

## Review of Chemical Vapour Deposition (CVD) Techniques

Anurag Prachis Bodra<sup>a</sup> and Won-Chun Oh\*

<sup>a</sup>Department of Advanced Materials Science & Engineering, Hanseo University, Chungnam 356-706, Korea

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**Abstract:** The purpose of this paper is to highlight CVD techniques and methods used in the synthesis of graphene. Graphene can be synthesized using this technique and by the help of carbon precursors that can either be in liquid form, gaseous form or solid form. Graphene produced by this technique is of the highest possible quality and gives the opportunity to grow large surface area graphene on the desired substrate. Several techniques involving CVD have been used over the years and as such the method of CVD keeps getting fine tuned and refined.

**Key words:** Graphene; Carbon Precursor; CVD; TEM; Raman.

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### 1. Introduction

Graphene is a two-dimensional allotrope of carbon which is made up of a one-atom-thick planar sheet of  $sp^2$ -bonded carbon atoms densely packed in a honeycomb crystal lattice. It has been attracting a lot of attention because of its excellent structural and electrical properties [1], such as high values of Young's modulus ( $\sim 1100$  GPa) [2], fracture strength (125 GPa) [2], thermal conductivity ( $\sim 5000$  W  $m^{-1}$  K $^{-1}$ ) [3], mobility of charge carriers (20,000  $cm^2$  V $^{-1}$ s $^{-1}$ ) [4] and specific surface area (2630  $m^2$  g $^{-1}$ ) [5]. Moreover, the high electron mobility and tuneable band gap of graphene makes it a potential material for electronics and sensing applications. It is well known that graphene is a semimetal where the charge carriers behave as Dirac fermions [1], which gives rise to extraordinary effects such as mobilities up to  $2 \times 10^5$   $cm^2$ /V s [4], ballistic transport distance of up to a micron at room temperature [6], half-integer quantum Hall effect [7] and absorption of only 2.3% of visible light [8]. These properties make graphene be widely applied in high frequency electronics [9], high performance chemical- and bio-sensors, transparent conductors, optoelectronics, flexible flat panel displays [10] etc. These applications urge establishing synthetic routes to obtain graphene films of large area and high quality (crystallinity). Several methods are used to produce graphene, e.g. chemical synthesized graphene, silicon sublimation from SiC crystals [11-13], and chemical vapor deposition (CVD) using transition metal catalysts [14-17]. The CVD method is the most suitable among them for commercial graphene production, as it can create large area graphene films with low cost. Copper

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\*Corresponding author: [wc\\_oh@hanseo.ac.kr](mailto:wc_oh@hanseo.ac.kr)

