

---

**SYNTHESIS OF GRAM-SCALE GRAPHENE  
FOR ENERGY-STORAGE APPLICATIONS**

**D. Pontiroli**<sup>a\*</sup>, **M. Riccò**<sup>a</sup>, **M. Mazzani**<sup>a</sup>,  
**S. Rols**<sup>b</sup> and **M. Choucair**<sup>c</sup>

<sup>a</sup>Dipartimento di Fisica, Università di Parma, Parma, Italy

<sup>b</sup>Institut Laue Langevin, Grenoble Cedex, France

<sup>c</sup>School of Chemistry, University of New South Wales, Sydney, Australia

\*Corresponding author: *Daniele.Pontiroli@fis.unipr.it*

The excellent mechanical, transport and thermal properties, together with the high specific surface area, make graphene as a promising material for a wide variety of applications: from the production of high-speed transistors [1] to the implementation of ultra-sensitive sensors [2]. From the construction of transparent and flexible electrodes [3], to the development of energy-storage systems [4,5].

For all these purposes, it is of great importance to develop a method to synthesize large amount of graphene, while keeping, however, a reasonable quality of the two-dimensional carbon hexagonal lattice, in order to preserve the electronic properties of the graphene samples.

We present here a novel chemical route for the production of large scale amount of graphene (of the order of grams), based on the thermal exfoliation of graphite-oxide (GO), obtained by annealing it at 1300 K for few minutes in inert atmosphere (see Figure 1). GO is obtained from modified Staudenmeier and Brodie method [6,7]. Partial chemical reduction of GO in NaBH<sub>4</sub> aqueous solution, possibly followed by a further treatment with concentrated sulphuric acid, allows to control the grade of defects in the exfoliated samples (which consist mainly of in-plane carbon vacancies and edge-like defects), by tuning the amount of carboxyl, hydroxyl and epoxy groups attached on the graphene-oxide planes.

Experimental characterisation via laboratory x-ray powder diffraction, SEM, TEM and SAED evidenced the almost complete exfoliation of the graphene planes in the samples (see Figure 2a). Further thermal treatments in hydrogen atmosphere proved effective in varying the hydrogen content in the samples, which is chemisorbed onto the graphene plane, as indicated also by neutron scattering experiments (shown in Figure 2b).

The study of the role of defects and the interaction between hydrogen and graphene are of great interest for controlling the electronic properties of these systems and for their future applications as hydrogen-storage materials [8].

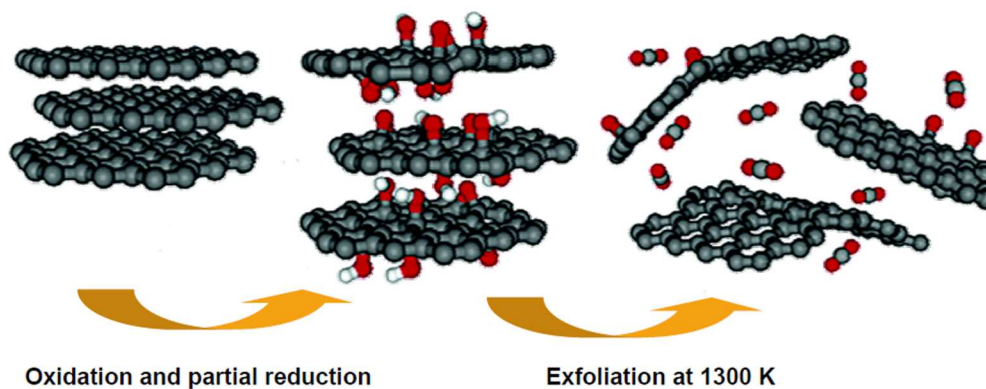


Figure 1: GO and partially reduced GO undergo a thermal annealing at 1300 K for 2 min. The decomposition rates of the epoxy and hydroxyl groups exceed the diffusion rate of the gases and they reach a pressure that exfoliates the single graphene sheets.

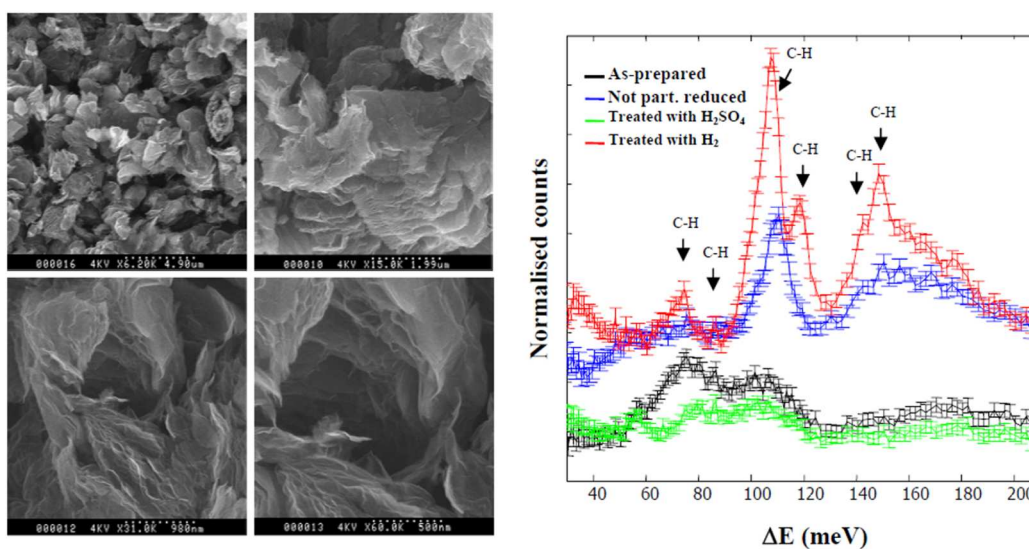


Figure 2: SEM images of graphene samples indicate the complete exfoliation of the planes. b) Inelastic neutron scattering at IN1BeF (ILL) on different samples shows the presence of residual hydrogen (out of plane C-H bending modes at 67, 82, 103, 118, 142, 149 meV). These modes are strongly suppressed in less defective samples, while are enhanced after treatment in H<sub>2</sub> at high temperature. This suggest that H could saturate defects on graphene.

**References**

- [1] Y.-M. Lin, C. Dimitrakopoulos *et al.*, *Science*, 327 (2010) 662.
- [2] F. Schedin *et al.*, *Nat. Mater.* 6 (2007) 652.
- [3] G. Eda, G. Fanchini and M. Chhowalla, *Nat. Nanotech.*, 3 (2008) 270.
- [4] M. D. Stoller, S. Park *et al.*, *Nano Lett.*, 8 (2008) 3498.
- [5] H. Lee, J. Ihm *et al.*, *Nano Lett.*, 10 (2010) 793.
- [6] L. Staudenmeier, *Ber. Deut. Chem. Ges.*, 31 (1898) 1481.
- [7] B. C. Brodie, *Ann. Chim. Phys.*, 59 (1860) 466.
- [8] This research is supported by the Sinergia Project "HyCarbo" (SNSF).