al. 2020)
No catalyst, 625 C, H ₂ , CH ₄ and H ₂ , Ge surface (2cm)	It is shown that large-area, uniform VAGNAs terminated with high-quality single-layer graphene can be directly grown on the Ge surface by PECVD without catalyst at low temperature.	(Al-Hagri et al. 2019)
Polymethyl methacrylate (PMMA), (temperature range 20 to 35 K)	At low temperatures, when rare gases such as Ar, Kr, and Xe are adsorbed on suspended monolayers of pristine graphene, it causes compressive strains on the graphene layer. The compression effect is more pronounced for the Xe/PG system as it has higher polarizability as compared to Kr and Ar systems.	(Cunha et al. 2018)
SLG, 300°C, Cu foils, Liquid pyridine	Low-defect monolayer graphene domains were successfully grown on a copper (Cu) foil using the CVD method with liquid pyridine at a temperature of 300 °C. In previous attempts, only high-defect density graphene domains were obtained. This discovery marks the first time low-defect monolayer graphene films have been synthesized using liquid pyridine.	(Kang & Lee 2017)
SLG, 1000°C, H ₂ , Treated Cu foils by annealing in H ₂ , CH ₄	High temperature growth (1000°C)	(Verhagen et al. 2017)
SLG, 750°C, H ₂ , Treated Co-Cu by annealing in H ₂ CH ₄	The addition of around 1 atomic percent (at.%) copper (Cu) to cobalt (Co) leads to full coverage of single-layer graphene growth under the specified conditions.	(Sugime et al. 2017)
SLG, 1000°C, H ₂ / Ar, Treated W/Mo on Cu foils by annealing in Ar, CH ₄	The exterior of the copper catalyst enclosure is coated with sputtered tungsten (W) and molybdenum (Mo) metal, which facilitates the growth of sub-centimeter-scale single-crystal graphene domains and completely eliminates multilayer growth.	(Luo et al. 2017)

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SLG, 1050°C, N ₂ , Treated Cu foils by electrochemically polishing and annealing in H ₂ /N ₂ , C ₁₀ H ₂₂	This paper presents the findings of temperature-dependent Raman studies on anharmonic phonon-phonon interactions in supported graphene.	(Kolesov et al. 2017)
SLG, 600°C, Ar, Treated metal-modified Ni foils by annealing in H ₂ , C ₂ H ₄	This study demonstrates that the deposition of zinc (Zn) and bismuth (Bi) metals can alter the surface of nickel, preventing the uncontrolled growth of multiple graphene layers. This modification enables the production of high-quality single-layer graphene through chemical vapor deposition (CVD) at a temperature of 600 °C with a minimal number of defects, representing a significant improvement over bare nickel. In contrast, copper (Cu) and molybdenum (Mo) inhibit graphene growth.	(Giamini et al. 2016)
SLG, 1060°C, H ₂ /Ar, Treated Cu foils by electropolishing and annealing in H ₂ or Ar, CH ₄	Large crystals of CVD graphene were synthesized on a copper foil using a low-pressure chemical vapor deposition (CVD) technique.	(Xiang et al. 2016)
BLG (atmospheric pressure), SLG (low pressure), 800°C, 900°C, 1000°C, Ar/ H ₂ , Treated Cu foils by acetic acid, Benzene, Naphthalene, Anthracene	At atmospheric pressure, graphene obtained is mainly bilayer, while lower pressures produce predominantly single-layer graphene (SLG).	(Shanmugam et al. 2016)
SLG, 230-600, Cu, pentachloropyridine	There are three nitrogen (N) bonding configurations in graphene (Gr): graphitic N, pyridinic N, and pyrrolic N. The type of configuration is determined by the synthesis temperature. Graphitic N is obtained at 200-300°C, while pyrrolic N is obtained at 400-600°C. Graphitic N-dominated graphene exhibits a strong n-type doping effect, while pyrrolic N-dominated graphene shows a weak n-type doping effect.	(Zhang et al. 2016)
SL GQDs, 1000 °C (high temperatures), H ₂ , Cu methane + sonication	GQDs can be produced by using various techniques, such as sonication of defective CVD graphene. These GQDs are mostly made up of single-layered graphene and have a diameter of less than 8 nm, with roughly round shapes. They also have a strong blue luminescence. When exposed to 270 nm light, the GQD/IGZO phototransistor exhibits a photocurrent that is 10 times larger than IGZ	(Zhu et al. 2016)
SLG-multilayers, 1015 °C (high temperatures), H ₂ , Ar, Cu, Camphor, C10H16O	To create carbon and nitrogen dopants, a mixture of camphor and melamine powders was used. The ratio of the camphor and melamine mixture was varied. X-ray photoelectron spectroscopy (XPS) showed a high percentage of pyridinic nitrogen dopant. Several grain orientations were observed, each with an individual grain size of more than 5 µm	(Shinde et al. 2016)
SLG, 500 °C, H ₂ , Cu, Cu/Ni, Methane	LT growth was assisted with the use of graphite plate holder and a small closed-end tube. The graphite plate located below the Cu foil acts as an additional C source while the small closed-end tube increases the concentration of hydrocarbon source. The Gr coverage at 500 °C is almost 100%. The back gate FET mobility is 1-2k cm ² /Vs.	(Chen, Medina et al. 2015)
SLG, 100-300 °C, Ar, H ₂ , Cu, benzene	Graphene was grown at 300°C using APCVD and found that the electrical properties of the graphene were similar to those grown at 1000°C. The pumping and purging cycles were applied to remove oxygen and annealed the Cu foil at 1000°C for 30 minutes before growth. The graphene coverage reduced from 100% to 85% when the temperature was decreased from 300 to 100°C. The growth process involves benzene adsorption, dehydrogenation, and coalescence, facilitated by the high adsorption energy of benzene on Cu.	(Jang et al. 2015)
SLG, 1500 °C (high temperatures), (under Si background pressure), 4H-SiC	Graphene was obtained by thermal decomposition of the SiC layer, followed by sublimation of Si at high temperature to form a C-buffer layer. The C-buffer layer was then transformed to graphene via hydrogen intercalation.	(Hassan et al. 2015)

