

JOINT DEVELOPMENT OPPORTUNITY

Methods of Growing Uniform, Large-Scale Multilayer Graphene Films



Schematic diagram of chemical vapor deposition reaction chamber and multilayer graphene films

From the top: graphene film put on substrate (*top left*); protective PMMA put over catalyst (*top right*); etching-removal of catalyst (*middle left*); rinsing of PMMA and graphene films (*middle right*); protective PMMA layer removed (*bottom*)

Ref: L. Gao, G.-X. Ni, Y. Liu, B. Liu, A. H. Castro Neto, and K. P. Loh, *Nature* 505, pp 190-194 doi: 10.1038/nature12763 (2014). Patent Pending: WO 2013/172792 A1

TECHNOLOGY BACKGROUND

One of the biggest hindrances to realizing graphene applications on an industrial-scale is the lack of a rigorous production method for the raw material, graphene. Such a method has to deliver high-quality graphene at a throughput that will meet the market's demands. Commercial requirements also include variability in sizes of raw materials and, preferably, large area graphene samples.

The widely accepted methods of synthesis of graphene in the laboratory are fraught with inherent limitations.

(a) Cleavage or exfoliation of highly oriented pyrolytic graphite (HOPG) produces small-area graphene films that are far from meeting industry needs.

(b) Another commonly used method, chemical reduction of exfoliated graphite oxide layers, produces graphene oxide which has poor electrical and structural properties.

(c) A lone bright spark comes from the large-area high quality graphene films from thermally annealing SiC at temperatures above 1,600°C in an ultra-high vacuum environment. However, separating and transferring the graphene film from the matrix to a substrate poses an intrinsic challenge in maintaining its fragile stability. The cost and physical constraints of SiC and the UHV conditions make this method commercial very unattractive.



(d) While chemical vapour deposition (CVD) of graphene on transition metals is more promising than the above methods, the published results on copper substrate self-limits the growth of graphene. This graphene also suffers from relatively low electronic mobility and conductivity. Other metals used like ruthenium and cobalt result in thicknesses that are non-uniform (with low optical transmittance) and structures that are low in crystallinity (hence high in electrical resistance).

SOLUTION

To produce uniform, high-quality, large-scale multilayer graphene films, we use a carbonizing catalyst in a CVD reaction chamber at appropriate temperatures and pressures. A gaseous carbon source and a weak oxidizing vapour are allowed to flow over the catalyst, upon which carbon atoms are deposited onto the catalyst surface. These atom forms a crystallized arrangement. Further cooling results in a multilayer graphene film on the carbonizing catalyst surface. (See figures at the top.)

BENEFITS

- Crystallinity of graphene films improves hence lowering sheet resistance.
- Graphene films obtained with lower number of defects than atmospheric pressure CVD-produced films
- Graphene films formed with uniform thickness continuous over a large area
- Number of layers of graphene film can be controlled and adjusted
- Electrical resistivity (sheet resistance in ohms per square) of multilayer graphene film (~ 200 Ω/\Box) is lower than those of conventional atmospheric pressure and low pressure CVD processes
- Optical transmittance of multilayer graphene obtained is measured at 86.7% (with a UV-VIS spectrophotometer) which compares very favorably with that of conventional CVD processes.

APPLICATION AREAS

- Flexible transparent electrodes and as ultrathin electrodes for Li-ion batteries in supercapacitors, interconnects of ICs, active layers for photodetectors, planar optical polarizers, and in biosensors.
- Graphene-on-silicon platform, e.g. gate-controlled Schottky barrier triode devices and optical modulators

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