PREPARATION AND CHARACTERIZATION OF TRANSPARENT AND CONDUCTING CARBON FILMS FROM SILYLATED GRAHITE OXIDE

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Introduction

Graphene based carbon thin films prepared from graphite oxide are promising for the transparent and conducting electrodes and many studies have been reported on them [1-12]. Graphite oxide thin films are usually prepared from the aqueous dispersion of it and then they are chemically or thermally reduced. The sheet resistance reaches 10^3 Ω/sq for 80% of transmittance, when the resulting films are appropriately treated [11]. However, it seems that the carbon films do not adhere well to the substrates because the interaction between carbon and substrates is weak. Therefore, we have recently introduced silylated graphite oxide as a starting material of the carbon thin films [13]. After the reduction of silylated graphite oxide film, organic components in the silylaitng reagents are removed and the silicon containing species remains at the interface between carbon and substrates. It worked as a "glue" and the carbon films well adhered to the glass substrates. In addition, the sheet resistance reached 700 Ω/sq for the film with 80 % of transmittance, when it was prepared at 900°C. It would be more interesting if the carbon films are obtained at lower temperatures.

In this study, therefore, graphene based carbon thin films were prepared from silylated graphite oxide thin films at lower temperatures than 100°C by UV light irradiation.

Experimental

Graphite oxide with the composition of $C_8O_{3.5}H_{2.4}$ was obtained from the synthetic graphite powder (Aldrich, 1-2 μm). It was silylated with octyltrichlorosilane, according to our previous papers [14,15]. Hereafter, the resulting sample is abbreviated as C_8SiGO. The composition of the C_8SiGO sample was $(C_8Si)_{0.72}GO$, based on the thermogravimetric analysis datum. Then, n-hexadecylamine molecules were intercalated into the resulting C_8SiGO sample. The product was dispersed in chloroform/cyclohexane (1:1 by volume) and homogenous solution was obtained. The resulting solution was cast on the quartz, glass or silicon substrates [16] and the solvent was evaporated slowly at 2°C. The n-hexadecylamine molecules were removed from the film samples by washing it with ethanol. The obtained C_8SiGO films were reduced by irradiating UV light using an ultra high pressure UV lamp (USHIO-500D) for 0.5-24 h. The distance of the sample from the lamp was about 5 cm. During irradiation of UV light, the temperature was lower than 100°C. For comparison, these films were pyrolyzed at 500°C under vacuum for 5h. The films samples were analyzed by X-ray diffractometry, IR spectroscopy and UV-Vis spectroscopy. The sheet resistance of the samples was measured by a potential sweep method at the sweep rate of 1 mV/s.

Results and Discussion

Fig.1 shows the UV-Vis spectra of C_8SiGO film and those after irradiation of UV light for various times. The absorption peak at 300 nm due to the carbonyl groups disappeared after 15 min of irradiation. The absorption peak at 223 nm observed for pristine C_8SiGO gradually shifted to longer wavelength and absorbance increased with the increase in the irradiation time. After 24h, the absorption peak reached 260 nm, which was similar to that observed for the C_8SiGO film heated at 500°C. This strongly indicates that the GO layer was reduced and π conjugating system is considerably recovered. The peak was sharper than those observed for C_8SiGO films reduced thermally or by hydrazine. Base on the Tauc's plot, the optical band gap decreased from 1.7 to 0.5 eV.

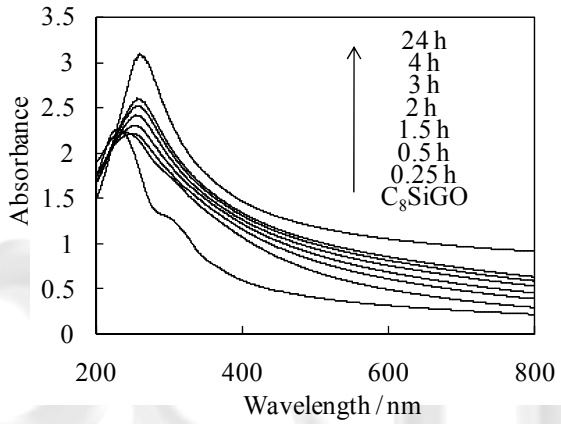

Fig. 1 UV-Vis spectra of C_8SiGO thin films before and after UV light irradiation.

