

*Full Length Research Paper*

# SiC substrate dependence epitaxial graphene growth: Effect of growth temperature and substrate off-cut angle

Dr. B. K. Daas

Abstract

Department of Electrical and  
Computer Engineering, University of  
South Carolina, 301 S. Main St,  
Columbia, SC 29208, USA

E-mail:

[biplobdaas\\_121@yahoo.com](mailto:biplobdaas_121@yahoo.com)

**We present epitaxial graphene (EG) growth on SiC substrate using solid-state decomposition method. The material characteristics are verified using Raman while surface topography analysis is done using AFM. Our investigation indicates that with the increasing growth temperature on SiC substrate, Si face steps become prominent while C-faces wrinkles starts invisible in EG layers due to thicker layer. This indicates step-flow mediated growth on Si face while C face has both step flow and C nucleation. Through validating step-flow growth on Si face, shows offcut dependency surface morphology while graphene quality is intact.**

**Keywords:** 6H-SiC, off-cut, Epitaxial Graphene.

## INTRODUCTION

Graphene is a two-dimensional (2D) carbon honeycomb crystal, the basic building block of other  $sp^2$  carbon nanomaterials, such as nanographite sheets and carbon nanotubes, which exhibits unusual electronic, optical and sensing properties (Saito et al., 2000; Castro et al., 2009; Novoselov et al., 2005; Daas BK et al., 2012; Daas BK et al., 2011; Berger C. et.al., 2004). The charge carriers are “massless” Dirac fermions resulting in high electron mobility (Novoselov et al., 2005). Additionally, other important characteristics such as high crystalline quality, high thermal conductivity and stiffness, plasmonics characteristics (Daas BK et. al., 2011), room temperature quantum hall effect and ballistic transport properties have generated enormous interest (Berger C. et.al., 2004). Since its discovery, graphene has been produced using mechanical exfoliation where graphene layers are peeled off layer by layer using scotch tape (Saito et al., 2000; Geim and Novoselov, 2007). This technique can provide single layer graphene with high structural and electronic properties, but is not suitable for large-scale production

due to poor yield, repeatability, lack of process control and small sample size.

As an alternative, large area epitaxial graphene (EG) is grown by thermal decomposition of the polar c-plane of 4H or 6H SiC in ultra high vacuum or Ar environment at high temperatures. In this technique, Si sublimates off from the SiC substrate, leaving behind carbon atoms which rearrange themselves into a graphene layer (Jahan et. al., 2008). The growth mechanism of EG on SiC is still a current issue of research because it involves high temperatures with three key steps: i) silicon desorption, ii) carbon diffusion and iii) island nucleation (Hupalo et al., 2009). It has been established regarding EG growth in Ar environment as well as ultra-high vacuum (UHV) environment, While our previous paper (Daas et al., 2012) justify this issue. In our previous paper, we show detail EG growth mechanism under UHV condition (Daas BK et. al., 2011; Daas et al., 2012), compared polar and non-polar face growth (Daas et al., 2012) in order to investigate EG growth mechanism. However, still

