Graphene growth studies in purified conditions

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Chemical vapor deposition (CVD) of graphene films on copper foils has achieved several milestones since it was first reported back in 2009. The field is mature enough, and tools for mass production are being developed. To reach the necessary standardization of the resulting material as required by the industry, and to further model and engineer the growth, some fundamental aspect of the growth still needs to be elucidated.
Our recent research revealed oxidizing impurities as a dominant factor governing the growth. For instance, oxygen impurities and not hydrogen, are responsible for graphene etching on copper and hydrogen is rehabilitated in its protecting role of counter balancing oxidizing impurities reactions. When the level of oxidizing impurity is minimized, continuous layer of graphene can be grown in the sole presence of methane [1, 2]. The kinetics of the reactions have been addressed and revealed a competitive action between precursor oxidation and carbon growth during graphene formation in the CVD reactor. Based on our experimental evidences a kinetic model including oxygen impurities has been developed. Within this model, a criterion is found for the O2/H2 partial pressure ratio that sets a limit between impurities limited growth and methane adsorption-dissociation limited growth. The level of purity required is such that most of the works to date are in the impurities limited growth regime. In this work, the adsorption-dissociation limited growth regime is explored in a series of growth experiments where the level of oxidizing impurity is strictly controlled. With this new control knob, we found that the growth is speeded up by a factor of x50 compared to the same recipes under UHP conditions without affecting graphene quality. In this purified regime the fundamental kinetic constant and activation energies are retrieved. This study further leads us to identify an origin for bilayer islands formation and gave us new insight on the copper surface oxidation effects on the resulting graphene grain sizes and morphology.
Our work in purified conditions allowed to acquire fundamental knowledge on graphene growth mechanisms and kinetics. This new understanding on the role of oxygen will enable new strategies to tailor the graphene growth and gives the consistency and speed required in a manufacturing context. In addition, the growth method shown in this report is straightforward and very simple to implement in industrial manufacturing processes making graphene sheets mass production economically feasible.

References

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