Systematic Study of Photo-thermal Chemical Vapor Deposition of Graphene on Copper

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Chemical vapor deposition (CVD) is considered as one of the most promising methods for the synthesis of high-quality single-layer graphene [1]. An alternative for typically used tubular furnaces with resistive heating is photo-thermal chemical vapor deposition (PTCVD) which is based on rapid thermal process (RTP). The PTCVD system used in this work includes a cold-wall reactor, pyrometer enabling real-time temperature measurement, and halogen lamps as a heat source. Compared to traditional tubular furnaces, the required growth time for single-layer graphene is significantly reduced in PTCVD due to very high growth rate. Typical growth time of graphene films utilizing tubular furnaces is around 20 - 30 minutes [2]. In contrast, uniform graphene films can be synthesized on copper using PTCVD in 15 - 60 seconds depending on growth parameters.
Various PTCVD process parameters were adjusted to assess their effects on the quality of graphene. The growth temperature plays a key role in graphene synthesis due to the hydrocarbon dissociation on the surface of the copper catalyst. Graphene films fabricated at different temperatures were examined by scanning electron microscopy (SEM) and confocal µ-Raman microscopy. The gas ratio (CH_4: H_2) has also significant influence on the quality of graphene. For example, the areal density of flakes (adlayer domains) can be decreased using a lower gas ratio. PTCVD was found to produce high-quality single-layer graphene at 935 - 950 °C using a growth time of 60 s with a gas ratio of 4 in low pressure (~10 mbar). The influence of the cooling rate from growth temperature was also studied by varying the rate between 3 °C/s and 15 °C/s. The results indicated that the areal density of flakes on graphene can be reduced by increasing the cooling rate. However, the size of the flakes seemed to be invariable although new flakes nucleated during a long cooling stage, resulting even in flake merging. Typical Raman histograms show intensity ratio of the D and G bands (I_D/I_G) around 0.2, which corresponds to very low defect density [3, 4]. The quality of the film was further improved with higher cooling rate as defect density (I_D/I_G ratio) was decreased and I_{2D}/I_G ratio increased.

References

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