CVD grown graphene evaluated with Raman and optical microscope

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This work has been aimed at developing a controllable process for the graphene growth via chemical vapour deposition (CVD) [1-3] using a home built thermal CVD system. Copper foil is used as catalytic substrates for graphene growth since it has been shown to be able to control the amount of layers of graphene [1-4]. During graphene growth, temperature has been held constant at 1000° C and methane gas was used as feedstock, combined with argon and hydrogen gas it has been tested in the pressure range of 1-10 Torr. After graphene growth on Cu foil, a well-known process for transferring graphene from Cu foil to 300nm SiO₂/Si was implemented [4-5]. Briefly the graphene on copper was coated with poly(methyl methacrylate) (PMMA) and cured in a refrigerator for at least 20 hours. The samples were subsequently soaked in an etchant consisting of sodium persulfate and water (1:5 wt), until the polymer film had been cleanly separated from the copper. The films are then cleaned in DI-water followed by transfer to SiO₂/Si. The end product is cleaned with warm acetone to remove the PMMA. Raman characterization was conducted on a Renishaw RamaScope with a 514nm laser [6].

After transferring graphene it is found that some of the samples are dominated by single layer graphene (SLG) on Cu foil based on the Raman spectroscopic results measured at different spots. A good coverage of SLG over a whole sample has been the major challenge. The problem has been the result from the need to control and limit the amount of multi-layer graphene (MLG) present in the samples, while getting a good coverage.

This graphene on copper foil is apparently distorted in a Raman spectrum due to the background signal originated from the Cu foil. This motivates why graphene has to be transferred to SiO_2 for Raman measurement. With graphene transferred onto the SiO_2/Si the Raman results (i.e. the blue curve in Figure 1) indicate the high quality of the SLG sample with I_{2D}/I_G approaching 3 and little D-peak at 1360 cm⁻¹. When measured at different spot, some areas show Raman signal with features as red line in Figure 1 which possibly is resulted from the influence of residual polymer on SLG.

The large portion of graphene film as single layer flake grown by CVD process can be corroborated by the images of optical microscope with a magnification of 5x-50x obtained with graphene transferred onto SiO_2/Si (Figure 2). The photographs in Figure 2 show that the majority of graphene comprises SLG. As shown in figure 2-a, some darker flakes are randomly distributed and covering about 5% of the graphene film which are probably due to the formation of MLG in the film [3]. This can be compared with Figure 2-b where no such darker fields are visible (the visible black spots in figure 2-b are smaller and darker suggesting them to be PMMA residues). The difference between these two samples are a growth time of four hours for 2-a while only one and half hour for 2-b. Based on Figure 2, it can be assumed that SLG was preferably grown over the Cu foil in the CVD process, followed by the occurrence of growth of MLG.

References

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Figure 1 Three different curves of graphene with different affecting backgrounds. The steepest curve is of, what probably is SLG, with residual PMMA on top of it. The middle curve is of MLG, on copper, indicated here as the shift in both the G and 2D peak. And the lowest one is of mostly SLG on SiO_2



Figure 2 Microscopic images of graphene at 50x magnifications, the size bar is 30µm.