

Growth of Graphene on Nickel using a Natural Carbon Source by Thermal Chemical Vapor Deposition

(Penumbuhan Grafir di atas Nikel dengan Menggunakan Sumber Karbon Semula
Jadi Melalui Pemendapan Berhaba Wap Kimia)

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ABSTRACT

We report the growth of few-layer graphene (FLG) on a nickel (Ni) substrate using palm oil as a single carbon source by thermal chemical vapor deposition (T-CVD). Using an ample amount of vaporized palm oil resulted in the formation of a thick, amorphous carbon film on the Ni surface. By decreasing the amount of the carbon source supply, thin films consisting of graphite grains and FLG were obtained at growth temperatures of 900 and 1000°C, confirming the potential of using palm oil as a source for graphene growth. The occasional voids were obtained on the transferred film and these voids were confirmed to be areas where FLGs are formed. These FLGs are likely removed during the transfer process. Further reducing the amount of palm oil suppressed the coverage of graphite grains and increased the void area formation on the transferred films, proving the increase of FLG coverage.

Keywords: Few layer graphene; nickel; palm oil; thermal chemical vapor deposition

ABSTRAK

Kami melaporkan penumbuhan beberapa lapisan grafir (FLG) di atas substrat nikel (Ni) dengan menggunakan minyak kelapa sawit sebagai sumber karbon tunggal melalui pemendapan berhaba wap kimia (T-CVD). Penggunaan jumlah wap minyak kelapa sawit berlebihan menghasilkan saput amorfus karbon yang tebal di atas permukaan nikel. Dengan mengurangkan jumlah bekalan sumber karbon, saput tipis yang mengandungi bijiran grafir dan FLG telah diperolehi pada suhu penumbuhan 900 dan 1000°C. Ini mengesahkan potensi menggunakan minyak kelapa sawit sebagai sumber untuk penumbuhan grafir. Beberapa lubang telah diperolehi atas saput yang dipindahkan dan lubang itu telah disahkan sebagai kawasan FLG terbentuk. FLG itu mungkin teralih keluar semasa proses pemindahan. Pengurangan selanjutnya jumlah minyak kelapa sawit menyekat pembentukan bijiran grafir dan menambahkan kawasan lubang pada saput yg telah dipindahkan. Ini membuktikan peningkatan liputan FLG.

Kata kunci: Beberapa lapisan grafir; minyak kelapa sawit; nikel; pemendapan berhaba wap kimia

INTRODUCTION

Graphene is known to possess superior and unique properties, such as ultra high carrier mobility (Bolotin et al. 2008; Novoselov et al. 2004), high optical transparency (Bae et al. 2010; Gunho et al. 2010), high thermal conductivity (Shahil & Balandin 2012) and large surface area (He et al. 2012). Because of these properties, graphene is considered to be a promising material for various electronic applications, including high frequency switching devices (Han et al. 2011; Yang et al. 2012), sensors (He et al. 2012), transparent electrodes (Bae et al. 2010; Gunho et al. 2010) and novel functional devices (Rahman et al. 2012). Since its discovery in 2004, extensive research has been performed not only on graphene's use in such applications but also on the technologies used to grow it (Berger et al. 2006; Gilje et al. 2007; Yu et al. 2008). The growth of large-area and high-quality single crystalline graphene film is essential for practical applications.

Among the studied growth methods, chemical vapor deposition (CVD) seems to be the most effective and reliable method for the formation of large-area graphene (Bae et al. 2010; Mattevi et al. 2011; Nandamuri et al. 2010; Reina et al. 2008; Yu et al. 2008). Recently, the growth of polycrystalline single-layer graphene film up to 30 inches diameter was successfully demonstrated and utilized as a transparent electrode (Bae et al. 2010).

One of the major issues with CVD growth is the precursor. The usage of toxic and explosive gases requires a growth system with a high degree of safety and handling precautions. Several attempts have been devoted to exploring novel non-toxic and non-explosive carbon sources (Byun et al. 2011; Guermoune et al. 2011; Kalita et al. 2010; Miyata et al. 2010; Zhang et al. 2012) to replace the typically used carbon precursors, i.e. methane (Nandamuri et al. 2010; Yu et al. 2008) and acetylene (Nandamuri et al. 2010). The feasibility of growing

graphene using alcohol and toluene has been demonstrated (Guermoune et al. 2011; Miyata et al. 2010; Zhang et al. 2012). To ease the dependence on fossil resources, the usage of camphor, which is a renewable natural carbon source, has also been proposed (Kalita et al. 2010).

