Systematic Raman analysis of graphene films grown on Cu foils by ethanol Chemical Vapor Deposition: effect of synthesis temperature

G.Faggio[1], N.Lisi[2], S. Santangelo[3], R.Giorgi[2], Th.Dikonimos[2], **F. Buonocore[2]**, V. Morandi[4], L. Ortolani[4], G. Messina[1]

[1]Dept. of Information Engineering, Infrastructure and Sustainable Energy, University "Mediterranea" of Reggio Calabria, Via Graziella Loc. Feo di Vito, 89122 Reggio Calabria, Italy
[2]ENEA, Materials Technology Unit, Surface Technology Laboratory Casaccia Research Centre, Roma, Italy
[3]Dept. of Civil Engineering, Energy, Environment and Materials, University "Mediterranea" of Reggio Calabria, Via Graziella Loc. Feo di Vito, 89122 Reggio Calabria, Italy
[4]Nat. Res. Council - Inst. of Microelectronics & Microsystems, Via Gobetti 101, 40129 Bologna, Italy

gfaggio@unirc.it

Abstract

Catalytic Chemical Vapor Deposition (C-CVD) is one of the most promising methods developed for large scale synthesis of graphene [1, 2]. Typically, graphene C-CVD takes place by exposure of a transition metal substrate to a hydrocarbon gas.

Inspired by the carbon nanotube growth using alcohol catalytic CVD [3-5], recently there have been a few attempts of graphene CVD growth with ethanol and other alcohol precursors. One advantage of using ethanol lies in its harmless, low cost, and easy-handling nature. In addition, other advantages could rise from a different growth kinetic due to its weakly oxidising nature [6].

As known [7, 8], Raman spectroscopy is a non-destructive and powerful technique for evaluating the structural properties of graphene providing useful information on the defects (D-band) and in-plane vibration of sp^2-carbon (G-band). The second order 2D Raman band has been used as a simple and efficient way to identify the number of graphene sheets.

In this work, we report on a systematic Raman analysis of graphene films grown by CVD on thin copper foil substrates using ethanol as precursor gas, with different dilution in hydrogen or argon. Aim of the work is optimizing the growth of CVD graphene films in terms of quality and number of graphene layers. For this purpose, we investigated the effect of synthesis temperature (860-1070°C) and hydrogen flow (0 or 100 sccm) by coupling Raman spectroscopy with Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM) to study morphology and spatial homogeneity of the films previously transferred to SiO_2/Si substrates.

Figure 1 shows Raman spectra of graphene films obtained at different hydrogen flow (Φ_{H2}) (0 sccm in Fig. 1a, and 100 sccm in Fig. 1b) by varying reaction temperatures in the range 860–1070 °C, for a growth duration of 10 min. Multilayer graphene films are obtained only for growth temperature greater than 930°C. Regardless of Φ_{H2} value, the D-band intensity reduces with increasing synthesis temperature, indicating that high quality graphene films can be obtained at high temperatures. A further reduction of defect density is achieved by adding hydrogen into growth mixture (Fig. 1b).

Figure 2 shows a typical SEM image of CVD graphene film grown at high temperature.

References

[1] X. Li, W. Cai, Jinho An, S. Kim, J. Nah, D. Yang, R. Piner, A. Velamakanni, I. Jung, E. Tutuc, S. K. Banerjee, L. Colombo, R.S. Ruoff, *Science,* 324 (2009) 1312.
[2] A. Reina, X. Jia, J. Ho, D. Nezich, H. Son, V. Bulovic, M. S. Dresselhaus and J. Kong, *Nano. Lett.,* 9 (2009) 30
[3] S. Maruyama, R. Kojima, Y. Miyauchi, S. Chiashi, M. Kohno, Chem. Phys. Lett., 360 (2002) 229.
[4] L.X. Zheng, M.J. O'Connell, S.K. Doorn, X.Z. Liao, Y.H. Zhao, E.A. Akhadov, M.A. Hoffbauer, B.J. Roop, Q.X. Jia, R.C. Dye, D.E. Peterson, S.M. Huang, J. Liu and Y.T. Zhu, *Nature Materials*, 3 (2004) 673.
[5] H.E. Unalan, M. Chhowalla, *Nanotechnology*, 16 (2005) 2153.
[6] Y. Guojun, G. Jinlong, W. Sen, Z. Dezhang, H. Suixia, Z. Zhiyuan, *Carbon*, 44 (2006) 1218.
[7] A.C. Ferrari, J. C. Meyer, V. Scardaci, C. Casiraghi, M. Lazzeri, F. Mauri, S. Piscanec, D. Jiang, K. S. Novoselov, S. Roth and A. K. Geim, *Phys. Rev. Lett*., 97 (2006) 187401.
[8] L.M. Malard, M. A. Pimenta, G. Dresselhaus, M. S. Dresselhaus, *Phys. Reports*, 473 (2009) 51.

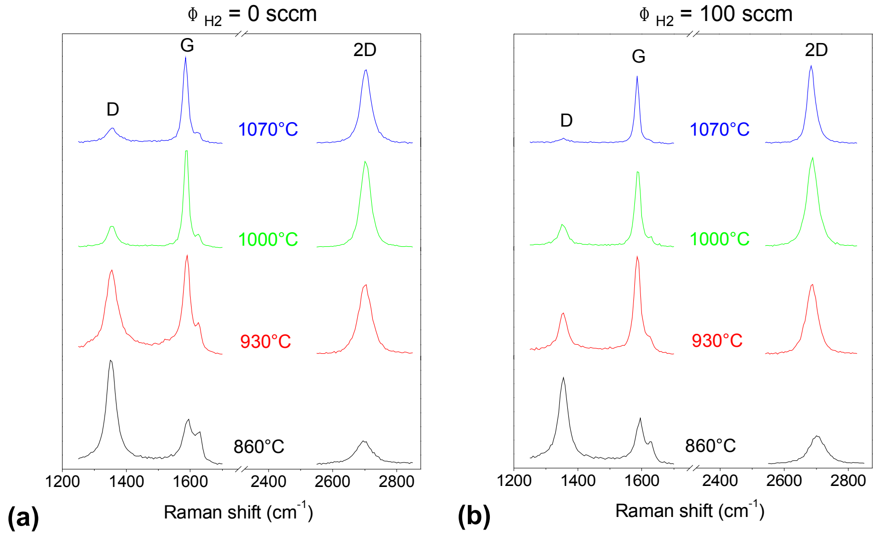

Figure1 Raman spectra of graphene films obtained at different hydrogen flow (Φ_{H2}= 0 sccm in Fig. 1a and (Φ_{H2}= 100 sccm in Fig. 1b) by varying reaction temperatures in the range 860–1070 °C for a growth duration of 10 min.

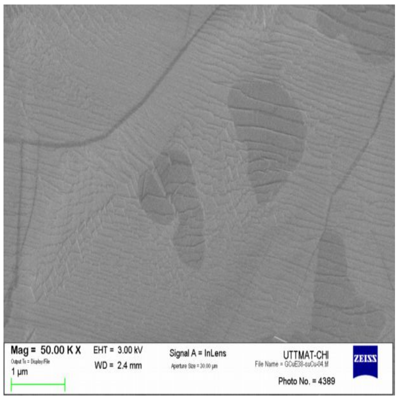

Figure2 SEM micrograph of a sample grown at high temperature by ethanol CVD