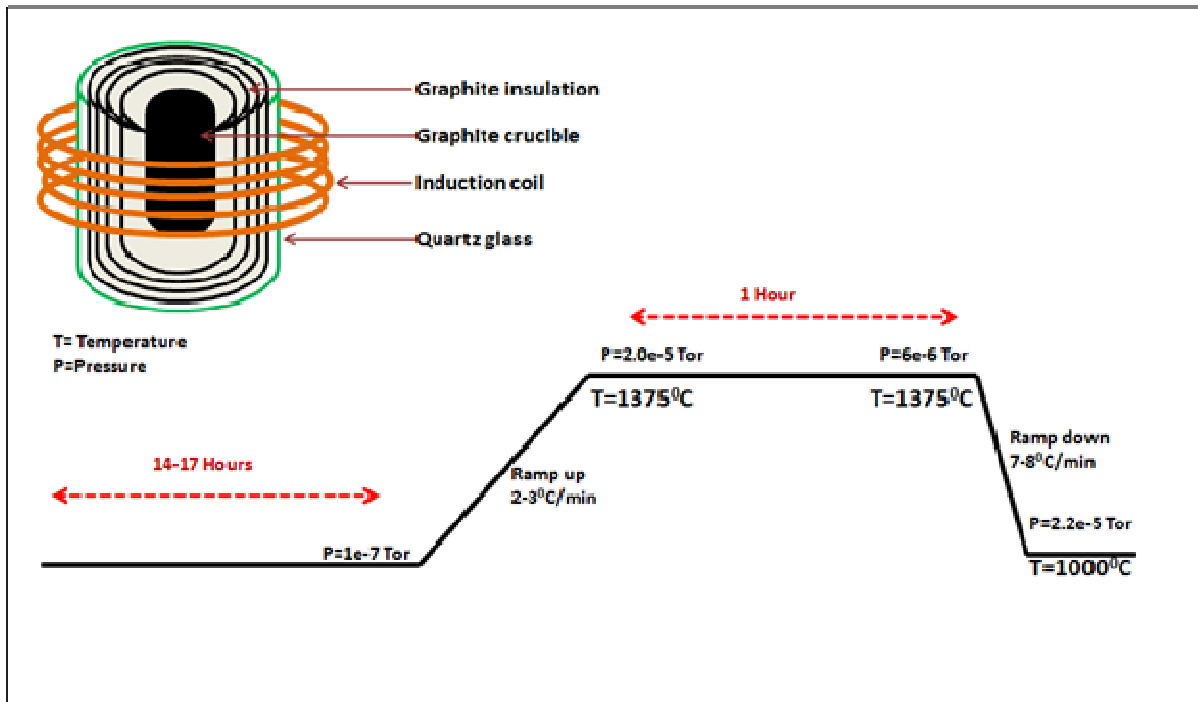


Figure 1. Temperature and pressure profile for EG growth using RF reactor at UHV.

question needs to be answered regarding temperature and SiC substrate off-cut dependency. In this article, we primarily document surface morphology investigation while varying growth temperature and substrate offcut angle to move another step ahead for EG growth mechanism under UHV.

