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SYNTHESIS OF GRAPHENE FROM ALTERNATIVE PRECURSORS AND THE USE OF QUARTZ PLATE FOR CARBON SOURCE MANIPULATION

MUHAMMAD IZHAR BIN KAIRI

UNIVERSITI SAINS MALAYSIA

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by

MUHAMMAD IZHAR BIN KAIRI

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TABLE OF CONTENTS

	Page
ACKNOWLEDGEMENTS	ii
TABLE OF CONTENTS	iii
LIST OF TABLES	vi
LIST OF FIGURES	viii
LIST OF ABBREVIATIONS	xiv
LIST OF SYMBOLS	xvi
ABSTRAK	xvii
ABSTRACT	xix
CHAPTER ONE: INTRODUCTION	1
1.1 Two-dimensional materials	1
1.2 Graphene	2
1.3 Chemical vapor deposition	4
1.4 Problem statement	5
1.5 Objectives	9
1.6 Scope of study	10
1.7 Thesis organization	11
CHAPTER TWO: LITERATURE REVIEW	13
2.1 Growth of graphene by CVD	13
2.1.1 Gaseous carbon precursors	15
2.1.2 Liquid carbon precursors	21
2.1.3 Solid carbon precursors	29
2.2 Mechanism involved in the use of carbon precursors for graphene synthesis by CVD	40
2.2.1 Aliphatic compound precursors	40
2.2.2 Aromatic compound precursors	43
2.2.3 Unzipping nanotubes	49
2.2.4 Other carbon precursors	50
2.3 Graphene vs. carbon nanotubes by CVD	55

2.4 Parameters for graphene growth via CVD 59
2.4.1 Annealing duration 64
2.4.2 Catalyst for simultaneous graphene and syngas production 65
2.5 Summary 67

CHAPTER THREE: MATERIALS AND METHODS 70
3.1 Materials and chemicals 70
3.2 Experimental equipment and test rig 71
3.2.1 Gas mixing section 78
3.2.2 Reaction section 79
3.3 Experimental steps 79
3.3.1 Thermal pyrolysis of rice straw 80
3.3.2 Synthesis of graphene from rice straw bio-oil 82
3.3.3 Synthesis of graphene via carbon trapping 83
3.3.4 Co-synthesis of graphene and syngas 84
3.4 Graphene transfer 84
3.5 Characterization techniques 85
3.5.1 Raman spectroscopy and mapping 90
3.5.2 High resolution transmission electron microscopy (HRTEM) 91
3.5.3 X-ray photoelectron spectroscopy (XPS) 92
3.5.4 Optical imaging 92
3.5.5 Gas chromatography (GC) 92
3.5.6 Proximate analysis 93
3.5.7 Elemental analysis 94
3.5.8 Bomb calorimeter 94

CHAPTER FOUR: RESULTS AND DISCUSSIONS 95
4.1 Preliminary studies 95
4.1.1 APCVD gas flow rate condition 95
4.1.2 Annealing duration 99
4.1.3 Preliminary studies of graphene transfer 100
4.1.4 Preliminary studies of fast cooling 103

4.2 Graphene growth from rice straw bio-oil 105
4.2.1 Thermal pyrolysis of rice straws 106
4.2.1(a) Properties of rice straw 106
4.2.1(b) Effect of reaction temperature 107
4.2.2 Graphene APCVD from rice straw bio-oil 109
4.2.2(a) APCVD mode 109
4.2.2(b) Effect of bio-oil volume 113
4.2.2(c) Discussion 116
4.2.3 Carbon trapping assisted graphene growth 119
4.2.3(a) Carbon trapping configuration 119
4.2.3(b) Carbon trapping material 122
4.2.3(c) Effect of rice straw bio-char weight 126
4.2.3(d) Effect of trapping techniques on metal catalyst 131
4.2.3(e) Discussion 133
4.3 Graphene and syngas production from greenhouse gases 135
4.3.1 Effect of reaction temperature 135
4.3.2 Discussion 141

CHAPTER FIVE: CONCLUSION AND RECOMMENDATION 144
5.1 Conclusion 144
5.2 Recommendations 146

