Optical analysis of grain size in graphene on copper revealed by wet chemical oxidation

A. Mameli, G.V. Bianco, M.M. Giangregorio, P. Capezzuto, M. Losurdo and G. Bruno*

CNR-IMIP, Institute of Inorganic Methodology and Plasmas,
Dept of Chemistry, Univ. Campus, Via Orabona 4, 70125 Bari, Italy
*giovanni.bruno@ba.imip.cnr.it

Abstract

Among the several synthesis methodologies for graphene, chemical vapor deposition (CVD) allows the production of large area graphene as required for its application as transparent conductive layer substituting ITO. CVD-graphene presents a polycrystalline structure that strongly influences both mechanical and electrical properties. Nowadays, it is well consolidated, among different users of graphene, that analyzing graphene grains after growth, is important for quality-control. In fact, graphene is a well ordered material and contains internal boundaries, commonly known as "grain boundaries". When graphene is grown, the carbon atoms within each growing grain are lined up in a specific pattern, depending on the crystal structure of sample. With growth, each grain impact others and forms interfaces where the atomic orientations differ. It has been established that the transport properties of the graphene improve as the grain size increases. Therefore, the growth conditions must be carefully controlled to obtain large grain size. Specifically, a strong correlation has been reported between grain sizes and sheet resistance in CVD-graphene[1]. Improving the quality of CVD-graphene requires the optimization of nucleation and growth rates in the synthesis process, in order to provide an increase in grain average size[1,2]. Therefore, diagnostic methodologies for the accurate monitoring of grain sizes and distribution directly on the growing substrate is needed.

In this work we present an effective methodology for analyzing grain size and distribution in graphene as grown on copper by optical microscopy. In contrast to transmission electron microscopy[3,4] and scanning tunneling microscopy[3,5], this optical technique is not expensive and time-consuming and, above all, effective for the diagnostic on large scale. We exploit a wet chemistry approach for the selective oxidation of the metal substrate through graphene grains making their boundaries visible by optical microscopy. Specifically, we use the Fenton's reaction as a source of hydroxyl radicals to functionalize graphene grain boundaries. This diagnostic approach has been optimized by using Raman spectroscopy to probe and confirm chemical and structural changes in graphene and the Cu substrate upon the wet treatment. We also demonstrate the feasibility of this methodology for highlighting defect sites in graphene and, hence, for probing the retention of its structural quality upon post-growth processing such as transferring on other substrate.

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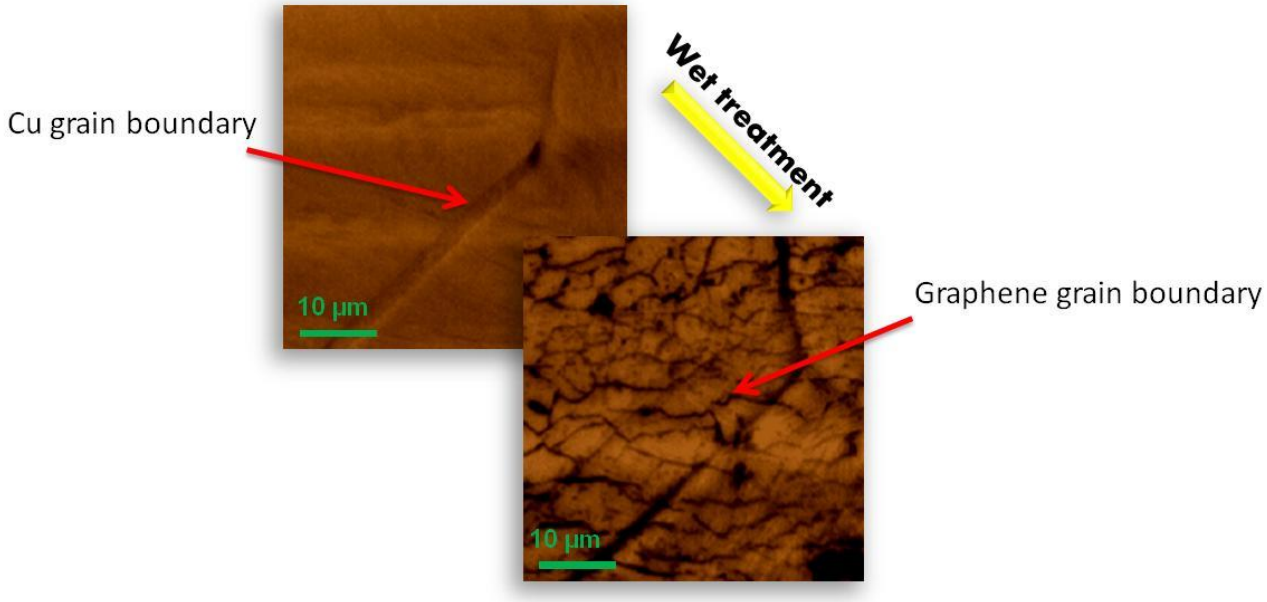

Fig. 1 - Optical images of CVD-graphene on copper before and after oxidation. The grain boundaries are clearly visible after treatment, it can be seen also the growth of graphene grain across the copper grain boundaries.