ELECTRICAL CHARACTERISATION OF MULTI-WALLED CARBON NANOTUBE/POLYDIMETHYLSILOXANE USING IMPROVED FACILE DISPERSION TECHNIQUE

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ABSTRACT

The growth of polymer composite in technological advancement has contributed to the development of electrocardiogram (ECG) electrode, which gives better wearable conformality on human skin. This gives a promising alternative to Multi-Walled Carbon Nanotube (MWCNT) and polydimethylsiloxane (PDMS) due to its low cost, ease of manufacturing and flexibility. However, with the high aspect ratio and strong Van der Waals interaction forces, MWCNT easily agglomerates, and bundling to each other. An effective dispersion technique of MWCNT/PDMS composite is essential to enhance the electrical conductivity by controlling MWCNT content and maintaining electrode flexibility. Thus, this study disperses the MWCNT/PDMS composite using a solution mixing method: sonication process and mechanical stirring. The MWCNT is dispersed using toluene solvent to achieve uniform dispersion, where the MWCNT content varies from 2 wt% to 10 wt% in PDMS matrix. As a result, the MWCNT/PDMS composite conductivity is in the range of 0.30×10^{-9} S/cm to 6.14×10^{-6} S/cm. At 4 wt%, the MWCNT/PDMS composite reached the percolation threshold region. The fabricated polymer composite was further characterised in Raman spectroscopy and the measurement shows vibration peaks in the D-band at 1349 cm^{-1} and G-band at 1585 cm^{-1}. This proves the dispersion of MWCNT in PDMS. In addition, the intensity of the D-band and G-band, I_D/I_G decreases when the MWCNT/PDMS concentration increases, indicating less defect of MWCNT during the sonication process. These findings show that the MWCNT/PDMS has the potential as an excellent polymer composite electrode.

ABSTRAK

Perkembangan komposit polimer dalam kemajuan teknologi telah menyumbang kepada pembinaan elektrod elektrokardiogram (ECG) yang memberikan kesesuaian boleh pakai yang lebih bagus pada kulit manusia. Ini telah memberikan alternatif yang baik kepada karbon nanotiub pelbagai dinding (MWCNT) dan polydimethylsiloxane (PDMS) kerana kosnya yang rendah, pembuatan yang mudah dan fleksibel. Walau bagaimanapun, dengan nisbah aspek yang tinggi dan daya interaksi Van der Waals yang kuat, MWCNT mudah teraglomerati dan bergabung sesama sendiri. Teknik penguraian MWCNT/PDMS komposit yang efektif sangat penting bagi meningkatkan kekonduksian elektrik dengan mengawal kepekatan MWCNT sekaligus mengekalkan daya kelenturan elektrod. Oleh itu, kajian ini telah menguraikan komposit MWCNT/PDMS dengan menggunakan kaedah pencampuran larutan: proses sonifikasi dan proses pencampuran secara mekanikal. Untuk mencapai penguraian yang seragam, MWCNT/PDMS diurai dengan menggunakan pelarut toluene, dimana kandungan MWCNT berbeza dari 2 wt% hingga 10 wt% dalam PDMS. Hasilnya, kekonduksian komposit MWCNT/PDMS adalah dalam lingkungan 0.30×10^{-9} S/cm to 6.14×10^{-6} S/cm. Pada 4 wt%, komposit MWCNT/PDMS mencapai kawasan ambang perkolasi. Komposit yang direka ini selanjutnya telah dicirikan dengan spektrokopi Raman dan bacaan data menunjukkan puncak getaran pada D-band di 1349 cm^{-1} dan G-band di 1585 cm^{-1}. Ini membuktikan proses penguraian MWCNT dalam PDMS. Di samping itu, intensiti D-band dan G-band, I_D/I_G berkurang ketika kepekatan MWCNT/PDMS bertambah, menunjukkan kurangnya kerosakan MWCNT semasa proses sonifikasi. Keputusan ini menunjukkan bahawa MWCNT/PDMS mempunyai potensi sebagai elektrod komposit polimer yang sangat baik.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

