The Growth of Millimeter-Scaled Graphene Single Crystal through Nucleation Density Control in Height-Confined Reaction Slits

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I. INTRODUCTION

In conventional CVD process, the as-grown continuous graphene film over entire Cu foil could be regarded as the coalescence via many small graphene grains with lateral size less than 50-60 µm. The dense graphene boundaries on continuous graphene films would introduce severe carrier scattering which degrades carrier mobility of graphene films, therefore retarding graphene-related applications. In this work, the growth of high-quality large graphene grains through the nucleation density control in confined reaction slits will effectively reduce the perimeter of graphene grain boundaries of graphene films and enhance its physical properties.

II. RESULTS

The height-confined slit positioned in an enclosed graphite cavity is designed for graphene growth. The Cu sublimation is suppressed within the confined slit, therefore reducing the surface roughness of Cu substrate and graphene nucleation density. The less dendrite-edged graphene single crystals were successfully grown in graphite cavity and height-confined graphite slit as shown in Fig 1 (c) and (d). Besides the control of the growth environment, the oxidized Cu foils are utilized for the fast growth due to the low reaction rate within the confined slit. The surface oxygen not only passivate the Cu active sites to diminish the graphene nucleation density, but also lower the surface reaction barrier to accelerate the growth rate. Fig. 2 (d) shows the size of graphene grains is up to millimeter scaled before coalescence.

III. CONCLUSION

In cooperation of $FeCl_3$/HCl surface pretreatment and surface pre-oxidation, the millimeter-scaled high-quality large graphene single crystals can be achieved in confined reaction slits. Without height-confined reaction slits, the graphene nucleation also can be retained less than 600 nuclei/cm^2. The success of this work would pave a new way for graphene industrial production and their potential applications in the future.

References

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Figures

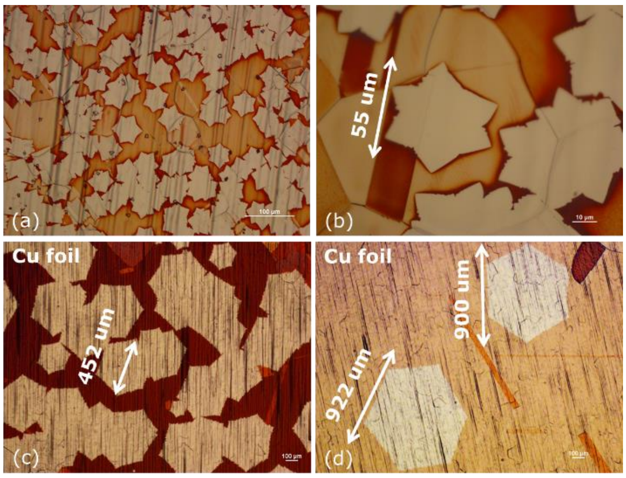

Figure 1. (a) Conventional CVD process. The size of graphene grains is normally less than 50-60 µm. (b) The close inspection of (a) shows graphene grains can grow across different Cu grains. (c) Cu surface pre-treatment in $FeCl_3$/HCl solution. About 600 nuclei/cm^2 has been successfully achieved. (d) the millimeter-scaled graphene single crystal grown on $FeCl_3$/HCl pretreated Cu foils can be further accomplished through a well-designed confined CVD reaction slit.