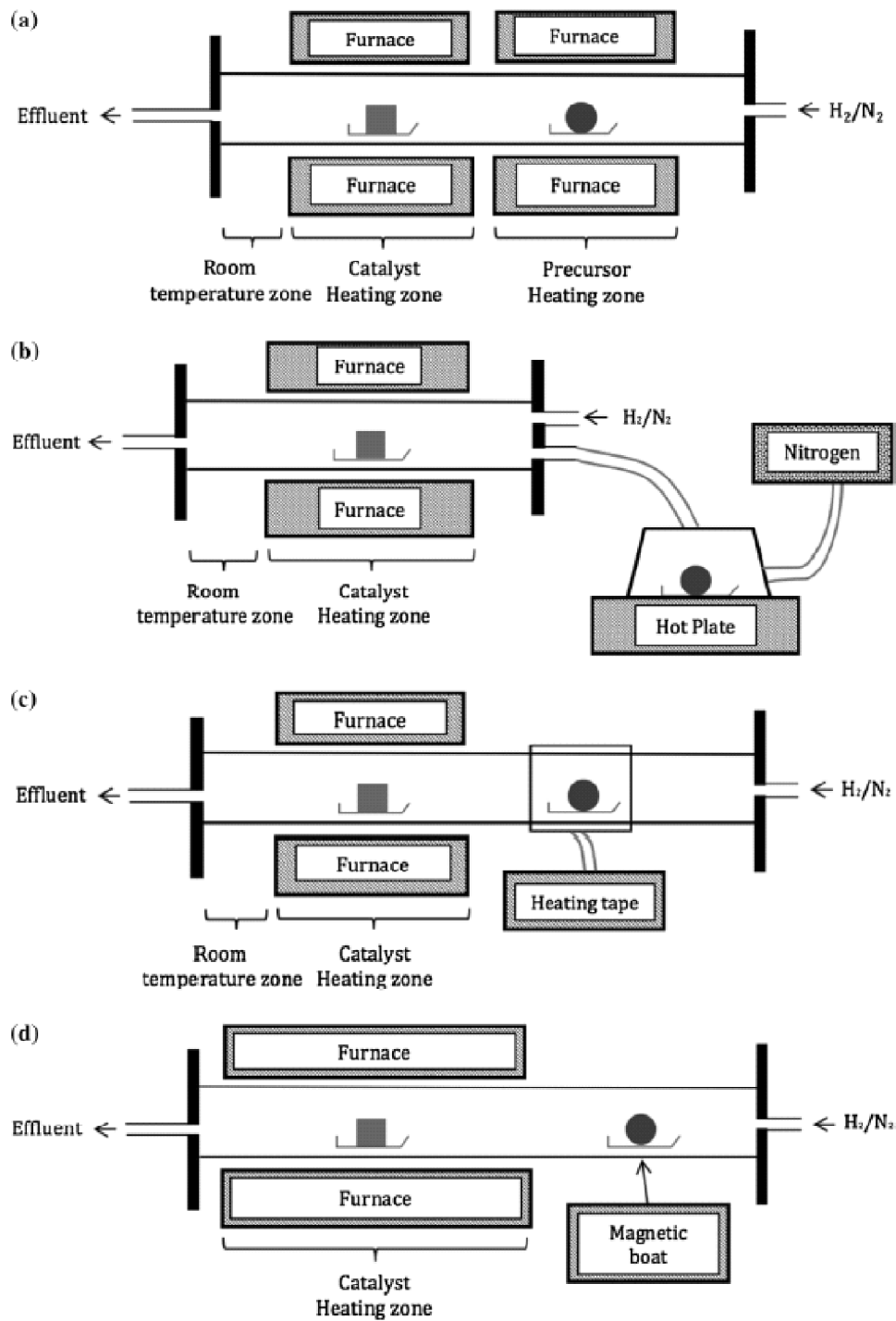
system is the most promising, as this system produces monolayer/multilayer graphene selectively, while many of the other transition metal systems create multilayer graphene films with un-uniform thickness which is intractable for device application [18-20 & 16]. Current key issues are how to form a single-crystalline graphene film by Cu-CVD, because a CVD graphene film is usually a patchwork of independently grown graphene islands taking random orientations [21-24]. A possible countermeasure to this problem is to carefully control carbon atom nucleation and graphene nucleus growth by process parameters of CVD, such as temperature, pressure, gas-flow, etc. [25, 26].

## 2. Experimental Details

In general, for any CVD technique following components are always used; (a) CVD furnace with inlets and outlets for gases, (b) piping for the supply of gases into the furnace, (c) flowmeters and flow control valves. Although, there are various combinations that are used, they all achieve the synthesis of graphene on a desired substrate. In 2009, it was the first time that CVD growth of monolayer graphene films was realized by using polycrystalline Cu foil as substrate [27]. CVD was recognized as a proficient method for graphene synthesis, and since then, there have been a lot of studies and experimentations aiming at further improving the process. Cu as catalyst has many advantages compared to other transition metals including ease of transfer, economical cost and good control of number of layers. Growth mechanism by Cu has been proven to be a surface adsorption-mediated process [28-30]. The used carbon precursor can be, a priori, in the form of gas, liquid or solid. However, it always needs to be in the form of gas as it reaches the metal catalyst surface. The most widely used gaseous carbon precursors are hydrocarbons such as methane (CH<sub>4</sub>) and acetylene (C<sub>2</sub>H<sub>2</sub>). The mechanism of graphene synthesis by Cu can be divided into three stages. At first stage, the carbon precursor molecules collide on the metal substrate surface. They can either adsorb on the surface, scatter back to the bulk gas phase or directly progress to the next stage of the reaction process. At second stage, the carbon precursor molecules dehydrogenate, partially dehydrogenate or remove noncarbon elements so that it can form active carbon species. Lastly, during third stage, these active species coalesce, nucleate and grow into graphene islands on top of the metal substrate [31]. These graphene islands enlarge until full coverage. If the used carbon precursor is under the form of liquid or solid, a supplementary stage precedes the collision phenomenon. Numerous CVD setups have been successfully developed to synthesize graphene, especially from gaseous carbon precursors. Following figure 1 illustrates the various types of CVD setups used in labs.

