Faculty of Mechanical Engineering

TRIBOLOGICAL PERFORMANCE OF GRAPHENE SYNTHESISED FROM SOLID WASTE PRODUCTS AS CARBON SOURCES

Noor Ayuma binti Mat Tahir

Doctor of Philosophy

2020

TRIBOLOGICAL PERFORMANCE OF GRAPHENE SYNTHESISED FROM SOLID WASTE PRODUCTS AS CARBON SOURCES

NOOR AYUMA BINTI MAT TAHIR

A thesis submitted
in fulfilment of the requirements for the degree of Doctor of Philosophy

Faculty of Mechanical Engineering

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

2020

DECLARATION

I declare that this thesis entitled "Tribological Performance of Graphene Synthesised from Solid Waste Products as Carbon Sources" is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not currently submitted in candidature of any other degree.

Signature	:	………..
Name	:	Noor Ayuma Binti Mat Tahir
Date	:	…………………………………..........

APPROVAL

I hereby declare that I have read this thesis and in my opinion this thesis is sufficient in terms of scope and quality for the award of Doctor of Philosophy.

Signature	:	……………………………………….
Supervisor Name	:	Prof. Madya Dr. Mohd Fadzli Bin Abdollah
Date	:	……………………………………….

DEDICATION

To my beloved family

ABSTRACT

Graphene is one of the most recent carbon nanomaterials that have attracted a widespread attention due to its excellent properties. Despite intense research on graphene for various applications has been conducted, the tribological properties as self-lubricants solid in coatings technology remains relatively unexplored. There are many studies showings that graphene can be synthesised from variety of carbon-containing sources including waste and bi-products. However, there are limited studies proposed solid waste product as a carbon source. If the synthesised graphene can be readily used without any treatment, the production cost can be lowered, and a good quality coating may be produced to face the demands and challenges in industries nowadays. The objectives of this study are to characterize the chemical bonding and determine the combustion point of fruit cover plastic waste and oil palm fiber. Then, to determine the optimum parameters to synthesise and investigate its tribological performances including comparing its performance with graphene from other studies. This study focused on fruit cover plastic waste and oil palm fiber as solid source by using chemical vapour deposition method. The chemical bonding characterization were conducted by using FTIR spectroscopy analysis meanwhile the combustion point was determined by using combustion in furnace. Based on the FTIR analysis, fruit cover plastic waste was dominated by C-H bond meanwhile C-O bond was dominating the oil palm fibre. The combustion point for fruit cover plastic waste were much lower (600 °C) compared to oil palm fibre (1000 °C). The optimisation was conducted based on Taguchi L9 arrays and Raman spectroscopy analysis were used as the response. The optimum parameters to synthesise graphene from fruit cover plastic waste source are by using Argon gas, at 1020 °C, for 90 minutes, and Hydrogen gas at 1000 °C, for 30 minutes for oil palm fiber. Both graphene coatings are classified under bi-layered and few-layered graphene and provides promising potentials as friction and wear reduction materials where the coefficient of friction obtained from dry sliding test are less than 0.1 for both coating and relatively low wear rate due to the formation of tribolayer on the counter surface. By comparing the coefficient of friction of the graphene synthesised in this study with others, both graphene coatings present lower coefficient of friction compared to the others.

