

# Study of Single Crystal Graphene Grown by Chemical Vapor Deposition on Copper

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Graphene has attracted great interest due to its intriguing electronic properties. Popular methods to realize monolayer graphene include exfoliation of graphite, epitaxial growth on SiC, chemical vapor deposition (CVD) on copper, and the reduction of graphite oxide. Among these methods, the CVD technique[1] has attracted considerable interest since it has already served to realize large sheets of monolayer, polycrystalline graphene. Graphene growth by CVD on copper starts with the formation of stable nuclei, these nuclei grow into grains and the grains coalesce at the grain boundaries, resulting in the polycrystalline graphene, with grain size often exceeding 100 microns. The polycrystalline morphology observed in CVD graphene degrades, however, the mechanical and electrical properties of CVD graphene. Thus, the development of growth techniques that reduce the density and increase the size of two-dimensional grains and, ultimately, helps to produce large single crystals of graphene, has become a topic of interest in CVD graphene growth.[2, 3]

The chemical vapor deposition (CVD) growth of single-crystal graphene on polycrystalline copper foils is a complex process affected by thermodynamics, kinetics, and growth conditions. These factors lead to a great diversity in the island shapes of single crystal graphene. Our low pressure CVD technique to produce graphene single crystals is illustrated in Fig. 1. Here, we present the results of an experimental atomic force microscopy (AFM) study of the different shapes of single-crystal graphene grown on the inner surface of closed copper enclosures using the low pressure CVD technique, see Fig. 1. Remarkably, our study indicates that graphene single crystals appear to form below the adjacent copper foil surface. This feature is revealed in cross sectional AFM scans of the height, which indicate that the graphene surface lies below the neighboring foil surface by 15- 30 nm. Our results also show that an impurity assisted growth mechanism governs the growth of single crystal graphene via isotropic diffusion, producing two-fold, four-fold, and six-fold symmetries in the resulting flakes. In addition, single crystal graphene produced via anisotropic diffusion also appear, but they do not exhibit signs of an impurity assisted growth mechanism. Finally, we find that strain relaxation in two-fold and four-fold symmetric graphene structures via isotropic diffusion is more involved than in the six-fold structures, which results in multiple step orientations in low symmetry structures.[4]

## References

- [1] X. S. Li et al.. *Science*, 324, 1312 (2009).
- [2] X. Chen et al. *Carbon*, 94, 810 (2015).
- [3] X. Wu et al. *Sci. Rep.* 6, 21152 (2016).
- [4] T. R. Nanayakkara et al., *Carbon*, 168, 684 (2021)

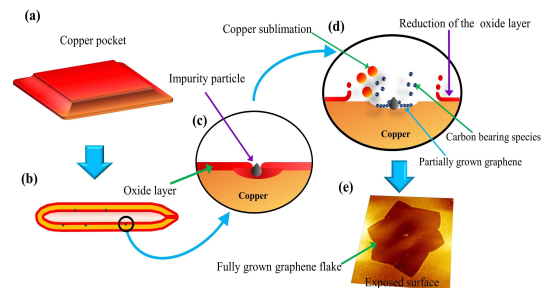


Fig. 1. Schematic diagram. (a) The oxidized copper foil is folded into a pocket. (b) Cross view of the copper pocket. The red outline represents the oxide layer in both the inner and outer surfaces. Black dots represent impurities. (c) A magnified view. (d) Nucleation could start at the impurity particle. (e) A single crystal graphene flake.