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Whelan, Patrick Rebsdorf; Kostesha, Natalie; Pizzocchero, Filippo; Bøggild, Peter; Booth, Tim

Published in: Proceedings of Graphene 2014

Publication date: 2014

Link back to DTU Orbit

Citation (APA):

Whelan, P. R., Kostesha, N., Pizzocchero, F., Bøggild, P., & Booth, T. (2014). Oxidative Decoupling Transfer: The influence of copper oxidation on CVD graphene transfer. In Proceedings of Graphene 2014

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Oxidative decoupling transfer: the influence of copper oxidation on CVD graphene transfer

Patrick R. Whelan, Natalie V. Kostesha, Filippo Pizzocchero, Peter Bøggild, Timothy J. Booth

DTU Nanotech - Department of Micro- and Nanotechnology Technical University of Denmark Ørsteds Plads, Building 345E 2800 Kgs. Lyngby Denmark patwhe@nanotech.dtu.dk

Abstract

A key challenge for many applications of chemical vapour deposited (CVD) graphene is the transfer of graphene from catalytic growth substrates to target substrates with a minimal number of defects, cracks, wrinkles and contaminants. Transferring large-area graphene is generally achieved by etching away the catalyst material [1,2,3]. In order to increase the quality of the graphene layers as well as lowering production cost and diminishing environmental implications, electrochemical bubbling methods for transferring graphene without completely etching away the catalyst material have been invented [4,5], thus allowing the catalyst to be re-used for graphene growth.

A recently developed Oxidative Decoupling Transfer (ODT) method for transferring CVD graphene delaminates a graphene/polymer layer from Cu substrates by an electrochemical induced, slow oxidation of the Cu surface [6].

A better understanding of the ODT mechanism itself and what factors influence the transfer time and the quality of the transferred graphene is important in order to make the process industrially relevant. Our hypothesis is that a key factor for the transfer process is the pre-existing oxidation of the Cu substrate that occurs when the sample is left in air before transfer. Here, we present a study of how Cu oxidation influences the ODT process as well as the quality of the transferred graphene layer.

Raman spectroscopy, scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX) were used to study the Cu oxidation near cracks in a graphene layer grown on Cu films using an Aixtron Black Magic CVD system. Graphene was transferred from Cu substrates to SiO₂ using the ODT method, where time-lapse recordings showed that the oxidation of Cu progresses from the edges and inwards. It was shown that the transfer process proceeded faster when the Cu surface was more oxidized. X-ray photoelectron spectroscopy showed the presence of Cu(I) oxide on the Cu substrate after transfer. We used optical microscopy and Raman spectroscopy to characterize the transferred graphene on SiO₂. The transfer from the oxidized samples to the SiO₂ substrates lead to more optically discontinuous graphene layers. In general, it was shown that the quality of graphene transferred with the ODT method depends critically on the treatment of the sample after graphene growth.

References

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Figures



Figure 1: SEM images of graphene on Cu films after growth. Cu grain boundaries and ridges/cracks in the graphene layer are clearly visible. The presence of copper oxide can be seen as the dark areas around graphene cracks and some oxidized areas are pointed out by white arrows. Insets show oxygen content determined by EDX.



Figure 2: Optical images of graphene on SiO₂ after transfer. Insets show Raman spectrum from sample.