Effect of Chemical Treatment and Thermal Annealing in \( \text{N}_2 \) Atmosphere on Copper Foil Surface for Graphene Growth by Direct-Liquid-Injection Chemical Vapor Deposition Process

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Abstract. Copper foils are widely used as a substrate for graphene grown by chemical vapor deposition method. The qualities of Cu foils can significantly affect the characters of resulting graphene films. Here, we systematically investigated the effects of chemical treatments and thermal annealing at high temperatures (890–950 °C) in \( \text{N}_2 \) atmosphere. We then compared the graphene quality grown by direct liquid injection chemical vapor deposition (DLI–CVD) method with cyclohexane (\( \text{C}_6\text{H}_{12} \)) precursor on un-treated and treated Cu foil. We found that the chemical treatment conditions can improve surface morphology of the Cu foil. In addition, the annealing process at 920 °C for 10 min in \( \text{N}_2 \) atmosphere can increase the grain size and lead to a favorable crystal orientation of (111) plane. Raman and microscopy analyses of the graphene film, show higher yields of monolayer graphene, while, at other annealing conditions and un-treated Cu foil, multilayer graphene is observed.

1. Introduction

In the recent years, several methods have been developed to synthesize graphene including mechanical cleavage, molecular beam epitaxy (MBE), chemical vapor deposition (CVD) \([1]\) and direct liquid injection chemical vapor deposition (DLI–CVD) \([2]\). There are several key factors that need to be considered for the growth of uniform graphene including thermodynamic and kinetic parameters as well as the type of precursor material and substrate in CVD process \([3]\). The choice of substrate is crucial to obtain a high quality graphene growth \([4]\). Cu foil is the most widely used metallic substrate to grow monolayer graphene for large area and high uniformity because of its low cost and scalability towards mass production. The key properties of the Cu foil needed for graphene growth are i) low surface roughness with the absence of impurities, oxides and grain boundaries, and ii) the crystal orientation.
Consequently, cleaning and improving the Cu foil surface are of critical importance towards obtaining high quality large area graphene.

There are many methods for preparing a clean Cu foil surface suitable for graphene growth. Generally, the most common method consists of washing the Cu foil surface with the sequence of acetone (C₃H₆O), ethanol (C₂H₆O) and deionized water (DI water). However, this method cannot remove the rolling lines and the inorganic impurities such as oxide from the foil surface. Hence, other different chemical treatments have been studied to remove the oxide, the rolling lines and other impurities from the foil surface in order to ensure a uniform growth of graphene. A significant fraction of these methods washed the copper foil with acetic acid (CH₃COOH), nitric acid (HNO₃), and hydrochloric acid (HCl) [5, 6]. It has been reported that cleaning Cu foil with HNO₃ could remove the rolling lines and produce smooth surfaces without impurities and oxide. However, due to its strong reactions with Cu, the process is difficult to control. In contrast, HCl and CH₃COOH could remove impurities and oxide, but they do not react with Cu [6, 7] leaving the rolling lines still intact. More recently, an electro-polishing process was used to lower the surface roughness. Kwon et al. [8] studied the electrochemical polishing of Cu foil surfaces at the elevated temperature in H₃PO₄ solutions of various concentrations. It was found that Cu foil electro-polished at 65 °C in 2.7 M H₃PO₄ exhibited a lower surface roughness and a higher percent specular reflection in comparison to those obtained from electro-polishing at room temperature. Awad et al. [9] studied the type and physical properties of electrolyte needed to eliminate the tarnishing layer of the Cu surface. They found that enhancements of the surface morphology and topography may be achieved by addition of soluble starch, ethylene glycol or methanol to the electrolytic solution. In addition to the surface roughness, the grain size and the crystal orientation of substrate are important factors for graphene growth. High quality graphene tends to grow on large grain facets of the Cu foil surface with low content of grain boundaries [10, 11]. Particularly, uniform single sheets of graphene usually grow on the Cu (111) crystal plane [12, 13], which promotes fast, monolayer graphene growth with few defects, while Cu (100) surface causes slow and multilayer graphene growth [14].

In this context, it appears that the thermal annealing process readily yields a smoother surface with larger grain size and increases abundance of the Cu (111) plane. Therefore, we focused on the preparation of the surface of Cu foil through a combined treatment of HCl etching, electro-polishing in phosphoric acid and annealing steps in order to achieve the most suitable substrate for growing monolayer graphene with high uniformity by DLI-CVD method with cyclohexane precursor in N₂ ambient [2].