REFERENCES 149
APPENDIX
LIST OF PUBLICATIONS

LIST OF TABLES

		Page
Table 1.1	List of large area graphene prices from various suppliers.	7
Table 2.1	Examples of graphene growth conditions using different gas as carbon precursors and their characteristics.	19
Table 2.2	Examples of graphene growth conditions using different liquid as carbon precursors and their characteristics.	26
Table 2.3	Examples of graphene growth conditions using different solid as carbon precursors and their characteristics.	36
Table 2.4	Estimated dehydrogenation time, tD for CH4, C6H6 and C18H14 at different temperatures (Choi et al. 2013).	44
Table 2.5	Observable mechanisms from various carbon precursors for graphene growth by CVD.	53
Table 2.6	Carbon nanotube grown by CVD using various carbon precursors.	57
Table 2.7	Previous reports on carbon precursors starting weight and CVD setup.	64
Table 2.8	Annealing conditions prior to graphene growth	65

Table 2.9	Recent progress on carbon dioxide reforming of methane using various kinds of catalyst.	67
Table 3.1	List of the chemicals and their properties used in this study.	70
Table 3.2	List of the gases and their characteristics used in this study.	71
Table 3.3	Details of major components in the experimental rig along with their function.	76
Table 3.4	Details of major characterization equipment that were used in this study along with their function.	86
Table 3.5	Graphene layer thickness based on the peak ratios from Raman spectroscopy (Hao et al. 2010; Matz et al. 2015; Park et al. 2009; Wu et al. 2012).	91
Table 4.1	Velocity of gas flowrate at different total gas flowrate.	98
Table 4.2	Mass of chemical compound required to get the same molarity and desired volume.	103
Table 4.3	Properties of rice straw.	107
Table 4.4	Properties of rice straw bio-oil and bio-char.	108

LIST OF FIGURES

		Page
Figure 2.1	Chemical vapor deposition setups: a) Two-furnace; b) Bubbling; c) Magnetic boat (Jacobberger et al. 2015; Kalita et al. 2011; Rosmi et al. 2016; Sharma et al. 2013).	14
Figure 2.2	Schematics of bilayer graphene growth using spin-coated polymer films via carbon diffusion through Ni layer (Peng et al. 2011).	32
Figure 2.3	Schematic illustration of graphene synthesis with PCBM as carbon precursor (Seo et al. 2013).	34
Figure 2.4	Diagram of methane dehydrogenation from the side view. Orange sphere = Transition metal atom; Green sphere = Hydrogen atom; Black sphere = Carbon atom; TS = Transition state (Gajewski & Pao 2011).	41
Figure 2.5	Schematic of graphene flake CVD growth by using hexachlorobenzene (Gan et al. 2012).	45
Figure 2.6	Schematic diagram of growth mechanism on a Cu surface from benzene by a) LPCVD; b) Normal APCVD; c) Oxygen-free APCVD (Jang et al. 2015).	47
Figure 2.7	Possible growth mechanism of 3D graphene grown by CVD from asphalt (Liu et al. 2014).	48
Figure 2.8	Scheme of rebar graphene synthesis on Cu foil from CNTs (Yan et al. 2014).	49

Figure 2.9	Suggested graphene CVD scheme with camphor as carbon precursor (Kumar et al. 2016b).	51
Figure 2.10	Graphical illustration of observable mechanisms from various carbon precursors for graphene growth by CVD (Jang et al. 2015; Yan et al. 2014).	54
Figure 2.11	Schematic diagram of graphene CVD gas flow from the (a) side view; (b) front view.	63
Figure 3.1	Photograph of the horizontal quartz tube reactor at School of Aerospace Engineering's MTDC lab.	72
Figure 3.2	Schematic diagram of the horizontal quartz tube reactor at School of Aerospace Engineering's MTDC lab.	73
Figure 3.3	Photograph of the vertical quartz tube reactor at School of Chemical Engineering's Petroleum lab.	74
Figure 3.4	Schematic diagram of the vertical quartz tube reactor at School of Chemical Engineering's Petroleum lab.	75
Figure 3.5	Flowchart of the research overview of this thesis.	80
Figure 3.6	Schematic diagram of thermal pyrolysis by using a movable furnace.	82
Figure 3.7	Chemical vapor deposition setup using a heating ring as the second energy source. Red square = metal catalyst, blue circle = rice straw bio-oil.	83
Figure 3.8	Schematic diagram of the carbon trapping setup used in conjunction with rice straw bio-char for graphene APCVD.	83
Figure 4.1	Overall process of APCVD in this thesis.	97

