

ABSTRACT**Novel hybrid materials based on graphene**

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In the first part of this dissertation, a straightforward method to deposit uniform single-layer graphene films on arbitrary substrates without size limitation and under ambient conditions was developed using a modified Lagmuir-Blodgett method. The fast high-yield method allows control of graphene coverage by simple adjustment of the applied surface pressure in a LB trough. Additionally, among all reported chemically exfoliated graphite, the flakes we obtain show one of the lowest resistivity at the Dirac charge neutrality point ( $\sim 65 \text{ k}\Omega$ ).

In the second part, a comparative study regarding the electrocatalytic activity of graphene oxide (GO), chemically-reduced graphene oxide (crGO) and graphene produced by direct liquid exfoliation (dG) was performed. Sensors were developed by modifying glassy carbon (GC) electrodes with GO, crGO and dG and ascorbic acid was used as a pilot analyte. GC/GO electrodes offer substantially lower oxidation overpotential, up to 350 mV, compared with GC/crGO, GC/dG and unmodified GC electrodes. In addition, the different carbon-to-oxygen atomic ratios in GO, as it occurs depending on the synthetic route, have a remarkable effect on the performance of the sensors.

In the third part, novel hybrid materials based on graphene were developed by combining organosilica precursors, magnetic nanoparticles and biomimetic complexes with graphene oxide matrices. Nanoporous hybrid materials based on graphene from silylated organoprecursors were produced using the sol-gel method. Subsequently, hybrid materials derived from decoration of graphene with magnetic

nanoparticles were prepared by a simple, versatile and reproducible approach. Towards this aim, GO was used as nanotemplate for the *in-situ* synthesis of a range of very small ferrimagnetic and/or anti-ferromagnetic iron oxide nanoparticles in different environments. Finally, a hybrid system consisting of a copper biomimetic complex covalently attached on the surfaces of GO was synthesized and studied. All the above hybrid materials were characterized by a combination of experimental techniques including X-ray diffraction, infrared,  $\mu$ -Raman, Mössbauer and Electron Paramagnetic Resonance spectroscopies, thermal analysis, surface area measurements and Transmission electron microscopy. The characterization techniques gave insight into the formation process and structural details of the produced hybrid structures.