High-quality bilayer graphene grown on softened copper foils by atmospheric pressure chemical vapor deposition

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ABSTRACT Bilayer graphene (BLG) shows great application prospect and potential in next-generation electronics because of its unique electrical and mechanical properties. However, the scalable synthesis of large-area high-quality BLG films is still a great challenge, despite the maturity of chemical vapor deposition (CVD) technique. In this study, we report a robust method to grow BLGs on flat, softened Cu foils by atmospheric pressure CVD. A moderate amount of residual oxygen accelerates the growth of BLG domains while suppressing the formation of multilayers. Raising the nucleation density at low hydrogen pressure efficiently increases the film continuity. Based on the optimized CVD process, the growth of graphene films on $4\times 4\text{ cm}^2$ Cu foils with an average BLG coverage of 76% is achieved. The morphology and structure characterizations demonstrate a high quality of the BLG. Dual gate field-effect transistors are investigated based on AB-stacked BLG, with a tunable bandgap and high carrier mobility of up to $6790\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ at room temperature.

Keywords: graphene, bilayer, chemical vapor deposition, Cu foil

INTRODUCTION

The realizations of tunable bandgap, unconventional superconductivity and ultrahigh hardness in bilayer graphene (BLG) make it fascinating for fundamental research as well as emerging electronic applications [1–3]. A scalable approach to synthesizing high-quality BLG is of great importance. Large-area single-layer graphene (SLG) can be achieved by chemical vapor deposition (CVD) on copper [4–6], thanks to the negligible solubility of carbon atoms in Cu and the self-limited growth me-

chanism [6,7]. However, it becomes a great challenge for BLG, owing to the complex manufacturing processes brought by the additional layers of graphene. Strategies such as using hazard gas (e.g., H_2) with high partial pressure [8–10], edge etching of the top graphene layer [11], and using alloy or multicomponent catalyst substrates [12–15] have been explored to synthesize BLG. Nevertheless, it is still imperative to develop a practical and industrially viable method for BLG preparation.

BLG can be prepared based on small ad-layered graphene (ALG), which is often formed beneath SLG synthesized on Cu [8–11,15–17]. Efforts have been made to investigate the nucleation and growth kinetics of ALG [18–21]. The understanding of the growth mechanism of ALG facilitates the layer uniformity of desired large-area products. For example, continuous SLG films have been achieved by simultaneous growth of SLG on both sides of the suspended Cu foil, which suppresses the ALG growth in a normal CVD process [22]. Different from SLG growth, BLG growth requires two unequal sides of Cu. It is essential to ensure the bulk diffusion of carbon atoms to the BLG growth side of the Cu foil, meanwhile to suppress graphene growth on the other side. Strategies such as the adoption of a Cu pocket to limit the carbon concentration inside the pocket have been successfully employed to grow BLG domains on the exterior surface [16,19]. However, drawbacks like low carbon diffusivity of the inside atmosphere and the changeable gap size of crimped edges undoubtedly limit the scalable use of Cu pockets. Furthermore, when both sides of the Cu foil are completely covered by graphene, forming a barrier for

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carbon diffusion, BLG stops to grow [19,23]. Therefore, the growth rate should be fast enough to prepare large BLG. BLG domains with size of hundreds of microns have been achieved after 6 h of growth with the assistance of oxygen to accelerate the carbon bulk diffusion process [16]. Another key factor for the preparation of uniform BLG is to suppress the growth of tri-layer graphene (TLG) and thicker multilayer graphene (MLG) [21,24,25]. As ALG often forms at the same initial nucleation centers and grows simultaneously with the first graphene layer [20], it is quite difficult to completely avoid undesired TLG or MLG.

In this work, we developed a robust and efficient method for atmospheric pressure CVD (AP-CVD) of high-quality BLG on Cu foils. The flat Cu foil was gradually softened and stacked onto the quartz tube/carrier surface during a designed heating process. The quartz significantly contributed to the formation of large BLG domains on the top side of Cu foil by ensuring the difference on two sides of Cu foil, suppressing the backside graphene nucleation density, and providing a continuous supply of oxygen during the CVD process. Ar flow was adopted to exhaust residual oxidative species in the AP-CVD chamber. Excessive time of Ar flow suppressed the growth of BLG, while shortening the Ar flow time greatly promoted the growth of excess TLG and MLG. The results were associated with the increased oxidation of Cu foil by system residual oxygen. Low hydrogen flow rate was efficient to promote the nucleation of BLG. With optimized parameters, continuous graphene films with average BLG covering of 76% were achieved. Detailed morphology and structure characterizations confirmed that the obtained BLG was of high quality. The AB-stacked BLGs exhibited tunable bandgaps with carrier mobility up to $6790 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature observed in the dual-gate field effect transistors (FETs).

