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SYNTHESIS OF GRAPHITE ULTRATHIN FILMS FROM SUCROSE

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Introduction

High quality graphite films, which are produced by carbonization and graphitization of polyimide film Kapton, are used as a heat dissipation material for the potable digital devices because of its flexibility, light weight and high thermal conductivity [1]. We have pursued the synthesis of graphite ultrathin films with thickness less than few hundreds nanometers using different types of precursor polymer films with spin coating process [2]. During the investigation, it was found that several precursor polymers for non-graphitizing carbons could give highly graphitized ultrathin films when the spin coated precursor polymers were carbonized with substrate to avoid the shrinkage. This phenomenon considered to be similar with the stress graphitization in the C/C composite fabrication using non-graphitizing matrix precursor.

In this study, we have examined a sucrose as a precursor polymer which is well known to give non-graphitizing carbon to produce graphite ultrathin films by applying the spin coating process and successive carbonization with substrate.

Materials and Methods

Sucrose (reagent grade) was dissolved in purified water (Millipore). The solution was heated at 50 ℃ and spin coated (1000-4000 rpm for 3sec) on a quartz glass substrate (2 inch) and heat treated in air at 180 ℃ for 2 hr (ramping rate of 1 ℃/min) for stabilization (caramelization). The stabilized sucrose film on the substrate was heat treated at 600 ℃ in nitrogen flow. Then, the carbonized film was peeled off from a substrate and further heat treated in between graphite plates under argon atmosphere. The samples obtained were characterized by XRD, SEM and Raman scattering measurements. The thickness and the electrical conductivity of the film was evaluated by the contact profilometer (Burker, Dektak) and the 4 points method, respectively.

Results and Discussion

Figure 1 shows photographs of sucrose film with different heat treatment temperatures. The original sucrose solution and spin-coating film on a quartz substrate were perfectly no colour and transparent. After the heating in air at 180 ℃, the film colour changed to right brown due to the caramelization of sucrose (**Fig. 1a**). Then, the film on a substrate was carbonized at 600 ℃ in nitrogen flow and become more brownish since the film was so thin so that one might not recognize black carbon colour (**Fig. 1b**). The carbonized film was peeled off from the quartz substrate after the making protecting PMMA layer. The carbon/PMMA film was shaping into 2cm square and further heat treated in between two graphite plates under argon atmosphere up to 2800 ℃ (**Fig. 1c**). X-ray diffraction lines (CuKα radiation) with 00*l* indices in reflection configuration to the film surface are shown in **Figure .2**. All 00*l* diffractions are very strong and split by Kα_1 and Kα_2

incident X-ray, indicating the markedly developed coherent length along the *c*-axis of graphite crystallite. The *hkl* diffractions with non-zero *h* and *k* were not observed by the same diffractogram, implying the high degree of preferential orientation of crystallite to the film surface. The thickness of graphitized film obtained was typically around 200 nm from 1g ml^{-1} sucrose solution and the in-plane electrical conductivity was about 12900 S cm^{-1}.

Figure 1 Photographs of the film produced from sucrose solution: a) caramelized with substrate at 180 ℃ in air, b) carbonized with substrate at 600 ℃ in nitrogen and c) graphitized in between graphite plates under argon atmosphere at 2800 ℃.

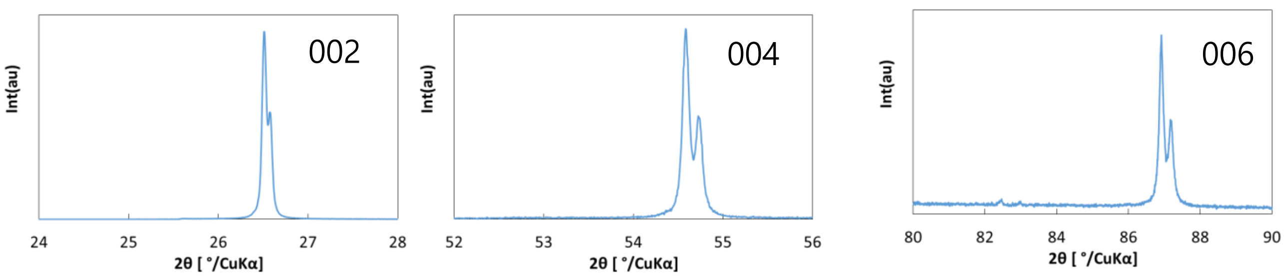

Figure 2 X-ray diffraction lines with 00L indices from the reflection configuration to the graphitized film surface.

From the SEM observation, electron channelling contrast (ECC) images were able to seen on the graphitized film surface, which is due to the flat surface and markedly grown graphite crystallite in carbon layer plane direction. The in-plane crystallite size evaluated by ECC reached ca. 1 μm for graphitized film with HTT2800.

Conclusions

Graphite ultrathin films with high degree of crystallinity and preferential orientation were synthesized from spin-coated sucrose film. It was suggested that the carbonization on the substrate resulted in the stress graphitization phenomenon even for typical non-graphitizing carbon precursor.

References

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