In this paper, we report the growth of graphene by utilizing palm oil as a carbon source. Palm oil is a non-toxic and non-explosive natural source that is inexpensive and abundant and readily available and cheap. In recent years, one of the co-authors' group has succeeded in growing carbon nanotubes (CNTs) from the palm oil (Suriani et al. 2010, 2009). In this work, we further apply this natural source for the growth of graphene on nickel by thermal-CVD method. The effects of growth temperature and amount of palm oil on the quality of grown graphene were investigated. Few-layer graphene has been successfully grown at temperatures in the range of 800 to 1000°C.

EXPERIMENTAL DETAILS

Nickel (Ni) foil with a thickness of 100 μm and a purity of 99% was used as a metal catalyst. Ni is known to have a relatively high solubility of carbon, which allows the diffusion of thermally decomposed carbon atoms into the substrate during the CVD process (Baraton et al. 2011; Lander et al. 1952). For the carbon source, commercially available refined cooking palm oil was used. Prior to the growth, the Ni substrate was cleaned using acetone, ethanol and de-ionized water inside an ultrasonic bath for 5 min to eliminate organic contaminants. Then, the Ni substrate was immersed in a nitric acid solution to remove native oxides that may have existed on the surface. The removal of contaminants and native oxides was expected to enhance the diffusion of carbon atoms into the Ni surface, thus allow the formation of graphene film.

Figure 1(a) shows the experimental setup of the CVD system consists of a single-zone horizontal tube furnace system as the CVD reactor. The tube furnace showed good temperature uniformity at the center of the tube, with a drastic temperature decrease at the edges of the tube. Ideally, during CVD growth, the precursor gases are introduced after the substrate reaches the desired growth

temperature. In case of using single-zone furnace, it is difficult to control the delivery of the evaporated palm oil because of large temperature difference between the smoke point of palm oil and the desired growth temperature. In this work, the growth was performed at 800, 900 and 1000°C. At such high temperatures, the palm oil is expected to be thermally decomposed and then diffused into the Ni substrate. In order to control the delivery of the carbon source at high growth temperature, we simply exploited the temperature difference between the center and the edge of the tube. An alumina boat filled with palm oil was placed at a distance of 15 cm from the center point of the tube. Using this setup, the palm oil was evaporated and delivered only when the center tube temperature exceed approximately 800°C. At growth temperature of 800°C, the temperature of alumina boat is 250°C. This temperature is above the smoke point of the cooking palm oil. At growth temperature of 900 and 1000°C, the temperature of alumina boat increases to 320 and 400°C, respectively.

Figure 1(b) shows the temperature profile of the growth process. First, the air in the tube was purged by flowing argon gas at 200 sccm for 30 min. Then, the sample was annealed at a setting temperature for a certain period of time, defined as the growth time. The grown samples were cooled down inside the furnace before they were removed. The cooling rate inside the furnace was 30°C/min. Based on the accepted growth mechanism on Ni, graphene is formed on the Ni surface as a result of the segregation and precipitation of the carbon atoms on the substrate during cooling (Baraton et al. 2011; Yu et al. 2008; Zhang et al. 2010). Throughout the CVD process, argon gas at 200 sccm was used continuously to create an inert environment, to carry the evaporated palm oil and to assist the cooling process.

The thin film that formed on the Ni substrate was then transferred onto 285 nm-thick SiO₂ on a Si substrate using the typical polymer-based method (Reina et al. 2008). The transfer process was conducted to enable the rapid characterization of the grown film using an optical microscope. As graphite becomes thinner, the surface color changes from yellow to light blue and then to a medium blue color. The graphene layer is purple color when there

