Rapid growth of large area graphene on glass from olive oil by laser irradiation

Yihe Huang1, Margherita Sepioni2, David Whitehead1, Zhu Liu1,3, Wei Guo1, Xiangli Zhong3, Heng Gu1 and Lin Li1

1 Laser Processing Research Centre, School of Mechanical, Aerospace and Civil Engineering, The University of Manchester, Manchester M13 9PL, United Kingdom
2 Graphene Enabled Systems Ltd, Core Technology Facility, 46 Grafton Street, Manchester M13 9NT, United Kingdom
3 School of Materials, The University of Manchester, Manchester M13 9PL, United Kingdom

E-mail: lin.li@manchester.ac.uk

Received 12 June 2019, revised 17 January 2020
Accepted for publication 11 March 2020
Published 6 April 2020

Abstract

Although homogeneous, high quality graphene can be fabricated on a Cu or Ni sheet using the traditional chemical vapour deposition method at high temperatures (over 1000 °C) under specific atmospheric conditions, their transfer to another substrate is difficult. In this paper a novel method of rapidly (i.e. 3–6 s of laser irradiation) producing a large area (>3 cm²) graphene film from olive oil on a glass surface (pre-coated with a 5–28 nm thick Ni film) with defocused, large area continuous laser irradiation is described. The turbostratic graphene film (6 layers) grown in such a way has shown high electrical conductivity (sheet resistance of around 20 Ω sq⁻¹) and an optical transmittance of 40–50%. With femtosecond laser patterning, 70% optical transparency was demonstrated. Continuous large area graphene was formed at relatively lower temperatures (<250 °C) and without the need for specific atmospheric conditions. The basic process characteristics and mechanisms involved are discussed.

Keywords: graphene, olive oil, low temperature, transparent, laser, electrical conductivity

(Some figures may appear in colour only in the online journal)

1. Introduction

Graphene is an attractive 2D material with outstanding properties including high electrical and thermal conductivities and it has very high tensile strengths [1, 2]. It is a single or a few layer sp² bonded carbon atoms arranged in a two dimensional hexagonal crystal lattice [3]. Chemical vapour deposition (CVD), operated at around 1000 °C temperature, is a widely adopted technique to synthesis large-area, high quality, homogeneous crystalline graphene on metallic foils [4]. Polycrystalline nickel, copper and copper–nickel alloy are widely used as substrates for CVD production of graphene [5–9]. High purity gases such as CH₄, H₂ and Ar, are commonly used as the carbon sources, which are processed at around 1000°C temperature for around 30 min. High vacuum processing condition is also often required [10–12].

Using liquid carbon source such as oil for CVD production of graphene was also demonstrated [13]. However, it required 30 min annealing and cooling time to form graphene. The main drawbacks of thermal CVD graphene growth are slow, costly and involve high temperatures that would not be suitable for temperature sensitive materials. Moreover, graphene transfer from nickel or Cu foil to another substrate has relatively low coherence surfaces and is costly [14, 15].

CVD growth of large area and high quality graphene on copper films for applications in nano and a micro electromechanical systems has been achieved [16]. It utilizes absorbance of carbon atoms from copper. Therefore a copper film about 1 µm thick is required to avoid the instability and dewetting of copper thin films in the graphene growth process at 1000 °C [4]. A nickel film has carbon solubility at a relatively lower
associated process mechanisms are presented and discussed. Characteristics, sheet resistances, optical transparency, and the inert gas environment. The basic fabrication process, material whole operation can be carried out without a vacuum or an fully submerged in oil without a contact with ambient air, the stainless vessel.

440 W cm$^{-1}$ 1070–1080 nm laser wavelengths, at a power density about a 16 KW IPG multi-mode Ytterbium fibre laser, emitting Laser heating in ambient condition was carried out using 2.2. Graphene growth Nickel coatings were sputtered at 25 nm min$^{-1}$ of layers by pulsed laser ablation or continuous CO$_2$ laser post processing 

2. Materials and experimental procedure

2.1. Metal deposition
Nickel coatings were sputtered at 25 nm min$^{-1}$ using a W150T Turbo-Pumped sputter coater on borosilicate glass sheets (20 mm × 15 mm × 1 mm thickness). The thicknesses of the deposited nickel layers were: 5 nm, 7 nm, 9 nm, 14 nm, 21 nm and 28 nm respectively.