2. Experiment
2.1. Chemical and electro-polishing treatments
A Cu foil with thickness of 130 µm (Ajax Finechem) was used as a substrate to study the effect of chemical treatment and thermal annealing in N₂ atmosphere. The surface of the as-purchased Cu foil had a lot of rolling lines, impurities and copper oxide. The copper foil was first cut into pieces of 4 × 5 cm². Two sets of substrates were used in our experiments with the details shown in Table 1.
Table 1 Conditions for preparation Cu foil substrate

| Conditions                  | Un-treated Cu foil                      | Treated Cu foil                      |
|-----------------------------|----------------------------------------|--------------------------------------|
| Cleaning                    | Acetone → Ethanol → DI-water → Dried by N2 | Acetone → Ethanol → DI-water → Dried by N2 |
| HCl etching                 | –                                      | HCl → Acetone → Ethanol → DI-water → Dried by N2 |
| Electro-polishing (EP)      | –                                      | H3PO4 → Acetone → Ethanol → DI-water → Dried by N2 |

2.2 Thermal annealing processes
We used a MOCVD model MC-050 from Annealsys Company for substrate annealing. A thermal annealing step was applied to both sets of substrates. The Cu foil was loaded into the chamber and heated up at a rate of 5 °C/min from room temperature to the annealing temperature which was in the 890–950 °C range. The annealing time was varied from 5 to 30 min. Heating and annealing of the substrates were carried out under a flow of N2 gas at a rate of 300 sccm, and total pressure of 2 mbar.

2.3 Graphene growth processes
After annealing process, the Cu foil was kept inside the process chamber and the substrate temperature was adjusted to a growth temperature. This is to prevent air exposure and degradation of the Cu foil which could affect the growth quality. We used liquid cyclohexane, C6H12, as a carbon precursor. An aerosol of liquid cyclohexane was introduced into the CVD chamber by injection head with controllable flow rate in N2 atmosphere. All parameters were shown in Table 2.

Table 2 Parameters used for graphene growth by DLI-CVD

| Parameters                               | values                      |
|------------------------------------------|-----------------------------|
| Annealing temperature and time           | 890–950 °C, 5–30 min        |
| Growth temperature and time              | 950 °C, 10 min              |
| Cyclohexane flow rate                    | 0.5 g/min                   |
| Working pressure                         | 2 mbar                      |
| Injection head opening time              | 1 ms                        |
| Solution frequency                       | 0.5 Hz                      |
| N2 carrier gas flow                      | 300 sccm                    |
| Heating and cooling rate                 | 5 °C/s                      |

2.4 Characterizations
The surface morphology of Cu foil substrates was first characterized by inspection with optical microscopy (OM), atomic force microscopy (AFM), and scanning electron microscopy (SEM). Electron dispersive X-ray spectroscopy (EDX) and X-ray diffraction (XRD) were used to determine the chemical composition of the Cu foil substrates and its crystal structure, respectively. In addition, Raman spectroscopy was performed to verify copper oxide on both the untreated and treated Cu foil surfaces. The grown graphene films on Cu foil substrates were characterized by OM and Raman spectroscopy.
3. Results and Discussion

3.1 Effects of HCl treatments on Cu foil substrates

We found that the untreated Cu foil surface was initially dominated by oxide, rolling lines and impurities in the form of particles containing C, O, and Al as shown in Figure 1(a)–(c). These impurities and substrate defects may act as nucleation sites or growth obstacles, precluding the growth of monolayer graphene [10]. In order to eliminate these problems, HCl was selected to clean the Cu foil surface because it readily dissolves the copper oxide as well as some other surface contaminants and impurities [5]. However, HCl could only remove impurities and oxide, but it do not react with Cu [9, 15] leaving the rolling lines intact.

Different HCl treatment periods were investigated to understand the effect of chemical cleaning process. We found that a 40 s exposure was sufficient to obtain a clean surface (Figure 1(b)), while with shorter period; impurities were still present on the foil surface.