## 3. Carbon Precursors

The most popular carbon precursors used are Methane (CH<sub>4</sub>), acetylene (C<sub>2</sub>H<sub>2</sub>) and ethylene (C<sub>2</sub>H<sub>4</sub>). One main reason for the popularity of methane is that it has strong C-H bonds that makes it less likely to undergo any chemical reaction. In the case of methane, operating temperature for synthesizing graphene is 1000 °C [32, 33]. Recent study in 2016 by Sun *et al.* [40] showed the feasibility of directly form large-area high-quality graphene on solid glasses using methane with I<sub>2D</sub>/I<sub>G</sub> value from 1.6 to 2.5. Even if the defect level [67] of the



**Figure 1:** Chemical vapor deposition setups: a two-furnace; b bubbling; c heating tape; d magnetic boat. Square = catalyst; circle = precursor

as-produced graphene was quite high ( $I_D/I_G < 1$ ), their work demonstrates the great potential of cheaper and scalable production route for graphene on insulating solid glasses.

All the setups have their own advantages and drawbacks depending on the used starting reactants and materials. For liquid precursors with elevated boiling point that do not readily evaporate like ethanol, an external source of heating is required to vaporize the precursor. The common procedure consists in heating up the catalyst until it reaches the desired reaction temperature; then, the region with the liquid precursor is heated above its boiling point. A flow of N<sub>2</sub> and H<sub>2</sub> carries the gaseous precursor to the metal substrate location for graphene growth. It is vital that the pathway from the external heating point to the heated catalyst zone is above the boiling point of the liquid precursor. Otherwise, the vaporized precursor might return to its original form and deposit onto the hot quartz tube. This represents an additional lead time as the contaminated quartz tube will need to be cleaned before a new experimental run can be performed.

#### 4. Liquid Carbon Precursors

In the early years of graphene research, aliphatic linear hydrocarbon like methane has become the mainstay as it is easy to set up and very stable at elevated temperature. Once the theoretical groundwork was confirmed, the emergence of liquid and solid precursors becomes more apparent. It is expected to change the landscape of the graphene synthesis using CVD method, as liquid carbon precursors are easy to use and relatively inexpensive as compared to conventional gaseous one [34]. Srivastava *et al.* [35] involved the use of hexane (C<sub>6</sub>H<sub>14</sub>) with which large-area continuous and uniform graphene films were formed. This novel method has also allowed an easier way to dope graphene by using various organic solvents as liquid precursors containing dopant atoms. Since monolayer graphene has zero band gap, many researchers have identified the use of dopant to construct a band gap of its own. The use of organic solvents as liquid precursors containing the desired dopant atoms would allow a better process maneuverability in addition to a safer and cheaper synthesis route. Li *et al.* [36] used benzene as liquid hydrocarbon source and obtained monolayer graphene flakes at a very low reaction temperature of 300 °C. Benzene molecules were suggested to only need dehydrogenation before it can interconnect with each other on top of the metal substrate Cu to form graphene. However, no graphene was found due to insufficient energy at 200 °C, probably temperature is too low to remove hydrogen and form any graphene. This finding implies that benzene basic structure has an instrumental impact toward the process of graphene synthesis. A recent publication by Seo *et al.* [37] has been touted to be the solution for low-cost synthesis of graphene. Here, soybean oil was used as the carbon source for graphene synthesis while the main emphasis was on exploiting the ambient air as the filler for the CVD. Normally, highly purified compressed gases such as N<sub>2</sub>, Ar and H<sub>2</sub> were used to create a highly controlled environment for graphene growth. However, single to few layers of continuous graphene films were converted from a renewable natural precursor in the form of soybean oil. The CVD technique is significant since it opens a new avenue to other types of renewable precursors that may also have the potential to be used similar to soybean oil. Other than

