



Precursor and pressure dependent 3D graphene: A study on layer formation and type of carbon material



Mohamed Salleh Mohamed Saheed, Norani Muti Mohamed*, Balbir Singh Mahinder Singh, Mohamed Shuaib Mohamed Saheed

Centre of Innovative Nanostructures and Nanodevices (COINN), Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak Darul Ridzuan, Malaysia
Department of Fundamental and Applied Sciences, Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak Darul Ridzuan, Malaysia

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ABSTRACT

The variation and structural properties of interconnected and freestanding 3D graphene grown in atmospheric (AP) and low pressure (LP) conditions with varying precursor concentrations (7–14%) is reported for the first time. LP grown 3D graphene at low precursor concentration (7 vol%) shows turbostratic signature whereas AP grown 3D graphene indicates a double layer signature in the same condition. The 2D band peaks splitting evaluation through Lorentzian fitting shows the distinct variations in the number of layers grown for each precursor concentrations. Extensive Raman analysis of the grown samples revealed the existence of disordered plane stacking of graphene layers, which decreases as the precursor concentration increases. In contrast, AP grown 3D graphene shows no disordered layers for any precursor concentration. The increase in a number of layers (2 to 5) of 3D graphene grown in both AP and LP conditions co-influenced by precursor concentrations is validated by TEM. Interestingly, it was discovered that both 3D graphene grown in different conditions exhibit sp^2 ring carbon material. Thus, control over growth kinetics by varying the precursor concentration in different chamber pressures allows for total manipulation of the number of layers of 3D graphene deposited on the template nickel catalyst.

1. Introduction

Since the successful isolation of monolayer graphene, extensive research has been carried out in order to gain an insight into the unusual intrinsic electrical properties of a monolayer, few layers of graphene and graphite [1]. This includes the extraordinary electron ballistic transport at room temperature, anomalous integer quantum Hall Effect (QHE), diamagnetism and many body effects of the synthesized graphene [1–4]. At neutral state, graphene electrons and holes electronic potential intersect exactly at the Fermi level with low energy excitation condition [1,2,5]. This uncommon behaviour of graphene massless electrons and holes complies with massless fermions quantum electrodynamics (QED), but at smaller speed v_F . These electrons and holes give rise to the most useful properties of high electrical mobility of $200,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature [6]. This high mobility is achieved via ballistic transport by massless electrons which are able to travel several hundred nanometers without scattering [7,8]. It is possible that the charge carriers can be transmitted to the forbidden region as described in Klein tunnelling [9]. Unlike the conventional semiconductor, the QHE of confined electrons in two-dimensional graphene

comes from its unique electronic structure. In graphene, electron-hole degeneracy and carrier mass vanished near charge neutrality point, giving rise to the peculiar integer QHE of electrons and holes [3,10]. These unusual phenomena of QHE and Berry phase where the magneto-oscillation attributed by linear dispersion and mass vanishing near Dirac point in graphene were observed by adjusting the potentials through electric field effect [4]. Besides the electronic properties, phonon scattering and vibrational modes also deemed to be the subject of interest in understanding the dynamics of quasiparticles and charge carriers. The optical phonons and electrons scattered by external optical excitation energy, reached another state called electron-phonon coupling (EPC). The Kohn anomalies observed in graphene was believed to be due to the effect of EPC on phonon frequency [11,12]. All these novel and remarkable properties set graphene to be the most capable material for future high performance devices.

