Growth mechanism of graphene film on Ni and Cu by alcohol catalytic chemical vapor deposition

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The mechanism of chemical vapor deposition (CVD) of graphene has not been totally clarified, especially when various transition metals are used as substrates for different carbon precursors.[1,2] Here we present CVD growths of graphene on Ni and Cu surface using ethanol as carbon precursor,[3] and investigate the growth mechanism based on parametric study on Cu and isotope labeling of ^{13}C-ethanol on Ni. Results show that during a low pressure alcohol catalytic CVD (LP-ACCVD) process, for both substrates a short growth time (30 s for Ni and 2 min for Cu) yields the formation of graphene films with high metal surface coverage (>80%), and the coverage and graphene layer numbers can increase with extended ethanol reaction time. As to their growth mechanisms, on Ni surface isotope labeling results show that the sequentially-introduced ^{12}C and ^{13}C ethanol sources are able to independently form graphene flakes other than a uniform mixture of two isotopes. It is interpreted by a direct surface-adsorptive growth model, in which the small carbon fragments catalyzed from ethanol first nucleate at the metal step edges and expand over the whole metal surface with different layer numbers. On the other hand, parametric studies on single- and poly-crystalline Cu show that graphene domains expand in a fashion without apparent growth frontiers, and the surface morphology of Cu plays a critical role in forming the second layer of graphene, which occurs soon after the formation of the first layer. Approaches of controllable growths of large-scale uniform single- or bi-layer graphene have also been discussed.

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