EXPERIMENTAL SECTION

Growth of BLG

The synthesis process was carried out in a thermal CVD furnace with a quartz tube (170 cm in length, 5 cm in diameter) at atmospheric pressure [26,27]. The quartz tube was sealed by two flanges to maintain the reactive atmosphere and avoid contamination. Cu foils (25 μm , 99.8%, Alfa Aesar) were used as the catalytic substrates. Acetone (99.5%, Peking Reagent) was used as the carbon source. The acetone was accurately fed to the CVD system with a sustained and controlled release approach, as described in our previous study [27]. A $4 \times 4 \text{ cm}^2$ Cu foil

was placed at the center reaction zone on a semicircular quartz carrier (see Fig. S1a). Ar (300 mL min^{-1}) was flowing for 40–300 min to exhaust the residual oxygen in the AP-CVD system. The furnace was heated to the reaction temperature (1040°C) at a speed of $10^\circ\text{C min}^{-1}$. It took 100 min to heat up the Cu foil, as shown in the schematic diagram (Fig. 1a). The growth time was 60 min with H_2 ($10\text{--}20 \text{ mL min}^{-1}$) and Ar (300 mL min^{-1}) flows. The Cu foil was rapidly cooled down after graphene growth.

Growth of SLG

As shown in Fig. S2, the Cu foil was rapidly pushed into the reaction zone when the furnace reached 1040°C . Besides the fast heating up process of the Cu foil (about 1 min), other growth parameters were the same as the preparation of BLG.

Growth of MLG

The quartz carrier was replaced by a quartz shelf support (Fig. S3) to maintain equal carbon precursor concentration on two sides of the Cu foil. Other growth parameters remained the same as the growth of BLG.

Transfer of graphene

Graphene films grown on Cu foils were wet-transferred onto SiO_2/Si (300 nm thick SiO_2) substrates by etching Cu with polymethylmethacrylate (PMMA) protection for subsequent characterizations or applications. The PMMA layer was removed by immersing in acetone and isopropanol.

Structural characterizations

The as-prepared graphene samples were characterized by scanning electron microscopy (SEM, MERLIN VP Compact, Carl Zeiss), transmission electron microscopy (TEM, JEM-2100F, JEOL), Raman spectroscopy (Horiba Evolution, 532 nm laser excitation), and atomic force microscopy (AFM, Dimension Edge, BRUKER, Tapping mode). Optical images were taken by an Axio scope A1 optical microscope and a Nikon Eclipse Ni-U optical microscope.

Fabrication and measurements of BLG-based FETs

BLG-based FETs were fabricated on 300 nm SiO_2/Si substrates using photolithography (ATD 1000). Cr (5 nm)/Au (50 nm) were deposited as contact electrodes using electron-beam evaporation (FU-121PEB). The channel length and width were set as 5 and 10 μm , respectively. The devices were thermally annealed in va-

Figure 1 (a) Schematic diagram of the time-dependent AP-CVD parameters. SEM images of graphene grown on (b) top side and (c) back side of the Cu foil. (d) Optical image of the top side graphene transferred on SiO₂/Si. (e) Corresponding Raman spectra of SLG (black) and BLG (red) recorded at the spots marked in (d). Raman 2D bands of (f) SLG and (g) BLG. The peak of BLG is well fitted with four Lorentzian functions (black dash lines).

cuum at 150°C for 40 min to improve the contact between BLG and the electrodes. Ionic liquid, 1-ethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide (99.9%, Solvionic) was used as the top gate dielectric material. A drop of ionic liquid was cast onto the substrate with graphene and the coplanar electrode to form the top gate. Electrical characterizations were performed with a Keysight B1500A semiconductor analyzer at room temperature. The effect of the dual-gate was investigated by measuring the current in vacuum (3×10^{-3} Torr) under a fixed drain bias (10 mV). The mobility of charge carriers was extracted with the equation:

$$\mu = \frac{dI_{DS}}{dV_{BG}} \cdot \frac{L}{W} \cdot \frac{1}{C_G V_{DS}},$$

where I_{DS} is the source-drain current, V_{BG} is the back-gate voltage, L and W are the device channel length and width, C_G is the gate capacitance per unit area (1.15×10^{-8} F cm⁻²), and V_{DS} is the voltage between source and drain electrodes.