Fig.2 shows the X-ray diffraction patterns of thick C_8SiGO film before and after UV irradiation for 24h. The diffraction peak at 2θ=5.66° (d=1.56 nm) observed for the pristine C_8SiGO film shifted to a higher angle of 2θ=10.94° (d=0.81 nm) after UV irradiation for 24h. This interlayer spacing was much larger than that of the graphite or the carbon films prepared, which means that some intercalated species remains between the carbon layers.

In the IR spectrum of the C_8SiGO film after UV irradiation, the absorption peak at 2920 and 1020 cm^{-1} due to the methylene groups in silylating reagents attached to the GO layers and epoxide groups on GO layers observed for the pristine C_8SiGO almost disappeared. This indicates that not only the reduction of oxygen containing groups on GO layers but also the removal of alkyl groups occurred. Considering the large interlayer spacing of 0.81 nm observed after UV irradiation, some silicon containing species should remain between the carbon layers. These suggest that the structure of the resulting material would be similar to that of the pillared

carbons which we prepared from highly silylated GO [17-19], though the silicon content was lower.

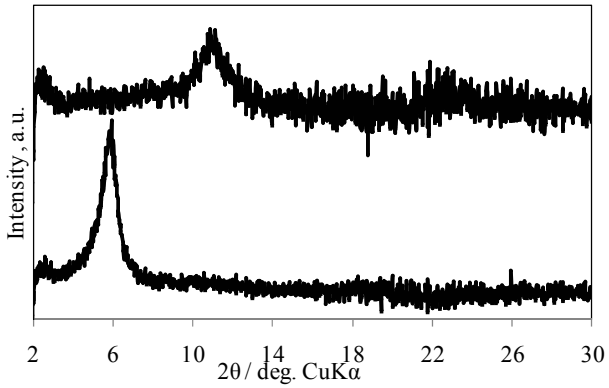

Fig. 2 X-ray diffraction patterns of C_8SiGO thin films before and after UV light irradiation for 24h.

Fig.3 shows the sheet resistance of the C_8SiGO films reduced by UV light irradiation for 24h as a function of transparency at 400 nm of them, together with those of the carbon films obtained from the pyrolysis of C_8SiGO at 500°C. It slightly decreased with the decrease in the transparency as was observed for thermally reduced films, however, the change was more moderate. The sheet resistance for the film with 80 % of transparency was slightly higher than 10 kΩ/sq.

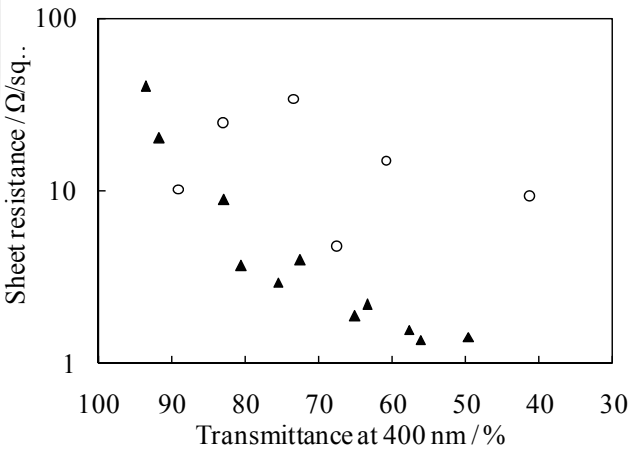

Fig. 3 Relationship between the transmittance at 400 nm and the sheet resistance of the carbon film obtained from C_8SiGO film: ○, together with those for C_8SiGO films heated at 500°C: ▲.

Conclusions

Silylated graphite oxide thin films were reduced by the UV light irradiation at the temperatures less than 100°C. It seems that GO layers were considerably reduced, though the interlayer spacing was still very large, 0.81 nm, indicating that some silicon containing species remained between the carbon layers. The sheet resistance of the resulting films was slightly higher than 10 kΩ/sq.

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