Figure 4.2	Raman spectroscopy of graphitic film produced at different rate of N_2 flow.	98
Figure 4.3	Optical micrograph of Cu catalyst surface with annealing time of (a, b) 15 minutes; (c, d) 0 minutes.	99
Figure 4.4	Picture of as-produced graphene on Cu substrate after being etched for (a) 5 min; (b) 10 min; (c) 15 min and (d) 20 min.	102
Figure 4.5	Fast cooling technique: (a) Mechanical displacement; (b) Movable furnace.	104
Figure 4.6	Fast cooling technique: Movable furnace vs. mechanical displacement.	105
Figure 4.7	Effect of pyrolysis temperature on product yield of rice straw at 200 sccm N_2 gas flow rate and 3 g of rice straw.	108
Figure 4.8	Schematic diagram of graphene growth via APCVD by (a) Heating ring setup; (b) Rapid heating of carbon precursor and metal catalyst; (c) Rapid heating of carbon precursor only. Red square is metal catalyst Cu; blue circle is rice straw bio-oil.	110
Figure 4.9	Typical Raman spectroscopy spectra of the deposit from rice straw bio-oil by using (a) heating ring setup; (b) rapid heating of carbon precursor and metal catalyst; (c) rapid heating of carbon precursor only.	112
Figure 4.10	Picture of the APCVD setup in the MTDC-Aero lab assisted with customized heating ring.	112

Figure 4.11	(a) Raman spectroscopy of graphitic material grown with different volume of bio-oil and Raman mapping of graphene grown from 0.5 mL of rice straw bio-oil, (b) I_{2D}/I_G and (c) I_D/I_G.	114
Figure 4.12	(a) HRTEM and (b) SAED of graphene grown from 0.5 mL of rice straw bio-oil. (c) XPS of the same graphene film and (d) deconvolution of C 1s peak.	115
Figure 4.13	Process flow diagram of graphene growth from rice straw via thermal pyrolysis and APCVD.	117
Figure 4.14	Image of (a) Cu foil enclosure (Li et al. 2011) and schematic diagram of (b) bimetallic Ni-Cu catalyst (Seah et al. 2016).	120
Figure 4.15	Picture of quartz plate and Cu plate used in this study.	121
Figure 4.16	Configuration of Cu catalyst with quartz plates used in this work.	122
Figure 4.17	(a) Raman spectroscopy of graphitic film produced from carbon trapping setup using Cu and quartz plate and (b) Images of Cu and quartz plate after carbon trapping assisted APCVD.	123
Figure 4.18	Optical micrograph of Cu plate: (a, b) after CVD and (c, d) after oxidization and (e, f) after reduction process. Images of (b), (d) and (f) are at higher magnification than (a), (c) and (e).	124
Figure 4.19	(a) Raman spectroscopy of graphene grown from bio-char of different weight. Raman mapping of graphene	127

growth from 2.5 mg of rice straw bio-char: (b, d) I_{2D}/I_G; (c, e) I_D/I_G.

Figure 4.20 HRTEM of graphene synthesized from (a) 2.5 mg (Enlarged images are cropped from the boxed region in respective graphene sheets) and (b) 10.0 mg of rice straw bio-char. SAED of graphene synthesized from (c) 2.5 mg and (d) 10.0 mg of rice straw bio-char. Optical micrograph of graphene synthesized from (e) 2.5 mg and (f) 10.0 mg of rice straw bio-char transferred onto silicon wafer with oxide layer. 129

Figure 4.21 (a) XPS of few-layer graphene obtained from graphene growth assisted by carbon trapping setup and the (b) C 1s spectra band highlighted. 130

Figure 4.22 Optical micrographs of (a, b) carbon trapping in this study, (d, e) bimetallic Ni-Cu catalyst and (g, h) Cu foil enclosure. Pictures of (c) carbon trapping in this study, (f) bimetallic Ni-Cu catalyst and (i) Cu foil enclosure. 132

Figure 4.23 Schematic diagram of graphene formation using (a) carbon trapping setup and (b) typical single foil setup. 134

Figure 4.24 Raman spectroscopy (a) and Raman mapping (b) I_{2D}/I_G and (c) I_D/I_G of the grown multilayer graphene. 137

