SYNTHESIS OF MONOLAYER GRAPHENE ON POLYCRYSTALLINE NICKEL AND NICKEL-COPPER BIMETALLIC CATALYST AND STUDY TOWARD THE REUSE OF NICKEL CATALYST

by

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LIST OF ABBREVIATIONS

μm	Micron
AFM	Atomic force microscopy
Ar	Argon
at%	Atomic percentage
atm	Atmospheric pressure
Au	Gold
C	Carbon
CH_4	Methane
C_2H_6	Ethane
C_3H_8	Propane
C_4H_{10}	Butane
Co	Cobalt
Cu	Copper
cm	Centimeter
CVD	Chem cal vapor deposition
C60	Buckminsterfullerene
CO_2	Carbon dioxide
CNT	Carbon nanotube
D	Diameter
DFT	Density functional theory
EDS	Energy dispersive spectrometer
EVA	Poly(ethylene co-vinyl acetate)
Fe	Iron
$fwhm_{2D}$	Full width half maximum of 2D-band
GC	Gas Chromatography
GO	Graphene oxide
H_2	Hydrogen gas
H_2S	Hydrogen Sulfide
He	Helium
HNO_3	Acid nitrate
HOPG	Highly ordered pyrolytic graphite

HRTEM	High resolution tramission electron microscope
ICT	Information and communication technology
I_{2D}/I_G	Intensity ratio of 2D-band over G-band
IPA	Isopropyl alcohol
Ir	Iridium
J	Joule
K	Kelvin
keV	Kilo electron Volt
KOH	Potassium hydroxide
kV	KiloVolt
L	Length
m	meter
MD	Molecular dynamics
min	Minute
MIT	Massachusetts Institute of Technology
MET	Mechano-electro-thermal
ML	Monolayer
mm	Milimeter
Mg	Magnesium
MOHE	Ministry of Higher Education
mol/L	Mol per liter
MOSTI	Ministry on Science, Technology & Innovation
MR	Member ring
mW	MegaWatt
N_2	Nitrogen
NaOH	Sodium hydroxide
Ne	Neon
Ni	Nickel
Ni_3C	Nickel carbide
Ni TFB	Transene Thin Film Nickel Etchants

nm	Nanometer
O	Oxiigen
O_2	Oxygen gas
OD	Outer diameter
Pa	Pascal
PBU	Polybutadien
PC	Poly(bisphenol-A-carbonate)
PDMS	Polydimethylsiloxane
PECVD	Plasma enhanced chemical vapor deposition
PET	Polyethylene terephthalate
PI	Polyimid
PMMA	Poly(methyl methacrylate),
Pt	Platinum
R2R	Roll-to-roll
R&D	Research and Development
Ru	Ruthenium
s	Second
sccm	Standard cubic centimeter
SiC	Silicon carbide
SiO_2	Silicon oxide
SSPO1	2-(diphenylphosphory) spirofluorene
SW	Stone−Wales
TCD	Thermal conductivity detector
TEM	Transmission electron microscopy
Tpa	Terapascal
TRT	Thermal release tape
UHV	Ultra high vacuum
USA	United State of America
V	Volt
Xe	Xenon
XRD	X-ray Di fraction

LIST OF SYMBOLS

%	Percent
π	pi
ºC	Degree Celcius
Å	Angstrom

PENUMBUHAN GRAFENA BERLAPIS TUNGGAL PADA NIKEL POLIHABLURAN DAN PEMANGKIN DWILOGAM NIKEL-KUPRUM DAN KAJIAN UNTUK PENGGUNAAN SEMULA PEMANGKIN NIKEL

