

# Remote Plasma Enhanced – Chemical Vapor Deposition (rPE-CVD) of Graphene on Various Substrates

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## Abstract

The electrical and mechanical properties of graphene, in particular ballistic electron transport properties, have opened up exciting possibilities for this material as a replacement for silicon. Graphene has a simple structure, consisting of a hexagonal arrangement of carbon atoms in a monoatomic layer (with a lattice parameter  $a_{gr} = 2,46 \text{ \AA}$ ), and is mechanically and chemically stable. Mechanical exfoliation of highly oriented pyrolytic graphite (HOPG) has been the most common method of producing single layers of this material. However, the lateral dimensions of monolayer samples are typically limited to the micro-scale. Since large area graphene films on insulating substrates are required for practical applications, several techniques have been explored such as chemical vapor deposition (CVD) on transition metals, graphitization of SiC wafers under high vacuum, and reduction of oxidized graphite films.

Recently, several authors [1-3] have reported new deposition methods using RF plasma as a way to decouple the dissociation process of the precursor gas from the graphene growth process onto the substrate via Plasma Enhanced Chemical Vapor Deposition (PE-CVD). It makes the method more tunable as it allows for an independent control of the reaction parameters and the growth parameters, which should lead to a better control of the size and shape of the nanostructures. Moreover, converting the precursor gas into a plasma state involves, by definition, a higher amount of active carbon radicals in the reaction process and thus enhances the deposition rate. By means of using a highly reactive deposition technique such as a hydrocarbon plasma, one can decrease the exposure time and/or decrease the substrate temperature. This latter feature opens the deposition process towards a wider variety of substrates with lower melting points. Last but not least, a remote plasma is – by definition - generated at a distance from the substrate, thus minimizing preferential perpendicular growth directions that the electrical fields may induce in a traditional plasma setup.

CELLS-ALBA together with ibss Group, Inc. has adapted the GV10x downstream inductively coupled RF plasma source typically used for cleaning hydrocarbon contaminated from SEM chambers to also remove carbon deposits on optical precision surfaces. For these applications the feedstock gas of the plasma consists of a chemically active agent such as oxygen or hydrogen converting carbon into CO<sub>2</sub>, CO, or hydrocarbons gas via a corresponding oxidation or reduction process, respectively.

Our goal in this work has been to “reverse” the working principle of the GV10x, in order to convert it into a thin film deposition tool instead of a cleaning tool. This has been achieved by simply exchanging the feedstock gases, moving from oxygen or hydrogen to hydrocarbon gases that work as carbon sources which therefore become the precursors for the graphene deposition process. Carbon and CH<sub>x</sub> ions as well as radicals bond with the thermally activated substrate surfaces via H-bond breaking and H<sub>2</sub> molecule formation, thereby eventually self-arranging in honeycomb graphite lattice geometry.

In order to accomplish the purpose of reversing the working principle of the GV10x from a

top-down cleaning tool to a bottom-up deposition tool, several routes for depositing graphene on different substrates (e.g., Ni foil, HOPG) have been explored. After that, the resulting samples were characterized following a systematic approach in order to verify that indeed graphene was deposited, and, if so, its thickness, shape, etc. First of all, Raman spectroscopy gives an idea of the kind of carbon allotrope and crystalline quality. Scanning Electron Microscopy (SEM) gives a direct picture of the sample with a nanometric resolution, and finally X-Ray Photoemission Spectroscopy (XPS) gives a detailed chemical analysis of the surface that allows calculating atomic concentrations and graphene film thicknesses in terms of deposited monolayers. The main point of this characterization procedure has been to cross-check results and to obtain a more complete picture of the characteristics of our graphene samples.[4]

**References**

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- [2] T. Otha, A. Bostwick, T. Seyller, and K. Horn, *Science* 313, (2006), 951.
- [3] Gopichand Nandamuri, Sergei Roumimov, and Raj Solanki. *App. Phys. Lett.* 96, (2010), 154101.
- [4] M. González Cuxart and E. Pellegrin, *rPECVD of graphene on various substrates*, thesis corresponding to the Master in “Nanotechnology and Materials Science”, Universitat Autònoma de Barcelona and ALBA-CELLS, (2014).

