

## PREPARATION OF FLEXIBLE CONDUCTIVE REDUCED GRAPHENE OXIDE PAPERS

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Graphite oxide/graphene oxide is produced in the reaction of graphite with strong oxidizers in water medium according to Hummer's method [1]. Graphite powders are oxidized in order to add oxygen-containing functional groups on the edges and basal planes of graphite flakes. These functional groups reduce the interplanar forces leading to an exfoliation of the graphite oxide into graphene oxide (GO) sheets in aqueous media.

GO sheets are composed of planar, graphene-like aromatic domains of random sizes interconnected by a network of cyclohexane-like units in chair configuration which are decorated by hydroxyl, epoxy, ether, diol, and ketone groups [2], imparting water solubility to these GO sheets. Solution-processable GO has been used to fabricate paper-like films with excellent mechanical properties [3], as well as electrically conductive polymeric [4, 5] and ceramic composites [6].

The electrical conductivity of GO is considerably enhanced through removal of the oxidized functionalities in the sheets by reduction. The reduced graphene oxide (RGO) sheets still contain lattice defects [7] that degrade its electrical properties compared to pristine graphene sheets obtained by the tape-peeling method [8]. Thus there is also a need for restoring the graphitization of the RGO sheets to enhance their electrical properties. Processing of RGO into free-standing papers is of great interest for flexible electronic or electrochemical devices. We report here the preparation of water-processable free-standing flexible conductive RGO papers and evaluate the effect of different chemical and thermal reduction treatments on the morphology and chemistry of the papers as effective means for enhancing the degree of graphitization and their electrical properties.

Starting point are flexible GO-papers prepared by vacuum filtration of aqueous solutions of graphene oxide sheets, which are obtained following the experimental details reported elsewhere [9]. After rinsing, drying and peeling-off the membrane, different reductive treatments are applied: i) hydrazine monohydrate vapor; ii) vapors of hydrazine monohydrate followed by a thermal annealing; iii) thermal treatment under an Ar flow; iv) thermal treatment under a H<sub>2</sub> flow.

All the obtained free-standing flexible papers, maintain a good structural integrity even after reduction. The morphology of the original GO-paper and the RGO-papers is observed by scanning electronic microscopy (SEM) showing a laminar structure, which is kept within reduction.

Different characterization techniques confirm the reduction of GO- into RGO-papers within the studied treatments. X-ray diffraction reveals the removal of oxygen-containing groups intercalated in the interlayer space in the original GO-paper. X-ray photoelectron

spectroscopy (XPS), which is used to probe the effect of the different reduction treatments on the chemical composition of the papers, reveals an increase of C/O atomic ratio, indicative of the effectiveness of the employed reduction methods for removing oxygen-containing groups.

The increased intensity and decreased width of the 2D-band in Raman spectra indicate the restoration of the graphitic structure ( $C_{sp^2}$ ) of the individual graphenes in the paper within reduction, which is confirmed by the measurement of the sheet resistances of the films. Due to the restoration of defects in the GO sheets and the decrease of interlayer distance between them, the interaction among the RGO sheets is increased and therefore, the electrical conductivity of the RGO papers improved.

Summarizing, in this communication we present a simple procedure for the preparation of flexible conductive RGO-papers through a range of chemical and thermal reduction treatments applied to a flexible GO-paper. We evaluate the studied reduction treatments on the basis of two independent important effects: i) the removal of chemical functional groups, and ii) the growth of graphitic nanodomains as the dominant process, in order to go towards the most appropriate graphene-based papers for electronic and electrochemical applications in terms of flexibility and conductivity.

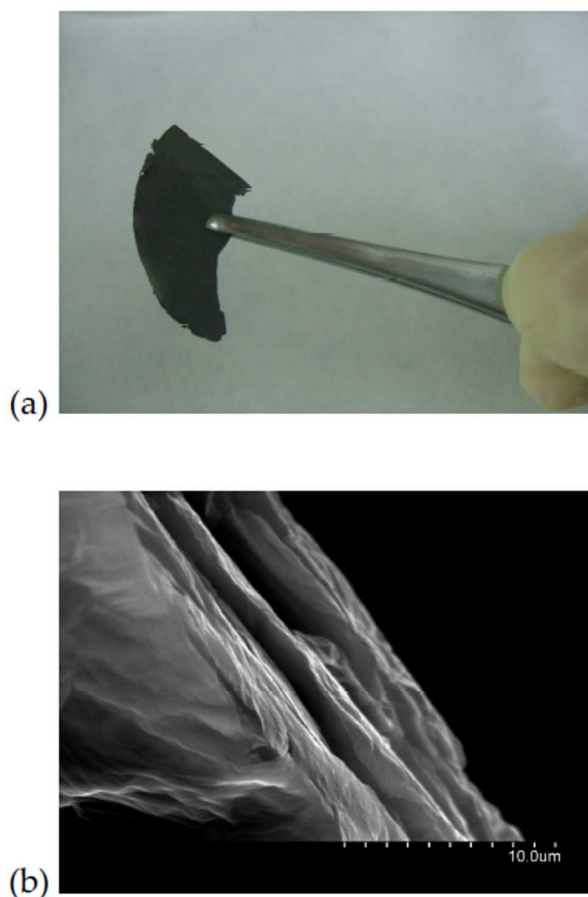


Figure 1: Photograph of a free-standing RGO-film (a); cross-section SEM image of a freestanding RGO-film (b).

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**References**

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