STRUCTURE AND ELECTRICAL CONDUCTIVITY OF POLYMER BASED GRAPHENE-CARBON NANOTUBES COMPOSITE

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DEDICATION

This thesis is dedicated to my parents for the blessing and doa'.

It is also dedicated to my family and friends.

I would not have made it this far without your love, support, and guidance.

May this knowledge be useful to others.

May Allah grant you Jannah.

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ABSTRACT

The enhancement of electrical conductivity of silicone rubber polymer has been reported when graphene and carbon nanotubes (CNTs) are added as separate carbon nanofillers into a polymer matrix. The concept of synergy through combining various types of carbon nanofillers is seen as one of the successful approaches to obtain desired properties of new materials. However, the electrical conductivity properties of graphene and CNTs combination as a single carbon nanofillers into polymer matrix are still unknown and has not been reported elsewhere. This study aims to synthesize polymer composites and compare their electrical conductivity and physical properties. The solution mixing technique was used to synthesize these polymer composites. The nanofillers' weight percentage (wt%) were varied, and the electrical conductivity test was performed using the Two-Point Probe system. Three sets of samples were prepared; (i) graphene-polymer composites, (ii) CNTs-polymer composites and (iii) graphene-CNTs-polymer composites. The highest electrical conductivity value recorded was 1.94×10^{-1} S/m when an applied voltage of 10 V was subjected to the 2 wt% of graphene mixed with 25 wt% of CNTs in the polymer matrix. The increment of electrical conductivity recorded by the Two-Point Probe system was supported by the field emission scanning electron microscope and high resolution transmission electron microscope structural images. The microstructural observation on the prepared conductive polymer composites samples indicated that graphene and CNTs were distributed homogeneously in the polymer matrix. The results also showed that graphene and CNTs created a conductive network in a polymer matrix, which greatly increased the electrical conductivity of polymer composites. Thermogravimetric analysis revealed that the thermal stability of graphene-CNTs-polymer composites was higher than the other prepared polymer composites and elastomer. At 400 ˚C, only 3.39% of weight loss for graphene-CNTs-polymer composites compared to elastomer where their weight loss was approximately 3.97%. This result shows that incorporation of graphene and CNTs in a polymer matrix enhanced the thermal properties of polymer composites.

ABSTRAK

Peningkatan keberaliran elektrik polimer getah silikon telah dilaporkan apabila grafin dan tiub nano karbon (CNTs) ditambah sebagai pengisi nano karbon secara berasingan di dalam matriks polimer. Konsep sinergi dengan menggabungkan pelbagai jenis pengisi karbon dilihat sebagai salah satu pendekatan yang berjaya untuk memperoleh sifat-sifat yang dikehendaki daripada bahan baharu. Walau bagaimana pun, sifat beraliran elektrik gabungan grafin dan CNTs sebagai pengisi nano karbon tunggal di dalam matriks polimer masih belum diketahui dan tidak dilaporkan dalam literatur. Tujuan kajian ini adalah untuk mensintesis komposit polimer dan membandingkan sifat keberaliran elektrik dan sifat fizikalnya. Teknik pencampuran larutan telah digunakan untuk mensintesis komposit polimer. Peratusan berat (wt%) pengisi nano diubah dan ujian keberaliran elektrik dijalankan menggunakan sistem Kuar Dua-Titik. Tiga set sampel telah disediakan; (i) komposit polimer-grafin, (ii) komposit polimer-CNTs dan (iii) komposit polimer-grafin-CNTs. Nilai keberaliran elektrik tertinggi yang direkodkan ialah 1.94×10^{-1} S/m apabila voltan 10 V dikenakan kepada matriks polimer dengan campuran 2 wt% grafin dengan 25 wt% CNTs. Peningkatan keberaliran elektrik yang dicatatkan oleh sistem Kuar Dua-Titik telah disokong dengan imej struktur mikroskop imbasan elektron pancaran medan dan mikroskop penghantaran elektron beresolusi tinggi. Pemerhatian mikrostruktur pada sampel komposit polimer beraliran yang disediakan telah menunjukkan bahawa grafin dan CNTs adalah teragih secara seragam dalam matriks polimer. Hasil juga menunjukkan bahawa grafin dan CNTs telah mewujudkan rangkaian beraliran dalam matriks polimer yang mana meningkatkan sifat keberaliran elektrik komposit polimer tersebut. Analisis termogravimetrik telah mendedahkan bahawa kestabilan haba komposit polimer grafin-CNTs adalah lebih tinggi berbanding dengan komposit polimer lain dan elastomer. Pada suhu 400°C, komposit polimer-grafin-CNTs kehilangan berat hanya sebanyak 3.39% berbanding dengan elastomer yang mana kehilangan berat kira-kira 3.97%. Keputusan ini menunjukkan bahawa gabungan grafin dan CNTs di dalam matriks polimer telah meningkatkan sifat haba komposit polimer.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