Ag	-	Silver
AV	-	Atrioventricular
CCB	-	Conductive Carbon Black
Cl	-	Chloride
CNF	-	Carbon Nanofiber
CNT	-	Carbon Nanotube
COOH	-	Carboxylic Group
CT	-	Computerized Tomography
CVD	-	Cardiovascular Disease
DMF	-	Dimethylformamide
ECG	-	Electrocardiogram
FPT	-	Four Point Probe
HCl	-	Hydrochloric Acid
HNO_3	-	Nitric Acid
H_2O_2	-	Hydrogen Peroxide
H_2SO_4	-	Sulphuric Acid
ID	-	Internal Diameter
IPA	-	Isopropyl Alcohol
IV	-	Current-Voltage
MWCNT	-	Multi-Walled Carbon Nanotube
NMP	-	N-methyl-2-pyrrolidone
OD	-	Outer Diameter
OH	-	Hydroxyl Group
PANI	-	Polyaniline
PDMS	-	Polydimethylsiloxane
PDLC	-	Polymer-Dispersed Liquid Crystal
PEDOT	-	Poly(3,4-ethylene dioxythiophene)
PET	-	Polyethylene Terephthalate
PMMA	-	Polymethyl Methacrylate
PPE	-	Personal Protective Equipment

Ppy	-	Polypyrrole
PSS	-	Polystyrene Sulfonate
PU	-	Polyurethane
RBM	-	Radial Breathing Mode
RC	-	Resistance-Capacitance
SA	-	Sinoatrial
SEM	-	Scanning Electron Microscopy
SC	-	Skin Capacitance
SCFNA	-	Spatially Confined Forced Network Assembly
SWCNT	-	Single-Walled Carbon Nanotube
THF	-	Tetrahydrofuran
TPP	-	Two Point Probe
UIRL	-	University Industry Research Lab
UTM	-	Universiti Teknologi Malaysia
WHO	-	World Health Organization

LIST OF SYMBOLS

A	-	Cross-sectional area
A	-	Ampere
$\vec{a}$	-	Base vector
C_C	-	Capacitance across the epidermis
C_d	-	Capacitance across the double layer of charge at electrode-electrolyte
$\overrightarrow{C_h}$	-	Chiral vector
C_p	-	Capacitance across the epidermis
D/d	-	Diameter
d_v	-	Density
E	-	Young's Modulus of elasticity
E	-	Energy
E_{hc}	-	Half-cell potential
E_p	-	Potential difference at sweat gland
E_{sc}	-	Potential different at dermis
f	-	Frequency
h	-	Plank constant
k	-	Thermal conductivity
L/l	-	Length
m/n	-	Integer
P	-	Power
p	-	Power fraction
p_c	-	Percolation threshold
R	-	Electrical resistance
R_c	-	Resistance across epidermis
R_d	-	Leakage resistance across double layer
R_p	-	Resistance across sweat gland
R_s	-	Series resistance in electrolyte
R_u	-	Resistance in dermis and subcutaneous layers
S	-	Tensile strength

T	-	Temperature
t	-	Times
t	-	Critical exponent
V	-	Volts
ν	-	Frequency of incident photon
v	-	Velocity/speed
wt%	-	Weight percentage
θ	-	Chiral angle
σ	-	Electrical conductivity
σ_s	-	Stress
ε	-	Strain
δ_g	-	Roller gap distance
Ω	-	Ohm
ρ	-	Electrical resistivity
λ	-	Wavelength

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Research Background

Recently, the demand for wearable medical electronic devices and sensors is growing due to the rise of cardiovascular disease. This market demand allows research and development of diagnostic tools in this area. Figure 1.1 clearly shows the need for medical wearable and sensors for Metabolic and Cardiovascular Monitoring (yellow colour) is expected to increase from 2018 to 2024 [1].