RESULTS AND DISCUSSION

Carrier gas Ar was commonly used to exhaust air in the AP-CVD system. In previous studies, oxygen had been intentionally introduced and showed complicated effects on graphene growth [16,28–32], such as the control of nucleation density, acceleration of growth rate, alteration

of domain shape and modulation of number of layers. However, the amount of residual oxidative species in the CVD chamber and its effects on graphene growth are not quite clear. To clarify this, we adopted 300 mL min⁻¹ of Ar to degas the AP-CVD system for different times (t) before the growth of graphene. Generally, Ar flow started simultaneously with the furnace heating (for example, $t = 100$ min, denoted as Ar-100 min).

Continuous graphene films were obtained on the top side of the Cu foil following the growth procedure illustrated in Fig. 1a. However, the back side of Cu foil was only covered with partially grown graphene (Fig. S1b, c). As observed by SEM, SLG film accompanying ALG sheets were formed on the top side of the Cu foil (Fig. 1b), while discrete domains were found on the back side of Cu foil (Fig. 1c). Optical image of the top side graphene film transferred on a SiO₂/Si substrate shows large BLG domains of about 50 μm (Fig. 1d). Raman spectrum of the as-prepared graphene film was recorded. As shown in Fig. 1e, the disorder-activated D peak (~ 1350 cm⁻¹) is almost negligible compared with the sharp G (~ 1583 cm⁻¹) and 2D (~ 2690 cm⁻¹) peaks, which demonstrates high crystallinity with minimal defects [33,34]. The single Lorentzian profile of 2D band with a full width at half-maximum (FWHM) of 35 cm⁻¹ (Fig. 1f) and high-intensity ratio of 2D/G ($I_{2D}/I_G > 2$) indicates the typical monolayer structure for the black spot marked in

Fig. 1d. Raman spectrum of the red spot marked in Fig. 1d shows the typical feature of AB-stacked BLG. The FWHM of the 2D peak of the red spot marked in Fig. 1d is about 50 cm^{-1} and the 2D band intensity is significantly reduced. Moreover, the 2D band is well fitted with four Lorentzian peaks centered at 2695, 2687, 2708 and 2731 cm^{-1} , which shows a typical AB-stacked BLG feature [33].

To understand the growth mechanism of BLG, a control experiment was performed by setting a fast heating process of Cu foil without any changes of other growth parameters, as shown in Fig. S2a. Interestingly, it turns out that uniform SLG film without obvious ALG forms on the top side of Cu foil (Fig. S2b). In addition, the back side of Cu foil is also nearly completely covered by SLG, as shown in Fig. S2c. The results demonstrate that the heating process of Cu foil has significant effects on the morphology of the as-prepared graphene. As discussed above, the key point of uniform SLG synthesis is the simultaneous growth of graphene on both sides of Cu foil [22]. Here, the fast heating process caused a rapidly starting of graphene growth once the Cu foil reached the reaction zone, when the conditions on both sides of the Cu foil were almost identical (Fig. S2d). For the slowly heating process, the Cu foil was gradually softened onto the quartz tube/carrier, as shown in Fig. 2a. The back side of Cu foil was consequently stacked on the quartz surface when it reached the growth temperature before the growth begins. This generated two unequal sides of the Cu foil, which is the key to prepare high-quality BLG. As further confirmed in Fig. S3, when a quartz shelf support was used to suspend Cu foil forming equal atmosphere on two sides with other growth parameters remaining un-

changed, only inhomogeneous graphene film with thick MLG formed on both sides of the Cu foil (Fig. S4).