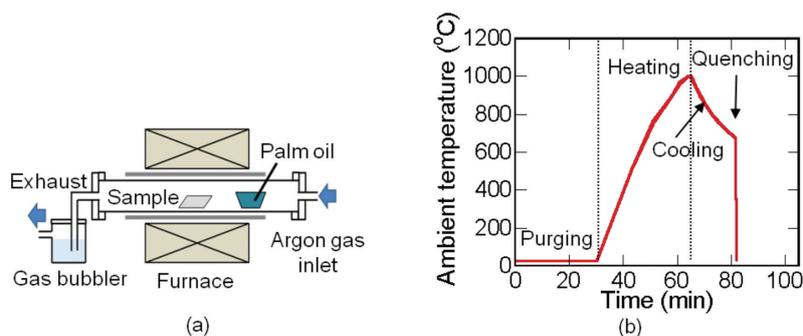


FIGURE 1. (a) Schematic of the CVD system setup and (b) ambient temperature profile during the CVD process for a growth temperature of 1000°C

are fewer than 10 layers (Ni et al. 2007). Based on the color transition, the graphite and graphene can be easily distinguished. Furthermore, the graphene on the insulator surface has better visibility (Blake et al. 2007; Ni et al. 2007; Rahman et al. 2012), thus aiding in the search for thin graphene film for Raman spectroscopy. The formed graphene layer was detached from the Ni substrate by a wet chemical etching process using an aqua regia solution (mixture of HNO_3 and HCl in 1:3 volume ratio) for 12 h. Here, the Ni substrate was pre-coated with a resist (ZEP520A) by a spin coater to give mechanical support to the graphene film. The detached ZEP520A/graphene layer was then transferred onto a new SiO_2/Si substrate. After the removal of the ZEP520A layer, the sample was ready for characterization. The grown graphene film was characterized using an optical microscope and micro Raman spectroscopy with an argon laser at a wavelength of 514.5 nm.

Raman spectroscopy was used to evaluate the structural properties of the grown film. In general, the Raman spectra of graphite and graphene show a pronounced peak and band at 1580 and 2700 cm^{-1} , which are known as the G peak and 2D band, respectively (Ferrari et al. 2006). Because the shape of the 2D band is sensitive to the number of layer, it can be used to distinguish between graphite and graphene. Thick graphite shows a broad, asymmetrical 2D band, while a thinner layer shows a more symmetrical and sharper peak (Ferrari et al. 2006; Gupta et al. 2006). Single-layer graphene is reported to show a sharp 2D peak with a full width at half maximum (FWHM) of approximately 30 cm^{-1} (Reina et al. 2008). In addition to the surface color and FWHM value, we calculated the G-to-2D peak intensity ratio (I_G/I_{2D}) to estimate the layer number of the graphene. A lower layer number is known to produce a lower G peak intensity and thus a lower I_G/I_{2D} (Graf et al. 2007; Reina et al. 2008). An I_G/I_{2D} value less than 1 indicates that there are fewer than 3 layers (Reina et al. 2008). In this study, we use the term 'few-layer graphene (FLG)' to refer to a film that is estimated to have fewer than 10 layers. A defect related peak at 1345 cm^{-1} (D-peak) in the Raman spectra is also considered when evaluating graphene film quality (Ferrari et al. 2006). The presence of the D-peak indicates a point defect or a disorder at the sp^2 bonded carbon atom network.

RESULTS AND DISCUSSION

GROWTH WITH AMPLE SUPPLY OF PALM OIL

For our first attempt at graphene growth, the CVD process was performed using 515 mg of palm oil at 1000°C for 1 min. During annealing, the palm oil was evaporated, with no remaining oil in the alumina boat. A black-colored thick film was deposited on the surface of the Ni substrate. The film was too thick to be transferred onto SiO_2/Si substrate. Figure 2 shows the Raman spectra of the deposited film. Broad D and G peaks were observed in the Raman spectra at 1350 and 1590 cm^{-1} , respectively.

The observed high and broad D and G peaks indicate the formation of amorphous carbon (Negishi et al. 2011). The obtained result was similar to what has been reported by Negishi et al. (2011) in whose study the CVD process on a non-catalytic substrate produced amorphous carbon when the temperature of the CVD reactor exceeded the thermal decomposition temperature of the carbon source. We believe that with a short growth time and an ample supply of carbon, the decomposed carbon source did not fully diffuse into the Ni substrate and thus accumulated on the surface. When the growth time was increased to 30 min, a 2D peak that related to a double resonance process of sp^2 -bonded carbon atoms was observed. The result indicated an improved graphite structure of the formed film. During the 30 min growth process, the carbon source was not delivered continuously because the 515 mg palm oil was likely to be completely evaporated and exhausted from the tube in the early time of the heating. Therefore, the growth process can be divided to two stages. The first stage is the deposition of carbon film which occurred during the short time where the Ni substrate was exposed to the evaporated palm oil. The second stage is the annealing of the deposited carbon film. The annealing process is believed to induce the graphitization of the deposited carbon film.