ABSTRAK

Grafena merupakan struktur karbon dengan ketebalan satu atom. Grafena terdedah semua atomnya ke medium sekitar. Selepas penemuan grafena pada 2004, ia menjadi subjek utama penyelidikan di seluruh dunia. Grafena mempunyai sifat-sifat yang luar biasa dari segi mekanikal, optik, haba dan elektrik. Sifat-sifat tersebut menjadikan grafena berpotensi digunakan dalam pelbagai aplikasi. Pemendapan wap kimia bermangkin (CVD) adalah saluran yang paling baik untuk menghasilkan grafena berskala wafer, kerana teknik ini mempunyai kelebihan dalam proses pemisahan grafena daripada pemangkin selepas CVD. Dengan bantuan penyejukan pantas, grafena berlapis tunggal berjaya dibentuk pada foil nikel polibabluran dibawah CVD tekanan atmosfera, dengan suhu 850 ºC, tekanan separa metana 0.2 atm and 5 min tempoh reaksi. Tetapi grafena berlapis tunggal gagal didentuk dengan menggunakan foil kuprum sahaja. Penyejukan pantas selepas CVD mendorongkan pelindapkejutan aktiviti pemangkin dan menghadkan kadar difusi karbon dalam nikel ke permukaan nikel. Proses ini memudahkan pembentukan grafena berlapis tunggal berskala wafer. Untuk meningkatkan keseragaman grafena berlapis tunggal, satu teknik mudah digunakan untuk menumbuh grafena berlapis tunggal secara serentak pada kedua-dua foil nikel polihabluran dan foil kuprum polihabluran, pada suhu 950 ºC, tekanan separa metana 0.2 atm and 5 min tempoh reaksi. Stuktur grafena yang seragam dan berkualiti tinggi dapat dibukti dengan spektroskopi Raman dan mikroskop transmisi electron resolusi tinggi. Sistem pemangkin dwilogam yang dicadang membolehkan pengawalan difusi karbon ke permukaan dalam foil Ni dan

Cu. Khususnya, kebolehcapaian karbon dapat dikurangkan pada permukaan Ni dalaman, manakala Cu memainkan peranan sebagai penghalang karbon. Mekanisme pertumbuhan grafena berlapis tunggal dapat dibantu denagn difusi karbon melalui bijian Ni dan sempadan bijian Ni. Daya penggerak untuk difusi karbon datang daripada kepekatan kecerunan karbon antara permukaan yang kaya dengan karbon dan permukaan kurang karbon. Sempadan bijian Ni telah terbukti memainkan peranan yang penting dalam kawalan difusi karbon semasa peringkat pertumbuhan. Dengan bantuan penyejukan pantas, proses pelindapkejut mengurangkan jumlah atom karbon diasing dari Ni, hanya atom karbon yang terletak berhampiran permukaan Ni mempunyai masa yang cukup untuk mengasing dan membentuk grafena. Sementara itu difusi atom karbon dalam tengah foil Ni telah dihalang dan lepas itu membentuk Ni_3C. Ni_3C dikenali sebagai perlindungan yang baik terhadap kakisan. Kehadiran Ni_3C digabungkan dengan penggunaan ferum nitrat (0.5mol/L) sebagai bahan punar lemah semasa pemisahan grafena, foil Ni boleh digunakan semula sehingga 6 kali tanpa menyebabkan sisihan yang besar terhadap kualiti dan keseragaman grafena berlapis dua. Ni_3C ternyata mampu untuk menghadkan kesan punaran foil Ni. Kerja-kerja ini telah berjaya mempamerkan cara yang mudah dan novel untuk mensintesis grafena berlapis tunggal dengan kualiti yang tinggi

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ABSTRACT

Graphene is a layer of sp^2 hybridized carbon atoms with a thickness of only one atom, which exposed most of its atoms to the surrounding medium. Since the discovery of graphene in 2004, it has become the main subject of research around the world. The attractiveness of graphene is mainly attributed to its remarkable mechanical, optical, thermal and electrical properties, enabling graphene to be potentially used in various applications. To date, CVD is the promising method to produce wafer-scale graphene, because it allows an easier separation of graphene from the catalytic substrate. With the assist of fast cooling, monolayer graphene was grown directly on polycrystalline Ni foil under atmospheric pressure CVD with temperature of 850 ºC, methane partial pressure of 0.2 atm and reaction duration of 5 min. However, monolayer graphene could not be formed on Cu under the chosen CVD conditions. Fast cooling after CVD allowed the quenching of the activity of the catalyst and limiting diffusion of dissolved carbon to the surface of Ni, which later facilitate the formation of predominantly wafer scale monolayer graphene. To further improve the uniformity of monolayer graphene, a facile technique was applied to grow monolayer graphene simultaneously on both polycrystalline Ni and Cu foils using a Ni-Cu bilayer catalyst at temperature of 950 ºC, methane partial pressure of 0.2 atm and reaction duration of 5 min. High uniformity and quality of the crystalline structure of the grown graphene was evidenced by Raman spectroscopy mapping and High Resolution Transmission Electron Microscope. The straightforward bimetallic catalytic system allows the control of carbon diffusion to the interface of Ni and Cu.