Controlling the number and stacking of layers in CVD graphene on Nickel

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Large area single layer graphene or few layer Bernal stacked (A-B) graphene is required for graphene device applications [1]. Chemical Vapour Deposition (CVD) provides a pathway to large area – high quality graphene growth, mainly on lower-cost Copper and Nickel substrates. Growth of graphene by CVD is a combination of surface growth and carbon diffusion from the substrate, making the control of the quality and number of layers challenging. Copper substrates are more suitable for predominantly single layer graphene growth while Ni substrates produce few or multi - layer graphene. Hydrogen is used in CVD graphene growth to regulate the catalytic dissociation of hydrocarbons but it can also be used as a graphene etchant [2]. We propose a method of controlling the number of graphene layers by a post growth annealing process in Hydrogen environment that reduces the number of layers without degradation of the graphene quality.

In this work, graphene is grown by CVD on Nickel substrates. Growth time and flow rate are intentionally increased in order to obtain multilayer graphene, where the top layers are preferably stacked in A-B order as verified by STM. Micro-RAMAN spectroscopy is performed in order to characterize graphene and the 2D peak is analysed to extract information about the number of layers and stacking order [3]. Knowing from the 2D peak analysis that graphene is in A-B stacking configuration an estimation of the number of layers is possible. On the other hand, in the case where turbostratic graphene is predominant, the number of layers cannot be revealed using this method [4]. A single step annealing process in Ar/H_2 at low temperature (450°C) is optimized as a graphene etching process. It is found that graphene etch can be achieved on metal catalysts at low temperature, without introducing defects into graphene.



Figure 1: RAMAN 2D-peaks of graphene of (a) initial substrate of multi-layer (>7 layer) graphene (A-B stacked), grown on Nickel foil (b) after 10-mins post-growth etching graphene is reduced to 3-4 layer with an A-B stacking order.



Figure 2: RAMAN spectrum after 12-mins of etching: (a) full spectrum showing the absence of D-peak (b) 2D-peak of after etching indicating a turbostratic graphene indicating the etching of the A-B stacked graphene overlayers.

References

[1] Novoselov et al, Science vol. 306, 666-669

- [2] Zhang et al, ACS Nano. 2012 Jul 24;6(7):6526
- [3] A. C. Ferrari et al, Nature Nanotechnology, vol 8, April 2013
- [4] Kwanpyo Kim et al., PRL 108, 246103 (2012)