Furthermore, the backside quartz tube/carrier is similar to a deliberately placed oxide support according to previous studies [23,31,35]. The backside quartz is determined to suppress the backside nucleation density of graphene, although the mechanism is debatable on the forming channel structure or oxygen release from the quartz [31,35]. This is why only discrete graphene domains are on the back side of Cu foil heated slowly with furnace. There are mainly two types of growth mechanisms of BLG: (i) a small fraction of carbon on the surface of Cu directly diffuses beneath the top graphene layer to nucleate BLG; (ii) carbon on the slower graphene growth side diffuses through the Cu to form BLG on the other side. Because of the barrier of top graphene layer, the growth rate of BLG based on surface diffusion is much lower [9,20]. Bulk diffusion hence dominates the growth of BLG. According to previous results, oxygen plays a key role for promoting BLG synthesis by accelerating the bulk diffusion process [16]. The backside quartz is found providing a continuous supply of oxygen during the CVD growth [31], which further contributes to the growth of the BLG domains.

Generally, the quartz tube/carrier as a backside oxide support suppresses the nucleation of the back-side graphene, and promotes carbon diffusing to the top side, significantly inducing large BLG growth on the top side of the flat Cu foil. In addition, due to the oxidation of Cu foil during the heating by residual oxidative species in AP-CVD system [36–38], the oxygen released from copper oxides also participates in the synthetic processes and generates huge impacts on the graphene synthesis.

Figure 2 (a) Photographs of a $4\times 4\text{ cm}^2$ Cu foil before and after heating. (b) Schematic illustrations of the growth mechanism for the BLG with the assistance of the back-side quartz tube/carrier.

Besides, the oxygen atoms in carbon source (acetone) are non-negligible, which might facilitate the crystallization of BLG [26,27]. Schematic illustration of the proposed mechanism of BLG growth is displayed in Fig. 2b: the solid green line with arrow represents the surface diffusion and the dashed green line with arrow represents the bulk diffusion of carbon atoms, while the red arrows indicate (i) the oxygen supplied by quartz, (ii) the surface copper oxide decomposition and (iii) the oxygenated functional groups in carbon source, respectively. Oxygen released from copper oxides helps graphene growth by promoting hydrocarbon dissociation [9,16,30]. Cu foils are oxidized at different levels with different times of Ar flow to exhaust the system residual oxygen, as illustrated in Fig. S5. The color changed gradually from yellow to

purple, indicating increased oxidation of Cu with shortened time of Ar flow. X-ray photoelectron spectroscopy (XPS) characterizations (Fig. S6) indicate that the copper oxides on the surface of Cu foil are mainly CuO. When the Ar flow time decreased from 300 to 40 min, the corresponding oxidation ratio of Cu increased from 2% to 46% (Table S1). Especially, when the Ar flow time was less than 100 min, the Cu oxidation ratio increased rapidly.

The increased amount of copper oxides was expected to further promote the growth of BLG. As shown in Fig. 3a, shortening the Ar flow time increases the amount of ALG on the top side of Cu foil. More carbon accumulations (strong contrast in optical images) deposited on the graphene films prepared with less Ar flow. A hexagonal

Figure 3 (a) Morphology characterizations of graphene grown with 100, 85, 70, and 40 min of Ar flow. From up to bottom, the SEM images of graphene on the top side of Cu foil, optical images of the top side graphene transferred on SiO₂/Si, and SEM images of graphene on the back side of Cu foil. (b) AFM image of a BLG domain. Inset shows the optical image (scale bar: 5 μm). (c) Corresponding height profile measured along the green dash line in (b). (d) The area percentage of BLG, TLG and MLG. (e) Correlation between the coverage of MLG and the Ar flow time.

BLG domain of about 20 μm was observed in the AFM image (Fig. 3b). The height of carbon accumulations in the center is about 2 nm higher than the BLG domain (Fig. 3c), which could be regarded as the undesired MLG. Moreover, with the decrease of the Ar flow time, the Cu foil surface becomes rougher, as shown in the SEM images. In particular, for the Ar-40 min sample the uneven morphology causes more wrinkles and breakages in the transferred graphene. Additionally, few back-side graphene domains were found for the Ar-40 min sample. Detailed statistics of the proportions of BLG, TLG and MLG are shown in Fig. 3d. The coverage of BLG are all around 45% with small variations, while the area percentage of the undesired TLG and MLG shows a clear increase with the decrease of Ar flow. As shown in Fig. 3e, the proportion of MLG is about 2% for the Ar-100 min samples, while it approaches 15% for the Ar-40 min samples. These results indicate that excessive copper oxides have little benefit for the growth of BLG, and even promote the growth of excess TLG and MLG, which sacrifices the morphology uniformity. Too much oxygen released from copper oxide promotes the pyrolysis of carbon precursors and accelerates the bulk diffusion of carbon [16]. This might cause a large amount of carbon atoms accumulating to TLG or MLG before reaching the BLG edges to form BLG.

