Graphene Synthesized by Microwave Plasma-Enhanced CVD

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Received 4 March 2017; Revised 6 September 2017; Accepted 10 September 2017

Abstract

This paper presents the synthesis of graphene by using microwave plasma-enhanced CVD on copper metal substrate. The graphene film was successfully created inside microwave oven by using the vapor of ethanol in atmosphere of argon. Scanning electron microscope and transmission electron microscope were used to observe the surface and morphology of created graphene film. UV-Vis spectra of graphene exhibit maximum absorption peak at about 243 nm attributable to $\pi - \pi^*$ transition of the atomic C-C bonds. Raman spectroscopy was used to measure the three major bands of graphene. The D, G and 2D-band located around 1,344, 1,573 and 2,679 cm^{-1}, respectively. The ratio of band intensity of 2D/G is about 1, it means that the graphene film has at least two layers.

KEYWORDS: Graphene; Microwave Enhanced Plasma CVD (MEPCVD); Scanning electron microscope; Transmission electron microscope; UV-Visible spectroscopy; Raman spectroscopy

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Introduction

Graphene is a new and an important material and one of the most expanding disciplines of contemporary research in applied physics and material science. A wide potential application of graphene in various areas have been utilized such as electronics, composite materials, sensors, energy storage, solar cell, fuel cells and etc. It is a two-dimensional sheet of sp^2-hybridized carbon nanomaterial which exhibits outstanding optics, electronics and mechanics properties. The Nobel Prize for physics in 2010 was awarded to Sir Professor Andrei Geim and Sir Professor Kostya Novosclov, from Manchester University, for their ground-breaking experiments regarding two-dimensional material graphene. Generally, the graphene community distinguishes between single-layer, bilayer and few-layer graphene, the latter of which refers to graphene with a number of less than 10, in the case more than 10 layers graphene, it can be considered as a graphite thin film [1]. Various methods have been reported for the synthesis exfoliation, such as mechanics exfoliation, chemical exfoliation and thermal decomposition. Since 2010, most notably advances have been made in the fabrication of large sheets of graphene by chemical vapor deposition (CVD) method. Currently CVD is most efficient in term of the energy consumption and yield technique for graphene synthesis [2 – 11]. In this work, we show indeed microwave plasma-enhanced CVD (MEPCVD) could be effective technique to deposit graphene sheet on copper substrate by employing an ethanol/ argon atmosphere in a few minutes.

In Raman spectroscopy of graphene, there are three response peaks of interest which correspond to different phonon modes in material. The D peak intensity (I_D) is used to consider the degree of general graphene disorder. The 2D peak (I_{2D}) is a harmonic of the D peak and the intensity ratio of 2D to G peak (I_{2D}/I_G) is used to characterize the number of graphene layer as shown in Table 1.

Table 1 Adopted criteria for graphene sample evaluation [2, 12].

I_D/I_G	I_{2D}/I_G	FWHM (cm^{-1})	Defect level	Number of layers
0.25	2	20 - 40	Low defect	1 (monolayer)
0.25 - 0.5	0.9 - 2	40 - 60	Medium defect	2 (bi-layer)
0.5	0.9	40 - 60	High defect	> 3 (Multilayer)

Materials and Methods

The synthesis was carried out using a MEPCVD setup. The construction of this system has been described [13]. The graphene sheet was synthesized on copper metal substrate directly. Schematic of the MEPCVD system used for the synthesis of graphene is shown in Fig.1. The MEPCVD is produced in reactor chamber by introducing 2.45 GHz of microwave frequency. The graphene film growth on the copper surface was obtained in 90 seconds.

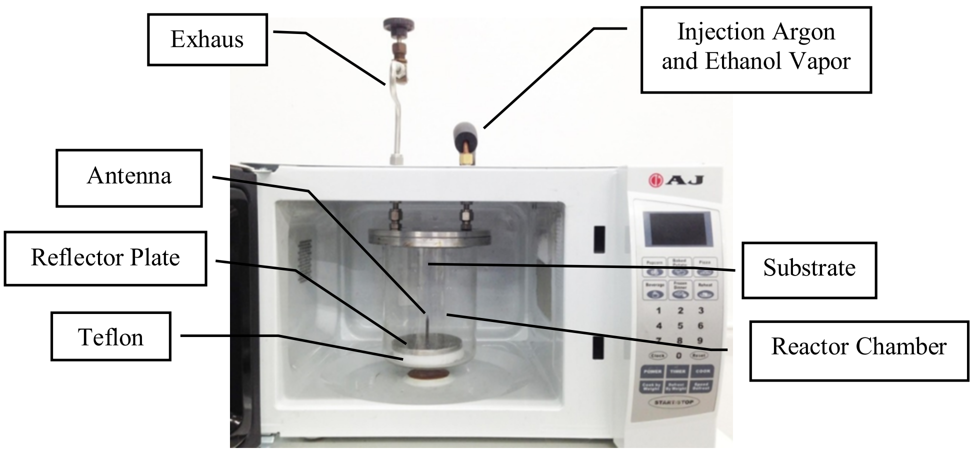

Fig. 1 Schematic of MEPCVD system for used the synthesis of graphene [13].