Table 1  
Examples of graphene growth by CVD using liquid carbon precursors

Carbon precursor	Growth substrates	Growth conditions				Graphene morphology			
		Method	Temperature (°C)	Pressure	Flow ratio	Reaction duration (min)	Type	Raman	FWHM (cm <sup>-1</sup> )
Ethanol, C <sub>2</sub> H <sub>5</sub> OH	Ni Thickness = 25 μm Size = 3 × 3 cm	CVD	850	1 Torr	H <sub>2</sub> = 10	5	Monolayer graphene	$I_{2D}/I_G = > 2$	
Ethanol	Stainless steel, SS304 Thickness = 0.1 mm Size = 1 × 1 cm	Direct thermal CVD	850	1 atm		10	Mono-, bi-, tri-layer graphene		
Ethanol	Ni foil Thickness = 10 μm Size = 1 × 1 cm	IR heating system	1000	1 atm		5	Monolayer graphene	$I_{2D}/I_G = 3.5$	30-35
Hexane, C <sub>6</sub> H <sub>14</sub>	Cu Thickness = 25 μm Size = 3.5 × 1.5 cm	CVD	950	500 mTorr		4	Monolayer graphene		40 ± 4
Pyridine, C <sub>5</sub> H <sub>5</sub> N	Cu	CVD	300	1 atm			Monolayer N-doped graphene	$I_D/I_G = 0.16$ $I_{2D}/I_G = 4.7$	
Methanol, CH <sub>3</sub> OH	Cu Thickness = 25 μm	CVD	850	1 Torr	H <sub>2</sub> = 10	5 min	Monolayer graphene	$I_{2D}/I_G = \sim 2-3$	
Ethanol, 1-propanol, C <sub>2</sub> H <sub>5</sub> OH	Cu Thickness = 50 μm	APCVD	900	1 atm	Ar:H <sub>2</sub> = 1000:10	30 min	Mono-, bi-, few-layer graphene		
Pentane, C <sub>5</sub> H <sub>12</sub>	Cu foil	CVD	300	8-15 Torr	H <sub>2</sub> = 50	15-30 min	Monolayer graphene flakes	$I_{2D}/I_G = 2$	38

Carbon precursor	Growth substrates				Growth conditions				Graphene morphology		
	Method	Temperature (°C)	Pressure	Flow ratio	Reaction duration (min)	Type	Raman	FWHM (cm <sup>-1</sup> )			
Benzene	Cu Thickness = 25 μm	Oxygen-free APCVD 300	1 atm	Ar:H <sub>2</sub> = 5:20	5 min	Monolayer graphene	$I_{2D}/I_G = > 1.8$				
	Cu Thickness = 10 μm	CVD 300	8–15 Torr	H <sub>2</sub> = 50	15 and 30 min	Monolayer graphene flakes		38			
Benzene	Cu (111) Size = 12 × 5 mm <sup>2</sup>	CVD 437	1 × 10 <sup>-10</sup> mbar		50 min	Multilayer graphene					
Soybean oil	Ni Thickness = 25 μm	APCVD 800 (30 °C/ min)	1 atm		3 min	Monolayer graphene	$I_{2D}/I_G = 0.95-1.50$ $I_D/I_G = 0.15-0.25$ $I_{2D}/I_G = \sim 4.8$				
Cyclohexane, C <sub>6</sub> H <sub>12</sub>	Cu <sub>2</sub> NiZn ternary alloy	LPCVD 1000	300–600 mTorr		3–20 min	Monolayer graphene					
	Commercial palm oil	APCVD 1100	1 atm			Multilayer graphene	$I_{2D}/I_G = 1.54$ $I_D/I_G = 0.73$	> 50			
Refined cooking palm oil	Thickness = 100 μm Size = 1 cm <sup>2</sup>	CVD 900		Ar:H <sub>2</sub> = 50:200	15 s	Mono-, bilayer graphene	$I_G/I_{2D} = 0.62$	27			
Waste chicken fat	Cu Thickness = 20 μm	LPCVD 1080	150 Pa	Ar:H <sub>2</sub> = 98:2	60 min	Monolayer graphene	$I_{2D}/I_G = > 3$ $I_D/I_G = < 0.1$	~ 34			

