

Kinetic Control of Catalytic CVD for High Quality Graphene at Low Temperatures

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Abstract

Chemical vapour deposition (CVD) is the most promising route towards scalable graphene production and integration, key requirements for the commercial exploitation of graphene's unique properties. The growth of high-quality, uniform monolayer graphene (MLG) has been well demonstrated on polycrystalline Cu foils, however this generally requires excessive temperature (~1000°C), and low pressures which result in significant catalyst sublimation.

Ni based catalysts have been shown to be effective for graphene growth at much lower temperatures (450-600°C).¹ We use in situ, time-, and depth-resolved X-ray photoelectron spectroscopy (XPS) and in-situ X-ray diffraction (XRD) to monitor graphene formation under realistic CVD conditions.^{1,2} In particular, we reveal that graphene growth occurs during isothermal hydrocarbon exposure, rather than being limited to a precipitation process upon cooling, and that carbon dissolved in to the catalyst sub-surface plays a critical role.

Here, we use this understanding to realise scalable graphene CVD on Ni-based catalysts at ~600°C with complete MLG coverage, achieving uniformity and quality that has hitherto only been reported for Cu-based CVD at >900°C.³ Raman mapping reveals a 2D/G ratio of >3.2, D/G ratio ≤ 0.08 and transport measurements show carrier mobilities of ≥3000 cm²V⁻¹s⁻¹ on SiO₂ supports.

We establish a kinetic growth model for graphene CVD based the balance of carbon fluxes at the catalyst surface,⁴ which is well supported by our systematic study of Ni-based polycrystalline catalysts. A finite carbon solubility of the catalyst is shown to be a key advantage, as it allows the catalyst bulk to act as a mediating carbon sink while optimized graphene growth occurs by only locally saturating the catalyst surface with carbon. This enables a route to the controlled formation of Bernal stacked bi- and few-layered graphene. The model developed is relevant to all catalyst materials and can readily serve as a general process rationale for optimised graphene CVD.

References

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