The EDX results show that peaks associated with impurities such as C, O, and Al, are significantly reduced after HCl treatment step (Figure 1(c)). Rolling lines and some impurities still remained on the Cu foil surface. In particular, copper oxide can be detected via Raman peaks attributed to CuO at 300–325 cm\(^{-1}\) and 636 cm\(^{-1}\), and Cu\(_2\)O at 150 cm\(^{-1}\), 217 cm\(^{-1}\) and 643 cm\(^{-1}\) [16, 17]. The Raman spectra shown in Figure 1(d) demonstrate the reduction of copper oxides after the HCl treatment as the peak intensities of copper oxide after treatment were reduced to about 10% of those before treatment. Although chemical etching have the effect to reduce the impurities and oxide on Cu foil surface, rolling lines and some impurities still remained on the Cu foil surface.
Figure 1 SEM, EDX and Raman analysis: (a) and (b) SEM images of Cu foil surface before and after HCl treatment, respectively. (c) EDX results of Cu foil surface before (red line) and after HCl treatment (blue line). (d) Raman spectra of copper oxide on Cu foil substrates before (red line) and after HCl treatment (blue line).

3.2 Effects of electro-polishing on Cu foil surface morphology

After chemical etching, the rolling lines and remaining particles of impurities are still intact. An electro-polishing (EP) process, often used to reduce surface roughness [8, 9], was conducted on the Cu foils. We used the EP process with 50% of H₃PO₄ to etch material away in order to produce a surface without scratches and rolling lines. Figures 2(a) and (b) show typical SEM images of the Cu foil surface before and after the EP treatment. The electro-polished surface shows a smoother appearance with a smaller number of rolling lines relative to that for the unpolished Cu foil surface. Figures 2(c) and (d) show the effects of the EP on the Raman spectra and its intensity for the polished Cu foils with various EP times in a range of 0 to 300 s. Before EP treatment, Raman peaks related to the copper oxide have the highest intensity. Clearly, during the EP process the copper oxide signals was reduced by 90% as the EP time increased up to 120 s with much smaller changes observed for longer times.

AFM micrographs in Figure 3 show height profiles of Cu foil surfaces before and after EP treatment in 50% of H₃PO₄. Initially, the unpolished surfaces show multiple rolling lines, and an RMS roughness of 25.5 ± 0.7 nm. After 40 s of electro-polishing, the number of rolling lines and the roughness were
reduced. The optimum conditions appear to be 120 s of electro-polishing which yields a roughness of ca. 0.9 ± 0.1 nm. It is found that longer EP times caused an increase of the roughness, as illustrated in Figures 3(g) and (h).

![Figure 2 SEM and Raman analyses of Cu foil surface](image)

**Figure 2** SEM and Raman analyses of Cu foil surface: (a) and (b) SEM images before and after EP in 50% of H₃PO₄ for 120 s, (c) Raman spectra of copper oxide on Cu foil surface at various polishing time, (d) Relative Raman intensity of a copper oxide peak as a function of EP time. Each data point is the average value at 4 different locations across 2 samples with an error bar representing the standard deviation.
Figure 3 AFM images and associated height profiles of Cu foil surfaces: (a) and (b) before EP treatment, (c) and (d) after EP for 40 s, (e) and (f) after EP for 120 s, and (g) and (h) after EP for 300 s.
3.3 Effects of thermal annealing

In addition to the surface roughness, the grain size and the crystal orientation of substrate are important for graphene growth. The dominant crystal orientation of the Cu foil from manufacturing is Cu (200) with abundant grain boundaries shown in Figure 4(a). These features increase the formation of defects, yielding multilayer structures and the lack of uniformity of graphene films grown on such copper surfaces.

![Figure 4](image)

**Figure 4** SEM images at two magnifications of Cu foil surface after annealing for 10 min: (a) and (b) after annealed at 890 °C, (c) and (d) after annealed at 920 °C, (e) and (f) after annealed at 950 °C.
To achieve a surface of lower roughness and larger grain size, the Cu foil substrates were thermally annealed after chemical and EP treatments. The Cu foils were loaded into CVD chamber for the annealing process immediately prior to the graphene growth stage. In order to optimize the annealing conditions, the Cu foils were annealed for varying periods of time at temperatures in the range of 890–980 °C under a stream of N₂ at 300 sccm, and a total pressure of 2 mbar.