FWHM full width at half maximum

soybean oil, there have been several other complex carbon precursors reported in the field of graphene synthesis such as waste chicken fat [38], commercial palm oil [39] and refined cooking palm oil [40]. Among them, waste chicken is very promising as it represents a huge portion of the waste from the poultry processing industries. The technology to convert waste materials to high-quality product is always a good way to maximize a process earning potential with existing resources. The use of liquid precursor is opening a new frontier in the field of graphene synthesis. By simple observation, although the most commonly used liquid carbon source for graphene growth is ethanol, the super-effectiveness of benzene has made it much more popular in recent times. It certainly raises some very interesting questions regarding the feasibility of liquid benzene as a solution for low-cost graphene production and the mechanism behind its unique properties compared to other carbon sources. Liquids require a means to convert the liquid precursor into gaseous form before it reaches the active metal catalyst region as opposed to gas precursors. Some of the liquid precursors are volatile organic compounds (VOC) and carcinogenic in nature making it quite harmful to human health. Hence, high-end safeguard system is also compulsory to safeguard the well-being of the operators. Liquid precursors such as benzene and pyridine can synthesize graphene at around 300 °C without the need of plasma-enhanced CVD. Reduction in the graphene growth reaction temperature directly lessens the cycle time of graphene CVD and the overall energy that is required to reach the standard reaction temperature for methane which is around 1000 °C, especially if it is a hot-wall CVD. Of course, benzene and pyridine also come with their own baggage in that they are human carcinogens. As long as the drawbacks can be managed, liquid precursors seem to have a bright future in low-cost largescale production of high-quality graphene.

From the above tables we can get a clear picture of the available liquid precursors for graphene synthesis. However, the mechanism by which actual growth of graphene takes place over the metal substrate can be understood by figure 2 which illustrates a simple schematic diagram for CVD of graphene from HCB. The presence of Cu in the reaction can efficiently enhance de-chlorination phenomenon during the process. The drastically decrease in the reaction temperature to 300 °C [41, 42] was attributed to its basic structure forming a C6 ring after de-chlorination.

We can see an almost similar mechanism while using simple benzene in CVD processes. This can be seen in figure 3.

## 5. Future Prospects

At the moment, many carbon source materials have been tested and they prove to be able to grow graphene. The expertise and knowledge regarding carbon precursors for graphene synthesis by CVD still has a long way to go. There are a number of issues and challenges that need to be continually investigated for the synthesis of graphene via the CVD method:

- I. The use of suitable carbon precursor with the right conditions can improve the production cost. Aromatic compounds epitomize the simplicity of solving the major stumbling block for graphene commercialization. If a similar setup were used for





