Controlled Growth of Graphene by Chemical Vapor Deposition

Interim Report

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Abstract

Graphene, firstly isolated in 2004, is a new type of carbon materials, which contains a single- or few-layered sheet of Sp2-bonded carbon atoms. This special atomic structure gives graphene rich physical properties and wide potential applications. It has excellent electrical, mechanical, thermal and optical properties and has wide applications in nano-electronic devices, transparent conductive films, power storage, composites materials et al. In our program, CVD method is used to fabricate large area and high quality graphene. The growth mechanism of graphene on different metal substrates is also investigated.

Experiment

Graphene Growing Steps

At first, the copper was put into the quartz tubes. Secondly, the growing time, the reacting temperature and the gas proportion were fixed. Next, the gas pressure –was adjusted by vacuuming. Then the samples were heated under H₂. When the temperature was up to the setup point, CH₄ and H₂ with the specific proportion in the tube to reacted at the specific growing time. After the end of the reaction, the samples were cooled under H₂. When the temperature was the same with room temperature, the copper was taken out. Finally, the graphene film grew on the copper.

Graphene Transference

In this experiment, the graphene film was transferred onto SiO₂/Si substrates by corrosion method. First, the PMMA was applied on copper to protect the graphene film. Next, the copper was put into the solution of FeCl₃. When the copper is disappeared completely, the films were put into the
clean water. Then they were transferred onto the SiO2/Si substrates.

**Controlling the Parameter**

Four main parameters including reacting temperature, gas proportion, growing time and reacting gas pressure affect the graphene structure. And we controlled the proportion of CH₄ and H₂ to prepare the graphene in this experiment.

![Figure 1](image1.png)

**Figure 1** Picture of CVD Furnace

**Characterization of Raman Spectrum**

Raman spectrum is used to investigate the layer number, defects and etc. The intension of peak D shows the defects of the sample, while it is the basic condition to prepare the single-layer graphene film whether the intention of peak G’ is much higher than the intention of peak G.

![Figure 2](image2.png)

**Figure 2** Horiba Jobin Yvon LabRAM HR800
Measuring Optical Properties

We measured the light transmittance of the graphene film by transferring the graphene to glass substrates.

Figure 3  UV-PDA Spectrophotometer

Electrical Properties

After transferring the graphene to SiO₂/Si substrates, we measured the conductivity of the graphene film by four point method.

Figure 4  Four-point-method Film Resistance Meter
**Current Progress**

Large area, high-quality graphene with good controllability on copper has been prepared by the CVD growth method.

Figure 5 shows the optical images of graphene films in different proportion of CH₄ and H₂. It is concluded that when the proportion of H₂ increases, the points on the sample are lighter and fewer. There are no points on the graphene films when the proportion of CH₄ and H₂ is 40:70.

![Optical Image of Graphene Film (1000°C, 1 Tor)](image)

Figure 5   Optical Image of Graphene Film (1000°C, 1 Tor)

(a) CH₄:H₂=60:50      (b) CH₄:H₂=60:55     (c) CH₄:H₂=40:60      (d) CH₄:H₂=40:70

Figure 6 gives the Raman spectrum of the graphene film with no points. It is seen from the spectrum that the measured graphene film is very thin with little defect.
Some of the samples are transferred well to study its optical properties. The light transmittance of the sample is 97% in visible range as shown in Figure 7, which is very close to 97.7%, the theoretical value of the single-layer graphene film’s light transmittance. So the sample is good in quality.

Figure 6  Raman Spectrum of Graphene Film ($1000^\circ \text{C}$, $\text{CH}_4: \text{H}_2 = 40:70$, 1 Torr)

Figure 7  Light Transmittance Image of Single-layer Graphene Film (300-800nm)
Figure 8 shows the picture of two samples for measuring resistivity. It is found that the resistivity of the left film is $23.1 \times 10^{-6} \, \Omega \cdot \text{cm}$, while the resistivity of another one is $9.11 \times 10^{-6} \, \Omega \cdot \text{cm}$. Their orders of magnitude are the same as the ideal graphene’s resistivity.

![Figure 8 Picture of Two Samples for Measuring Resistivity](image)

**Future work**

Based on the fact that graphene can be grown on the top of the copper surface and the copper sputtered onto the oxide silicon, it is predicted that graphene may form under the copper film. In this case, we plan to change the four parameters to get high-quality and continuous graphene. As thicker copper film may lead to amorphous carbon film, a proper copper thickness will result in a mono-layer graphene. In addition, if PMMA will not be used, we may get a cleaner graphene. The superior properties of graphene are beneficial for all electronic devices.
**Financial Statement**

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