ABS	-	Acrylonitrile Butadiene Styrene
CNTs	-	Carbon nanotubes
CPs	-	Conductive polymers
FESEM	-	Field Emission Electron Microscopy
FPP	-	Four Point Probe
Gra	-	Graphene
HR-TEM	-	High Resolution-Transmission Electron Microscopy
LiPo	-	Lithium-ion polymer battery
PLA	-	Polylactic Acid
TGA	-	Thermogravimetric Analysis
TPP	-	Two Point Probe
DCM	-	Dichloromethane
wt%	-	Weight percentage
v%	-	Volume percentage
SWCNTs	-	Single-Walled Carbon Nanotubes
MWCNTs	-	Multi-Walled Carbon Nanotubes
CVD	-	Chemical Vapour Deposition
Ppy	-	Polypyrrole
EEG	-	Electroencephalography
Ag/AgCl	-	Silver/silver chloride
MRI	-	Magnetic resonance imaging
PEDOT:PSS	-	Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)
PLA	-	Poly(lactic acid)
PETI	-	Phenylethynyl terminated imides

LIST OF SYMBOLS

δ	-	Conductivity
˚C	-	Degree Celsius
ρ	-	Resistivity
Ω	-	Ohm
S/m	-	Siemens per metre
S/cm	-	Siemens per centimetre

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Research Background

Electrode is an electrical conductor used as a medium to make contact and allow current flow to a non-metallic part of the circuit. Electrodes are typically good electric conductors, but not necessarily metals materials. It can be a semiconductor, an electrolyte, vacuum or air. Electrodes are used in various application such as medical purposes, energy storage, electroplating, arc welding, grounding, and chemical analysis.

There are several types of present metal electrodes such as tin plated, pure tin, silver, silver/silver chloride (Ag/AgCl) and gold plated that are used for medical purposes, especially in electroencephalography (EEG) for recording brain activity. Magnetic resonance imaging (MRI) compatible EEG electrodes commonly used gold plated or Ag/AgCl. The problems of present metal based electrodes are it involves a complex casting process, requires high temperature, high cost of the manufacturing process, limited electrodes shapes and sizes, low surface contact, and poor flexibility. To enhance its electrical conductivity, the procedure of metal electrodes requires using hydrogel or conductive paste for skin preparation. This will lead to irritation, discomfort, skin abrasion to the patients, and leads to stress environment.

Nowadays, there are many hospitals using polymer electrodes. As Schultz (2012) wrote in his survey report in The Neurodiagnostic Journal, they get positive feedback from patients at the EEG centre based on survey responses. From 54 surveys returned, 17 hospitals utilised conductive polymer electrodes at the workplace. Polymer base electrodes have proven improved patient safety and care due to less electrode fixing and maintenance, less skin deterioration, and lessen patient exposure to irritation and irritants. (Salvo *et al.*, 2012). Hydrogel or conductive paste are not required anymore. The flexibility of polymer electrodes further enhanced the surface contact and the electrical conductivity.