- benzene and methane, the graphene growth reaction temperature used for benzene would be significantly lower than that used for methane, reducing the amount of energy, lead time and cost of the overall process. However, aromatic compounds are well known to be carcinogenic in nature. Hence, the safety aspect of the production setup needs to be in top-notch condition. This issue is something that can be solved with continuous effort of research. After all, there are a lot of chemical processes which involved the use of hazardous materials.
- II. Another issue is regarding the transport of carbon precursor through the reaction chamber. A two-furnace setup for graphene synthesis required while using liquid and solid carbon precursors are not an ideal setup as it reduces the efficiency of the process considering the length of the quartz tube that is longer to accommodate the extra furnace. Some researchers have proposed the use of spray CVD for liquid precursors. Though, this method requires an extremely high-precision spray mechanism considering the amount of carbon precursor required for graphene growth since 1 atom thick is considerably low. Not to mention the gas that will be used as the external force needs to be inert and not react chemically once it has gone inside the reaction chamber. A more innovative solution is required so that graphene can reach its full potential as the next-generation material of the future.
  - III. Recently, new techniques of graphene synthesis besides CVD have been reported. Laser ablation-induced transformation of graphite into graphene [43], electrochemical exfoliation [44] and plasma-enhanced arc discharge [45] are some of the most popular new techniques for graphene synthesis. Is it the sign of CVD is getting less popular? On the contrary, the rise of new techniques other than CVD seems to stem from the saturation of CVD research and development. CVD has been explored for quite some time now. In fact, there are already few papers that have investigated the feasibility of large-scale production of graphene via CVD [46]. Development of new techniques allows identifying their advantages and disadvantages compared to CVD, and this is the whole graphene production area that makes progress. In the end, the best graphene synthesis method would probably be a combination of these new techniques with CVD.
  - IV. The research in this field is somehow overlooked as most researchers are concentrating efforts on research areas such as plasma etching, laser ablation, catalyst modification and electrochemical bubbling for graphene transfer. The research regarding carbon precursors for graphene growth by CVD might seem simple, but their impact on the graphene growth is not negligible. Recent study has shown that oxygen-free APCVD using benzene as carbon precursor can grow graphene ( $\sim 1 \text{ cm}^2$ ) at  $300 \text{ }^\circ\text{C}$  with a surface coverage of 100%, a field-effect mobility of  $1900\text{--}2500 \text{ cm}^2/\text{V}/\text{s}$  and an optical transmittance of 97.6% [47]. This result is remarkably better than most graphene produced by plasma-assisted CVD, which are normally damaged by the bombardment of the plasma energy. Moreover, plasma-enhanced CVD incurs some cost increase due to the energy that it consumes.

Unlike PECVD, natural carbon precursors are available in abundance and a cheaper solution in every sense.

## 6. Conclusion

Graphene is a super material with extraordinary properties that can be the catalyst for the evolution of the next-generation devices, and CVD seems to be the most prominent production route for large-scale production of graphene. After an extensive and comprehensive investigation on various carbon precursors used to grow graphene, it appears that the state (liquid, solid and gas) of the carbon precursor does not fully correlate with the graphene growth conditions. Instead, the chemical structure of the precursor seems to play the vital role in determining the graphene synthesis. In particular, aromatic compounds for which the structure is similar to the fundamental network of graphene appear as interesting precursors. However, more studies that are more thorough need to be performed so that precise control of graphene growth can be achieved.

## References

- [1] A.H. Castro Neto, N.M.R. Peres, K.S. Novoselov, and A.K. Geim. The electronic properties of graphene, *Rev. Mod. Phys.* 81, 109 (2009).
- [2] C. Lee, X. Wei, J.W. Kysar and J. Hone. Measurement of the Elastic Properties and Intrinsic Strength of Monolayer Graphene, *Science* 321, 385–388 (2008).
- [3] A.A. Balandin, S. Ghosh, W. Bao, I. Calizo, D. Teweldebrhan, F. Miao and C.N. Lau. Superior Thermal Conductivity of Single-Layer Graphene, *Nano Lett.* 8, 902–907 (2008).
- [4] K.I. Bolotin, K.J. Sikes, Z. Jiang, M. Klima, G. Fudenberg, J. Hone, P. Kim and H.L. Stormer. Ultrahigh electron mobility in suspended graphene, *Solid State Commun.* 146, 351–355 (2008).
- [5] M.D. Stoller, S. Park, Y. Zhu, J. An and R.S. Ruoff. Graphene-Based Ultracapacitors, *Nano Lett.* 8, 3498–502 (2008).
- [6] X. Du, I. Skachko, A. Barker and E.Y. Andrei. Approaching ballistic transport in suspended graphene, *Nature Nanotechnology* 3, 491–495 (2008).
- [7] K.S. Novoselov *et al.* Two-Dimensional gas of massless Dirac fermions in graphene, *Nature* 438, 197–200 (2005).
- [8] R.R. Nair *et al.* Fine Structure Constant Defines Visual Transparency of Graphene, *Science* 320, 1308 (2008).
- [9] Y.M. Lin, C. Dimitrakopoulos, K.A. Jenkins, D.B. Farmer, H.Y. Chiu and A. Grill. 100-GHz transistors from wafer-scale epitaxial graphene, *Science* 327, 662 (2010).
- [10] J.K. Wassei and R.B. Kaner. Graphene a promising transparent conductor, *materialstoday* 13, 52–59 (2010).
- [11] K.V. Emtsev, A. Bostwick and K. Horn. Towards wafer-size graphene layers by atmospheric pressure graphitization of silicon carbide, *Nat. Mater.* 8, 203–207 (2009).
- [12] H. Wang. Controllable synthesis of submillimeter single-crystal monolayer graphene domains on copper foils by suppressing nucleation, *J. Am. Chem. Soc.* 134: 3627–3630 (2012).
- [13] J. Zhao, S. Pei, W. Ren, L. Gao and H.M. Cheng. Efficient preparation of large-area graphene oxide sheets for transparent conductive films, *ACS Nano* 4, 5245–5252 (2010).
- [14] A. Kasry, M.A. Kuroda, G.J. Martyna, G.S. Tulevski and A.A. Bol. Chemical doping of large-area stacked graphene films for use as transparent, conducting electrodes. *ACS Nano* 4, 3839–3844 (2010).

