Effects of substrate temperature on graphene grown on Ni foil via atmospheric pressure chemical vapor deposition

QunfeiZou[1,a],GuangzhiYin[2], YueliangGu[2], Deming Xie[1,b]

[1]zhejiang university of technology,hangzhou,zhejiang,310014,China.

[2]Shanghai Synchrotron Radiation Facility, Shanghai Institute of Applied Physics, Chinese Academy of Sciences , Shanghai , 201203 , China

[a]zqf1221@163.com ,[b]xiedeming@sina.com

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Abstract. Multilayer graphene was grown on Ni foil(30μm) via atmospheric pressure chemical vapor deposition (APCVD) at different temperature of 800℃,900℃,1000℃, we observe structural difference between the three process by in situ X-ray diffraction and Raman measurements, our experimental results show that temperature plays an important role on graphene growth, the growth rate is increased with the increasing temperature. Moreover, we found that at growth temperature of 900℃, graphene has the best quality. These results will be helpful to better understanding the growth mechanism in the graphene growth process.

Introduction

Since its discovery,graphene has attracted more and more interest as apotential candidate for next generation electronic devices[1,2] for its remarkable electronic properties[3], Many efforts have been made to produce graphene using a variety of method,suchas reduction of graphene oxide[4], chemical vapor deposition (CVD)[5],mechanical exfoliation of highly oriented pyrolytic graphite (HOPG)[6]and epitaxial growth on SiC substrates[7,8],To date,large-Area and high-qualitygraphene has been reported by CVD, which uses transition metalsubstrates, such as Cu and Ni as a catalyst to grow graphene[9-11]. However, the growth mechanism of graphene on catalyst is still not very clear, it is necessary to studythe effect of substrate temperature on graphene grown.

This paper present few layer graphene (FLG) growth on Ni foil(30μm) by CVD at three different temperature including 800℃,900℃and 1000℃.X-ray diffraction(XRD) is a nondestructive method of characterization which can provide structure information ofthe interlayer spacing and strain,the whole experiment process was observed using in situ XRD in Shanghai Synchrotron Radiation Facility(SSRF). The surface structure information was also characterized by Raman spectroscopy.From these results,we found that temperature plays an important role in graphene growth and will affect the growth process, such as growth rateand crystalline quality.

Experimental and results

For in situ observation of the whole graphene growth,a home-built reactor consists of Mass-flow Controller (flowmeter), Vacuum pump, Temperature Controller,Water Cooling Circuitsand ,Vacuometer were prepared, Ni foil(30μm)with a purity>99.99% purchased from HeFeiKeJing Materials Technology Co.Ltd. was cutted into 1x1cm^2, and ultrasonic cleaned by acetone, absolute ethyl alcohol, deionized water for 5minutes respectively, Figure.1 shows the illustration of the graphene growth process and its different stages,the CVD chamber for graphene growth was pumped to approximately 400 Pa , then the Ni foil was heated up to 900℃in 28 minutes for high temperature heat-treatment for15 minutes, at the same time, 180sccm H_2(10%) /Ar and 60sccm Ar mixed gaswas introduced during the whole process until the heat treatment ends, the sample was heated up to or cool down to designated growth temperature, XRD equipment starts to collect data once 150sccm CH_4, 10sccmH_2mixed gas was introduced. CH_4 was removed once the intensity of graphene peak(002) increased to a designated value, and replaced by 180sccm

H_2(10%)/Ar,60sccm Ar mixed gas.

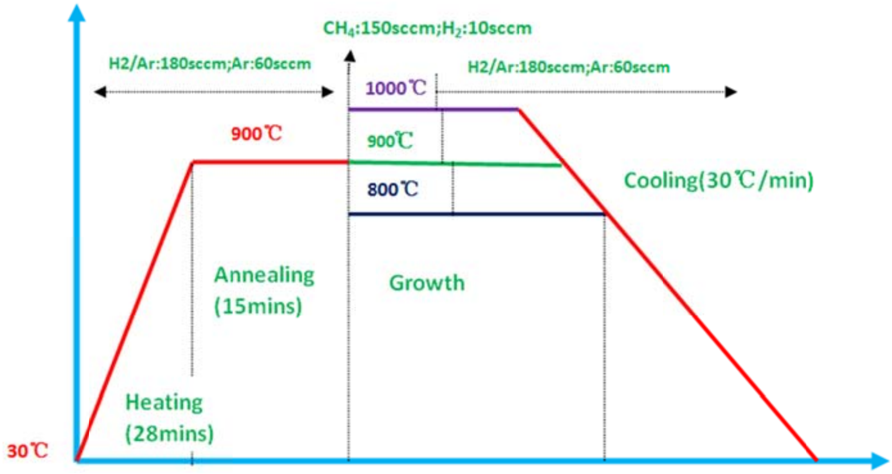

Figure.1. Illustration of the graphene growth process and its different stages.