2.2. Graphene growth
Laser heating in ambient condition was carried out using a 16 KW IPG multi-mode Ytterbium fibre laser, emitting 1070–1080 nm laser wavelengths, at a power density about 440 W cm$^{-2}$. In order to achieve a large area and a relatively uniform power distribution, a 60 mm diameter defocused laser beam was directed to the experimental specimen. As illustrated in figure 1, the specimen was submerged in olive oil in a stainless vessel.

In this paper we report, for the first time, the growth of large area (>5 cm$^2$) multiple layer (6 layers) graphene films from a liquid precursor (extra virgin olive oil) through laser irradiation of an ultra-thin nickel film (<30 nm) on glass submerged in the oil. The nickel film allowed the absorption of the laser beam at the interface of oil and the metal. Dissolution of oil occurs adjacent to the nickel film. The unsaturated fatty acid in the extra virgin olive oil with the lower smoke point was easily thermally dissociated into carbon at relatively low temperatures of 150 °C–200 °C [24]. As the nickel surface was fully submerged in oil without a contact with ambient air, the whole operation can be carried out without a vacuum or an inert gas environment. The basic fabrication process, material characteristics, sheet resistances, optical transparency, and the associated process mechanisms are presented and discussed.

3. Result and discussion

3.1. Surface morphology and single wavelength Raman spectra
The typical morphology of the surfaces after laser irradiation of the nickel coated glass in olive oil for 4–6 s is shown in figure 2 taken with a FEG–SEM. A thin film with paper wrinkle morphology was formed and covered nano-islands of white protrusions.

In order to understand whether the grown coatings were graphene or not, Raman spectrometry at a 514 nm stimulating temperature (e.g. 500 °C), based on carbon segregation after carbon diffusion in the metal [17, 18]. Until now, there was no graphene growth on nickel films with a thickness less than 50 nm.

Laser chemical vapor deposition (LCVD) has high flexibility for arbitrary graphene patterning via non-contact and maskless fabrication processes. Hence, high efficiency and high binding forces between graphene and substrates could be achieved in a single-step combining the graphene growth and patterning [19]. CH$_4$ and H$_2$ gas carbon sources at a 5:2 ratios was commonly used in LCVD processes, and the number of graphene layers could be controlled by a pulsed or continuous wave laser through controlling the laser scanning speed. However, LCVD processes suffer from low uniformity and the use of hazardous gases [15, 20, 21]. Solid carbon precursors such as amorphous carbon coating on Ni, Cu films or Ni/Cu alloys have been developed [9, 18, 19]. The graphene quality could be controlled by the thickness of carbon coating and the percentage of copper in the Ni/Cu alloy [9, 18, 19]. In addition, multilayer graphene coatings can be reduced to fewer number of layers by pulsed laser ablation or continuous CO$_2$ laser post processing [22, 23].

