Edge-Controlled Growth and Kinetics of Single-Crystal Graphene Domains by Chemical Vapor Deposition

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Abstract

The controlled growth of large-area, high-quality, single-crystal graphene is highly desired for applications in electronics and optoelectronics; however, the production of this material remains challenging because the atomistic mechanism that governs graphene growth is not well understood. The edges of graphene, which are the sites at which carbon accumulates in the sp2 lattice, influence many properties, including the electronic properties and chemical reactivity of graphene,[1-5] and they are expected to significantly influence its growth. However, this influence of edge structure has not been verified experimentally due to the difficulty of growing graphene with controlled edges. For instance, only zigzag or randomly oriented edges have been fabricated by chemical vapor deposition (CVD). Here, we demonstrate the growth of single-crystal graphene domains with controlled edges that range from zigzag to armchair orientations via growth–etching–regrowth in a CVD process (Figure 1).[6] We have observed that both the growth and etching rates of a single-crystal graphene domain increase linearly with the slanted angle of its edges from 0 to ~19° and that the rates for an armchair edge are faster than those for a zigzag edge (Figure 2). Such edge-structure-dependent growth/etching kinetics of graphene can be well explained at the atomic level based on the concentrations of the kinks on various edges. In addition, we found that the graphene edges and morphology are not determined by the nucleation but are kinetically controlled, following the classical kinetic Wulff construction theory, during the CVD process, and that defects in the graphene can be healed through an etching–regrowth process (Figure 3). Using these findings, we propose several strategies for the fabrication of wafer-sized, high-quality, single-crystal graphene.

References

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Figures

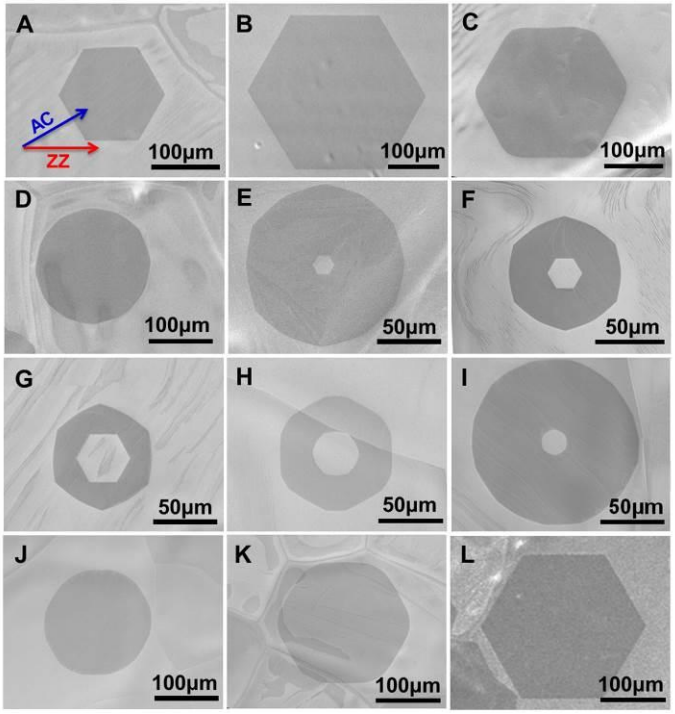

Figure 1. Morphology and edge evolution of single-crystal graphene domains grown on a Pt surface during the growth-etching-regrowth process.

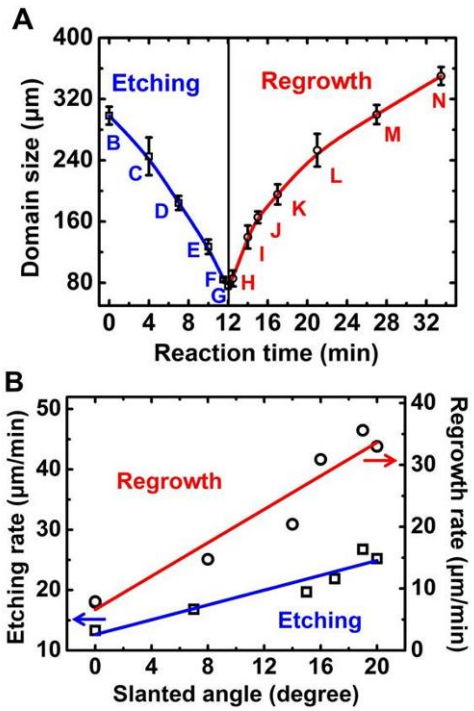

Figure 2. Edge-structure-dependent growth/etching of single-crystal graphene domains.

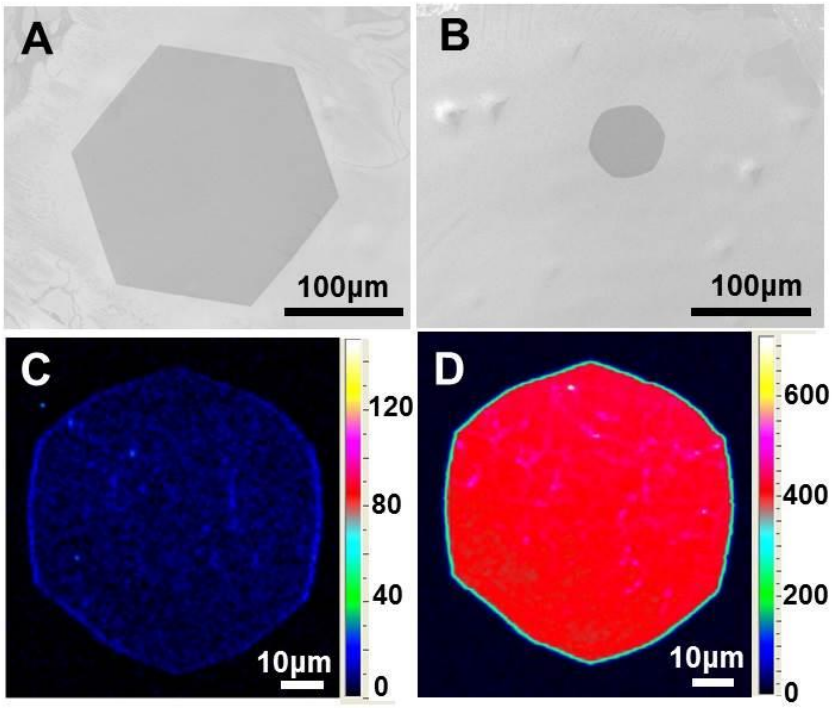

Figure 3. Regrowth of defect-free single-crystal graphene domains.