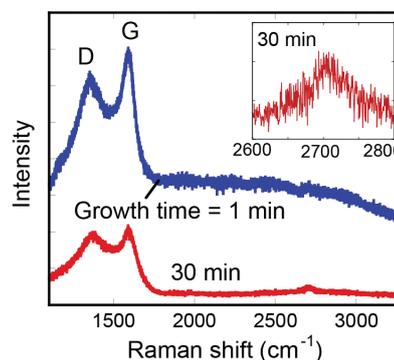


FIGURE 2. Raman spectra of the thin film grown at 1000°C by using 515 mg of palm oil for 1 and 30 min

GROWTH WITH A REDUCED SUPPLY OF PALM OIL

The quantity of the carbon source was reduced to 29 mg, approximately 1/17 of the previous amount. The growth time was fixed at 1 min, while the growth temperatures varied from 800 to 1000°C. Figure 3 shows the optical microscope images and Raman spectra of the transferred grown film on the SiO_2 surface. The Raman spectra shown in this figure was obtained after baseline correction and normalization of the raw Raman spectra. Optical images of the transferred film grown at 1000°C are shown in Figure 3(a) and 3(b). The transferred films were dominated by the surface having a mixture of light blue and yellow colors which likely corresponded to thicker and thinner graphite, respectively (Ni et al. 2007). Similar surface as shown in Figure 3(d) and 3(e) was also observed for the transferred

film grown at 900°C. Figure 3(c) and 3(f) show Raman spectra obtained from different spots in the film grown at 1000 and 900°C, respectively. Spot A, B, C and D are spots from the film grown at 1000°C. Spot E, F and G are spots from the film grown at 900°C. Yellow-colored A and E spots showed broad and asymmetrical 2D peaks, confirming that the regions were graphite (Ferrari et al. 2006). Clear view of the 2D band of A and E peak was shown in the inset of Figure 3(c) and 3(f). The medium blue-colored B spot, which was expected to be thinner graphite, showed a more symmetrical and sharper 2D band. FWHM of the 2D band and I_G/I_{2D} for B spot was 80 cm^{-1} and 3.7, respectively.

Occasional voids (C and F spots) were observed in the optical image of the transferred film. The normalized Raman spectrum from C spots still shows G and 2D peaks, indicating the existence of graphite cluster. However, the intensity of these peaks in the raw Raman spectra (not shown here) was negligible. On the other hand, no prominent peaks related to graphite structure could be seen at Raman spectra of F spot. Interestingly, purple-colored flakes (D and G spots) that corresponded to FLGs could be found around the void areas. The Raman spectrum of the purple-colored D spot showed an FWHM of 2D band of 73 cm^{-1} and an I_G/I_{2D} of 2.6. Compared to the medium blue-colored B spot, D spot has lower FWHM of 2D band and smaller I_G/I_{2D} . This indicates that the D spot had a lower

layer number in relative to the B spot. It is worth noting that there was only small difference in the FWHM of the 2D band between these two spots. For CVD-grown graphene, possible stacking faults between layers resulted in less dependence on the 2D band shape as an indicator of the layer number (Reina et al. 2008). Conversely, the G spot showed a lower I_G/I_{2D} of 1.1 and a sharper 2D with an FWHM of 45 cm^{-1} . The calculated values were consistent with the reported values by A. Reira et al. (2008) for three-layer graphene.

For the film grown at 800°C, a grainy black-colored surface was observed. However, the Raman spectrum of the sample (H spot) still showed a pronounced G peak and 2D band, indicating the formation of a graphite structure with a relatively high D peak. The high D peak could be interpreted as a consequence of a disordered carbon network or the small grain size of graphite. Because the observed G peak was relatively sharp compared to the peak in Figure 2, the Raman spectrum can be interpreted as a result of the graphite's small grain size. At lower growth temperature, the reconstruction of Ni grain boundaries did not occur as much as at higher temperatures and hence created a rough surface, which is expected to contribute to the formation of small graphite crystallite.

The graphite grain formation observed in Figure 3 was believed to be a result of excessive diffusion of the carbon source into the Ni substrate during the CVD process.