## Experiment

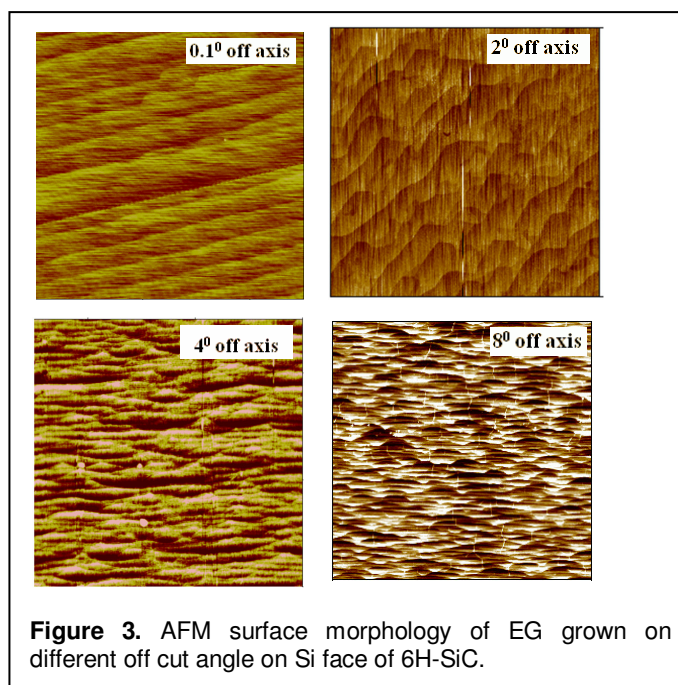
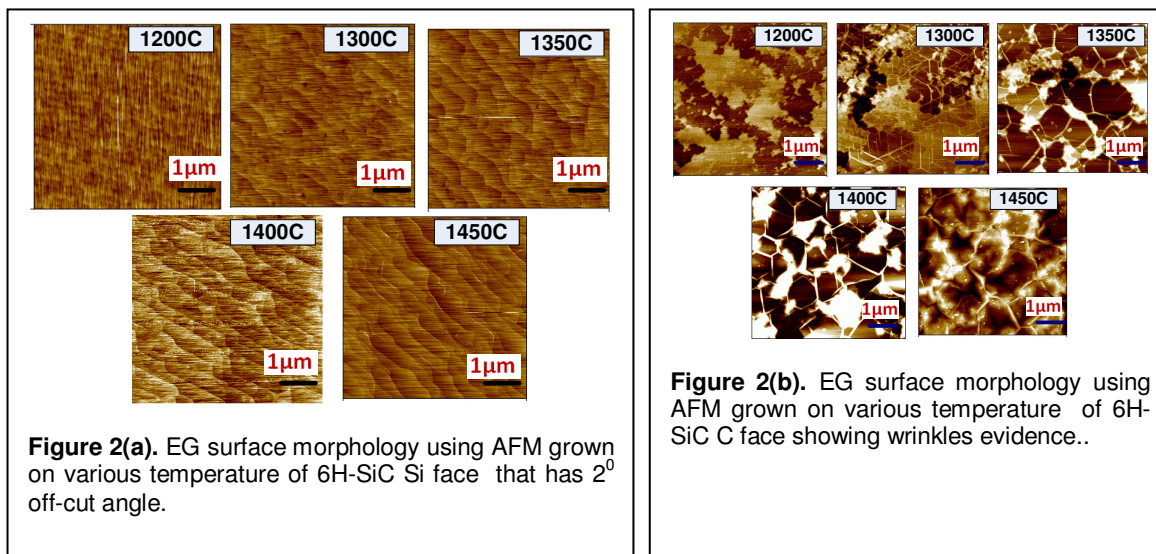
Epitaxial growth of large-area graphene by thermal decomposition of commercial <0001> 4H and 6H SiC substrates at high temperature and vacuum (Berger C. et. al., 2004; Jahan et.al., 2008) produces EG a few monolayers (ML) to >50 ML thick, depending on growth conditions and substrate. In our experiments, EG was grown on commercial, nitrogen doped, 6H-SiC surfaces prepared by chemical mechanical polishing (CMP). The 1cmX1cm samples were degreased using Trichloroethylene (TCE), acetone and methanol respectively. They were then rinsed in DI water for three minutes. The samples were finally dipped in HF to remove native oxide and rinsed with DI water before being blown dry. They were then set in a graphite crucible in an inductively heater furnace where high vacuum was maintained ( $<10^{-6}$  Torr) and baked out overnight at  $1000^{\circ}\text{C}$ . Figure 1 shows the details of susceptor environment with growth parameter (temperature and pressure) curve. No graphitization occurs during  $1000^{\circ}\text{C}$  bake out. The temperature was

then slowly raised to the growth temperature ( $1200$ - $1450^{\circ}\text{C}$ ). All growths were performed for 60 minutes before cooling to  $1000^{\circ}\text{C}$  at a ramp rate of  $7\sim 8^{\circ}\text{C}/\text{min}$  and eventually to room temperature. Slow temperature ramps were utilized to minimize thermal stress on the samples (Daas BK et. al., 2011; Kevin et al., 2012). We also confirmed EG quality through Raman and surface topography through AFM while XPS analysis gives us EG thickness.

## RESULT AND DISCUSSION

### a) Growth temperature dependency

At this point we investigate the effect of growth temperature while growth time remains fixed to 1 hour, samples were placed both face up and face down orientation. Detail experimental explanation is the same as our previous paper (Daas et al., 2012). So the other parameters affecting growth condition remain same for all the case. Figure 2(a) shows the AFM surface morphology of EG grown on 6H-SiC Si-face which has an offcut angle of  $2^{\circ}$ . It is to be noted that at  $1200^{\circ}\text{C}$  no EG was grown, confirmed through Raman analysis while other it has EG evidence while growth temperature  $>1200^{\circ}\text{C}$ . At  $1200^{\circ}\text{C}$  there is minimal steps (no EG grown) are seen while steps becomes more prominent with increasing growth temperature (EG evidence), indicating growth is step flow mediated (supported by (Joseph et al., 2010)) and with



increasing growth temperature EG is being thicker as confirmed through XPS analysis. Si face EG thickness variation is ~1ML while temperature varies from 1200°C to 1450°C. In this document we only focus on the surface morphology but do not comment regarding EG growth mechanism.