The results of the annealing experiments are demonstrated in Figures 4 and 5. Note that, before annealing process the Cu foil surface appeared as in Figure 2(b). Compared to those before annealing, the Cu foil surfaces after the thermal annealing (Figure 4) showed clear grain boundaries, consistent with a recrystallization of the material. It is believed that heating of the Cu foil substrate to near the bulk melting point causes surface protrusions and stressed points to melt. As a result, it leads to atomic rearrangements to form more stable crystals [18]. When the annealing temperature was increased, we found that the grain size was increased and showed clear grain boundaries and a smooth surface.

**Figure 5** XRD results: (a)−(c) XRD spectra of Cu foil substrate before (black) and after annealing at 890 °C (red), 920 °C (blue), and 950 °C (green) for 10, 20, and 30 min respectively. (d) XRD intensity ratio between (111) reflection to (200) of Cu foil substrates as a function of annealing temperature and time. Each data point is the average value at 3 different locations with an error bar representing the standard deviation.
Moreover, analyzation using XRD results shown in Figure 5 also suggests that during the annealing the Cu foil surface recrystallized and led to a noticeable increase of the abundance of ratio the (111) with others crystal orientation. Specifically, before the thermal annealing, the Cu foils displayed three diffraction peaks that were identified as Cu (111), Cu (200) and Cu (220), which are consistent with a polycrystalline structure. The most intense diffraction peak corresponded to the Cu (200) reflection, followed by the Cu (111) and a weaker Cu (220) signals, as shown in Figure 5(a)–(c).

After the thermal annealing at 920 °C for 10 min, the same three diffraction signals were still observed in most samples, but their relative intensities had changed significantly. Namely, the most intense diffraction peak associated to the Cu (111) orientation, while the Cu (200) signal was the second biggest. As a way of quantifying the abundance of the crystal orientations present on the Cu foil after the annealing process, Figure 5(d) present the XRD intensity ratio between the Cu (111) reflection and the next strongest (h, k, l) reflection obtained after annealing at different temperatures and times. The largest increase of the Cu (111) orientation resulted from annealing at 920 °C for 10 min. These conditions were taken as the optima for the annealing process. Therefore, the annealing in N2 atmosphere can increase the grain size and the crystal orientation of Cu foil.

3.4 Graphene growth
In order to establish the effects of the chemical treatments and the thermal annealing process on the graphene growth, the graphene films were grown by DLI–CVD on both the untreated and treated Cu foils right after annealing the substrate. In this series of experiments, the annealing temperature covered the 890–950 °C range and varied in duration from 5 to 30 min.

Raman spectroscopy with 473 nm laser excitation wavelength was used to determine the number of layers and the properties of the grown graphene films. The Raman spectrum of graphene shows two main peaks, namely G and 2D bands. The relative intensity (I_{2D}/I_G) ratio of these bands provides an assessment for the number layer of graphene [19]. Moreover, a peak position and a full width at half maximum (FWHM) of the 2D band can also be used to verify the quality of graphene [19]. The I_{2D}/I_G ratio greater than 2.0 is a signature of monolayer graphene, while the I_{2D}/I_G ratio in the range between 1.0 and 2.0 indicates the presence of a bi-layer and the I_{2D}/I_G ratio < 1.0 is an indication of three or more layers [20, 21].

Furthermore, the D peak, absent in pristine graphene, is typically present in CVD grown graphene with varying intensities. The magnitude of the D peak correlates with the concentration of defects in the graphene structure [22]. Thus, the intensity ratio of the D and G peaks (I_D/I_G) is a measure of the disorder or abundance of defects [23]. Then, an increase in the I_D/I_G ratio indicates an increase in the disorder of the graphene film. Typically, monolayer graphene exhibits the FWHM of the 2D band in the range between 30 to 40 cm⁻¹, which is smaller than that for multilayer graphene (> 40 cm⁻¹) [24]. Also, the position of the 2D peak shifts towards higher frequencies as the number of graphene layers increases [19]. We observed that graphene films were formed on Cu foils in all the growth conditions but with different number of layers and film qualities.
Figure 6 Raman spectra of graphene films on treated Cu foil substrates: (a) different annealing temperatures on Cu foil, (c) different annealing times on Cu foil, (b) and (d) the I\textsubscript{2D}/I\textsubscript{G} of graphene films on Cu foil substrates as a function of annealing temperature and time, respectively. Each data point is the average value at 5 different locations across 3 samples with an error bar representing the standard deviation.