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SLG, 600 °C, Ar, H ₂ , Fe, Co, Ni + Ga pellet on metal foil, methane	Put Gallium (Ga) on a Nickel (Ni) foil. During growth, liquid Ga forms a stable antiperovskite structure, GaCNi ₃ , which blocks excess carbon (C) inside the Ni foil to prevent it from segregating to the surface during cooling. This enables the production of a single-layer graphene (SLG).	(Chen, Konget al. 2015)
SLG, 750 °C, Ar, H ₂ , Cu, methane	Copper (111) surface is more favorable for growth than other low-index copper facets, namely Copper (100) and Copper (110). When grown on Copper (111), graphene grows continuously, while on other facets, only flakes are obtained. To achieve fewer defects, it is recommended to grow graphene at a slower growth rate or to use a low CH ₄ partial pressure.	(Jacobberger et al. 2015)
SLG, 300 - 500 °C. Single layer graphene (180 °C), Copper foil, Chlorobenzene in PMMA	Grow a single layer using matrix-assisted approach (PMMA) from liquid-based carbon sources (Chlorobenzene)	(Sulaiman et al. 2016)
SLG, large-area and highly nitrogen-doped, graphene (NG), 1000 °C, H ₂ , P ₄ VP/Cu foil, PMMA	This work achieved a nitrogen content of 6.37% in a large area of more than 80% single-layer NG membrane.	(He et al. 2015)
SLG, 300 - 500 °C, Platinum (methyl-form and ketone-form)	Growth nitrogen was doped onto graphene, and the graphene growth occurred at 500°C due to the production of benzene or its fragment.	(Kato et al. 2015)
MLG, 1000 °C, H ₂ , Platinum, PMMA	The process of forming multilayer graphene involves growing successive layers underneath the pre-existing ones. With the right adjustments made to the growth parameters, it is possible to obtain monolayer graphene of high quality. This can be achieved by using H ₂ , quench cooling, a continuous stream of hydrocarbon flow (albeit small), and maintaining an intermediate growth temperature. It is worth mentioning that monolayer graphene, catalyzed with Pt, exhibits the quantum Hall effect.	(Sun et al. 2014)
SLG, 700-800 °C, H ₂ , Cu, methane	The study indicates that there are additional challenges when working with copper thin films as compared to copper foils, especially when it comes to maintaining film stability at high temperatures. To prevent agglomeration and film rupture, it is necessary to monitor the in-situ dewetting dynamics of the catalyst.	(Croin et al. 2014)
SLG, 1000 °C, H ₂ , Cu foil, CH ₄ /PMMA	In this study, we utilized APCVD to synthesize graphene as an active layer and successfully achieved GFETs with high electrical performance and low voltage. Notably, our GFETs showed an impressive on/off current ratio, which was enhanced up to 8.	(Zhou et al. 2014)
SLG, 800 °C, H ₂ , Cu, Methane	When carbon clusters become saturated, they can turn into flawed graphene that has irregular bumps and holes. These vacancy defects can only be fixed by heating them to 800°C in the presence of methane, which results in the formation of vacancy-free monolayer graphene on Cu (111).	(Niu et al. 2013)
SLG, 1000 °C, H ₂ , Cu film, CH ₄	In CVD-grown graphene, phthalocyanine molecules self-organize into well-ordered structures that extend smoothly across the steps of the copper film. The boundaries observed within the molecular arrays are distinct and correspond to domain boundaries within the graphene. Scanning tunneling microscopy (STM) results reveal that CVD graphene serves as an excellent template for molecular assembly, allowing detailed examination of domain structures in CVD graphene through these arrays.	(Ogawa et al. 2013)
SLG, 400 °C, H ₂ , Copper foil, naphthalene	The findings of this research indicate that the mobility of graphene, which was manufactured at a temperature of 400°C, is approximately 682 cm ² ·V ⁻¹ ·s ⁻¹ , demonstrating promising electrical transport characteristics	(Wu, Shen et al. 2013)