Conductive polymers are described as organic polymers which can conduct electricity or allow current to flow. Researchers have dedicated enormous attention to study about conductive polymers because of their capability to conduct electric similar to metals and semiconductors, but at the same time, retaining polymers properties. As properties of conductive polymers are lightweight, high flexibility, easy to manufacture and inexpensive, conductive polymers have become the most preferred alternative in materials research field nowadays. Since 1980, various conductive polymers have been investigated, for example, polypyrrole (PPy), polyaniline (PANI), polyacetylene (PA) (Kumar and Sharma, 1998; Singh *et al.*, 2012), and poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) for intrinsically conductive polymer and for extrinsically conductive polymers such as graphene-polymer composites, CNTs-polymer composites, PLA/graphene nanocomposites, and polypyrrole/graphene nanocomposites.

Materials which are made from at least two different materials to obtain desired properties are called composites materials. A polymer composite material is formed from a combination of different materials which reinforcing nanofillers are integrated with a polymer matrix (Dantas de Oliveira and Augusto Gonçalves Beatrice, 2019). Each material has its own characteristic with significantly different physical or chemical properties. When combining the positive qualities of both materials, composites materials will produce a material that has the characteristics different from the individual materials (Balint *et al.*, 2014). Synthesis of composite materials enhance the characteristics of materials itself. The materials will become stronger, lighter, stable, high conductivity, high thermal stability, high flexibility, and less expensive.

Carbon based materials such as carbon black, fullerenes, graphite fibres, graphene, and carbon nanotubes (CNTs) act as conductive nanofillers for the synthesis of conductive polymers composite materials (Nambiar and Yeow, 2011). Usually, conductive polymer composites consist of one or more non-conducting polymers and the conductive nanofillers materials which are dispersed throughout the polymer matrix using several techniques. The introduction of nanofillers into conventional polymer composites can significantly enhance the electrical conductivity properties of polymer composites (Barkoula *et al.*, 2008). Therefore, the combination of graphene, CNTs and polymer matrix gives significant effect to enhance electrical conductivity properties (Bose *et al.*, 2010; Soldano *et al.*, 2010; Potts *et al.*, 2011).

Conductive polymers draw a vast interest to improve the mechanical (Spanos *et al.*, 2015), electrical (Bose *et al.*, 2010), and chemical (Potts *et al.*, 2011) properties of polymeric materials. It is applied for various application such as photovoltaic devices (Kim *et al.*, 2010; Huang *et al.*, 2012), thermal interphase materials (Potts *et al.*, 2011; Hu *et al.*, 2014), energy storage or power delivery materials (Singh *et al.*, 2012; Zhang *et al.*, 2015), and biomedical materials (Nambiar and Yeow, 2011; Balint *et al.*, 2014).

Nowadays, the concept of synergy through combining the various types of nanomaterials is seen as the most successful approach to obtain desired properties of selected new materials. Therefore, it has become the current trend to incorporate nanofillers (graphene and CNTs) into elastomer polymers (silicone rubber) to produce conductive polymers composites with high electrical conductivity and high flexibility as compared to neat polymers. Also, the incorporation of graphene and CNTs into a polymer matrix can dramatically enhance the electrical conductivity and physical properties of polymer composites even with a small wt% of nanofillers (Galpaya *et al.*, 2012). On the other hand, conductive polymers composites are the most widely utilise classes of new materials due to their incredible variety of physical structures and properties, together with their relatively low cost (Khan *et al.*, 2016; Phiri *et al.*, 2017), easy processing (Balint *et al.*, 2014), and high potential for recycling and serve as sustainable materials (Zhong *et al.*, 2017) which make them more favourable in the industries.

