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Graphene Layers on Sapphire Substrates Grown by Alcohol CVD method

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1. Introduction

Graphene, a two-dimensional one-atom-thick of sp^2-bonded carbon atoms consisting of honeycomb crystal structure, has been attracted great interest due to its high electron mobility, thermal conductivity , mechanical properties and transparency. It is a promising candidate for FET transistors, transparent electrodes, liquid crystal display and solar cells as graphene applications. There are several methods for fabricating graphene including exfoliation of HOPG (high oriented pyrolysis graphite), SiC film decomposition at high temperature in vacuum, segregation and accumulation on catalyst metals by CVD (chemical vapour deposition) and so forth. Recently, optical properties of graphene have been well reported [1-3]. However, many of report have been discussed suspended graphene. In this report, we synthesized graphene layers on sapphire substrates and evaluate by the optical properties in conjunction with Raman spectra which is often used for evaluation of quality of graphene networks. The properties show excellent conductivity with high transparency. The result indicates the possibility of graphene application for transparent electrodes as a replacement of ITO (indium tin oxide).

2. Experimental

Graphene layers were deposited on as cut *a*-sapphire (11-20) substrates by Alcohol CVD technique under Ar and H_2 atmosphere at the pressure of 10 torr with the ethanol gas at a rate of 50sccm (sccm denotes standard cubic meter per minutes) at the temperature range of 600 to 1000°C for 15min. After the growth, Visible-Ultraviolet (UV-VIS) spectra transmittances measurement were carried out in the range of 300-700 nm wavelength to evaluate optical properties of graphene layers. Raman spectroscopy with excitation wavelength of 532 nm YAG laser was also employed to estimate the domain size of graphene network. Transmittance spectra of amorphous carbon films on sapphire substrates deposited using DC discharge sputtering at room temperature were also evaluated for the comparison purpose by the UV-VIS spectrophotometer. The thicknesses were controlled by times of DC discharge flashing. Sheet resistances of the synthesized graphene layers were measured by four probe method. Thickness of graphene layers were evaluated by AFM (atomic force microscopy). The layer number of the graphene sheets were estimated by dividing the thickness by the distance between each layer (c_o = 0.335 nm).

3. Results and discussion

Transmittance spectra of carbon films sputtered by DC discharge flashing, Fig. 1 (a-c) gradually decrease in the wavelength 700 to 300 nm range. On the other hand, transmittance spectra of carbon films synthesized by Alcohol CVD method show distinct difference from sputtered carbon films. The synthesized carbon film at 800°C, Fig 2. (e), holds the transmittance thorough 300-700nm. When the growth temperature is higher than 800°C, the spectra, Fig. 2 (d, f-h), hold the transmittances in the wavelength range of around 500-700nm and drastically decrease around 400nm. The property well corresponds to the reported optical property of graphene layers obtained by exfoliation technique[2]. These results show that growth temperature is crucial factor for the determination of optical properties of carbon films by CVD. It is assumed that to form six-membered ring structure of carbon atoms an amount of thermal energy is essential. Therefore, the transmittance spectrum of synthesized carbon film at the temperature of 800°C, Fig. 2 (e), might include defection such as five- and seven-membered ring since the spectrum does not show the graphene-like property. Here, note that carbon film synthesis by CVD was recognized not only the front side but also the back side of the substrates because flow of electricity was measured by four probe method, although the sheet resistances of back sides were much higher than that of the front sides. Thus, the transmittance properties are considered as sum of the carbon films on both sides. At this point, it is not adequate to estimate layer number of graphene by transmission attenuation. Fig.3 shows temperature dependence of Raman spectra of carbon films on sapphire substrates by CVD at the temperature range of 800-1000°C. Formation of graphene layers can be confirmed from the peaks of D-band (around 1350cm^{-1}) G-band (around 1600cm^{-1}) and 2D-band (around 2700cm^{-1}) at 850°C and over substrate temperatures. In contrast, weak intensities of D-,G- and 2D-band are observed at 800°C, thus the carbon film is considered as amorphous carbon. The results well correspond to the temperature dependence of opacity properties of carbon films in Fig. 2. Fig. 4 shows temperature dependence of the size of graphene network (La) by an estimation model proposed by Cançado, *et.al.*, [4]. La is inversely proportional to the intensity ratio of D-band and G-band (I_D/I_G). Larger graphene network was formed as the increase of substrate temperature. It is because honeycomb crystal structures of graphene networks highly crystallized with high thermal energy. In the same way, it is assumed that a certain amount of thermal energy is essential for carbon films to transform the structure form amorphous to crystalline. Fig. 5 shows the thickness of graphene sheet dependence of the sheet resistance. The sheet resistance decreases as an increase of the growth temperature. The synthesized graphene layers at 900°C shows 9.79 kΩ/sq with the transmittance at 84.6% at 550nm wavelength.