- [15] L.G. Arco, Y. Zhang, C.W. Schlenker, K. Ryu, M.E. Thompson and C. Zhou. Continuous, highly flexible, and transparent graphene films by chemical vapor deposition for organic photovoltaics, *ACS Nano* 4, 2865-2873 (2010).
- [16] X. Wang, L. Zhi, N. Tsao, Z. Tomovic, J. Li and K. Mullen. Transparent carbon films as electrodes in organic solar cells. *Angew Chem Int Ed* 47, 2990-2992 (2008).
- [17] X. Qi, K. Pu, H. Li, X. Zhou, S. Wu, Q. Fan *et al.* Amphiphilic graphene composites. *Angew Chem Int Ed* 49, 9426-9429 (2010).
- [18] P. Sutter. Epitaxial graphene: How silicon leaves the scene, *Nature Materials* 8, 171-172 (2009).
- [19] Wilfrid Poirier and Félicien Schopfer. Can graphene set new standards, *Nature Nanotechnology* 5, 171-172 (2010).
- [20] Patrick Maher *et al.* Tunable fractional quantum Hall phases in bilayer graphene, *Science* 345, 61-64 (2014).
- [21] K.S. Kim *et al.* Large-scale pattern growth of graphene films for stretchable transparent electrodes, *Nature* 457, 706-710 (2009).
- [22] X. Li *et al.* Large-area synthesis of high-quality and uniform graphene films on copper foils, *Science* 324, 1312-1314 (2009).
- [23] L. Gao *et al.* Repeated growth and bubbling transfer of graphene with millimeter size single-crystal grains using platinum, *Nature Commun.* 3, 699 (2012).
- [24] P.W. Sutter, J.I. Flege and E.A. Sutter. Epitaxial graphene on ruthenium, *Nat. Mater.* 7, 406-411 (2008).
- [25] Q. Yu, J. Lian, S. Siriponglert *et al.* Graphene segregated on Ni surfaces and transferred to insulators, *Appl. Phys. Lett.* 93, 113103 (2008).
- [26] A. Reina, X. Jia, J. Ho *et al.* Large Area, Few-layer graphene films on arbitrary substrates by chemical vapor deposition, *Nano Lett.* 9, 30-35 (2009).
- [27] H. Wang, L. Cui, Y. Yang, H.S. Casalongue, J.T. Robinson, Y. Liang, *et al.*  $Mn_3O_4$ -graphene hybrid as a high-capacity anode material for lithium ion batteries, *J Am Chem Soc* 132, 13978-13980 (2010).
- [28] H. Sun, J. Xu, C. Wang *et al.* Synthesis of large-area monolayer and bilayer graphene using solid coronene by chemical vapor deposition. *Carbon* 108, 356-362 (2016).
- [29] J.W. Suk, A. Kitt, C.W. Magnuson *et al.* Transfer of CVD-grown monolayer graphene onto arbitrary substrates. *ACS Nano* 5, 6916-6924 (2011).
- [30] X. Li, W. Cai, L. Colombo, R.S. Ruoff. Evolution of graphene growth on Ni and Cu by carbon isotope labelling. *Nano Lett* 9, 4268-4272 (2009).
- [31] Z. Li, P. Wu, C. Wang *et al.* Low-temperature growth of graphene by chemical vapor deposition using solid and liquid carbon sources. *ACS Nano* 5, 3385-3390 (2011).
- [32] X. Li, W. Cai, J. An *et al.* Large area synthesis of high quality and uniform graphene films on copper foils. *Science* 324, 1312-1314 (2009).
- [33] A. Reina, X. Jia, J. Ho *et al.* Large area, few-layer graphene films on arbitrary substrates by chemical vapor deposition. *Nano Lett* 9, 30-35 (2009).
- [34] A. Guermoune, T. Chari, F. Popescu *et al.* Chemical vapor deposition synthesis of graphene on copper with methanol, ethanol, and propanol precursors. *Carbon* 49, 4204-4210 (2011).
- [35] A. Srivastava, C. Galande, L. Ci *et al.* Novel liquid precursor-based facile synthesis of large-area continuous, single, and few-layer graphene films. *Chem Mater* 22, 3457-3461 (2010).
- [36] Z. Li, P. Wu, C. Wang *et al.* Low-temperature growth of graphene by chemical vapor deposition using solid and liquid carbon sources. *ACS Nano* 5, 3385-3390 (2011).
- [37] D.H. Seo, S. Pineda, J. Fang *et al.* Single-step ambient air synthesis of graphene from renewable precursors as electrochemical geno-sensor. *Nat Commun* 8, 14217 (2017).