XRD measurements at grazing incidence werecarried out bysynchrotron radiation apparatus at the beam-line BL14B1 of Shanghai(SSRF), the wavelength of X-ray is 1.239Å[12]. The beam size on the sample was approximately 0.2×0.2 mm2 and was confined by vertical and horizontal slits. A MarCCD detector was used to acquire two-dimensional X-ray diffraction signals with sub-second time resolution, which means we can obtain information of the whole growth process,a two-dimensional GIXRD patterns can obtained every 13 seconds, Figure.2 illustrates (a)the peak area of graphene(002)diffraction peak dependence of time during the whole process at different temperature of 800℃,900℃,1000℃, (b)XRD profile of graphene(002) correspond to the peak of dot curve in (a) at different temperature. it is clearly to see that the graphene peak appear earlier at 1000℃ than 900℃ or 800℃, Which may due to a higher catalytic activity of Ni foil at higher temperature. In the meantime, the maximum Peak Area of graphene(002) was much higher at 1000℃ than grown at 900℃,800℃, indicating that the growth rate of graphene is increasedwith the increasing temperature. Finally, 180sccm Ar/H_2(10%),60sccm Ar mixed gas was introduced,it was surprised to found that the peak area of graphene(002) started to be weaken after a smooth stage, which indicating that graphene can be removed from Ni substrate gradually by Ar/H_2 mixed gas. Moreover, we found that the graphene grown disappears faster at higher temperature. Which further indicating the effectof growth temperature.

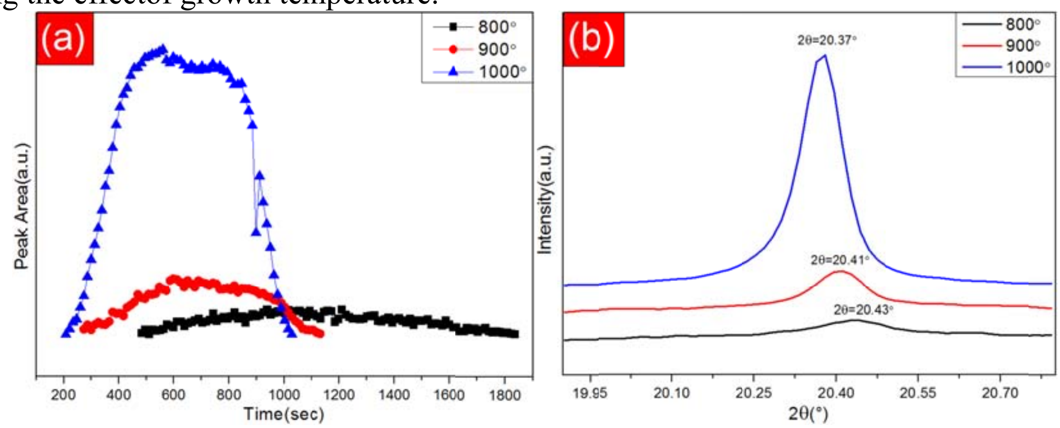

Figure.2.(a)the Peak Area of graphene(002) diffraction peak dependence of time during the whole process at different temperature of 800℃,900℃,1000℃,(b)XRD profile of graphene(002) correspond to the peak of the curves in (a) at different temperatureat 800℃,900℃,1000℃.

Raman spectroscopy(633nm excitation) and optical microscope were used for graphene characterization.Figure.3 showed Raman spectrum of graphene films. From this figure,graphene with G peak at~1582cm^{-1},2D peak at~ 2730cm^{-1} was obtained.which is listed in table.1, As we known, I_D/I_G value can represents the quality of graphene, we found that with the temperature increasing,the quality of FLG increases, the I_D/I_G≈0.0295 at 900℃reveals a best quality among the three temperature,At higher temperature (1000℃), the quality of the FLG would be deteriorate,which consists with the article reported by Zhongliang Liu[13].optical microscopespectrum of FLG at different temperature were showed in Figure.4,which reveals a best quality at temperature of 900℃corresponds to the result of Raman spectrum.

