

CVD Graphene shape evolution during growth: Impact of the *in-situ* increased hydrogen partial pressure

Zewdu M. Gebeyehu^{1,2}, Marius V. Costache¹,
Maria J. Esplandiú¹, Sergio O. Valenzuela^{1,3}

¹Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and The Barcelona Institute of Science and Technology, Campus UAB, Bellaterra, 08193 Barcelona, Spain

²Universitat Autònoma de Barcelona (UAB), Bellaterra, E-08193, Spain

³Institució Catalana de Recerca i Estudis Avançats (ICREA), 08070 Barcelona, Spain

zewdu.messele@icn2.cat

Abstract

It is well known that exposing chemical vapor deposited (CVD) graphene to hydrogen gas can yield dendritic graphene shapes due to an etching process [1,2]. Here, we demonstrate that similar dendritic structures can be achieved at long growth times without the external addition of hydrogen. These structures are not a result of a diffusion controlled growth as reported before [3,4] but to the competing backward etching reaction which starts dominating over the growth during long growth times due to an *in-situ* increase of the hydrogen partial pressure. We have performed a systematic study on the growth of graphene as a function of time to identify the onset and evolution of graphene etching (Fig 1). During hydrogen partial pressure balancing to find a process window for keeping growth ongoing, two kinds of etching mechanism (oxidative and reductive) were observed (Fig. 2). In addition, we have found that the *in-situ* increased hydrogen graphene etching is dependent on the copper foil geometrical arrangement. Highly etched graphene with dendritic shapes was observed on unconfined copper foil regions which highlight the importance of the reactant

distribution in activating the etching process during growth.

References

- [1] Zhang, Y.; Li, Z.; Kim, P.; Zhang, L.; Zhou, *ACS Nano*, 1 (2012) 126–132.
- [2] Geng, D.; Wu, B.; Guo, Y.; Luo, B.; Xue, Y.; Chen, J.; Yu, G.; Liu, Y. *J Am Chem Soc*, 17 (2013) 6431–6434.
- [3] Zhang, Y.; Zhang, L.; Kim, P.; Ge, M.; Li, Z.; Zhou, C. *Nano Lett*, 6 (2012) 2810–2816.
- [4] Meca, E.; Lowengrub, J.; Kim, H.; Mattevi, C.; Shenoy, V. B. *Nano Lett*, 13 (2013) 5692–5697.

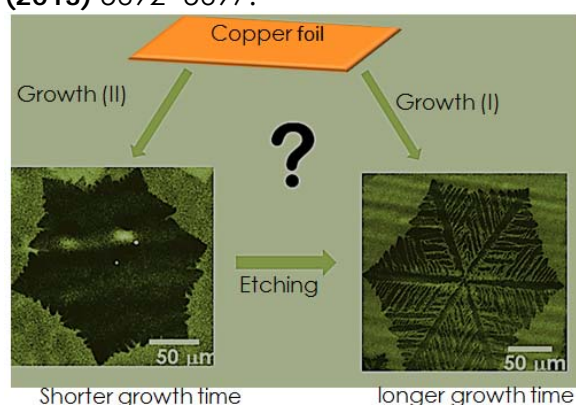


Figure 1: SEM images of graphene on copper foil showing the two possible mechanisms of graphene domain shape evolution.

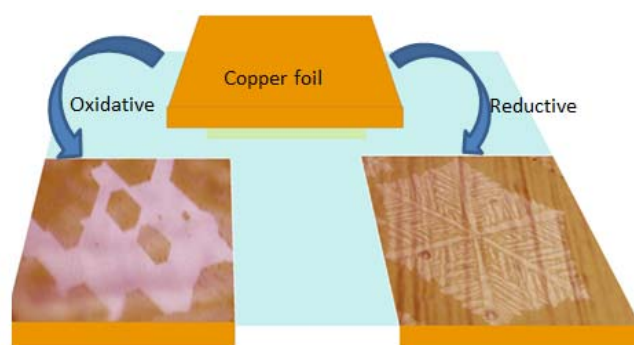


Figure 2: Optical microscope image showing etching at low hydrogen flow (oxidative etching, left) and high hydrogen flow (reductive etching, right).