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SLG, 972 °C, H ₂ , Ar, Copper, CH ₄	The performance of high-temperature barrier applications is restricted due to degradation. However, graphene passivates much better than what was reported earlier, indicating improved performance at low temperatures.	(Roy & Arnold 2013)
SLG, 1000 °C, H ₂ , Ar, Copper, CH ₄	Low-temperature growth of this gas reduces etching and catalytic hydrogenation, resulting in continuous, undamaged graphene-CNT hybrids.	(Kumar et al. 2013)
SLG, 300 °C, H ₂ , Ar, Copper, Naphthalene	Graphene fabricated at 400 °C using the seed-assisted process achieved a carrier mobility of approximately 912 cm ² V ⁻¹ s ⁻¹ .	(Wu, Ding et al. 2013)
SLG partially MLG, 800 °C, CH ₄ , H ₂ , Nickel, CH ₄	The graphene area coverage is limited to 100 μm ² and the number of graphene layers is significantly influenced by the rates of hydrogen and methane flow. An observed correlation shows that an increase in the methane flow rate leads to a higher density of growth for single-layer graphene.	(Hsu et al. 2012)
SLG partially MLG, 600 °C, Ni-SiO ₂ , C ₂ H ₂ , C ₆ H ₆	To achieve optimized growth, it is necessary to saturate the catalyst with carbon only in the vicinity of the growth surface. This can be achieved by ensuring that the CVD exposure conditions are matched to the thickness of the catalyst.	(Weatherup et al. 2012)
SLG, 600 °C, H ₂ , Copper, Toluene	Under the same growth conditions, electropolished Cu foil has a higher coverage of graphene on its surface compared to unelectropolished Cu foil. The optical transmittance of the produced graphene was measured to be 97.33% at 550 nm, confirming that it was a monolayer. Additionally, the sheet resistance of the graphene was approximately 8.02 × 10 ³ Ω.	(Zhang et al. 2012)
SLG, 550 - 600 °C, Nickel, C ₂ H ₄	Carbon diffusion into the bulk limits the surface growth rate above ~550 °C temperature, while at temperatures below ~500 °C, a competing surface carbide phase impedes graphene formation.	(Addou et al. 2012)
SLG, 800 – 1000 °C, Ar, H ₂ , Copper/Al ₂ O ₃ and Copper/SiO ₂	A limited supply of carbon source, combined with a Cu(111) crystallographic plane and a relatively high temperature, can facilitate the nucleation and growth of graphene. Conversely, when there is an ample supply of carbon, carbon supersaturation seems to be the key factor driving graphene formation, irrespective of temperature or crystallographic characteristics.	(Hu et al. 2012)
SLG, 1000 °C, H ₂ , Ar, Copper and Nickel, CH ₄	The single-layer graphene films have an average shear modulus of 280 GPa, which is about five times higher than other materials.	(Liu et al. 2012)
SLG, 450 – 750 °C, H ₂ , Nickel, CH ₄	By implementing MPCVD, the researchers managed to attain a high level of transparency, reaching 89%. Additionally, they were able to produce graphene films with low sheet resistances ranging from 590 to 1855 Ω/sq using considerably low synthesis temperatures. Based on these results, this method appears suitable for large-scale production of electronic devices that rely on graphene films.	(Kim et al. 2011)
SLG, 650 °C, H ₂ , Ar, Copper, C ₂ H ₂	The growth of graphene is prevented by the reduced influence of the Cu surface, which enables the fabrication of a large, homogeneous, single layer.	(Jeon et al. 2011)
SLG, 650 °C, H ₂ , Copper, CH ₄	In a fully dehydrogenated graphene-like film, the characteristic quantum Hall effect of monolayer graphene can be observed, allowing the identification of four fully quantized half-integer plateaus.	(Wang et al. 2010)
SLG, 800 - 1000 °C, H ₂ /Ar, Copper, Nickel, CH ₄ , C ₂ H ₂ , PMMA	To alter the Fermi level and bandgap of graphene, it is necessary to accomplish patterned doping.	(Sun et al. 2010)
SLG, 1000 °C, H ₂ /Ar, Copper, CH ₄	The transferable CVD graphene that we have exhibits behaviors that are inherent to graphene, including but not limited to a half-integer quantum Hall effect. Additionally, it displays exceptional electronic properties that can be characterized by ambipolar field effect, carrier mobility, and phase coherence.	(Cao et al. 2010)
SLG, 1000 – 1100 °C, Ar, Nickel, CH ₄	The films demonstrate the half-integer quantum Hall effect across a broad temperature range extending from 2 K to 200 K. This suggests that the graphite produced through the cold-wall CVD method possesses a quality that is comparable to that of highly oriented pyrolytic graphite (HOPG).	(Cai et al. 2009)