1.2 Problem Statement

To date, the synthesis of graphene/CNTs-polymer composites with enhanced electrical conductivity and physical properties are still limited in commercial success. This limitation is mainly due to the inability to control the desired microstructures, economically or practically. Generally, research efforts are addressing the following fundamental questions. How do polymers assemble at the nanofillers-matrix interface? What is the interphase structure of prepared polymer composites? The answers to at least some of these questions will build an understanding and investigative approach for studying the fundamental interactions of the intermediate-scale structures of polymer composites.

Polymer has played an important role in various application, however, it is a non-conductive material. To further improve their electrical conductivity and expand their applications, nanofillers such as CNTs are often incorporated to synthesise CNTs-polymer composites. CNTs have been widely studied as nanofillers due to its electrical conductivity properties. After the discovery of graphene, a lot of effort has been recently attracted to synthesise graphene-polymer composites to produce high-performance conductive polymer composites. Between graphene-polymer composites and CNTs-polymer composites, the former has more disadvantages over the latter. The electrical conductivity of CNTs-polymer composites is much higher as compared to graphene-polymer composites at the same wt% of nanofillers (Sun *et al.*, 2013). As an individual material, graphene shows an electrical conductivity of 10^6 S/cm, higher than 10^5 S/cm of the CNTs (Sun *et al.*, 2013). However, both nanofillers are still the most effective nanofillers materials to enhance electrical conductivity and physical properties of the polymer matrix. This raises a new thought on the development of polymer composites materials. Why do graphene and CNTs are not being incorporated into polymers to synthesise new polymer composite materials? Although either graphene-polymer composites or CNTs-polymer composites have been widely investigated, the simultaneous incorporation of graphene and CNTs into polymers is still limited.

HR-TEM has recently arose as an advance structural characterisation tool. HR-TEM covers a range of low magnification imaging as well as atomic-scale details. Based on literature review, there is no accurate detailed study of the interphase structural of graphene and CNTs in the polymer matrix of graphene/CNTs-polymer composites observed directly using high resolution-transmission electron microscope (HR-TEM). Graphene is one of the most promising materials in nanotechnology. The electronic and physical properties of graphene samples with high perfection of the atomic lattice are outstanding, but the structural defects, which may appear during growth or processing, deteriorate the performance of graphene-based devices. However, deviations from perfection can be useful in some applications, as they make it possible to achieve new functionalities of graphene (Banhart *et al.*, 2010; Liu *et al.*, 2015). Current HR-TEM is capable of observing the defect in graphene which quite interesting to study. Therefore, by using suitable methods for the preparation of sample, it is possible to observe the atomic structure of graphene defects and the atomic interaction under HR-TEM.

Even though conductive polymer composites have been widely reported (Kim *et al.*, 2010; Potts *et al.*, 2011; Hu *et al.*, 2014), there is still lack of data in terms of the electrical conductivity of graphene/CNTs-polymer composites that are synthesised in this study. Through a combination of graphene and CNT at various wt%, it may significantly enhance the electrical conductivity of polymer composites. It is expected that the incorporation of nanofillers (graphene and CNTs) and polymer matrix would enhance the electrical conductivity properties of the graphene/CNTs-polymer composites based on the formation of conductive networks.

By introducing electrical conductivity properties in polymer, it will be useful in various applications. For example, the advancement of 3D printer technology signifies that polymer-based electrodes have great adaptable potential as an alternative electrode with an unlimited design for substituting the metal-based electrodes (Salvo *et al.*, 2012). There is a commercially available polymer such as silicone rubber, which is generally non-conductive. Therefore, it will be embedded with graphene and CNTs to enhance its conductivity properties. This graphene/CNTs-polymer composites could be used as conductive electrode materials (Sun *et al.*, 2013).

The development of materials which incorporate graphene and CNTs into polymer matrix (elastomer) is believed could maximise the benefits which can be applied as polymer based electrodes. Polymer based electrodes are the promising alternative electrodes with outstanding properties in flexibility, lightweight and high electrical conductivity (Lee *et al.*, 2013; Hu *et al.*, 2014; Sadasivuni *et al.*, 2014) to substitute the conventional metal based electrodes.