For graphene to improve the performance and efficiency of current materials and devices, in-depth understanding of its basic properties is very much needed. Thus synthesis of good quality monolayer to few layers of graphene sheet become critical where methods such as sol-gel, template-assisted and chemical vapour deposition (CVD) are

* Corresponding author at: Centre of Innovative Nanostructures and Nanodevices (COINN), Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak Darul Ridzuan, Malaysia.
E-mail address: noranimuti_mohamed@utp.edu.my (N.M. Mohamed).

extensively exploited [13–21]. For CVD process, conditions such as gas volume, pressure, temperature, and time duration should be adhered stringently in order to produce good quality graphene. By good quality means that graphene should be devoid of defects, impurities, grain boundaries, and structural disorders as they can have an adverse effect on its electronic and optical properties. Recently, another type of graphene derivative, the porous three-dimensional graphene (3DG) structure was produced and found to exhibit similar electronic properties of monolayer and few layers graphene [22]. It is reported that unlike the monolayer graphene, the 3DG show better robustness due to the stacked multilayers that can withstand the mechanical stress and has higher specific surface area contributed by its nanoporous structure [17,22–24].

Most of the past studies focused specifically on the 2D graphene structure using low-pressure CVD method [16]. The intention is to avoid the unwanted doping and have better control of the number of graphene layers. Although a comparative study on the effect of growth pressure such as low and atmosphere pressure are many for 2D graphene, it is still lacking for 3DG. This study may shed light on the correlation between the growth pressure and precursor concentrations as well as the resulted 3DG quality.

Detailed non-destructive and fast characterizations are needed to analyse the quality of as-grown graphene structures. Raman spectroscopy has proven to be the most sought characterization tool given its non-destructive method and the deep analysis ability to reveal the graphene structure, doping level, and defects [25,26]. It is also known that the Raman peaks are closely associated with the phonons at the longitudinal optical (iLO) and transverse optical (iT0) energies and found to be highest at the Brillouin and *K* points [27,28]. In Raman, the two main bands, *G* band is dependent on the iLO phonon while the second order band known as *2D* or *D** is the second order resonance which arises from the phonon scattering due to the laser excitation energy [29,30].

The *G* band peak of Raman spectrum is contributed by the in-plane sp^2 carbon atom vibrations in the graphene layers [31,32]. When the graphene layer increases, the intensity of *G* peak also increases with responding to Raman laser excitation energies within the visible light. Samples containing sp^2 carbon will give rise to *G* peak at 1582 cm^{-1} whereas samples with sp^3 will have the *G* peak appeared at 1333 cm^{-1} [30]. As the number of graphene layer stacking increases, the intensity of *G* band becomes more prominent, appearing as a higher peak.

The other two bands are *D* band, which requires defects for its activation and *2D* band, the second order of Brillouin zone with boundary phonon of $q \neq 0$. As for the crystalline graphene, the defect induced *D* bands are absent. The integrated ratio of I_D/I_G is used to determine the crystallite size, L_a and quantifying the defects in graphene [33]. Notably, the graphite *G* band peak is quite similar to single layer graphene but with $3\text{--}5\text{ cm}^{-1}$ *G* band position shift compared to the one of the bulk graphite. The *G* band intensity is found to be uniformly distributed at the graphene surface, but the *D* band appeared at the imperfect crystalline structure commonly at the edges and defects at the lattices of graphene [34–36]. On the other hand, the *2D* band dispersion of graphene multilayer which contributes to the band splitting was different compared to the bulk graphite [37]. Even without any defects, the Raman exhibits the *2D* band in most of graphene and graphite materials. The *2D* band shape obtained from the Raman measurement can be used to determine the number of layers as the second order band is sensitive to the graphene stacking order along *c* axis [30,31]. The graphene layer stacking evolution from single to five layers was able to be differentiated via the *2D* band splitting while the higher layer stacking is attributed to the graphite [30,36]. In some cases, the Raman peak of *2D* may only have a single intense peak for multilayer graphene even with different thickness but with shifted position of full width at half-maximum (FWHM) function. The dispersive behaviour of *D* and *2D* band was reported by Vidano and Fischbach [38]. They have found that the *D* bands are linearly dispersive against the incident laser excitation

within the visible range with the slope of $50\text{ cm}^{-1}/\text{eV}$. The same dispersive behaviour was observed for the *2D* band but with twice of the *D* band slope, $100\text{ cm}^{-1}/\text{eV}$. All these are the Raman analysis for 2D monolayer and few layers of graphene. However, detailed Raman analysis of 3DG is still lacking and is very much needed to develop interesting applications where more advantages such as high surface area, flexibility, and elasticity can be added to the current demand for ultra-light, highly conductive graphene-based materials with exceptional mechanical strength. Thus, the work here involved the investigation of the structure and intrinsic properties of 3DG grown via atmospheric and low pressure CVD by using Raman spectroscopy.