Figure 4.25 Typical HRTEM images of graphene edge synthesized from CO_2 and CH_4 (a) and from CH_4 (c), respectively. Electron diffraction of graphene synthesized from carbon dioxide and CH_4 (b). 139

Figure 4.26 Gas conversion of CH_4 to H_2 and carbon dioxide to CO over CVD at 900 ^oC for 60 min and the ratio of H_2:CO produced. 141

Figure 4.27 Schematic representation of the graphene and syngas co-synthesis CVD mechanism. 142

LIST OF ABBREVIATIONS

2D	Two-dimensional
3D	Three-dimensional
ABS	Acrylonitrile-butadiene-styrene
APCVD	Atmospheric pressure chemical vapor deposition
Ar	Argon
BNNTs	Boron nitride nanotubes
C	Carbon
CH_4	Methane
C_2H_2	Acetylene
C_2H_4	Ethylene
C_2H_5OH	Ethanol
C_3H_6	Propene
C_3H_7OH	Isopropyl alcohol
$C_3N_6H_6$	Melamine
C_5H_5N	Pyridine
C_6H_{14}	Hexane
$C_{12}H_{22}O_{11}$	Sucrose
$C_{13}H_{10}$	Fluorene
CNTs	Carbon nanotubes
CRM	Carbon dioxide reforming of methane
Cu	Copper
CVD	Chemical vapor deposition
DFT	Density functional theory

EA	Elemental analysis
FWHM	Full width half maximum
GC	Gas chromatography
GNCs	Graphene nanoclusters
H_2	Hydrogen
HBC	Hexabenzocoronene
HCB	Hexachlorobenzene
HHV	High heating values
HIPS	High-impact polystyrene
HOPG	Highly oriented pyrolytic graphite
HRTEM	High resolution transmission electron microscope
LPCVD	Low pressure chemical vapor deposition
N	Nitrogen
Ni	Nickel
PCBM	Phenyl-C_{61}-butyric acid methyl ester
PhD	Doctor of Philosophy
PMMA	Poly(methyl methacrylate)
RFCVD	Radio frequency chemical vapor deposition
RM	Ringgit Malaysia
SAED	Selected area electron diffraction
sccm	Standard cubic centimeter per minute
TST	Transition state theory
UHV-CVD	Ultra-high vacuum chemical vapor deposition
VOC	Volatile organic compounds
XPS	X-ray photoelectron spectroscopy

LIST OF SYMBOLS

€	Euro
%	Percent
π	Pi
$	US dollar
I_{2D}/I_G	Ratio of 2D-peak over G-peak in a Raman spectra
I_D/I_G	Ratio of D-peak over G-peak in a Raman spectra
RM	Ringgit Malaysia
t_D	Dehydrogenation time
wt. %	Weight percent

SINTESIS GRAFENA DARIPADA PENDAHULU ALTERNATIF DAN PENGGUNAAN KEPINGAN KUARZA UNTUK MEMANIPULASI SUMBER KARBON

ABSTRAK

Grafena ialah satu lapisan atom karbon yang tersusun dalam struktur sp^2-hibridisasi dengan sifat yang jauh lebih hebat berbanding dengan bahan lain. Penyelidikan dan pembangunan di dalam bidang sintesis grafena telah berkembang dengan pesat beberapa tahun kebelakangan ini terutamanya dengan menggunakan kaedah pemendapan wap kimia (CVD). Walau bagaimanapun, untuk membolehkan grafena bergerak dari skala makmal ke domain komersial; kos untuk sintesis graphene perlu terus dikurangkan dari masa ke masa. Di dalam tesis ini, masalah ini disentuh daripada beberapa sudut. Salah-satu daripada kajian yang dilakukan dalam tesis ini menunjukkan yang jerami padi dan biogas sintetik; sisa buangan daripada industri padi dan kelapa sawit boleh digunakan sebagai sumber karbon murah untuk penghasilan grafena. Dalam kes jerami padi, pertama sekali ia akan melalui proses pirolisis untuk memperoleh minyak-bio dan arang-bio. Hanya 0.50 mL minyak-bio kemudiannya digunakan sebagai sumber karbon untuk mensintesis 1 cm x 1 cm grafena filem dengan kualiti yang bagus (I_D/I_G = ~0.55) melalui kaedah dua pemanas pemendapan wap kimia pada tekanan atmosfera (APCVD). Dengan melakukan pirolisis, kuantiti sumber karbon yang keluar sebagai efluen dapat dikurangkan memandangkan kebanyakannya telah ditukar kepada produk sampingan sebelum pertumbuhan grafena. Arang-bio juga digunakan sebagai sumber karbon untuk mensintesis grafena tetapi dengan proses APCVD yang dibantu dengan kaedah memerangkap karbon yang baru dan mudah