2.3. Material characterisation
The laser processed samples were analysed using Raman spectrometry with multiple wavelengths (514 nm and 633 nm) (inVia Qontor confocal Raman microscope; Renishaw), field emission gun–scanning electron microscopy (FEG–SEM, Sigma VP field emission gun scanning electron microscope; Zeiss), transmission electron microscopy (Titan Chemi STEM; Zeiss), atomic force microscopy (AFM; Bruker), and energy disperse spectroscopy (EDS, Merlin SEM with Gemini II; Zeiss) were used for material characterisation. The AFM was used to determine the surface morphology and x-ray photon-spectrometry (XPS; Kratos) was used to understand the atomic structures of the materials. Sheet resistances were determined using the 4-point probe method and recorded by a 6220 current source (Keithley Instruments, Cleveland, OH, USA). The optical transmittance of the materials was examined using an optical spectrometer (300–1100 nm). The temperature of the materials during laser processing was measured using a thermocouple mounted on the bottom centre of the stainless steel vessel and the surface temperature was estimated using computational fluid dynamic modelling using ANSYS.
wavelength was used for the 9 nm nickel coated samples after 1–6 s of laser irradiation. From figure 2(b), it can be seen that olive oil was disintegrated into carbon chains when the specimen was laser irradiated for 1 s, which was demonstrated by the Raman peak close to 3000 cm$^{-1}$. The G and D peaks appeared when the laser irradiation time passed 2 s. A short 2D peak indicates the possibility of crystallization of carbon atoms segregated from nickel. After 3 to 4 s of laser irradiation, G and 2D peaks were clearly seen. The $I(2D)/I(G)$ ratio increased gradually with increasing laser irradiation time indicating increasing nano-crystallization. The best crystallization condition of the carbon was at 4 s when the ratio of $I(2D)/I(G)$ was about 0.75. For laser irradiation time between 3.5 and 4.5 s, a decrease of $I(D)/I(G)$ demonstrates a reduction of defects at grain boundaries, because of sufficient crystallite length were present in the detected area [3, 25, 26]. Both $I(D)/I(G)$ and $I(2D)/I(G)$ ratios decreased with laser irradiation time of 5–6 s compared with those with 4 s laser irradiation as shown in figure 2. After 4 s of laser irradiation, carbon atoms segregation from the nickel film may have surpassed the speed of carbon crystallization due to the excessive carbon dissolution in the nickel film at high temperatures. Amorphous carbon generation would dominate the carbon film generation during cooling after 5 s of laser irradiation as shown in figure 2(a). The thicker and darker paper like wrinkles on thin carbon film in figure 2 showed the possibility of a mixture of amorphous carbon with a carbon crystal structure.

3.2. Multi-wavelength Raman spectra and cross section characteristics

Multiple wavelengths Raman spectroscopic analysis (at 514 nm and 633 nm) was carried out to further understand the coating material formation mechanism. The results are shown in figure 3. After 3–6 s of laser irradiation, the G peak position shifted from 1588 to 1600 cm$^{-1}$, indicating a transition of the carbon film to a nano-crystallized structure. If there were sp$^2$ rings in the material, the G peak dispersion would saturate at a maximum of 1600 cm$^{-1}$. The dispersion rate would increase with disorder [27]. The D peak always disperses with the excitation energy in all carbons; however, the more disorder the less dispersion will be, which is opposite to the behaviour of the G peak. In our experiments, the D peak shifted from 23 cm$^{-1}$ to 33 cm$^{-1}$ and rebounded back to 23 cm$^{-1}$ as the laser irradiation time increased from 3 to 5 s. This phenomenon proved the production of less disordered structures in the carbon film when the irradiation time increased to 4 s. The weaker crystallized structure was equalized to a more disordered structure, which induced less D peak shifts. On the contrary, the G peak shifted from 12 cm$^{-1}$ to 9 cm$^{-1}$ and rebounded to 15 cm$^{-1}$ as the laser irradiation time increased from 3 s, through 4 s to 5 s.

In the work of Tuinstra and Koenig [26, 28] $I(D)/I(G)$ was used to estimate the sp$^2$ crystallite length ($L_a$). In the crystallization stage, the ratio is proportional to $L_a$, which can be expressed as [23, 26],

$$\frac{I(D)}{I(G)} = C' (\lambda) L_a^2 \quad (1)$$

where $C'$ is a constant depending on the wavelength ($\lambda$) of the laser. Therefore, with the increase of $I(D)/I(G)$ for laser irradiation time of 3.5 s, the crystallite length would grow to 11.83 nm, according to the $I(D)/I(G)$ ratio shown in figure 2. Defects of carbon crystallization were reduced when the laser irradiation time increased from 3.5 s to 4.5 s. In this stage $L_a$ was proportional to $I(D)/I(G)$. An empirical relationship developed by Cancado et al. relates $L_a$ directly with the excitation wavelength, and $L_a$ was interpreted as the distance over which phonons (atomic vibrations) get de-correlated [29, 30]. It is expressed as [27, 28]

$$L_a (\text{nm}) = 2.4 \times 10^{-10} \lambda^4 \left(\frac{A_D}{A_G}\right)^{-1} \quad (2)$$

where $A$ is the integrated Raman peak intensity and $\lambda$ is the exciting laser wavelength in nm. The crystallite length ($L_a$)
Figure 3. G and D peak shifts between 514 nm and 633 nm Raman spectrometry spectra for specimens with 9 nm nickel coating irradiated by the laser in olive oil for (a) 5 s, (b) 4 s and (c) 3 s.