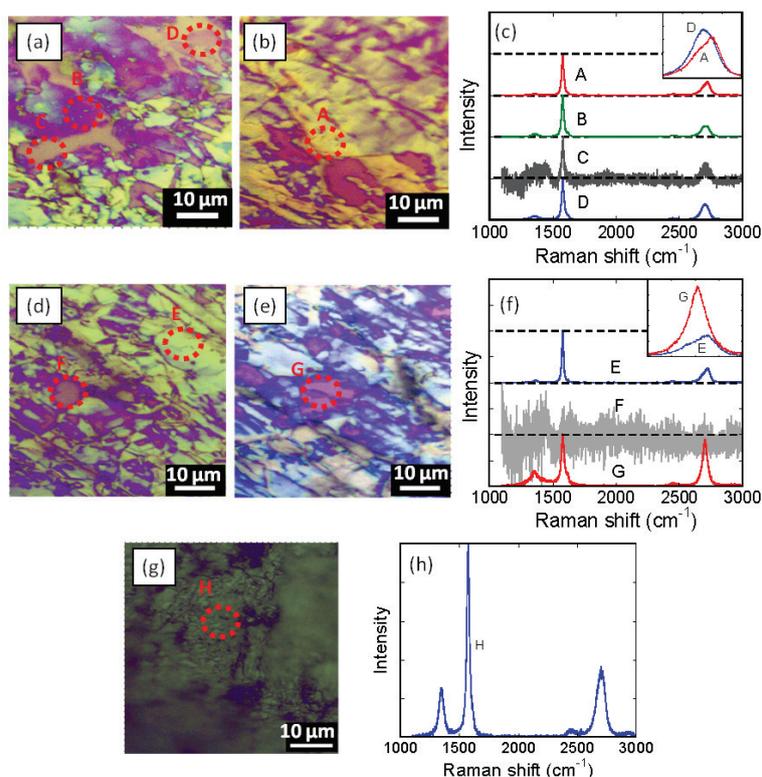


FIGURE 3. Optical microscope images of the transferred films grown at (a and b) 1000, (d and e) 900 and (g) 800°C. Raman spectra data of the film grown at (c) 1000, (f) 900 and (h) 800°C.

Inset of (c) and (f) show 2D band for A, D, E and G spots

Miyata et al. (2010) have described the process as graphite sedimentation. To suppress the graphite sedimentation, we further decreased the amount of palm oil to 6 mg to limit the amount of diffused carbon atoms. The CVD process was conducted at 900°C for 1 min. Figure 4 shows the optical images and Raman spectra data of the transferred film. Yellow-colored graphite and purple-colored FLG grains were grown and transferred onto the SiO₂ surface. Raman spectra confirmed the formation of FLGs at the I and J spots. The background signal in the raw Raman spectra in Figure 4(c) is believed to be from the ZEP520 polymer that was not completely removed during the transfer process. Figure 4(d) shows the baseline-corrected and normalized Raman spectra. Compared to the optical image of graphene grown using 29 mg, it was observed that the coverage of the graphite region decreased. The result showed that by limiting the amount of the carbon source supply, the graphite sedimentation can be suppressed, thus allowing FLG formation.

The transferred films in this experiment exhibited poor film continuity due to the occurrence of voids in the film. We believe that the voids were created by the removal of the grown FLG during the transfer process. The observed FLGs near voids were the remaining films after the transfer process. To confirm this, we conducted Raman spectroscopy for the Ni substrate just after the CVD growth. Figure 5 shows the optical image and Raman spectra of the Ni surface. The Ni surface showed different color contrast in different regions. The grey-colored L spot in Figure 5(a) was an area on which graphite was grown. Here, because the surface color contrast can be used as an indicator to justify the presence of the graphite/graphene film, we can expect no growth at the areas that have the same surface color as the original Ni surface. However, the silver-colored

K spot still showed Raman spectra of FLG. The FWHM of the 2D peak and I_G/I_{2D} value for the K spot were 40 cm⁻¹ and 0.6, respectively. FWHM of 2D band of 30 - 40 cm⁻¹ and I_G/I_{2D} less than 1 are understood to be two characteristics feature of Raman spectra for one- and two-layer graphene. (Reina et al. 2008). Reina et al. (2008) have shown that one- and two-layer graphene gave I_G/I_{2D} of 0.35 and 0.18, respectively. Based on these reported value, K value could be estimated as two-layer. It was difficult to determine the presence of graphene by optical microscope alone because the graphene layer is transparent. Raman spectroscopy characterization at different spots, especially the silver-colored regions, showed the formation of continuous graphite and graphene film on the Ni surface. It can be concluded that the poor continuity of the transferred film was a result of the imperfect transfer process.