A mixture of ethanol vapor and argon gas was used as reactants. The microwave power was maintained at 800 W during growth. The graphene sheets were characterized by using scanning electron microscope (SEM), LEO model LEO1450VP, transmission electron microscope (TEM) was using Philips model TECNAI 20, UV-Visible spectrometer, Thermo Scientific model GENESYS 10S UV-Vis Spectrophotometer and visible Raman spectroscopy with a 532 nm laser wavelength.

Results and Discussion

During the growth process, vapor of ethanol was carried by itself to the reactor chamber with argon. After 90 seconds of reaction time, the reactor was cooled to room temperature. The prepared sample was moved from the reactor and used directly for characterization. SEM photograph was taken at room temperature. Fig. 2 shows the graphene surface by the SEM. Fig. 3 shows the graphene film taken by TEM. The grain size of film under argon atmosphere was measured by using program image-J, the diameter is 0.32 ± 0.03.

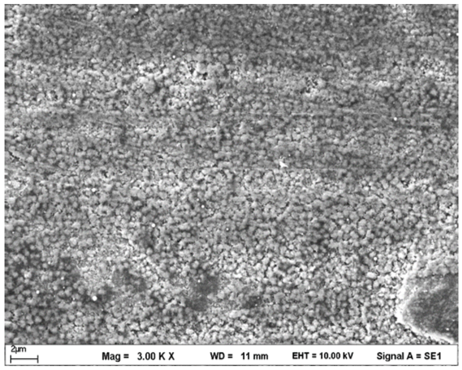

Fig. 2 SEM image of a graphene on a Cu substrate

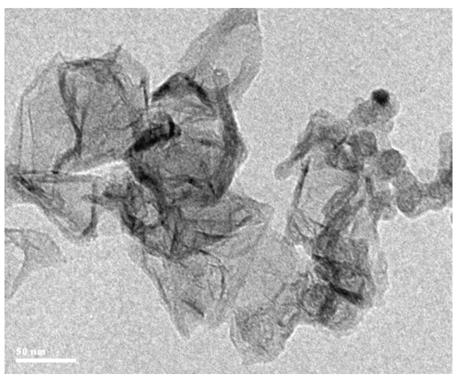

Fig. 3 TEM of sample graphene sheet

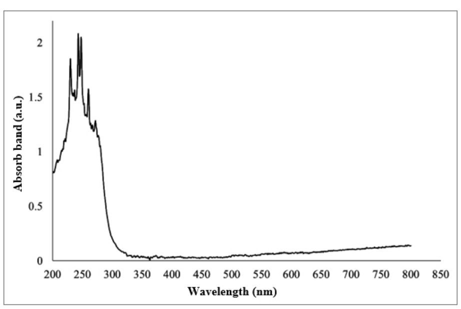

Fig. 4 UV-Vis spectrum

From the Fig. 4, it is observed that graphene shows maximum absorption peak at ~ 243 nm attributable to $\pi-\pi^*$ transition of the atomic C-C bonds.

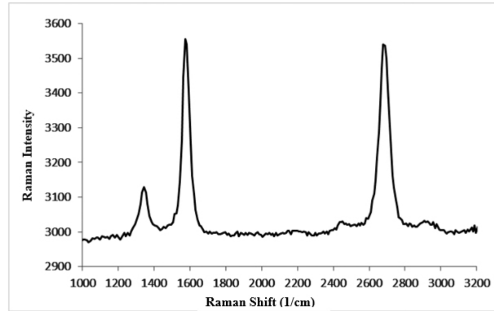

Fig. 5 Raman spectrum with 532 nm excitation laser wavelength

Fig. 5 shows a Raman spectrum of prepared sample performed at room temperature. It can be seen that there are three peaks centered at 1,344.28, 1,573.72 and 2,687.71 cm^{-1}. These are identified as the Raman fingerprints of graphene namely D, G and 2D (named traditionally as G*) peaks. There is G* peak appeared at 2,420 cm^{-1}. For the prepared sample, ratio of I_D/I_G = 0.24, I_{2D}/I_G = 1.064, therefore it can be determined that the graphene film has medium defect level. Correspond with the value of the full with at half maximum (FWHM) of 2D peak of 60, it can be classified that the prepared graphene has 1 – 2 layers.

Conclusion

In summary, UV- Vis spectra of graphene exhibit maximum absorption peak at ~243 nm attributable to $\pi-\pi^*$ transition of the atomic C-C bonds. Raman spectroscopy was used to measure the three major bands of graphene. The D, G and 2D-band located around 1344, 1573 and 2679 cm^{-1}, respectively. The ratio of band intensity of 2D/G is about 1, it means that the graphene film has at least two layers. it can be concluded that synthesized graphene sheet 1–2 layers can be prepared by MEPCVD in a mixture of ethanol and argon gas on copper substrate directly at low temperature (500 – 700 °C). The graphene sheets can be possibly grown in a short process within 90 seconds.

Acknowledgements

The authors gratefully acknowledge the financial support provided by Thammasat University Research Fund under the TU Research Scholar, Contract No.TN 22/2558

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