1.3 Research Objectives

Based on the problem statement, a lot of efforts are still needed to turn graphene/CNTs-polymer composites into practical applications. Thus, this study aims to investigate the detailed analysis of the electrical conductivity and physical properties of graphene/CNTs-polymer composites that will be synthesised using a new combination of graphene, CNTs and elastomer (silicone rubber).

Therefore, this study has two main objectives, which are:

(a) To synthesise the graphene/CNTs-polymer composites and characterise its structural properties;

(b) To investigate the electrical conductivity and physical properties of the electrolytic graphene/CNTs-polymer composites at various weight percentage (wt%) composition.

1.4 Research Scope

The objectives of this study can be obtained through the research scopes outlined as follow:

1. To achieve homogeneous dispersed graphene and CNTs in a polymer matrix, simple solution mixing technique has been carried out for the synthesis of graphene/CNTs-polymer composites proposed by Zhang *et al.*, (2015) and Phiri *et al.,* (2017). This technique is a well-known strategy to prevent graphene/CNTs agglomerates or stacking in the polymer matrix.

2. This research was focusing on employing HR-TEM to characterise in detail the interphase structure of prepared graphene/CNTs-polymer composites.

3 Parameter of the mixture graphene and CNTs were varied to optimise the electrical conductivity. The range of wt% of graphene nanofillers is 0.5-2.5 wt% (Saravanan *et al.*, 2014; Bafana *et al.*, 2017) and for CNTs are 5-25 wt% (Ghose *et al*, 2015) and the mixture were mixed with elastomer (silicone rubber) as a based substrate.

4. Electrical conductivity test was conducted by using Two Point Probe (TPP) systems to investigate the effect of varying composition of graphene and CNTs. Other instruments that were used are RAMAN spectroscopy, FESEM, and thermogravimetric analysis (TGA).

Finally, the most significant results of the outstanding electrical conductivity and physical properties of graphene and CNTs-polymer composites, and some fundamental properties will be discussed in chapter 5; results and discussion.

1.5 Significance of This Study

This study was a challenge to develop desirable properties of conducting polymer composites such as high electrical conductivity, high flexibility, high physical properties, lightweight, easy to manufacture and retaining polymer properties by combining graphene, CNTs and elastomer. The findings of this research will contribute to the benefit of medical application.

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LIST OF PUBLICATIONS

Journal with Impact Factor

1. **Nur Farhana Hasmuni**, Aini Zuhra Abdul Kadir, Izman Sudin, Madzlan Aziz, Tanemura Masaki, & Mohd Zamri Mohd Yusop (2018). Structural and Electrical Properties of Graphene/CNTs-Polymer Composites Observed Under Transmission Electron Microscope. *Japanese Journal of Applied Physics (JJAP).* **(Q3, IF:1.452)-Submitted**

Indexed Journal

1. **Nur Farhana Hasmuni**, Suhaili Ismail, Aini Zuhra Abdul Kadir, Izman Sudin, Madzlan Aziz & Mohd Zamri Mohd Yusop (2018). Fabrication of Polymer-Based Graphene Composites as Highly Conductive Polymer Electrode. *American Institute of Physics (AIP).* **(Indexed by SCOPUS)-Accepted**

Non-Indexed Conference Proceeding

1. **Nur Farhana Hasmuni**, Suhaili Ismail, Aini Zuhra Abdul Kadir, Izman Sudin, Madzlan Aziz & Mohd Zamri Mohd Yusop (2018). Fabrication of Polymer-Based Graphene Composites as Highly Conductive Polymer Electrode. In *2018 International Conference on X-Rays & Related Techniques in Research & Industry 2018 (ICXRI 2018)*. (pp. 102-103).