Selective Graphene Growth from DLC Thin Film Patterned by Focused-ion-beam Chemical Vapor Deposition

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Graphene, a single layer of graphite, has been gathering attention of researchers throughout the world because of outstanding electrical and mechanical properties. A large-scale graphene with high quality is fabricated by two major methods: decomposition of SiC and chemical vapor deposition on metal such as Ni and Cu. The former method is, however, limited to SiC substrate, and the latter method requires etching and transfer process owing to metal layer. Furthermore, both the methods need lithography and etching processes to obtain proper shape for devices. Thus, a simpler and substrate independent process is required. Here, we report the patterned graphene growth from diamond-like carbon (DLC) using focused-ion-beam chemical vapor deposition (FIB-CVD) and gallium catalyst.

Figure 1 shows our process to fabricate graphene. Firstly, DLC was deposited on Si substrate with 280 nm SiO_2 by FIB-CVD which can form arbitrary two dimensional DLC structure. Secondly, gallium was evaporated and deposited onto the substrate. The thickness of gallium was 40 nm. And lastly, the substrate was annealed at 900-1100°C under vacuum. While annealing, Ga graphitized DLC and evaporated. Graphitization was confirmed using Raman spectroscopy.

As shown in Fig. 2, square pattern annealed at 1100°C had 2D-band which represents the presence of sp^2-bonded carbons. In contrast, Raman spectra of DLC fabricated by FIB-CVD and annealed DLC did not have 2D-band as shown in Fig. 3. Hence, Ga had catalytic effect on graphitization of DLC. Although carbon is generally insoluble in gallium, it has been reported that reaction between liquid Ga and amorphous carbon occurs at narrow interface region, and Ga works as a catalyst to form thin graphite layers[1]. Therefore, our annealing process changed gallium into liquid which reacts with DLC for graphitization and finally evaporate. It indicates that graphitization would not occur when Ga is evaporated without getting liquid state.

Figure 3 shows Raman spectra of graphene sheets with different thickness. All samples were annealed at 1100°C for 30 minutes. We found that thicker DLC film tends to become graphene with better quality. It is attributed that the amount of carbon in thin DLC film was not enough to rearrange graphite structure. Figure 4 shows minimum annealing time to graphitize DLC. We also succeeded in graphitization at 900, 1000 and 1100°C. We will report detailed relationship between thickness of graphene and graphitization.

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[1] J. Fujita *et al*., J. Vac. Sci. Technol. B **27**, 3063 (2009).

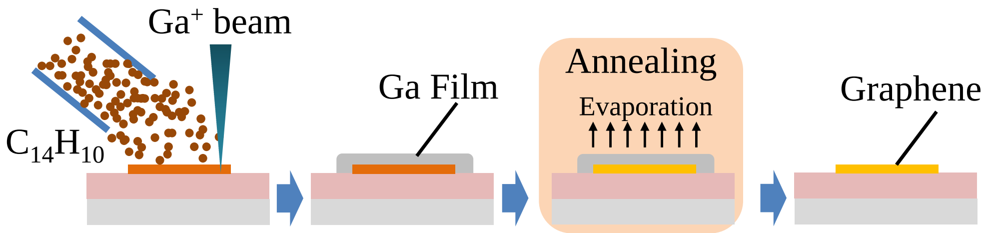

Figure 1: Process of Selective Graphene Growth

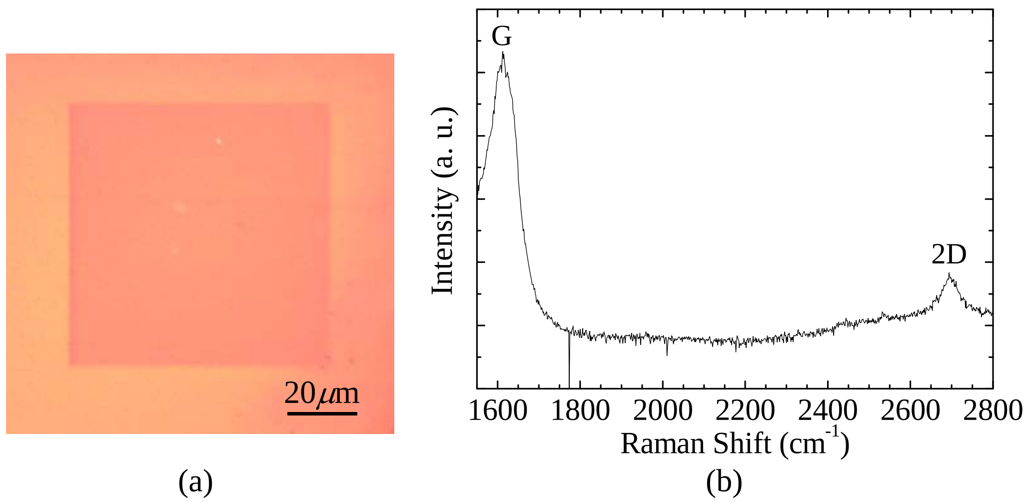

Figure 2: Graphitized Pattern: (a) Optical micrograph of patterned graphene and (b) its Raman spectrum.

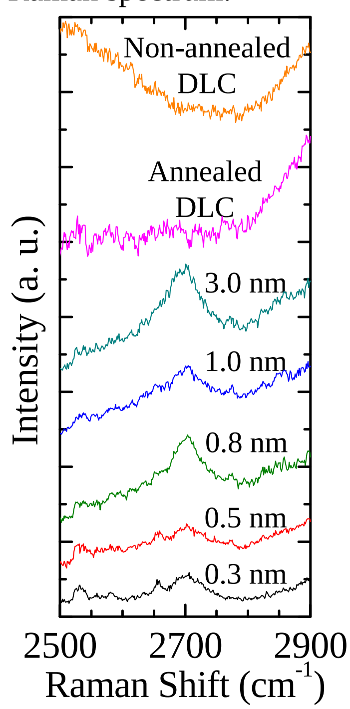

Figure 3: Thickness dependence: Raman spectra of non-annealed and annealed DLC without Ga and 2D bands of graphene with different thickness on the same substrate.

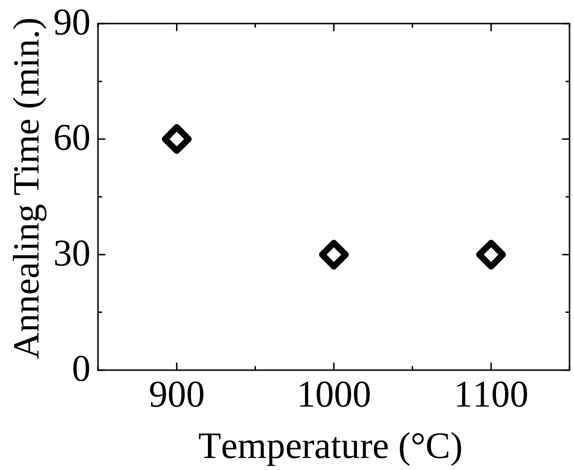

Figure 4: Temperature dependence: Minimum annealing time to graphitize DLC. Thicker graphene tends to have stronger peak.