A higher coverage of FLG was obtained by limiting the amount of palm oil. The results indicated that further improvement of the graphene coverage percentage and suppression of graphite sedimentation can be achieved by a small, constant supply of the carbon source. We speculate that the simultaneous graphite sedimentation and FLG formation were due to the difference in carbon solubility of the surface (Miyata et al. 2010). Therefore, the pre-treatment procedure to remove the oxide layer on the Ni substrate must also be reconsidered to enhance the carbon solubility of the Ni surface.

CONCLUSION

We have investigated the possibility of using palm oil as the carbon source for the formation of graphene. Using an ample supply of palm oil lead to the formation of thick amorphous carbon on the Ni surface. After reducing the

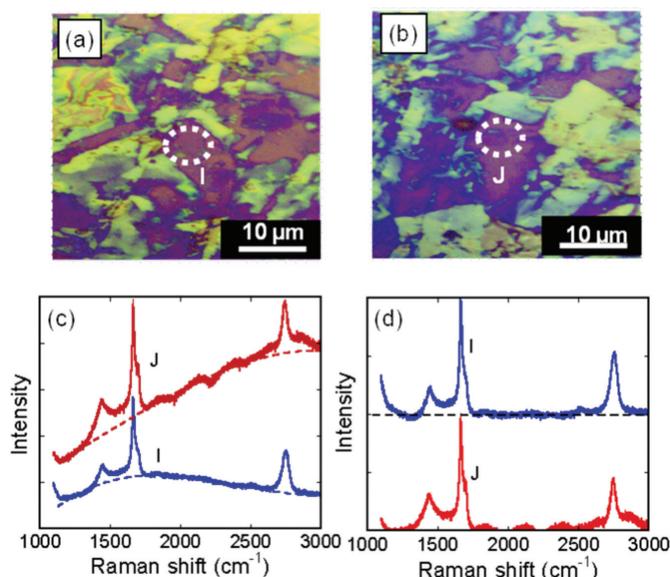


FIGURE 4. (a and b) Optical microscope images, (c) raw Raman spectra data of the transferred films grown using 6 mg of palm oil at 900°C for 1 min and (d) the baseline-corrected and normalized Raman spectra

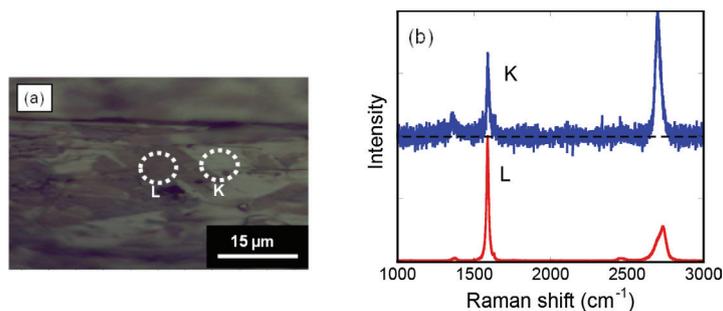


FIGURE 5. (a) Optical microscope images and (b) Raman spectra data of the Ni surface after the CVD process using 6 mg of palm oil at 900°C for 1 min

amount of palm oil to 29 mg, films consisting of graphite grains and FLG with occasional voids were obtained at growth temperatures of 900 and 1000°C. Further decreasing the carbon source supply suppressed the so-called graphite sedimentation and increased the coverage of the FLG. While the obtained result confirms the feasibility of synthesizing graphene film from palm oil, further optimization of the CVD process and growth conditions as well as the substrate pre-treatment are required to improve the uniformity of the film. Besides, the further decrease of carbon source supply, controlling the cooling rate can be performed to suppress the formation of graphite grain (Umair & Raza 2012; Zhang et al. 2013). Fast cooling rate is expected to minimize the amount of carbon atoms which diffuse out and subsequently form the carbon film. In order to improve the film uniformity, pre-annealing of Ni substrate was shown to be an effective approach (Zhang et al. 2013, 2010). Pre-annealing at high temperature reduces the Ni grain boundary. This lead to the decrease in the nucleation of multilayer graphene. In addition, the transfer process needs to be optimized to improve the continuity of the transferred graphene film.

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