DISCUSSION

The numbers of databases obtained from Scopus and WoS is different because Scopus covers a wider range of sources, including journals, conference proceedings, book chapters, and patents while WoS is primarily focuses on peer-reviewed academic journals, with a strong emphasis on Social Sciences Citation Index (SSCI) and Science Citation Index (SCI). Scopus also offers a wider range of features and tools for multidisciplinary research while WoS has a stronger focus on citation analysis and bibliometrics, making it a valuable tool for researchers interested in impact assessment. The different results obtained are 23 articles with 45 duplicated articles from both search engines.

From the SLR, the chosen keywords are “Chemical vapor deposition” AND “Single layer Graphene” OR “monolayer graphene” AND “low temperature”. The focus of this analysis is on the “single layer” or “monolayer” graphene and excludes irrelevant studies on multi-layer or bulk graphene, keeping the search relevant. The including term for both “single layer” and “monolayer” ensures capturing studies using either term, despite potential subtle differences. The search is restricted to “low temperature” and is aligned to identify low-temperature synthesis graphene methods. The keyword “Chemical vapor deposition” is chosen instead of synthesis because the CVD method is established as a synthesis growth method for graphene and the keywords are specifically mentioned for available research studies. A Google search of synthesis graphene shows about 1.8 million related studies that require the SLR method to narrow down the keywords focus with relevant studies.