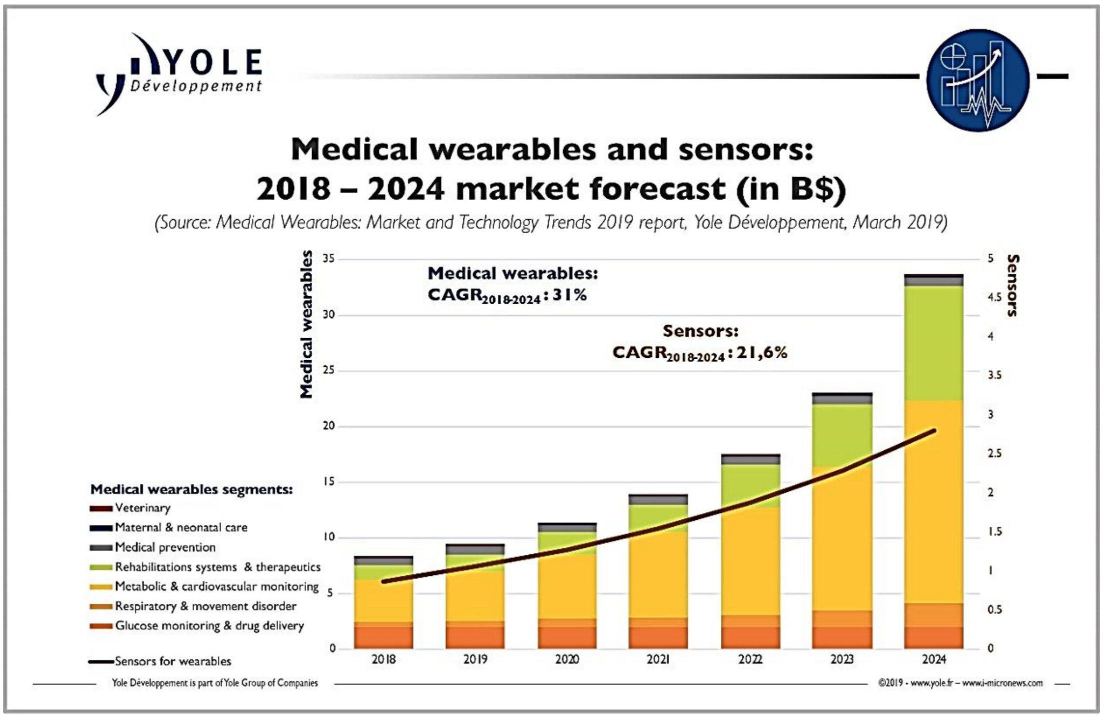

Figure 1.1 Market forecast for medical wearable and sensor [1].

Cardiovascular disease (CVD) refers to a condition that involves the heart and blood vessels such as atherosclerosis (heart disease), ischaemic disease, heart attack, stroke, and diabetes. According to the World Health Organization (WHO), CVD kills almost 17.9 million people every year around the world [2]. Besides, the Department of Statistics Malaysia (DOSM) also reports 8776 deaths in 2007 due to ischaemic disease. This mortality rate has increased by 54% after 10 years to 13 505 deaths in 2017 [3]. This situation is a bit worrying because the initial symptom is frequently ignored, and thus many patients are treated only after suffering a heart attack or stroke. To prevent this situation, preliminary action is needed to diagnose CVD's patients.

Figure 1.2 shows the list of medical technologies used to diagnose cardiovascular diseases, such as a physical exam, imaging tests such as chest x-ray or CT scan, echocardiogram, blood tests such as ELISA, and electrocardiogram. Among existing medical techniques, electrocardiogram (ECG) is the most popular technique used to monitor CVD patients' health [4]. Electrocardiogram (ECG) is a method of collecting electrical signals from the heart. The electrical signals generated by the muscle depolarization of the heart is measured in minimal voltage in microvolt by placing the ECG electrode on the skin.