Figure 4. XPS analysis of the specimen with a 9 nm nickel coating laser irradiated for 4 s in olive oil.

was calculated to be about 44 nm for the specimen laser irradiated for 4 s. From the XPS analysis (figure 4) of the film produced for laser irradiation time of 4 s, it can be found that the crystallite carbon film was deconvoluted into 92.8% sp² (284.45 eV), 3.9% C–O (286.7 eV), 4.43% C=O (288.7 eV). The sp² ring dominated the film structure and the nano-crystallization graphene showed defeats or edges of the oxygen group (C–O and C=O). From the TEM image of the cross section of the graphene coating (using Titan ChemiSTM) few layers graphene (6 layers) was found as shown in figure 5. One area of graphene was segregated from the nickel grains and the other area showed precipitated graphene from the nickel grain boundary to parallel layers originated from nickel grains.

As laser irradiation time increased, La reduced to 7.3 nm according to equation (1), which gradually reduced and turned to be amorphous carbon as the laser irradiation time increased from 1 s to 4–5 s.

3.3. The effect of nickel coating thickness on graphene growth characteristics

With an increase of nickel coating thickness from 5 nm to 11 nm, less defect and larger La values from 18 nm to 44 nm were developed, calculated according to the ratios between I(D)/I(G). There were many pores and rougher surfaces on the 5 nm nickel film coated on the glass surface. With the increase of nickel coating thickness, the thin carbon film grew more continuously and the nano islands of protrusions were larger, smoother with a lower density (i.e. larger separations), as shown in figure 6(a). The mechanism of white spot protrusion, as shown in figure 6(c) EDS mapping, may be related to balling effect of the nickel sputter coating during annealing from laser irradiation. Using multi-mode atomic force microscopy (AFM) and energy disperse spectrum (EDS), it can be seen that (figure 6(b)) the surface topography of the
Figure 5. TEM image of the graphene film cross-section grown on a 21 nm nickel coating on borosilicate glass.

Figure 6. (a) SEM and (b) AFM images of surface morphology of laser irradiated nickel coated glass specimens in olive oil for different nickel coating thicknesses, and (c) EDS maps of a protrusion area and (d) 514 nm Raman spectrometry results of $I(D)/I(G)$ and $I(2D)/I(G)$ ratio changes.
transient film on the spherical protrusion was successive and smooth with a 8.3 nm height, which demonstrated a continuous graphene film growth on both protrusions and areas between them. There were more pores and protrusions generation because of the balling effect of the nano-nickel in water from the thermal effect of laser irradiation as shown in figure 7.

As the illustration in figure 8, during the laser irradiation, precipitation of carbon from nickel and the thermal energy would cause crystallization of carbon at 150–200 °C before balling effect of nano-nickel coating and would also cause rotational fault during crystallization [31]. So there was no defined positional relationship between atom planes. This lattice structure is called a turbostratic structure. This is different from the Bernal structure and leads to electronic decoupling of the layers due to the lack of a three-dimensional order [31]. The I(2D)/I(G) ratios and symmetrical 2D peaks shown in figures 2(a) and 6(d) demonstrated some feature of monolayer graphene as also shown in the TEM image. This indicated that turbostratic graphene may exist and is similar to multilayer single graphene. The turbostratic graphene was produced because the rotations between graphene layers overcame the restrictions of interfacial interactions. It enabled the ballistic transport of electrons through decoupled graphene layers [32].