We first focused on the effects of the substrate annealing temperature. Figure 6(a) shows the Raman spectra for a series of graphene specimens grown immediately after 10 min annealing of the Cu foil at various temperatures. The D peak intensity appears relatively high in all the samples, indicating of rather large amounts of defects. We observed that the intensity of the D band is smallest for the samples prepared on substrates annealed at 920 °C. The intensities of G and 2D bands increase as the annealing temperature increases but fluctuations in intensity are observed from batch to batch, which arise from the spatial inhomogeneity within the samples. More robust trends become apparent in the behavior of the intensity ratio I\textsubscript{2D}/I\textsubscript{G} as a function of the substrate annealing conditions. Figure 6(b) shows that this ratio has a maximum in the vicinity of the annealing temperature at 920 °C with I\textsubscript{2D}/I\textsubscript{G} ~ 4. In addition, graphene films synthesized at this temperature exhibits lower D peaks. In contrast, for graphene grown on Cu foil substrates annealed at other temperatures, the Raman spectra show slightly lower I\textsubscript{2D}/I\textsubscript{G} ratio of about 2–4 with higher D peak, suggesting the formation of mono or few layers of graphene with
higher defect concentration. Additionally, the substrates annealed at those temperatures showed rougher Cu foil surfaces that were clearly more polycrystalline with abundant grain boundaries. Qualitatively, with smaller grain size, there is higher number of nucleation sites which cause neighboring graphene flakes to overlap and form bi- or few-layer graphene.

Our results suggest that the annealing temperature of 920 °C is the optimum for the growth of single layer graphene via DLI−CVD from cyclohexane. To gain further insight into the effects of the annealing treatment, we investigated the effect of the different annealing times (5−30 min). Figure 6(c) shows the Raman spectra of the resulting graphene on Cu foil substrates annealed at 920 °C with annealing time from 5 to 30 min. When the substrates were annealed for only 5 min, the Raman spectrum shows a value of \( I_{2D}/I_G \) and \( I_D/I_G \) ratios about 2.60 and 0.33, respectively. This annealing time showed mainly bi- and few-layer graphene with high defect concentration, while monolayer graphene was observed at relatively few areas. When the annealing time increased to 10 min, the Raman spectra show the \( I_{2D}/I_G \) ratio increase to about 3.00 and the \( I_D/I_G \) ratio decrease to about 0.11, suggesting a predominance of monolayer and low defect graphene with a few small areas showing lower \( I_{2D}/I_G \) ratios. Annealing the substrate for more than 10 min yields graphene with lower \( I_{2D}/I_G \) ratio in a range of 1.0−3.0 and the \( I_D/I_G \) ratio about 0.2−0.4, which this indicates the formation of bi- or few-layer graphene with higher number of defects. Figure 6(d) shows that this ratio has a maximum in the vicinity of the annealing temperature at 920 °C for 10 min with \( I_{2D}/I_G \approx 3.0 \). In addition, graphene films synthesized at this time exhibits lower D peaks.

After graphene growth process, we reidentified the crystal orientation of the Cu foil substrate again. We found that Cu foil substrates after annealed at 920 °C for 10 min and growth temperature of 950 °C for 10 min, show smooth surface with only Cu (111) domains (Figure 7). Under this condition, we obtained the highest \( I_{2D}/I_G \) ratio, which suggests that monolayer graphene is mostly grown on Cu (111) plane, consistent with previous reports [12, 13]. It is interesting to note the effects of substrate rolling lines on the resulting graphene. This may be done crudely by comparing between graphene films grown on untreated Cu foil substrate and those grown on treated Cu foil substrate. Figures 8(a) and (b) shows optical images of graphene on untreated and treated Cu foil surfaces, respectively. Analysis of the Raman spectra for the samples shown on Figures 8(c) and (d) confirmed these observations. Graphene films prepared on untreated Cu foil substrate exhibits the \( I_{2D}/I_G \) ratio of 1.27 with peak position and FWHM of the 2D band of 2712 cm\(^{-1}\) and 56.8 cm\(^{-1}\), respectively. On the other hand, for graphene films prepared on treated Cu foil substrate, the \( I_{2D}/I_G \) ratio was examined to be 3.16 with a 2D peak position of 2706 cm\(^{-1}\) and a FWHM of 32.8 cm\(^{-1}\). In addition, lower intensity ratio of \( I_D/I_G \) was observed for the graphene film on treated Cu foil substrate, indicating lower defect concentration. Since both the untreated and treated Cu foil substrates were subject to annealing, it must be concluded that the improved graphene quality is due primarily to the electro-polishing process.