Obviously, the optimization of the oxygen content

alone has reached its limit in the growth of large-area continuous BLG. Besides accelerating the growth rate to increase the size of BLG domains, high nucleation density also plays an important role for preparing large-area BLG films. Hydrogen is commonly introduced to maintain the catalytic activity of Cu and meanwhile to etch disorder carbon structure on the Cu surface, ensuring the growth of graphene. Because of the etching effect of hydrogen on graphene at high temperature, the nucleation of graphene was also suppressed [39]. The nucleation density of graphene was low at hydrogen-rich atmosphere during the CVD process [40,41]. ALG was found sharing the same nucleation center with the top SLG on Cu [20]. To synthesize large BLG films, the hydrogen flow rate was reduced to 10 mL min^{-1} . As shown in Fig. 4a, when the Ar flow is 100 min, reducing the hydrogen flow rate significantly increases the nucleation of ALG. The productivity comparison of different layers of graphene grown under 20 and 10 mL min^{-1} of hydrogen is shown in Fig. 4b. The productivities of BLG, TLG and MLG were all improved at low hydrogen flow rate, and the corresponding proportion of SLG decreased by about 30%. The total coverage of BLG and ALG reached over 85%. As discussed above, the growth of excess MLG might be related to the excessive residual oxidative species in the AP-CVD system. To further improve the uniformity of BLG, the Ar flow time was increased. For the Ar-160 min

Figure 4 (a) Optical images of the top side graphene grown with 100, 160, and 300 min of Ar flow (H_2 flow: 10 mL min^{-1}). (b) The coverage comparison of SLG, BLG, TLG, MLG with 10 and 20 mL min^{-1} of H_2 flow (Ar flow: 100 min). (c) Correlation between the coverage of TLG and MLG and the Ar flow time.

samples, the growth of MLG was reduced, and BLG coverage in the graphene films was about 76% (Table S2). Further increasing the Ar flow time to 300 min sharply suppressed the growth of ALG, forming SLG dominated films. The area percentage of TLG and MLG also decreased with the increase of Ar flow at low hydrogen flow rates. The results further confirm the important effect of residual oxygen in the CVD system on the growth of BLG.

Therefore, the preparation of large continuous BLG films requires sufficient nucleation sites and enough growth rates. The growth kinetics is a complex interaction between carbon precursor, catalyst, oxygen and hydrogen. By adjusting the growth parameters to balance

the effects of hydrogen and system residual oxygen, the productivity and uniformity of BLG can be effectively optimized. Raman mapping and TEM characterizations were conducted to demonstrate the quality of the obtained BLG. As shown in Fig. 5a, a clear straight BLG edge with optimal crystallinity is in the high-resolution TEM (HRTEM) image. Corresponding selected-area electron diffraction (SAED) pattern shown in Fig. 5b displays a typical hexagonal crystalline structure. The intensity of {1100} diffraction spot is significantly lower than {2110}, further demonstrating the BLG feature [42]. Raman mapping was measured over a randomly selected $20 \times 20 \mu\text{m}^2$ region of BLG. The corresponding optical image and typical Raman spectra are shown in Fig. S7.

Figure 5 (a) HRTEM image and (b) SAED pattern of BLG. The inset shows the corresponding intensity profile of diffraction spots indicated by the green dash line. (c–f) Raman mappings of G peak intensity, the intensity ratio of D/G, 2D/G and 2D peak FWHM measured over a $20 \times 20 \mu\text{m}^2$ region (scale bars: $2 \mu\text{m}$). (g) Schematic illustration and (h) transport characteristic of the dual-gated BLG FET device. The inset shows the optical image of the device (scale bar: $5 \mu\text{m}$). BLG is indicated by a black dashed square. (i) Series of the channel resistance R plotted as a function of the top-gate voltage V_{TG} at different fixed back-gate voltages V_{BG} from -20 to 20 V, with 10 V increment.