3.4. Electrical conductivity characteristics

In a graphene structure, each carbon atom is bonded to three neighbour carbon atoms by the sp² bond. The low
energy electronic structure of the graphene has two $\pi$ orbitals with one lower energy valence bond and one higher energy conduction bond. The momentum of charge carrier and its pseudospin are reversed by backscattering. Therefore, for graphene, the backscattering of charge carriers is suppressed and electrical mobility is high leading to very high electrical conductivities [33].

Nickel sputter coatings initially appeared as separate particles on borosilicate glass after sputtering. During laser irradiation process, the nickel coating was annealed and more connection among particles took place to form a continuous film. However, there were many pores in the nickel coatings. The specimens with graphene layers have shown much lower sheet resistances about 1/5 than that of nickel coating for specimens annealed through laser irradiation in water using the same irradiation parameters as the ones in oil as shown in figure 9.

The minimum sheet resistance for a single-layer graphene (SLG) is reported to be about 15–30 $\Omega$ sq$^{-1}$ [34]. The minimum sheet resistance for commercial Ag nanowires printed on substrate with 70% transmittance is about 10–20 $\Omega$ sq$^{-1}$. The sheet resistance for commercial indium tin-oxide (ITO) is about 6–10 $\Omega$ sq$^{-1}$ at 200 nm thickness [35]. Our multiple layer graphene film coating by laser irradiation of a 7 nm nickel coating on glass in olive oil showed about 20 $\Omega$ sq$^{-1}$ sheet resistances, which has the potential to compete with commercial ITO.

3.5. Optical transparency

According to the optical absorbance spectrum shown in figure 10, the nickel coating thicknesses of 5 nm, 7 nm and 9 nm on glass specimen had transparency was between 50 and 60%. After laser irradiation, the optical transparency dropped to 40–50%, which is much lower than that of commercial ITO, thus more effort would be needed to increase the optical transparency for certain applications. The transmittance of glass sample with 9 nm nickel coating was improved back to about 70% after 50% material removal by 800 nm femtosecond laser processing with 1 kHz frequency, 1 mW laser power and 12 mm s$^{-1}$ scanning speed.

3.6. Processing temperature

During the laser irradiation, the temperature changes on the bottom of the stainless steel vessel were recorded with a thermocouple, which was located at the centre of the stainless steel vessel bottom. The thickness of the stainless steel vessel wall was 1 mm. Figure 11 shows the recorded temperatures, which shows lower than 300 $^\circ$C in all the experiments.

The temperatures on specimen surfaces were estimated using computational fluid dynamic modelling from the knowledge of the stainless steel vessel temperature at the bottom centre of the vessel. Isotropic thermal conductivity, specific heat and density of the olive oil (0.17 W m$^{-1}$ K$^{-1}$,
Figure 10. Optical absorption and transmission of the specimen before and after laser treatment. (a) Olive oil transmittance, (b) optical transmittance of the borosilicate glass with a 5 nm nickel coating, thickness, (c) optical transmittance of the borosilicate glass with a 7 nm nickel coating, and (d) optical transmittance of the borosilicate glass with a 9 nm nickel coating and femtosecond laser patterning.

Figure 11. Temperatures the stainless steel vessel for various laser irradiation time from 2 to 6 s with a nickel coating thickness of 9 nm.

0.97 J kg\(^{-1}\) K\(^{-1}\) and 0.91 g cm\(^{-3}\), borosilicate glass (1.14 W m\(^{-1}\) K\(^{-1}\), 830 J kg\(^{-1}\) K\(^{-1}\) and 2.23 g cm\(^{-3}\)) and 304 l stainless steel (16 W m\(^{-1}\) K\(^{-1}\), 480 J kg\(^{-1}\) K\(^{-1}\) and 8 g cm\(^{-3}\)) were used in the temperature simulation using Ansys.

The 3D continuity equation is given as [36]:

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0 \quad (3)
\]

where \(\rho\) is density of the material in a calculating cell, \(t\) is time, \(u\), \(v\) and \(w\) are the component velocities in the \(x\), \(y\) and \(z\)-directions of resultant velocity \(\mathbf{v}\).

