Improving the cleanliness of graphene grown on copper by chemical vapor deposition for biosensing applications

M. Pittori^{1,2}, R. Brajpuriya^{3,4}, E. Leoni³, A. Capasso⁵, T. Dikonimos³, F. Buonocore³, R. Mazzaro^{2,6}, V. Morandi², R. Rizzoli², M.G. Santonicola¹, N. Lisi³

¹Department of Chemical Materials Environmental Engineering, Sapienza University of Rome, Via del Castro Laurenziano 7, 00161 Rome, Italy Institute for Microelectronics and Microsystems (IMM) Bologna, Consiglio Nazionale delle Ricerche, Via Gobetti 101, 40129 Bologna, Italy ³Surface Technology Laboratory, Materials Technology Unit, Casaccia Research Centre, ENEA, Via Anguillarese 301, 00123 Rome, Italy ⁴Amity Institute of Nanotechnology, Amity University Haryana, Manesar, Gurgaon, 122413 Haryana, India ⁵Graphene Labs, Istituto Italiano di Tecnologia – IIT, Via Morego 30, 16163 Genova, Italy ⁶Department of Chemistry "G. Ciamician", University of Bologna, Via Selmi 2, 40126 Bologna, Italy pittori@bo.imm.cnr.it

Abstract

Since graphene (G) was first isolated by mechanical exfoliation [1], several methods have been explored for producing G, in order to exploit its extraordinary properties in many applications, including biomedical and biosensing ones. G preparation methods should be selected according to the specific sensing target and mechanism to be utilized, with a balanced consideration on performances (e.g., detection limit and dynamic range), reproducibility, cost and manufacturability. For a wide range of biomedical and biosensing applications, the chemical vapor deposition (CVD) method is considered to be the most promising approach to synthesize large-area, high-guality G, with material properties approaching the theoretical predictions. It is expected that CVD-G will exhibit fascinating electrochemical properties, including wide electrochemical potential windows and low charge-transfer resistance, which can enable the study and exploitation of redox-active biological processes from new perspectives. In our work, we investigate the fabrication of electrochemical biosensors in which G has the role of the transducer and the biosensing element is a redox-active membrane protein embedded in a supported lipid bilayer (SLB) mimicking the cell membrane (Fig. 1). In fact, membrane proteins maintain their functionalities only if they are embedded in their native environment, and so the integration of SLBs in such biosensors is a prerequisite for their functionality. [2] In order to provide a useful support for lipid bilayers, the G surface should be as clean and uniform as possible. In addition to altering significantly the G properties, contaminants generate defects in SLBs that compromise their fluidity and integrity. When using a traditional quartz tube furnace, heavy contaminations can be observed on G films grown by CVD on Cu substrates [3]. The contaminants are mainly SiO₂ clusters, deriving from the quartz walls when Cu atoms diffuse inside them. This effect is linked to the aging of the quartz tube, especially in the presence of oxidants, such as when using methane, ethanol, hydrogen and hydrocarbons. In order to reduce and avoid these contaminations, we designed a novel reactor, based on an alumina shielding tube placed coaxially in the guartz one. The G guality has been characterized both by SEM and by TEM equipped with an energy-dispersive X-ray spectrometer (EDX). Fig. 2 shows the SEM morphological observations of G grown using the old and the novel reactor setup. The G sample shown in Fig. 2b is clearly free of all the contaminations appearing as white nanoparticles in Fig. 2a. Fig. 3 shows a TEM image of a G layer with contaminants on the surface (black dots) and the EDX spectrum evidencing the presence of Si and O in such contaminants. In this work, results obtained for CVD-G synthesized using the novel reactor will be discussed in view of its potential application as a clean and smooth support for lipid bilayers in the fabrication of reliable electrochemical biosensors.

References

[1] K. S. Novoselov et al., Science, 306 (2004) 666.

[2] Castellana, E. T. et al., Surface Science Report, 61 (2006), 429.

[3] N. Lisi et al., Thin Solid Films, 571 (2014) 139.

Figures



Fig. 1: Electrochemical biosensor with supported protein/lipid membrane.



Fig. 2: SEM images before and after the use of the new quartz

tube.



Fig. 3: EDX spectrum (left) and TEM image (right) of graphene on nichel TEM grid