The intensity of G band (I_G) is uniformly distributed around 200 in Fig. 5c, indicating an optimal continuity. The undetectable I_D/I_G ratio shown in Fig. 5d indicates a low density of defect. The I_{2D}/I_G values are mainly between 0.5 and 1 (Fig. 5e), and the FWHM of the 2D band in Fig. 5f is about 50 cm^{-1} , corresponding to a high-quality continuous AB stacked BLG. To further analyze the stacking order of the BLG film, Raman measurements were conducted on randomly selected fifty BLG regions (Fig. S8). The results show that 75% of the I_{2D}/I_G ratio are ~ 1 , and 62% of 2D FWHM are $\sim 50\text{ cm}^{-1}$. The scatter plot where the y -axis is the FWHM of the 2D peak and the x -axis is the I_{2D}/I_G ratio (Fig. S8d) indicates 60% of the BLG are AB-stacked.

As shown in Fig. 5g, a BLG-based dual-gate FET was fabricated on a 300 nm SiO_2/Si substrate with Cr/Au as the source/drain electrode, SiO_2 as the back-gate dielectric and ionic-liquid forming the top gate. Electrical measurements were performed on the device and Fig. 5h shows plots of I_{DS} versus V_{BG} of the device. The carrier mobility reaches $6790\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ at room temperature, which is better than most of previously reported CVD BLG [8,10,11,17,25,43] and mechanically exfoliated BLG [1,44] (500 to $5000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$). A perpendicular electric field on the BLG was used to demonstrate the bandgap tunability by adjusting the applied voltages to the two gate electrodes. When sweeping the top gate voltage (V_{TG}) from -2 to 2 V with V_{BG} fixed between -20 to 20 V , the channel resistance (R) versus V_{TG} transfer curves varied with different V_{BG} (Fig. 5i), which was consistent with the behavior of AB stacked BLG devices [8,10,17]. Due to the use of ionic liquid as the top-gate dielectric, a typical gate hysteresis behavior [8] between two consecutive transfer characteristics (I_{DS} - V_{BG}) was found in the device (Fig. S9a). This strong doping effect induced by ionic liquid can be inhibited by delaying the second transfer characteristics scanning after 1 h of the first scan, as shown in Fig. S9b, which demonstrates the reliability of the dual-gate devices. The characterization results reveal the high quality of the BLG prepared by AP-CVD.

CONCLUSIONS

In summary, by comprehensively understanding the growth process in AP-CVD, we developed a simple yet efficient approach to synthesize BLG on Cu foil which was softened and stacked onto the back-side quartz during the heating up process. The quartz significantly promoted the formation of large BLG domains on the top side of Cu foil. Oxidized Cu foil by the residual oxygen was a key accelerator for the BLG growth. Reduction with

hydrogen increased the nucleation density, and thus optimized the uniformity of BLG. Finally, large area continuous graphene films with a BLG coverage of 76% were obtained by optimizing residual oxygen content and nucleation density. The BLG exhibited tunable bandgaps with high carrier mobility up to $6790\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ at room temperature. The investigation of BLG growth in this study will be helpful for the controllable synthesis of graphene with specific number of layers.

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常压化学气相沉积法在软化铜箔上生长高质量双层石墨烯

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摘要 双层石墨烯因其独特的物理性质在新型电子器件等领域具有广阔的应用前景. 大面积高质量双层石墨烯的批量化制备是实现其后续应用的关键. 目前, 基于铜表面自限制催化的化学气相沉积法可有效实现单层石墨烯的生长, 但由于第二层石墨烯结构导致更复杂的生长过程, 双层石墨烯的可控制备极具挑战性. 本文系统研究了石墨烯的常压化学气相沉积制备过程, 提出了一种在软化铜箔基底上生长高质量双层石墨烯的方法. 铜箔在随炉升温过程中软化并贴合至背面的石英舟/管表面, 形成具有差异反应条件的铜箔双面, 从而促使双层石墨烯在其正面生长. 反应系统中的残余氧气有效加快了双层石墨烯的生长. 同时, 适量的残余氧气可抑制三层及少数层石墨烯的形成, 提高双层石墨烯产物的均匀性. 基于优化的生长条件, 在 $4\times 4\text{ cm}^2$ 铜箔上实现了双层覆盖率达76%的高质量石墨烯薄膜的生长. 基于AB堆垛双层石墨烯的双栅场效应晶体管, 室温载流子迁移率达 $6790\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$. 本研究有助于推动石墨烯等二维材料的层数可控合成技术的发展.