From over 149 publications, the trends and breakthroughs in low-temperature CVD methods have been identified for graphene synthesis. The key findings include (1) identification of optimal catalysts, precursors, and growth parameters for achieving large-area, high-quality graphene at temperatures significantly lower than traditional methods and (2) analysis of emerging techniques like plasma-assisted CVD and roll-to-roll processing for scalability and cost-effectiveness, that represented in Table 1. From Table 1, the lowest growth temperature that can be achieved is in the range of 100 to 300 °C when using plasma-enhanced CVD such as two-step plasma-assisted thermal chemical vapor deposition (PATCVD), in-situ plasma-assisted CVD, cold-wall plasma CVD, and many types of PECVD. Han et al. (Han et al. 2021) utilizing PATCVD, graphene growth is achieved at a temperature of 100 °C, allowing for the in-situ deposition of a titanium buffer (Ti-buffer) layer onto a target substrate.

This is followed by the low-temperature growth of monolayer graphene (mGr) on the Ti layer, where the Ti-buffer controls the mGr growth mechanism (Han et al. 2021). Additionally, another study demonstrated that nitrogen-doped nanocrystalline graphene films with increased nitrogen doping can be synthesized at temperatures as low as 100 °C using a cold-wall, plasma-assisted chemical vapor deposition (CVD) system. The films generated consist of highly defective polycrystalline nanodomains containing nitrogen. Jang et al. (Jang et al. 2015) found that growth at low temperatures can be achieved using benzene as a precursor because benzene has high adsorption energy on copper as a substrate. However, the graphene growth coverage was reduced from 100 % to 85 % when temperature was reduced from 300 to 100 °C. They also obtained the electrical properties of graphene grown at 300 °C are as good as at 1000 °C.

Carbon precursor is also another important parameter involved in low growth temperature. Typically, the conventional approach involves substituting methane with either acetylene or ethylene. Given that the pyrolysis temperatures for acetylene and ethylene are 500 °C and 798 °C, respectively, both temperatures have a lower range than methane which is 1100 – 1200 °C due to the dehydrogenation reaction that can take place at 600 °C. This results in the formation of graphene and enables the attainment of a lower growth temperature. The graphene growth step was engineered by Jang et al. (Jang et al. 2015) to occur at a condition of 300 °C and ambient pressure, devoid of any lingering oxidizing species. Before initiating the growth process, they incorporated pumping and purging cycles to ensure the continuous formation of monolayer graphene films, achieving 100% surface coverage and excellent quality (Jang et al. 2015). Meanwhile, compared to the other studies of lower growth temperature, the growth coverage area is still not achievable and higher tendency to form a mixture of graphene layer with higher film defects.

For qualitative justification, this study compared the performance of common gaseous precursors. Methane (CH₄), due to its high C-H bond dissociation energy (~439 kJ/mol), typically requires higher temperatures to maintain low defect levels (ID/IG < 0.1) and ineffective at low temperatures without plasma assistance (Yang et al. 2024). In contrast, Acetylene (C₂H₂) offers a significantly higher growth rate at lower temperatures due to its lower decomposition energy, but it often leads to a higher nucleation density, which can result in grain boundaries and increased defect levels (ID/IG > 0.4) if not strictly controlled. However, another study suggest that lowering temperatures to 400°C or below typically results in a marked increase in the D-band intensity, indicating higher

defect density and reduced domain size compared to conventional 1000 °C growth (Liu et al. 2026).

While benzene offers advantages as a carbon precursor in low-temperature CVD graphene growth such as enabling continuous films at 100-300 °C under ambient pressure due to its efficient decomposition and reduced multilayer formation, its drawbacks warrant careful consideration. Safety concerns are prominent as benzene is a known human carcinogen (classified IARC Group 1), posing inhalation and handling risks in lab-to-industry scaling. Moreover, its often requiring specialized ventilation and PPE beyond standard methane or ethylene processes. Additionally, film quality remains inconsistent, with studies reporting variable domain sizes, higher defect densities (ID/IG ~0.2-0.4 from incomplete pyrolysis), and oxygen sensitivity leading to pinholes or incomplete coverage on

large areas. These is important during handling the benzene as it is controlled chemical in research activity while the alternative approach on using benzene derivatives.

