In-situ Observation of Graphene Growth Dynamics by Environmental Scanning Electron Microscopy

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We report on in-situ graphene growth studies on nickel, copper and platinum catalysts inside a modified environmental scanning electron microscope (ESEM). The method enables a direct observation of the dynamic processes that occur during a complete CVD process, involving substrate annealing, graphene nucleation and growth and finally, substrate cooling. Watching the formation of single atom thin sheets of carbon enables direct kinetic and mechanistic studies in a novel and unparalleled way.

INTRODUCTION

Most promising approaches for industrial scale production of graphene are based on metal catalyzed chemical vapor deposition (CVD). Although improvements in graphene quality and yield have been achieved, there remains a lack in the mechanistic understanding of graphene formation. This lack of understanding is due to the fact that most insights on graphene growth have been derived from post growth characterizations, which are in principle incapable of capturing the dynamics of a CVD process.

EXPERIMENTAL/THEORETICAL STUDY

Graphene growth was performed inside a commercial FEI Quanta ESEM. The instrument is equipped with mass flow meters for gas dosing, a mass spectrometer for the analysis of the chamber atmosphere and a home built laser-heating stage for sample heating. CVD growth experiments were conducted at pressures in the range between $4x10^{-2}$ Pa and several 10 Pa at substrate temperatures of up to 1000 °C.

RESULTS AND DISCUSSION

The in-situ experiments presented here reveal the dynamic nature of the process and provide important and real-time insights on the growth kinetics and the substrate-film interactions at the micron to nanometer scale (Figure 1). In the case of growth on nickel, temperature and atmosphere induced dissolution and precipitation dynamics can be observed. For the case of copper, it is found that graphene growth above 850°C occurs on a pre-melted, highly mobile surface. The nucleation and growth behavior will be discussed and the influence of grain dependent surface dynamics presented. Furthermore, we show that graphene induced copper surface reconstructions occur during cooling.

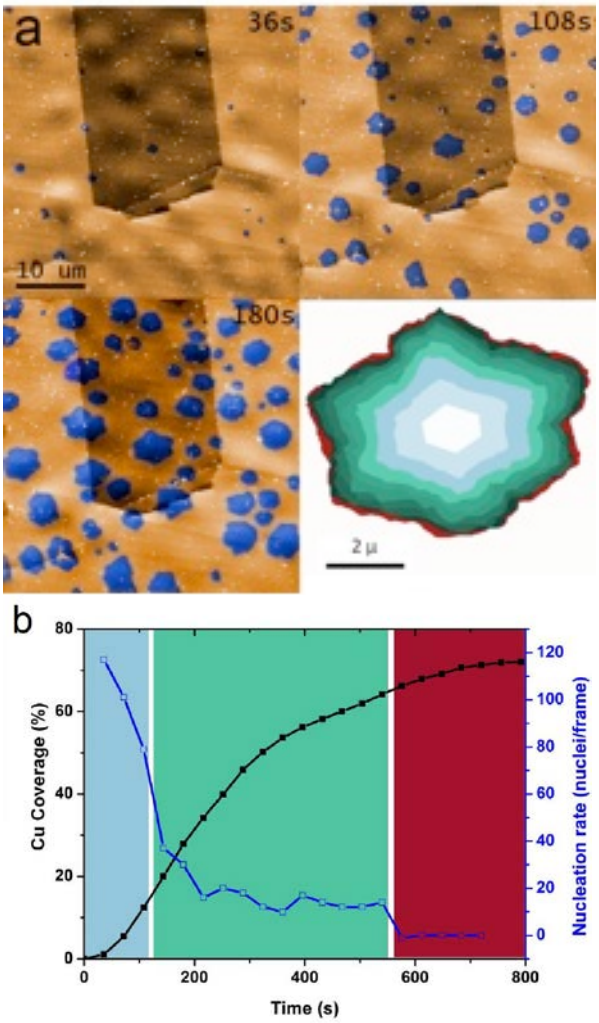

Fig. 1 a) shows colorized snapshots taken during low-pressure CVD growth of graphene on copper at 1000°C. The growth and nucleation behavior can directly be abstracted from the recorded images as shown in b).

CONCLUSION

The study represents a very rare case in catalysis where the dynamics of a catalyst can be studied while at the same time, the product of the reaction can be seen. Direct imaging of the forming graphene at variable magnifications and pressures opens a door for artifact free and real-time observation of the nucleation and growth process.