melalui plat kuarza yang ditempah khas. Di sini, 2.5 mg arang-bio telah dikelilingi dengan plat kuarza dan kepingan kuprum dalam susunan yang khusus untuk bertindak sebagai perisai karbon bagi memerangkap spesies aktif karbon pada suhu tinggi dalam ruangan yang sempit. Dengan melaksanakan perangkap karbon, kuantiti sumber karbon yang diperlukan untuk sintesis grafena pada kadar saiz yang sama adalah lebih rendah; keupayaan pertumbuhan grafena meningkat sekitar 400 %. Akhir sekali, efluen kepada proses pertumbuhan grafena telah ditukarkan menjadi syngas apabila biogas sintetik digunakan sebagai sumber karbon bersama dengan nikel sebagai pemangkin pada 900 ^{o}C dalam proses yang sama; menukar sisa kepada harta.

SYNTHESIS OF GRAPHENE FROM ALTERNATIVE PRECURSORS AND THE USE OF QUARTZ PLATE FOR CARBON SOURCE MANIPULATION

ABSTRACT

Graphene is a single layer of carbon atoms arranged in an sp^2-hybridized structure with properties far superior compared to other materials. Research and development in graphene synthesis have been rapidly growing the past few years especially using chemical vapor deposition (CVD). However, in order to move graphene from laboratory scale to commercial domain; graphene synthesis cost need to be continually reduced. In this thesis, this problem was approached from a few angles. One of the study in this thesis demonstrate that rice straw and synthetic biogas, typical wastes from the rice and palm oil production industries can be used as inexpensive carbon source for the production of graphene. In the case of rice straw, the lignocellulosic biomass was first put through thermal pyrolysis in order to obtain the bio-oil and bio-char. Only 0.50 mL of bio-oil was then used as carbon source to sufficiently synthesized 1 cm x 1 cm of large area graphene with good quality (I_D/I_G = ~0.55) via a two-heating source setup ambient pressure CVD (APCVD) . By doing this, the amount of carbon source going out as effluent is reduced as it has been recovered as side-products prior to graphene growth. The bio-char was also used as carbon source for graphene growth but with a novel and facile carbon trapping assisted APCVD involving customized quartz plate. Here, 2.5 mg of bio-char was encapsulated with quartz plate and Cu foil in a specific arrangement which acted as carbon shields and trapped the carbon active species at high temperature within the confined space of the setup. By implementing carbon trapping, lesser amount of carbon source was needed for graphene growth of the same size; graphene growth efficiency increased

around 400 %. Lastly, the effluent of the graphene growth process was converted into syngas when synthetic biogas was used as the carbon precursor with Ni as the catalyst at 900 ^{o}C in a one-step process; turning waste into wealth.

CHAPTER ONE

INTRODUCTION

This chapter provides an overall introduction to the PhD research project. A brief introduction on two-dimensional materials and graphene as the main theme of this thesis are outlined at the beginning of the chapter followed by a section on chemical vapor deposition (CVD) as the tool to synthesis graphene. At the end of this chapter, the problem statement, objectives, scope of work and thesis organization are laid out.

1.1 Two-dimensional materials

The introduction of new materials has enabled the growth of new technologies that have beneficial impact on society. Currently, the world is in the precipice of a new age of 2-dimensional (2D) materials. Boron nitride (BN) (Örnek et al. 2018; Wang et al. 2018b; Zhuang et al. 2017), bismuth telluride (Bi_2Te_3) (Termsaithong & Rodchanarowan 2017; Wada et al. 2017), bismuth(III) selenide (Bi_2Se_3) (Desai et al. 2018), molybdenum disulfide (MoS_2) (Jiang et al. 2017; Solanki et al. 2017), molybdenum diselenide ($MoSe_2$) (Sathe et al. 2013; Wang et al. 2017), molybdenum ditelluride ($MoTe_2$) (Park et al. 2015), tungsten disulfide (WS_2) (Choi et al. 2017), tungsten diselenide (WSe_2) (Chen et al. 2015; Yun et al. 2016), silicone (De Padova et al. 2017; Vogt et al. 2014), phosphorene (Khandelwal et al. 2017; Woomer et al. 2015), bismuthene (Aktürk et al. 2016), graphyne (Dearden & Crean 2014; Kehoe et al. 2000), graphane (Arguilla et al. 2014) and graphene (Ali et al. 2016; Murdock et al. 2017) are among the 2D materials that have been investigated. Within that group, graphene is the most-researched material since its discovery in 2004 (Geim & Novoselov 2007; Novoselov et al. 2004). It is investigated for use in the next