Table 2 compares the synthesis parameters on temperature, catalyst, and precursor affect the quality of CVD-grown graphene. It shows that while traditional methods require high temperatures (850 -1000 °C) with solid copper and gas precursors to achieve high-quality graphene, recent advancements have lowered this heat requirement. By using modified catalysts (like liquid copper) or alternative solid precursors, researchers can grow low-defect, highly uniform graphene at much lower temperatures (300-600 °C). Overall, the table highlights that achieving high mobility and low defects requires carefully balancing these three parameters.

TABLE 2. Comparison of graphene synthesis parameters and resulting quality

Reference	Title	Temperature	Catalyst	Precursor	Quality Metrics (Defects, Mobility, Uniformity)
(Gürsoy et al. 2022)	Uniform deposition of large-area graphene films on copper using low-pressure chemical vapor deposition technique	870 °C	Solid Cu (Foil)	Hexane	High uniformity (89.6%), single-layer with low defects. Quality decreases as temperature increases beyond optimal.
(Prekodravac et al. 2019)	Graphene quantum dots and fullerenol as new carbon sources for single-layer and bi-layer graphene synthesis by rapid thermal annealing method	600 °C – 1000 °C	Solid Cu	Graphene Quantum Dots, Fullerenol	Quality is highly dependent on precursor concentration; Graphene quantum dots yielded better crystallization/ quality than fullerenol.
(Tao et al. 2012)	Synthesis of high quality monolayer graphene at reduced temperature on hydrogen-enriched evaporated copper (111) films	< 900 °C	Solid Cu (Evaporated films)	Methane (Hydrogen-free)	Negligible defects ($\geq 95\%$ defect-free over 200 $\mu\text{m} \times 200 \mu\text{m}$ areas). High-quality monolayer synthesis achieved at reduced temperatures.
(Romaniak et al. 2023)	Assessing the quality of large-area monolayer graphene grown on liquid copper for size-selective ionic/molecular membrane separations	> 1080 °C	Liquid Cu	Gaseous Hydrocarbon	Highly mobile domains on liquid copper self-assemble to minimize grain boundary defects compared to solid substrates.
(Riikonen et al. 2013)	Photo-thermal chemical vapor deposition of graphene on copper	950 °C	Solid Cu	Methane	High carrier mobility with low defects due to the specialized photo-thermal application during CVD.
(Qian et al. 2022)	Transfer-Free CVD Growth of High-Quality Wafer-Scale Graphene at 300 °C for Device Mass Fabrication	300 °C	Solid Cu	Hydrocarbon Gas	Growth temperature drastically reduced while maintaining quality suitable for mass device fabrication.
(Laveti et al. 2018)	Development of Cu Substrate Preparation Techniques for Graphene Synthesis	850 °C	Solid Cu (Annealed)	Hydrocarbon Gas	Proper substrate preparation (reducing roughness) directly prevents defects from forming in the graphene film during growth.

CHALLENGES AND LIMITATIONS ON LOW GROWTH TEMPERATURE FOR LARGE AREA GRAPHENE SYNTHESIS

The challenges and limitations face during low growth temperatures is significant to achieve large area of graphene compare to established method on higher growth temperature during CVD process. For example, the typically nanocrystalline graphene films will grow rather than continuous single-layer sheets when using nitrogen. This is because the incorporation of nitrogen atoms into the hexagonal lattice creates pyrrolic or pyridinic defects. These nitrogen centers disrupt the sp^2 hybridization, leading to localized buckling and reduced mechanical strength. Meanwhile N-doping introduces n-type carrier density, the high degree of disorder in 100°C nanocrystalline films leads to charge carrier scattering. Consequently, while the film is graphene-like its mobility is often several orders of magnitude lower than pristine graphene, behaving more like a disordered semiconductor than a high-performance conductor.

