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Variation of Band Gap in Graphene Grown by Plasma Enhanced Chemical Vapor Deposition

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Abstract

We report the transport behavior of graphene grown by plasma enhanced chemical vapor deposition (PECVD). The graphene films were grown at 950 °C for 5, 10, 30 and 60min, respectively. Raman spectra showed that the synthesized films are bilayer with strong defect peaks. The temperature dependent conductivity of graphene films showed the band gap modulation with increasing growth times. Thermally activated (TA) conduction model showed that the values of band gap of graphene films are 95, 73, 48 and 36meV for 5, 10, 30 and 60min growth times, respectively.

Keywords: Graphene; Raman; Band gap; PECVD; Defect

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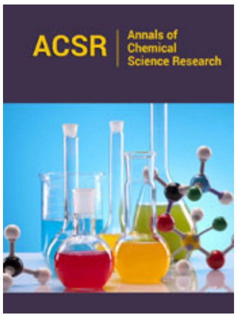

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Introduction

Graphene is a two-dimensional monolayer with linear dispersion relation near the Dirac point and has attracted much interest owing to its superb material properties, such as very high mobility of 10,000 cm^2/Vs at room temperature and quantum hall effect to promise for the potential in nanoelectronics [1-4]. However, graphene is a semimetal without an energy band gap which is required to be used as an electronic transistor. Recent studies have shown that band gap is formed at various situation, in nanoribbons, [5] bias-applied, and [6] strained or molecule-doped graphene [7,8]. Though the band gaps were generated in various structures, the control of the electrical properties is still a challenge. On the other hand, defects and impurities in graphene are important in electrical transport, because scattering is a hurdle in carrier transport in graphene, making the modulation of graphene transport quite important for physical and device application. In particular, defects in graphene have been intensely studied because quasi-localized states near the Fermi energy can be induced due to vacancy [9,10]. Here, we report that band gap engineering has been achieved by PECVD growth and plasma treatment can induce modulation of the defective properties of graphene. The graphene films were synthesized by plasma-enhanced chemical vapor deposition (PECVD) at 950 °C on sapphire substrate without catalyst, and the radio frequency plasma power was fixed at 150W. Carrier gas was argon (300sccm) and hydrogen (30sccm). The hydrocarbon, CH_4 (1sccm) was used as a source precursor during the growth. Growth time was adjusted from 5min to 60min. Pristine graphene was identified as a bilayer using an optical microscope and Raman signal. Raman spectra of the graphene films were measured at an excitation of 514.5nm at room temperature using a spectrometer (Horiba Jobin-Yvon, HR800UV). The electrical transport was characterized as a function of temperature using low temperature measurement system (Sungwoo Instrument & Sumitomo).

Figure 1 shows the Raman spectra of graphene film grown on sapphire substrate for 5min by PECVD. The D band (defect) intensity of the graphene grown by PECVD is relatively very high compared with that of usual graphene grown by thermal CVD methods. The defect peaks were strong at 1350cm^{-1} due to plasma effect and the ratio of I2D/IG is approximately close to unity as shown in Figure 1, indicating that this sample is, in fact, the bilayer graphene [11].

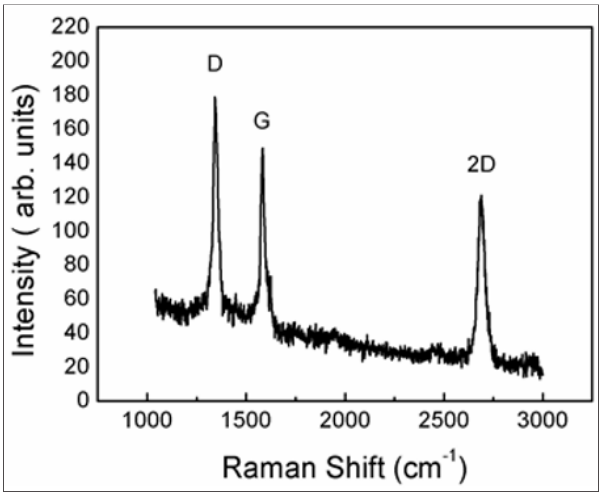

Figure 1: Raman spectra of graphene synthesized by PECVD for 5min.

Figure 2 shows the temperature dependent conductivity of graphene synthesized by PECVD for 5(a), 10(b), 30(c) and 60min(d), respectively. The conductivity increased with increasing temperature, like a usual semiconductor, for the graphene films grown at 950 °C. According to the previous work, the temperature-dependent conductivity of graphene can be well explained by the thermally activated (TA) conduction equation. The TA model can be described as: $\sigma=\sigma_0\exp(-E_g/2k_BT)$, where E_g is the band gap energy, k_B is the Boltzmann constant, and T is the absolute temperature in Kelvin, for high temperature regions over 100K, whereas variable range hopping mechanism is effective at low temperature [12]. The bandgap energy of 0.095eV was observed as shown in Figure 2 for the graphene synthesized for 5min, and then the value of band gap decreased gradually. It is well known that plasma irradiation generates many vacancies during the formation of graphene film [13]. Such vacancies in the graphene film synthesized by using PECVD result in defects. The origin of band gap formation is resulted from defects in the graphene [14]. Also, previous theoretical report predicted that the defects could induce a band gap in graphene. The formation of the band gap is owing to the breaking of sublattice and molecular symmetry in the graphene [15]. In this study, the vacancies in graphene could be generated during the growth by plasma radiation. The values of band gap of films are 95, 73, 48 and 36meV for 5, 10, 30 and 60min growth times, respectively. This means that the defects of graphene grown by PECVD were decreased as the growth time increases and the sample quality was improved by long time growth.

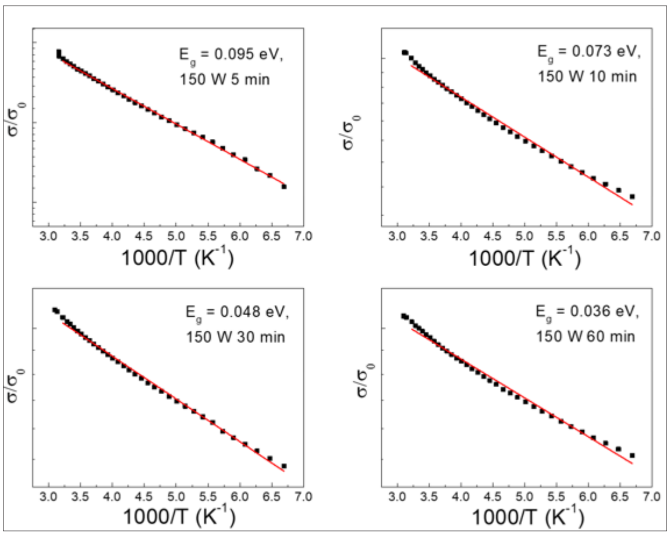

Figure 2: Temperature dependent conductivity of graphene synthesized by PECVD for 5(a), 10(b), 30(c) and 60(d) min, respectively. Red line means the fitting by conductivity equation.

Conclusion

In conclusion, we have studied the transport behaviors of graphene grown by PECVD. Band gap engineering was achieved in the temperature dependence of conductivity. TA conduction mechanism of transport was described for the band gap evaluation. TA conduction model showed that the values of band gap of films are 95, 73, 48 and 36meV for 5, 10, 30 and 60min growth times, respectively. Therefore, the band gap energy of graphene can be modulated by growth times, which enables graphene to have promising properties for electrical applications.

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