Momentum conservation equation is expressed as [36]:

\[
\frac{\partial}{\partial t} (\rho \mathbf{v}) + \nabla \cdot (\rho \mathbf{v} \mathbf{v}) = -\nabla p + \nabla \cdot (\mathbf{\tau}) + \rho \mathbf{g} + S_{\text{mom}} \quad (4)
\]

where \(p\) is static pressure, \(\mathbf{\tau}\) is the stress tensor due to the viscous stress on element surface as a result of viscosity between molecules, \(\rho \mathbf{g}\) is gravitational body force while \(S_{\text{mom}}\) is additional momentum source.

Energy conservation equation is described as [37]:

\[
\frac{\partial (\rho H)}{\partial t} + \nabla \cdot (\rho \mathbf{v} H) = \nabla \cdot (k \nabla T) + S_h \quad (5)
\]

where \(H\) is enthalpy, \(k\) is thermal conductivity and \(S_h\) is user-defined Gaussian distributed heat source term.

Due to the temperature gradient caused by the laser heat input, density difference would occur which would contribute to the appearance of buoyancy force. The direction of convection flow induced by thermal expansion is vertically upward against the gravity direction. A buoyancy source term was therefore added to the momentum conservation equation to simulate the thermal expansion induced convection flow. Material density was considered as a constant in other conservation equations apart from the buoyancy term in the momentum equation to enable a faster convergence during the calculation. Boussinesq approximation model [38] can be expressed as:
\[ S_{\text{buoyancy}} = (\rho - \rho_m) g \approx -\rho_m \beta (T - T_m) g \]  
where \( \rho_m \) is density of the material at melting temperature \( T_m \); \( \beta \) is the thermal expansion coefficient. An approximation was made where \( \rho = \rho_m (1 - \beta \Delta T) \) to eliminate the temperature dependent material density property.

3.6.1 Boundary conditions. According to figure 9(d), 45\% of the laser energy was absorbed by the glass top surface coated with nickel. Therefore, boundary condition for glass top surface is given as:

\[
k \cdot \frac{\partial T}{\partial n} = 3P \eta \frac{\pi r_0^2}{r_n^2} \exp \left[-\frac{3r_n^2}{r_0^2}\right] \times 0.45 - h_{c,\text{oil}} (T - T_{oill}) - \sigma \varepsilon (T^4 - T_{env}^4) \tag{7}
\]

where \( P \) is laser power, \( \eta \) is laser efficiency, is the laser beam radius, \( r_n \) is radial distance from the laser beam centre, \( h_{c,\text{oil}} \) is heat convection coefficient, \( \sigma \) is Boltzmann constant, \( \varepsilon \) is Radiation emissivity, \( T_{oill} \) is oil temperature and \( T_{env} \) is environment temperature.

Within the projection of glass area, the rest of 55\% of laser energy was assumed to be 80\% [39] absorbed by the stainless steel vessel which is in contact with the bottom surface of glass. Since the glass bottom connected with stainless steel surface and there is limited contact between small amount of oil and steel on this surface, heat transfer through convection is not taken into consideration for this part of steel. For the area beyond the projection of glass but still within the effect of laser beam radius, boundary conditions including laser heat input, heat conduction, convection and radiation were applied. The combined boundary condition for top steel surface can be expressed as:

\[
k \cdot \frac{\partial T}{\partial n} = \begin{cases} 
\frac{3P \eta}{\pi r_0^2} \exp \left[-\frac{3r_n^2}{r_0^2}\right] \times 0.44 - \sigma \varepsilon (T^4 - T_{env}^4) & (|x| \leq 0.01, |y| \leq 0.0075) \\
\frac{3P \eta}{\pi r_0^2} \exp \left[-\frac{3r_n^2}{r_0^2}\right] - h_{c,\text{oil}} (T - T_{oill}) - \sigma \varepsilon (T^4 - T_{oill}^4) & (|x| \geq 0.01, |y| \geq 0.0075)
\end{cases} 
\tag{8}
\]

Boundary condition for the side and bottom of steel surface is described as:

\[
k \cdot \frac{\partial T}{\partial n} = -h_{c,\text{air}} (T - T_{env}) - \sigma \varepsilon (T^4 - T_{env}^4) \tag{9}
\]

Marangoni stress on tangential direction of oil top surface induced by the temperature dependent surface tension coefficient can be presented as:

\[
\tau_{\text{Marangoni}} = \frac{\partial \gamma}{\partial T} \frac{\partial T}{\partial n} \tag{10}
\]
where $\gamma$ is surface tension coefficient and $n$ is surface normal. $\gamma$ was adopted as $-8.6 \times 10^{-5}$ N m$^{-1}$ K$^{-1}$ according to the work from Dikko [40].