Figure 2 (b) shows EG grown on 6H-SiC substrate C face while C face was placed as face down position in the susceptor. Raman characterization was done to confirm graphene on these samples. It is also visible from the carbon face AFM images that growth are defects

mediated whereas wrinkles are formed as silicon sublimates off from the defects positions. At 1200°C it is visible while increasing growth temperature, creates thicker layer and wrinkles are less visible, confirmed defect mediated growth, supported by ref (Hupalo et al., 2009). Through several experiments we confirmed that on Si face at 1350°C shows good Raman characterization with disorder ratio ( $I_d/I_g$ ) 0.005 while for C face it is at 1400°C. We attribute this temperature as the high quality EG growth condition for our case, based on Raman analysis. For more investigation of substrate

offcut dependency we use 1350<sup>0</sup>C as the growth temperature for Si face.

### b) Substrate off-cut dependency

Difference in growth mechanism between silicon and carbon face initiates us to grow graphene on SiC substrates of varying off-cuts. The following Figure-3 shows the AFM surface morphology of EG on silicon face with growth temperature 1350<sup>0</sup>C when substrate off cut angle was 0.1, 2,4 and 8<sup>0</sup>. On Si face EG growth mechanism occurs through C nucleation and steps merging with each other. So for the lower offcut angle EG steps are visible due to wider step size of SiC while higher off cut angle shows zig-zag feacture due to steps merging with each other as they are close to each other. At lower off-cut angles, we find no wrinkles while steps become wavy and wrinkles are visible at higher off cut angle, confirming growth is more likely to enhance through defects at higher off cut angles. Raman characterization performed on these substrates confirms graphene quality while no significant difference in material Varity was observed.

### CONCLUSION

Through this study of surface morphology analysis, we find that Si face EG growth is likely to be step flow

mediated at lower offcut angle while it is both defect and stepflow mediated at higher offcut angle. C-face EG growth most likely tends to be through defect and stepflow mediated. However, this observation is based on surface topography analysis while more investigation is required to explicitly document this hypothesis.

### REFERENCES

- Berger C (2004). J. Phys. Chem. B 108, 19912  
Castro NAH, F Guinea, NMR, Peres KS, Novoselov, AK Geim (2009). Rev. Mod. Phys. 81, 109  
Daas BK, KM Daniels, S Shetu, MVS chandeeshekhar (2012). Materials Science Forum, 717-720, 665  
Daas BK, KM Daniels, S Shetu, MVS Chandeeshekhar (2011). J. Appl. Phys. Vol 110, issue 11 ,113114  
Daas BK, Sabih UO, S Shetu, Kevin MD, S Ma, TS Sudarshan, MVS (2012). Chandrashekhar *Cryst. Growth Des.*, 12 (7), pp 3379–3387  
Daas BK (2012). Materials Science Forum, 717-720, 633  
Geim AK, KS Novoselov (2007). Nat. Matter. 6 183  
Hupalo M, E Conrad, MC Tringides (2009). Phys. Rev. B 80, 041401(R)  
Jahan M. Dawlaty (2008). Appl. Phys. Lett. 93,131905  
Joseph LT (2010). App. Phy. Lett. Vol 96 page 222103  
Kevin MD, BK Daas, N Srivastava, C Williams, RM Feenstra, TS Sudarshan, MVS Chandrashekhar (2012). J. Appl. Phys. Vol 111, issue 11  
Novoselov KS, AK Geim, SV Morozov, D Jiang, MI Katsnelson, IV Grigorieva, SV Dubonos, AA Firsov (2005). Nature-London, 438, 197-201  
Saito R, G Dresselhaus, MS Dresselhaus (2000). Phy.Rev.B 61, 2981