**Figure 7** XRD spectra of Cu foil surface after graphene growth
Figure 8  Effect of rolling lines on the quality of graphene: (a) and (b) Optical images of graphene on untreated and treated Cu foil, respectively, (c) and (d) Raman spectra of graphene films on untreated and treated Cu foil substrates. Circles inserted in (a) and (b) indicate the measured locations for Raman spectra shown in (c) and (d), respectively.

To determine homogeneity of our DLI–CVD graphene, we study the spatial distribution of the $I_{2D}/I_G$ ratio, FWHM and peak position of the 2D band. Figure 9 shows Raman mappings and histograms of an area of 30–30 μm$^2$ using a pixel size of 1.5 μm for graphene films grown on untreated and treated Cu foil substrates, respectively. In Figure 9(a), the $I_{2D}/I_G$ ratio of graphene film grown on untreated Cu foil substrate spans the range of 0.5 to 3.0. Most of the scanned area are of bi- or few-layer graphene ($I_{2D}/I_G$ ratio < 2) with small islands of monolayer. Additionally, Figures 9(b) and (c) shows relatively high values of the FWHM and position of the 2D band. The FWHM of the 2D peak varies between 45 and 90 cm$^{-1}$ with an average of 70 cm$^{-1}$, while its position is ca. 2722 cm$^{-1}$, suggesting that about 98.8% of the scanned area are multilayer graphene. In contrast, monolayer graphene was observed to occur on 96.0% of the scanned area for treated Cu foil substrate as shown in Figure 9(d). In this case, the Raman mapping of the scanned area shows abundant zones containing monolayers of graphene signaled by values of the $I_{2D}/I_G$ ratio > 2, and by a 2D peak position of 2705 cm$^{-1}$ with smaller values of the FWHM in a range of 24–44 cm$^{-1}$ with a typical value of 34 cm$^{-1}$. This narrower distribution of peak widths is a strong indication of a high degree of uniformity of the graphene film.
Figure 9 Raman mapping results: (a)–(c) and (d)–(f) are mapping of the I$_{2D}$/I$_G$ ratio, FWHM, and position of 2D band for the graphene films on untreated and treated Cu foil substrates, respectively. (g)–(i) are histograms of the I$_{2D}$/I$_G$ ratio, FWHM of 2D peak, and position of 2D band of graphene films on untreated (blue) and treated substrates (red), respectively.

Figure 9 (g)–(i) show the histograms of the I$_{2D}$/I$_G$ ratio, FWHM and peak position of the 2D band for graphene synthesized on untreated and treated Cu foil substrates. It was found that the most scanned area of graphene on untreated Cu foil has values of the I$_{2D}$/I$_G$ ratio < 2. The position of 2D peak is widely distributed (2707 to 2740 cm$^{-1}$) and it exhibits higher FWHM values. In contrast, the graphene that was prepared on treated Cu foil substrate shows the I$_{2D}$/I$_G$ ratio > 2.0 and 2D peaks localized at ca. 2705 cm$^{-1}$. Most of the scanned area in this case shows the 2D peaks at lower wave number (cm$^{-1}$), and lower FWHM values with a narrower distribution. Therefore, it can be concluded that the graphene grown on treated Cu foil surface shows higher uniformity and higher yields of monolayer graphene, but graphene on untreated surface show low uniformity and typically exhibits multilayer graphene.

4. Conclusions
We have studied the effect of preparing Cu foil surfaces process, which focus on chemical treatment and thermal annealing in N$_2$ for graphene growth via DL$_1$–CVD. Chemical and electro-polishing treatments allow us to achieve very low roughness of Cu foil surfaces with minimized scratches and rolling lines. Further lowering of the substrate roughness can be obtained by annealing the Cu foil substrate with the additional advantage of increasing the grain size as well as the abundance of the Cu
(111) crystal orientation. It is possible to control the number of layers of graphene by selecting the annealing conditions for the Cu foil substrates. Monolayer graphene tends to form in higher abundance on the substrates annealed at 920 °C for 10 min, while double and few layered graphene appears to form on substrates annealed at temperatures lower than 900 °C or for those annealed at 950 °C. We demonstrated that DLI−CVD from cyclohexane in N2 ambient produces graphene films of quality comparable to that prepared from gas precursors, with the advantage of a simpler precursor handling.

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