- [38] M.S. Rosmi, S.M. Shinde, N.D.A. Rahman *et al.* Synthesis of uniform monolayer graphene on re-solidified copper from waste chicken fat by low pressure chemical vapor deposition. *Mater Res Bull* 83, 573–580 (2016).
- [39] M.J. Salifairus, S.B. Abd Hamid, T. Soga *et al.* Structural and optical properties of graphene from green carbon source via thermal chemical vapor deposition. *J Mater Res* 31, 1–10 (2016).
- [40] D. Jalani, S.F.A. Rahman, A.M. Hashim. Defect-free mixed mono- and bi-layer graphene synthesized from refined palm oil by thermal chemical vapor deposition. *Mater Lett* 182, 168–172 (2016).
- [41] X. Gan, H. Zhou, B. Zhu *et al.* A simple method to synthesize graphene at 633 K by de-chlorination of hexachlorobenzene on Cu foils. *Carbon* 50, 306–310 (2012).
- [42] X. Gao, W. Wang, X. Liu. Low-temperature dichlorination of hexachlorobenzene on solid supports and the pathway hypothesis. *Chemosphere* 71, 1093–1099 (2008).
- [43] R. Kumar, R.K. Singh, D.P. Singh *et al.* Laser-assisted synthesis, reduction and micro-patterning of graphene: recent progress and applications. *Coord Chem Rev* 342, 34–79 (2017).
- [44] S.A. Sharief, R.A. Susantyoko, M. Alhashem, S. Almheiri. Synthesis of few-layer graphene-like sheets from carbon-based powders via electrochemical exfoliation, using carbon black as an example. *J Mater Sci* 52, 11004–11013 (2017).
- [45] R. Kumar, R.K. Singh, P.K. Dubey *et al.* Pressure dependent synthesis of high-quality few-layer graphene by plasma-enhanced arc discharge and their thermal stability. *J Nanopart Res* 15:1847 (2013).
- [46] E.S. Polsen, D.Q. McNerny, B. Viswanath *et al.* Highspeed roll-to-roll manufacturing of graphene using a concentric tube CVD reactor. *Sci Rep* 5, 10257 (2015).
- [47] J. Jang, M. Son, S. Chung *et al.* Low-temperature grown continuous graphene films from benzene by chemical vapor deposition at ambient pressure. *Sci Rep* 5, 17955 (2015).