As shown in figure 12, the calculated temperatures on the specimen surface were higher than that on bottom of stainless bowl. The temperature was close to 400 °C after 2 s of laser irradiation. Then, the temperature difference between top surface of specimen and bottom of stainless steel vessel gradually reduced to a constant value of about 90 °C. When the specimen surface temperature was over the smoke point of olive oil about 200 °C, thermal dissolution of unsaturated fat acid in olive oil would occur and the flow of boiling oil increased heat convection, which would help balancing the heat as shown in figure 12.

4. Conclusion

This paper has demonstrated a single-step few-layers (6 layers) turbostratic graphene growth on nickel ultra-thin film coated glass by laser direct writing from an olive oil source. For a laser irradiation time of more than 2 s, the olive oil was dissociated into carbon atoms, water and gaseous species and the carbon was dissolved into nickel. Carbon atoms recrystallization on the nickel surface occurred through segregation from Ni grains and precipitation from the Ni grain boundaries. Different directional growth of grains and suboptimal crystallization of carbon atoms induced some defects. The defects could be improved through the increase of the thickness of nanonickel coating. When the specimen were laser irradiated for 4 s, double $\pi$ bond and a multi single layer graphene structure of large areas ($>5$ cm$^2$) was grown and demonstrated low sheet resistances (e.g. 20 $\Omega$ sq$^{-1}$) with a 40–50% transparency when a 5 nm nickel coating was deposited on the borosilicate glass, which has the potential to complete with some of commercial transparent conductive thin film coating products that have relatively thicker films (e.g. around 200 nm for ITO coatings).