4. Conclusion

We performed graphene layers synthesis on sapphire substrates by alcohol CVD technique and show the validity of evaluation of the carbon film qualities by UV-VIS spectrophotometer in conjunction with Raman spectroscopy. It was shown that to synthesize graphene layers an amount of thermal energy was essential, thus in case of the scarce thermal energy, carbon films showed the transmittance property of an amorphous carbon. The results indicate the way of graphene application as transparent electrodes without any etching process.

Reference

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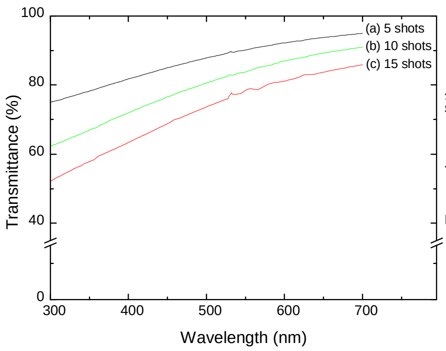

Fig.1 Tranmittance spectra of carbon films by DC discharge sputtering on sapphire substrates. Shots represent times of DC discharge flashing.

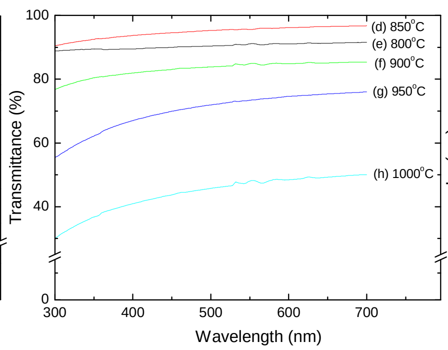

Fig2. Transmittance spectra of carbon films by Alcohol CVD on sapphire substrates at the 50sccm ehtanol gas flow for 15min.

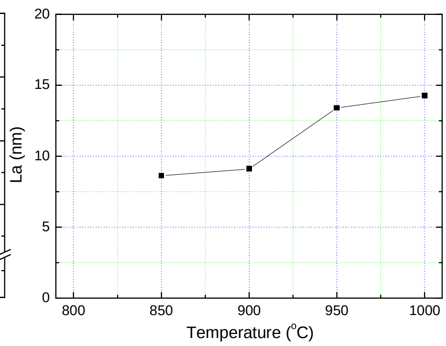

Fig.4 Temperature dependence of the domain size of graphene network (La) on sapphire substrates.

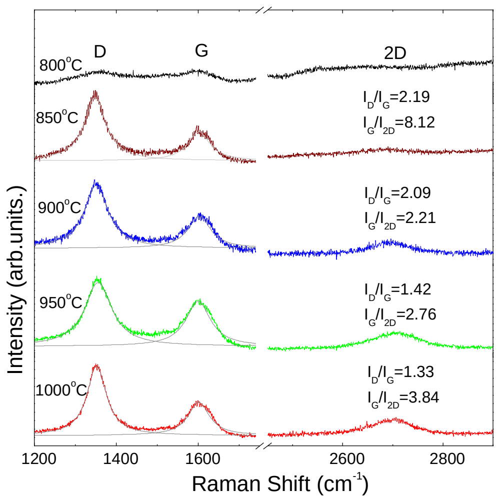

Fig.3 Temperature dependence of Raman spectra of carbon films on sapphire substrates at the 50sccm ehtanol gas flow for 15min.

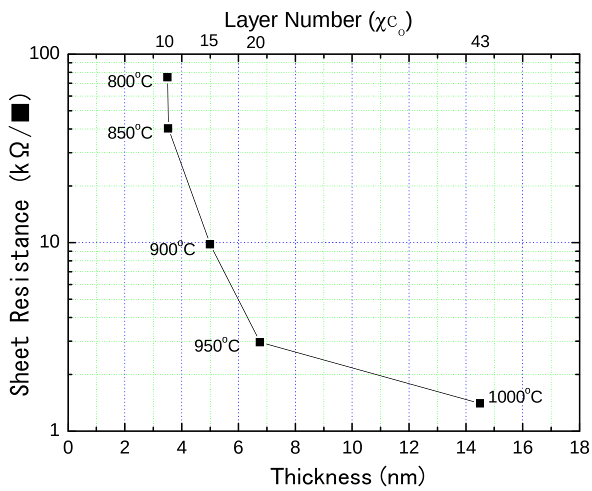

Fig.5 Thickness and number of layers dependence of sheet resistivity of carbon films on sapphire substrates.