Controlling the thickness and coverage of multilayer graphene on copper in the chemical vapour deposition process

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Abstract

Among several methods for production of Graphene, CVD method has the highest potential for large area production of graphene with adequate crystalline quality, especially for the optoelectronic applications. In order to achieve a better conductivity, a practical approach can be usage of multilayer graphene while the slight reduction of transparency can be in the acceptable limits. Accordingly, multilayer graphene seems to be closer to meet the conductivity-transparency requirement for a variety of applications ranging from solar cells to displays or touch screens.
Although, low carbon solubility in the copper impose the growth of graphene to predominantly single layer film and this growth is mainly due to the surface adsorption of carbon atoms on the surface [1], However recently, it has been reported that multi-layer graphene films can be grown under certain CVD conditions [2].

Towards the objective of large area high quality graphene growth, with controllable thickness, in this work we investigated the multilayer graphene formation on the surface of a copper foil by changing the growth time during the CVD process. The growth has been performed within an intermediate pressure range which has makes a significant impact on the growth kinetics especially in the presence of impurities. It has been observed that both the average thickness and coverage are linearly proportional to the growth time. Optical microscopy, Scanning Electron Microscopy and Raman analysis together with Optical Transmittance and Sheet Resistance measurements are consistently demonstrate extension of multilayer islands with growth time.

A qualitative model (Figure A) is suggested to explain the microscopic mechanism of the multilayer growth on copper. In this model Impurity particles, which exist within the copper surface, act as the catalysts and the seeds for creation of multilayer graphene islands. The relation between the distribution of impurities on the surface and molecular mean free path define the growth regime. Although graphene growth on copper at low pressure is known to be mediated by surface adsorption, we claim that the multilayer growth can be enhanced in presence of impurities and an optimized process pressure. When the pressure is adjusted to an intermediate level so that the mean free path and average distance between the activation sites are comparable, the formation of multilayer graphene islands can be improved to achieve full surface coverage. Under such conditions, the multilayer coverage and average graphene thickness increase monotonously as the growth time is increased in a wide range of time, indicating the non-localized nature of the multilayer growth.

References

[1] Li, X. S.; Cai, W. W.; An, J. H.; Kim, S.; Nah, J.; Yang, D. X.; Piner, R. D.; Velamakanni, A.; Jung, I.; Tutuc, E.; Banerjee, S. K.; Colombo, L.; Ruoff, R. S, Science, **324** (2009) 1312
[2] Tu Z, Liu Z, Li Y, Yang F, Zhang L, Zhao Z, Xu C, Wu S, Liu H, Yang H,Richard P, Carbon, **73** (2014) 252-258.

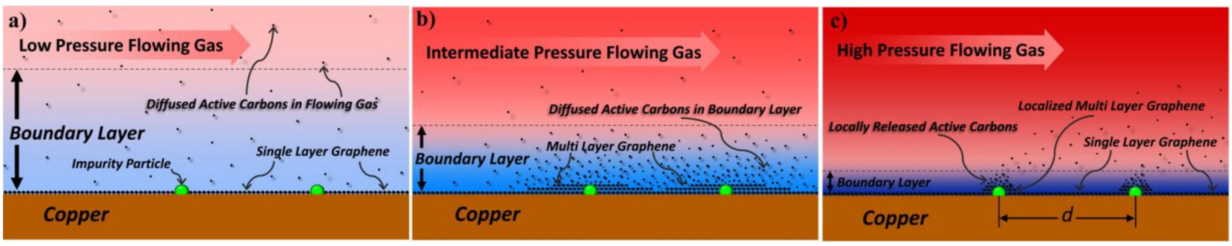

Figure A