**Figures**

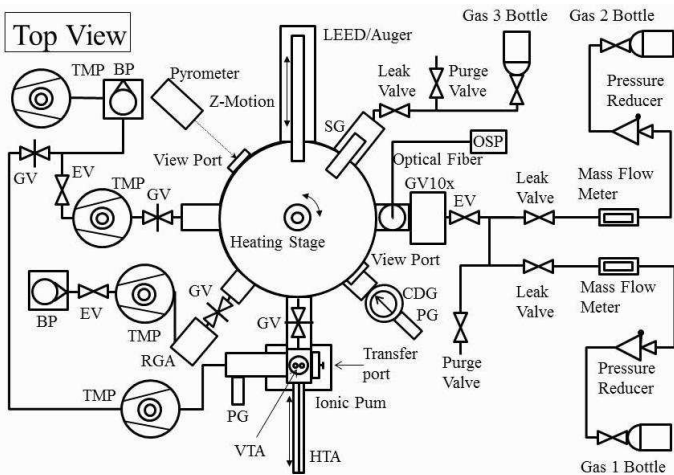


Figure 1: Conceptual layout of the RF plasma deposition chamber.

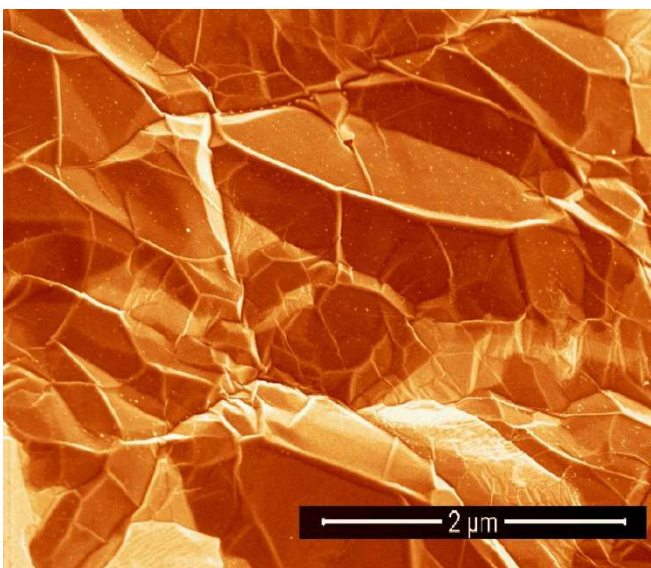


Figure 2: SEM image of one of our graphene samples.

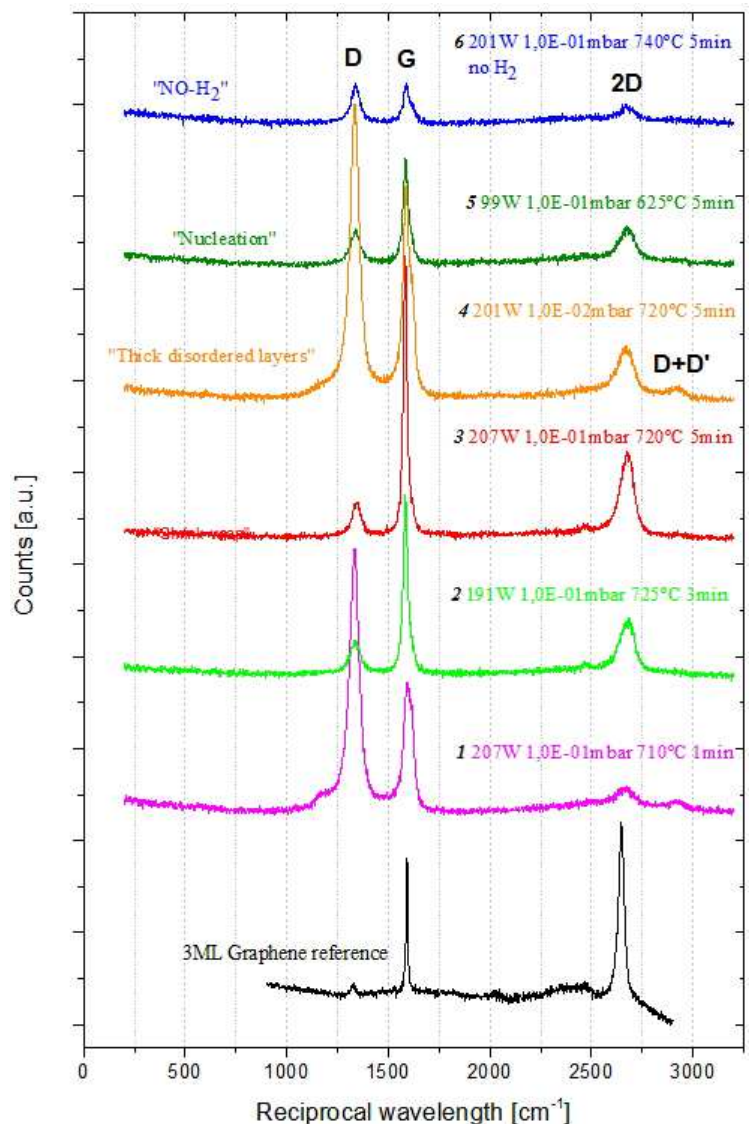


Figure 3: Raman spectra corresponding to some of our graphene samples.