

Materials Science & Engineering Doctoral Defense

Nucleation and Growth of Single Layer Graphene on Supported Cu Catalysts by Cold Wall Chemical Vapor Deposition

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abstract

Chemical Vapor Deposition (CVD) is the most widely used method to grow large-scale single layer graphene. However, a systematic experimental study of the relationship between growth parameters and graphene film morphology, especially in the industrially preferred cold wall CVD, has not been undertaken previously. This research endeavored to address this and provide comprehensive insight into the growth physics of graphene on supported solid and liquid Cu films using cold wall CVD.

A multi-chamber UHV system was customized and transformed into a cold wall CVD system to perform experiments. The versatile growth process was completely custom-automated by controlling the process parameters with LabVIEW. Graphene growth was explored on solid electrodeposited, recrystallized and thin sputter deposited Cu films as well as on liquid Cu supported on W/Mo refractory substrates under ambient pressure using Ar, H₂ and CH₄ mixtures. The results indicate that graphene grown on Cu films using cold wall CVD follows a classical two-dimensional nucleation and growth mechanism. The nucleation density decreases and average size of graphene crystallites increases with increasing dilution of the CH₄/H₂ mixture by Ar, decrease in total flow rate and decrease in CH₄:H₂ ratio at a fixed substrate temperature and chamber pressure. Thus, the resulting morphological changes correspond with those that would be expected if the precursor deposition rate was varied at a fixed substrate temperature for physical deposition using thermal evaporation. The evolution of graphene crystallite boundary morphology with decreasing effective C deposition rate indicates the effect of edge diffusion of C atoms along the crystallite boundaries, in addition to H₂ etching, on graphene crystallite shape.

The roles of temperature gradient, chamber pressure and rapid thermal heating in C precursor-rich environment on graphene growth morphology on thin sputtered Cu films were explained. The growth mechanisms of graphene on substrates annealed under reducing and non-reducing environment were explained from the scaling functions of graphene island size distribution in the pre-coalescence regime. It is anticipated that applying the pre-coalescence size distribution method presented in this work to other 2D material systems may be useful for elucidating atomistic mechanisms of film growth that are otherwise difficult to obtain.

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