2. Material and experimental work

The nickel foam purchased from Shanghai Winfay Industry Ltd. Acetone and 2-propanol were purchased from Merck KGaA for nickel cleaning purpose. Poly(methyl methacrylate) and surfactant anisole were purchased from Sigma Aldrich for the support layer of 3D graphene. Nickel etching agent, iron (III) chloride (FeCl_3) were purchased from Merck KGaA. All the chemicals were used without further purification. Methane gas (99.9%), argon gas (99.9%) and hydrogen gas (99.9%) as carbon source, carrier gas and activation gas were used, respectively.

The growth of 3DG was carried out using CVD system that was maintained at atmospheric (760 Torr) and low (3 Torr) pressure. For the template, nickel foam with the thickness of 0.5 mm was chosen in order to get graphene to emulate the microporous structure of nickel foam to form 3D structure. As for atmospheric pressure CVD (APCVD) graphene synthesis, the chamber pressure (quartz tube) was maintained at 1 atm. The cleaned and dried microporous nickel template ($4 \times 4\text{ cm}^2$) was loaded into the chamber where constant flow of argon (Ar) and hydrogen (H_2) gas (ratio of 5:1) were introduced with simultaneous temperature increase. The nickel foam template was annealed up to $1000\text{ }^\circ\text{C}$ at the rate of $25\text{ }^\circ\text{C}/\text{min}$ for 40 min and temperature stabilized for further 10 min before carbon source of methane (CH_4) gas was flowed in. At $1000\text{ }^\circ\text{C}$, CH_4 was flowed in which decomposes and form $\pi\text{-}\pi^*$ carbon interactions on the surface of nickel which acts as the catalyst. After the graphene synthesis, the CH_4 were immediately cut-off and the grown graphene was cooled down rapidly ($\sim 100\text{ }^\circ\text{C}/\text{min}$) to room temperature in the presence of Ar and H_2 gas for the next process to obtain free-standing 3-dimensional graphene. The only difference for the low-pressure CVD (LPCVD) graphene synthesis is the chamber pressure evacuated to 3.3 mTorr by using vacuum pump and maintained at 3 Torr with constant flow of mixture Ar and H_2 gas during the synthesis of graphene. Samples were prepared with different CH_4 concentrations, ranging from 7 to 14 vol% for both atmospheric and low-pressure condition with 20 min deposition period. Poly(methyl methacrylate), hereafter abbreviated as PMMA is used as the support layer for graphene during the nickel etching process. The PMMA was spin-coated at 1000 rpm for 30 s on the nickel/graphene structure and baked at $80\text{ }^\circ\text{C}$ for 1 h and cooled down at room temperature. The sacrificial nickel template was then etched by immersing nickel/3D graphene/PMMA into etching solution, 30% iron (III) chloride in distilled water. For proper nickel etching, the template edges were trimmed to allow etching solution to penetrate. Graphene/PMMA structure was left to dry at room temperature before the final removal of the PMMA coating using the hot acetone at $55\text{ }^\circ\text{C}$. The hot acetone 3D graphene purification was repeated thrice for complete PMMA removal. The 3D graphene later scooped carefully on micro slide and left to dry in room temperature to obtain free standing 3D graphene.

Field emission scanning electron microscope (FESEM) Zeiss Supra55 VP was used to determine the material composition and the morphology of the free-standing graphene grown in atmospheric and low-pressure condition. X-ray photoelectron spectroscopy (XPS) were characterized using K-Alpha, Thermo Scientific. All spectra were

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