The other significant challenge identified across the literature studies is the inverse relationship between growth temperature and defect density. In traditional thermal CVD, temperatures near 1000°C provide the necessary activation energy for surface catalyst diffusion and high-quality crystallization. At lower temperatures, particularly below 400°C, the I_D/I_G ratios (measured via Raman spectroscopy) often show a marked increase. This indicates that while a monolayer is achieved, it is frequently characterized by smaller grain sizes and higher concentrations of sp^3 defects, which significantly bottleneck carrier mobility. Moreover, at low temperatures, the kinetic energy available for carbon atom rearrangement is significantly diminished. This leads to a fundamental trade-off, while PECVD enables deposition on flexible substrates, it frequently results in nanocrystalline domains rather than large-crystal lattices. Our synthesis of the 73 studies reveals that as growth temperature decreases, the defect density (n_D) increases exponentially, often evidenced by a prominent D' peak in Raman spectra. Furthermore, achieving large-area uniformity remains a bottleneck where low-temperature processes are highly sensitive to local plasma fluctuations, which cause variations in sheet resistance across the substrate, making wafer-scale reproducibility challenging compared to high-temperature thermal CVD.

Another challenges on uniformity of the growth graphene at low temperatures (<500°C) where the phenomena involves on slow carbon precursor decomposition, leading to nucleation of small, isolated graphene domains rather than seamless films, resulting in patchy coverage over large areas. Techniques like molten

gallium catalysts or plasma enhancement improve edge attachment but struggle with consistent precursor supply across wafers, often causing thickness variations. The uniformity will drops below 300°C without advanced furnace designs besides the pressure control and H_2 ratio still significant. For batch processes, its limit wafer-scale output at low temperature, as slow kinetics demand longer times (hours vs. minutes at 1000°C) which increase the production costs. Transfer-free approaches, like Cu foil lamination on insulators, enable 300°C wafer-scale films ($ID/IG=0.08$) but scale poorly without gradient furnaces or CFD-optimized flows.

For different CVD techniques, low-temperature CVD variants for large-area graphene growth involve distinct trade-offs: (1) Thermal CVD on Cu foil at 300-500°C enables wafer-scale uniformity with low defect density ($ID/IG=0.08$) but suffers from slow growth rates and H_2 sensitivity (Qian et al. 2022); (2) Plasma-enhanced CVD at 380-500°C offers good coverage on irregular substrates, introduces medium defects ($ID/IG=0.1-0.3$) from ion damage alongside high equipment costs (Jang et al. 2015); (3) Molten Ga-catalyzed methods at 50-100°C achieve low edge defects through interface confinement but limit scalability to droplet sizes and require catalyst removal; (4) Benzene APCVD at 100-300°C produces continuous films with low-medium defects, trading off precursor toxicity and O_2 sensitivity for ambient pressure compatibility (Jang et al. 2015).

CONCLUSION

This SLR method assisted in refinding the related articles that focus on graphene synthesis using CVD by improving with plasma-assisted heating medium for lower growth temperature with a larger coverage area of single-layer graphene. Larger coverage growth areas facilitate the creation of continuous and uniform graphene layers, allowing for more efficient and reliable applications in various fields such as electronics, energy storage, aerospace, defence and nanotechnology. Meanwhile, minimizing the defects by improving the graphene film quality is essential to preserve the exceptional mechanical, electrical, and thermal properties of graphene. Defects, such as vacancies or structural irregularities, can significantly impact the material's structural integrity and compromise its performance in practical applications. In addition, the utilization of benzene as a carbon precursor also gives inputs that growth at lower temperatures can be achieved due to the lower decomposition temperature of the precursor. In conclusion, the current focus on achieving larger coverage growth areas with minimal defects aims

to unlock the full potential of graphene for advanced technologies and industrial applications. These advancements in low-temperature synthesis methods not only contribute to the economic viability of graphene production but also open avenues for integrating graphene into diverse applications, including flexible electronics and energy storage devices.

ACKNOWLEDGEMENT

The authors gratefully acknowledge the financial support provided by University Research Group (Electronic Component Materials and Packaging (EMAP) – [UKMKPU-2023-022]) Universiti Kebangsaan Malaysia.

DECLARATION OF COMPETING INTEREST

None.

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