PRESTASI TRIBOLOGI GRAPHENE YANG DISINTESIS DARIPADA PRODUK SISA BUANGAN PEPEJAL SEBAGAI SUMBER KARBON

ABSTRAK

Graphene merupakan salah satu bahan nano-karbon terkini yang telah menarik perhatian ramai kerana sifatnya yang sangat baik. Walaupun penyelidikan intensif terhadap graphene untuk pelbagai aplikasi telah dibuat, sifat-sifat tribologi sebagai pepejal berpelincir sendiri dengan menggunakan teknologi salutan masih belum diterokai secara menyeluruh. Terdapat banyak kajian menunjukkan bahawa graphene boleh dihasilkan melalui pelbagai sumber yang mengandungi unsur karbon termasuklah bahan buangan dan sisa pengeluaran. Walau bagaimanapun, kajian untuk menggunakan sisa buangan pepejal sebagai sumber karbon adalah terhad. Jika graphene yang disintesis boleh digunakan dengan mudah tanpa sebarang rawatan, kos penghasilan graphene yang berkualiti baik boleh diturunkan. Objektif kajian ini adalah untuk mengenalpasti jalinan kimia dan menemukan titik pembakaran bagi sampah plastik pembalut buah dan serat buah kelapa sawit. Selain itu, kajian ini juga bertujuan untuk mencari parameter optimum bagi penghasilan graphene serta mengenalpasti prestasi tribologi graphene yang dihasilkan dengan perbandingan terhadap prestasi graphene dari kajian lain. Kajian ini menumpukan kepada dua jenis bahan sisa pepejal iaitu sampah plastik pembalut buah-buahan dan serat kelapa sawit dengan menggunakan kaedah yang dikenali sebagai 'chemical vapour deposition' (CVD). Analisis jalinan kimia dilakukan menggunakan 'fourier-transform infrared spektroskopy' (FTIR) manakala analisis titik pembakaran dilakukan menggunakan kaedah pembakaran di dalam relau. Berdasarkan analisis FTIR, plastik pembalut buah-buahan didominasi oleh jalinan C-H manakala jalinan C-O mendominasi serat buah kelapa sawit. Titik pembakaran bagi plastik pembalut buah-buahan mempunyai titik pembakaran yang jauh lebih rendah (600 °C) berbanding serat kelapa sawit (1000 °C). Kaedah pengoptimuman dijalankan menggunakan kaedah Taguchi untuk menghasilkan susun atur L9 manakala analisis Raman dipilih sebagai tindak balas. Parameter optimum untuk menghasilkan graphene dari sampah plastik pembalut buah-buahan adalah dengan menggunakan gas Argon, pada 1020 °C, selama 90 minit, dan gas Hidrogen pada 1000 °C, selama 30 minit untuk serat kelapa sawit. Kedua-dua salutan graphene tersebut diklasifikasikan sebagai 'bi-layered dan few-layered graphene' dan ianya berpotensi sebagai bahan pengurangan geseran dimana pekali geserannya kurang dari 0.1 dan rendah kadar kehausan. Dengan membandingkan pekali geseran graphene di dalam kajian ini terhadap yang kajian lain, nilai yang dihasilkan bagi kedua-dua graphene dalam kajian ini adalah lebih rendah berbanding kajian yang lain.

ACKNOWLEDGEMENTS

First and foremost, I would like to take this opportunity to express my sincere gratitude to my supervisor, Associate Professor Dr. Mohd Fadzli Bin Abdollah for his non-stop supervision, support, motivation, encouragement and immense knowledge towards the completion of research and thesis writing. I would also like to express my gratitude to Professor Ts. Dr. Noreffendy Bin Tamaldin as my co-supervisor and deepest gratitude to Dr. Mohd Rody Bin Mohamad Zin for his generous ideas and helps.

Huge gratitude to the Ministry of Education Malaysia for supporting this research by the grant (Grant No.: FRGS/1/2016/TK10/FKM-CARE/F00315) and also to Universiti Teknikal Malaysia Melaka for the Zamalah Scheme Scholarship (2016 – 2018) and grant research (Grant No: PJP/2019/FKM(11A)/S01684).

Tremendous thank you to Japan Science and Technology Agency (JST) for the opportunity given on the Japan-Asia Youth Exchange Program in Science (nicknamed as SAKURA Exchange Program in Science) to experience the advanced scientific technology of Japan. Warmest thanks to Professor Dr. Noritsugu Umehara and Associate Professor Dr. Takayuki Tokoroyama for the hospitality, guidance and lectures.