generation devices due to its outstanding combination of properties not observed in any other type of materials. Its magnificent properties are attributed to the strong bonding between the hexagonal arrangement of carbon atoms that make up graphene.

Graphene can be produced in several forms which include flakes (Chae et al. 2017; Ghaemi et al. 2017; Nizam et al. 2017), ribbons (An et al. 2012; Liu et al. 2016), and large-area sheets (Bhaviripudi et al. 2010; Yan et al. 2012). They differ in lateral dimensions; flakes with limited lateral dimensions (from several nanometers to micrometers), large-area sheets possess macroscopic and extended lateral dimensions, while ribbons have one lateral dimension that is at least one order of magnitude larger than the other (Bianco et al. 2013). These differences allow graphene to be used in various types of application; certain physical forms are more compatible for particular applications. For example, large-area graphene sheets are more suited for wafer-scale thin film-like application such as transparent conductive electrode (Li et al. 2009c) while graphene flakes (GFs) are investigated for conductive ink applications (Arapov et al. 2016; Yang & Wang 2016).

1.2 Graphene

The unique properties of graphene have triggered a sudden drive of graphene researches all over the world. Graphene is expected to be the next generation two-dimensional materials having the potential to enhance current technology and create a new one in the near future. Graphene is a thermodynamically stable species of pure carbon atoms bonded in a single sheet of hexagonal sp^2 structure (Gilje et al. 2007). It represents the base structure of carbon nanotubes (CNTs), graphite and fullerenes. Electron configuration of the carbon atoms is the key factor for the incredible properties of graphene such as its high mechanical strength (Ahmad et al. 2015; Wang

et al. 2014), high surface area (Gadipelli & Guo 2015; Huang et al. 2012), extraordinary thermal conductivity (Wang & Tsai 2016; Wang et al. 2016), ultra-high elasticity (Geim 2009) and high optically transparency (Weber et al. 2014; Zhu et al. 2014). Due to its outstanding properties, graphene has immense potential in many applications. Even if properties of graphene depend on its structure and form (Lee et al. 2017; Pang et al. 2016) and the intended applications depend on graphene characteristics, its potential to improve performances in numerous materials and processes is widely recognized. Among the numerous applications targeted, graphene could be used as transparent conducting electrodes (Casaluci et al. 2016), fillers in reinforced polymer nanocomposites (Kumar et al. 2017b; Mittal et al. 2015; Young et al. 2012), super-capacitors (Ke & Wang 2016; Kumar et al. 2017a, 2017d), lithium ion batteries (Kheirabadi & Shafiekhani 2012; Kucinskis et al. 2013; Zhao et al. 2016a), fuel cells (Devrim & Albostan 2016; Najafabadi et al. 2016), solar cells (Kuhn & Gorji 2016), photo-catalysis (Putri et al. 2015), biosensors (Park et al. 2016), chemical sensors (Zhao et al. 2016b), purification of water (Aghigh et al. 2015) and optoelectronics (Jin et al. 2015; Polat et al. 2016). To face the demand for both fundamental studies and developments for its practical utilization, development of large-scale synthesis methods has never been more vital (Bae et al. 2010; Levchenko et al. 2016).

Since the discovery of graphene through highly oriented pyrolytic graphite (HOPG) mechanical exfoliation (Geim & Novoselov 2007; Seah et al. 2014), a number of alternative methods for graphene synthesis have been developed. Wet chemical exfoliation is one of the most widely used techniques to prepare graphene nowadays but it does have some drawbacks (Botas et al. 2013) including numerous lattice defects, multiple grain boundaries and oxidative traps, which increases its electrical