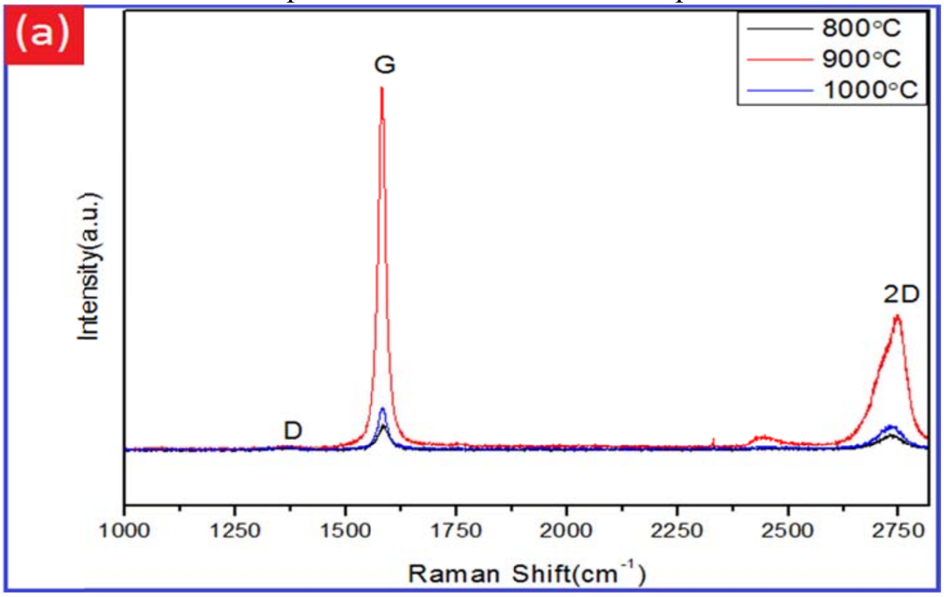

Figure.3.(a)Typical Raman spectra of graphene growth on Ni foil for 13mins at different temperature for 800℃,900℃,1000℃

Table 1 parameter of the Raman spectra at different temperature

	Position of D peak (cm^{-1})	Position of D peak(cm^{-1})	Position of D peak(cm^{-1})	I_D/I_G	I_{2D}/I_G
800°	1365	1583	2732	0.273	0.72
900°	1365	1582	2736	0.0295	0.579
1000°	1365	1583	2730	0.18415	0.636

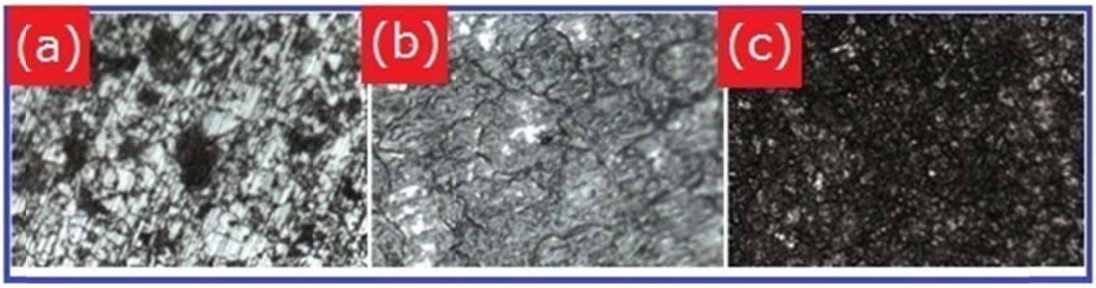

Figure.4 Optical microscope of graphene grown on Ni foil for 13 mins at temperature of (a)800℃,(b)900℃,(c)1000℃.

Conclusions

In summary,multi-graphene layer was obtained on Ni foil by atmospheric pressure CVD,in-situ XRD was used to observation of the whole growth process,samples were characterized by Raman spectroscopy and optical microscope,we found that temperature plays an import role on the FLGlayer formation on Ni foil,thegrowth rate has significant positive correlation with the temperature,among three temperature, we found 900℃ is the best temperature for graphene growth with better quality,at a higher temperature (1000℃), the quality of the FLG would be deteriorate.

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Preference

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