**In-situ Observations of Gas Phase Dynamics During Graphene Growth Using Solid-State Carbon Sources**

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A single-layer graphene has been uniformly grown on a Cu surface at elevated temperatures by thermally processing a poly(methyl methacrylate) (PMMA) film in a rapid thermal annealing (RTA) system under vacuum. The detailed chemistry of the transition from solid-state carbon to graphene on the catalytic Cu surface was investigated by performing in-situ residual gas analysis while PMMA/Cu-foil samples being heated, in conjunction with interrupted growth studies to reconstruct ex-situ the heating process. The data clearly show that the formation of graphene occurs with hydrocarbon molecules vaporized from PMMA, such as methane and/or methyl radicals, as precursors rather than by the direct graphitization of solid-state carbon. We also found that the temperature for vaporizing hydrocarbon molecules from PMMA and the length of time the gaseous hydrocarbon atmosphere is maintained, which are dependent on both the heating temperature profile and the amount of a solid carbon feedstock are the dominant factors to determine the crystalline quality of the resulting graphene film. Under optimal growth conditions, the PMMA-derived graphene was found to have a carrier (hole) mobility as high as $\sim 2,700 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ at room temperature, superior to common graphene converted from solid carbon.

**Keywords:** graphene, RTA, solid carbon source, QMS, Cu foil