ORCID ID

Yihe Huang https://orcid.org/0000-0002-5514-0460

References

[1] Lee C et al 2008 Measurement of the elastic properties and intrinsic strength of monolayer graphene Science 321 385–8
[2] Bunch J S et al 2007 Electromechanical resonators from graphene sheets Science 315 490–3
[3] Ferrari A C and Basko D M 2013 Raman spectroscopy as a versatile tool for studying the properties of graphene Nat. Nanotechnol. 8 235–46
[4] Cho J H et al 2017 Growth of monolayer graphene on nanoscale copper–nickel alloy thin films Carbon 115 441–8
[5] Chae S J et al 2009 Synthesis of large-area graphene layers on poly-nickel substrate by chemical vapor deposition: wrinkle formation Adv. Mater. 21 2328
[6] Chen S S et al 2011 Synthesis and characterization of large-area graphene and graphite films on commercial Cu–Ni alloy foils Nano Lett. 11 3519–25
[7] Li X S et al 2009 Large-area synthesis of high-quality and uniform graphene films on copper foils Science 324 1312–14
[8] Reina A et al 2009 Large area, few-layer graphene films on arbitrary substrates by chemical vapor deposition Nano Lett. 9 30–35
[9] Ye X et al 2015 Laser controllable growth of graphene via Ni–Cu alloy composition modulation Lasers Manuf. Mater. Process. 2 219–30
[10] Ismach A et al 2010 Direct chemical vapor deposition of graphene on dielectric surfaces Nano Lett. 10 1542–8
[11] Kumar A et al 2017 Low temperature synthesis and field emission characteristics of single to few layered graphene grown using PECVD Appl. Surf. Sci. 402 161–7
[12] Fan L S et al 2013 Laser direct writing graphene patterns on SiO$\text{2}$/Si substrates Laser-Based Micro- and Nanopackaging and Assembly VII vol 8608
[13] Seo D H et al 2017 Single-step ambient-air synthesis of graphene from renewable precursors as electrochemical genosensor Nat. Commun. 8 14217
[14] Maghsoumi A et al 2015 Overtone and combination features of G and D peaks in resonance Raman spectroscopy of the C$_6$H$_n$ polycyclic aromatic hydrocarbon J. Raman Spectrosc. 46 757–64
[15] Jiang J et al 2014 Graphene synthesis by laser-assisted chemical vapor deposition on Ni plate and the effect of process parameters on uniform graphene growth Thin Solid Films 556 206–10
[16] Cullinan M A and Gorman J J 2013 Transfer-free, wafer-scale fabrication of graphene-based nanoelectromechanical resonators. 2013 Microsystems for Measurement and Instrumentation (Mamma) pp 3–6
[17] Baraton L et al 2011 On the mechanisms of precipitation of graphene on nickel thin films Europhys. Lett. 96 46003
[18] Xiong W et al 2013 Single-step formation of graphene on dielectric surfaces Adv. Mater. 25 630–4
[19] Xiong W et al 2014 Direct writing of graphene patterns on insulating substrates under ambient conditions Sci. Rep. 4 4892
[20] Park J B et al 2011 Transparent interconnections formed by rapid single-step fabrication of graphene patterns Appl. Phys. Lett. 99 053103
[21] Park J B et al 2011 Fast growth of graphene patterns by laser direct writing Appl. Phys. Lett. 98 123109
[22] Lin Z et al 2013 Thinning of large-area graphene film from multilayer to bilayer with a low-power CO$_2$ laser Nanotechnology 24 275302
[23] Lin Z et al 2015 Precise control of the number of layers of graphene by picosecond laser thinning Sci. Rep. 5 11662
[24] Moreno M C M M et al 1999 Analytical evaluation of polyunsaturated fatty acids degradation during thermal oxidation of edible oils by Fourier transform infrared spectroscopy Talanta 50 269–75
[25] Ferrari A C and Robertson J 2000 Interpretation of Raman spectra of disordered and amorphous carbon Phys. Rev. B 61 14095–107
[26] Tuinstra F and Koenig J L 1970 Raman spectra of graphite Bull. Am. Phys. Soc. 15 296
[27] Budde H et al 2016 Raman radiation patterns of graphene ACS Nano 10 1736–63
[28] Tuinstra F and Koenig J L 1970 Raman spectrum of graphite J. Chem. Phys. 53 1126
[29] Cancado L G et al 2006 General equation for the determination of the crystallite size L-a of nanographite by Raman spectroscopy Appl. Phys. Lett. 88 163106
[30] Eapen J et al 2014 Early damage mechanisms in nuclear grade graphite under irradiation Mater. Res. Lett. 2 43–50
[31] Bianco A et al 2013 All in the graphene family—a recommended nomenclature for two-dimensional carbon materials Carbon 65 1–6
[32] Garlow J A et al 2016 Large-area growth of turbostratic graphene on Ni(111) via physical vapor deposition Sci. Rep. 6 19804
[33] Fuhrer M S, Lau C N and MacDonald A H 2010 Graphene: materially better carbon MRS Bull. 35 289–95
[34] Jo G et al 2012 The application of graphene as electrodes in electrical and optical devices Nanotechnology 23 112001
[35] Bonaccorso F et al 2010 Graphene photonics and optoelectronics Nat. Photon. 4 611–22
[36] Squire L C and Batchelor G K 1967 An introduction to fluid dynamics J. R. Aeronaut. Soc. 71 801
[37] Vollor V R and Prakash C 1987 A fixed grid numerical modeling methodology for convection diffusion mushy region phase-change problems Int. J. Heat Mass Transfer 30 1709–19
[38] Spiegel E A and Veronis G 1960 On the Boussinesq approximation for a compressible fluid Astrophys. J. 131 442–7
[39] Kawahito Y et al 2013 Laser absorption characteristics in high-power fibre laser welding of stainless steel Weld. Int. 27 129–35
[40] Dikko A 2015 Density and surface tension relationship of olive oil and carrot oil at different temperatures Worldw. J. Multidiscip. Res. Dev. 1 3