I am also gratefully acknowledged the contributions of the Green Tribology and Engine Performance (G-Tribo-E) research group members, assistance engineers and laboratory technicians. Warmest and deepest thank you to all my postgraduate fellows and faculty's members for their direct and indirect assist.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

0D	-	zero-dimension
1D	-	one-dimension
2D	-	two-dimension
3D	-	three-dimension
AFM	-	atomic force microscope
Al_2O_3	-	aluminium oxide
AP-CVD	-	atmospheric pressure chemical vapor deposition
Ar	-	argon gas
BF_4	-	1-butyl-3-methylimidazolium tetrafluoroborate
BL	-	boundary layer
BLG	-	bi-layered graphene
CH_4	-	methane
Co	-	cobalt
CO	-	carbon monoxide
CO_2	-	carbon dioxide
COF	-	coefficient of friction
CVD	-	chemical vapor deposition
Cu	-	copper
DOE	-	design of experiment
DLC	-	diamond like carbon

ECR-CVD	-	electron cyclotron resonance chemical vapor deposition
EDX	-	energy dispersive x-ray
EHD	-	elasto-hydrodynamic
EG	-	expanded graphite
FCPW	-	fruit cover plastic waste
FLG	-	few layered-graphene
FQM	-	fluorescence quenching microscopy
FTIR	-	Fourier-transform infrared spectroscopy
FWHM	-	full-width half maximum
GNP	-	graphene nano-plates
GO	-	graphene oxide
H	-	hydrogen atom
H_2	-	hydrogen gas
H_2O	-	water
HD	-	hydrodynamic lubrication
HfO_2	-	hafnium oxide
HOPG	-	highly oriented pyrolytic graphite
LP-CVD	-	low pressure chemical vapor deposition
ME	-	mechanical exfoliation
MgO	-	magnesium oxide
MLG	-	multi-layered graphene
MoO_3	-	molybdenum trioxide
MoS_2	-	molybdenum disulphate
MPOB	-	Malaysian Palm Oil Board
MWNT	-	multi-walled nanotubes

NaOH	-	sodium hydroxide
Ni	-	nickel
NMSC	-	Ni_3Al matrix self-lubricating composites
NP	-	nano-platlets
OIT	-	optical imaging technique
OPF	-	oil palm fiber
PBM	-	polymer-based material
PB-CVD	-	pulse-biased chemical vapor deposition
PDMS	-	polydimethylsiloxane
PE-CVD	-	plasma enhanced chemical vapor deposition
PET	-	polyethylene terephthalate
PF_6	-	1-butyl-3- methylimidazolium hexafluorophosphate
PKAC	-	palm kernel activated carbon
PKAC-E	-	palm kernel activated carbon epoxy
PMMA	-	polymethyl methacrylate
PSAC	-	palm shell activated carbon
PTFE	-	polytetraflurouthylene
PVD	-	pressured vapor deposition
QHE	-	quantum hall effect
rGO	-	reduced-graphene oxide
RBC	-	rice bran ceramics
RH-CVD	-	rapid heating chemical vapor deposition
RS	-	raman spectroscopy
SEM	-	scanning electron microscope
Si	-	silicon

SiO	-	silicon oxide
Si_3N_4	-	silicon nitride
SLG	-	single-layered graphene
SPGF	-	solution processed graphene flakes
SPGO	-	solution processed graphene oxide
SWNT	-	single-walled nanotubes
T-CVD	-	thermal chemical vapor deposition
TEM	-	transmission electron microscope
TPES	-	total primary energy supply
TRT	-	thermal released tapes

LIST OF SYMBOLS

°C	-	degree Celsius
μ	-	coefficient of friction
α	-	outside angle
φ	-	tip angle
σ	-	in-plane bond
π	-	out-of-plane bond
%	-	percentage
ρ	-	density
a	-	wear scar radius
h	-	wear depth
I_{2D}/I_G	-	intensity of 2D/G peak
I_D/I_G	-	intensity of D/G peak
k	-	wear rate
L	-	sliding distance
m_{loss}	-	mass loss
r	-	ball bearing radius
v	-	sliding speed
V_{loss}	-	volume loss
W	-	applied load
wt%	-	weight percentage