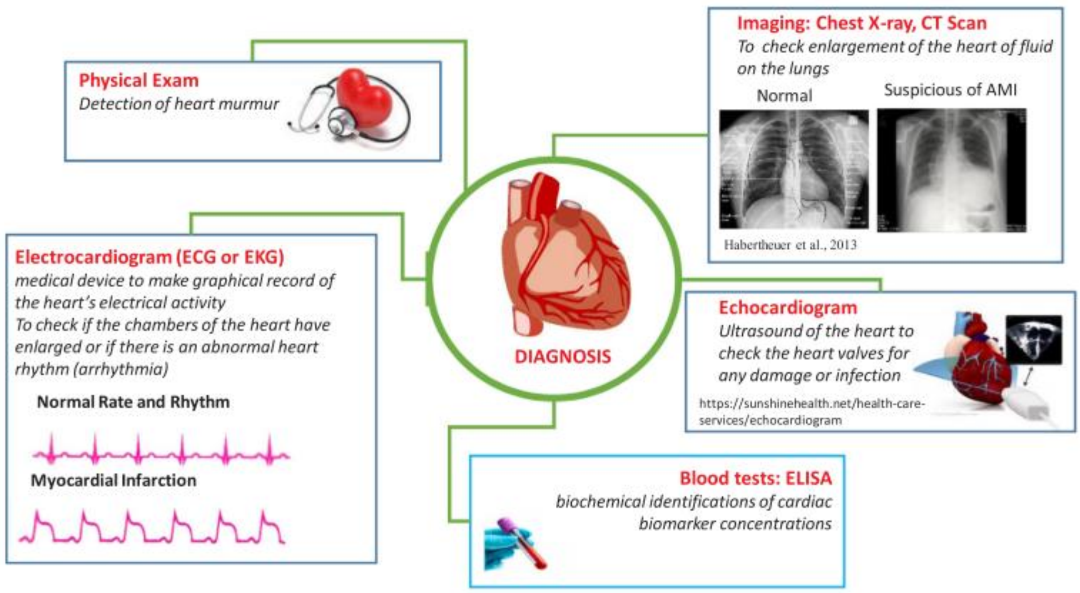

Figure 1.2 Current diagnostic tools to diagnose Cardiovascular Disease [4]

An electrode is used to record the heart's electrical activity from human skin and interpret the signal into graphical form. This electrode can be classified into two types: wet electrode and dry electrode. It is imperative that the electrode types only referring to the electrical conductor from the electrochemical properties.

Conventionally, wet electrodes or silver-silver chloride (Ag/AgCl) electrodes are commonly used in the clinical field. This is due to the electrolyte gel which helps to increase skin hydration, reduces the impedance of the skin, and produces good ECG signal quality. However, the electrolyte gel has a limitation, especially in the long-term monitoring. The signal degrades because the gel dried over time [5]. In addition, the electrolyte gel causes skin irritation and redness [6]. Besides, the wet electrode also consumes a long period of skin preparation, such as dirt-removing and hair cutting [7].

To address these problems, dry electrodes are developed by removing the electrolyte part without reducing the signal quality. Dry electrodes can be categorised into three types; metal electrode, textile electrode, and polymer electrode. Among these three, this study focuses on developing dry polymer electrodes due to its flexibility and wearability in ECG performance. Although the polymers are widely used in the sensor field, but the electrical conductivity of polymer is low to achieve the requirement of ECG sensors. In order to achieve high electrical conductivity of polymer, a conductive filler is used. Conductive filler reinforces with the polymer matrices known as polymer composite.

Previous studies have shown that the polymer composite is the most prominent feature of carbon nanotubes based devices, especially in wearable medical sensors [8]. The characteristics of carbon nanotube are high aspect ratio, high electrical conductivity, and large surface area. However, carbon nanotube materials have an issue of dispersing with polymer matrix. Thus, it is essential to disperse the carbon nanotube with polymer matrix in an efficient method. In conclusion, the type of dry electrodes, material challenges, an efficient dispersion method and electrical characterisations are discussed in this study.

1.2 Problem Statement

Many polymer composites have been produced to encounter the demand in flexible and wearable devices to the human skin. Multi-Walled Carbon Nanotube (MWCNT) is a highly conductive material to develop conductive polymer composite. However, due to the high aspect ratio and strong Van der Waals forces, MWCNT seems inclined to bundling, aggregation and agglomeration [9].

Previous studies showed that the exfoliation of MWCNT and polymer in surfactant liquid could improve the bundles of MWCNT into individual tubes. Many dispersion methods have been explored recently, including the melt mixing method, in-situ polymerisation and solution mixing method. Studies found that solution mixing methods are more facile in fabrication and low cost.

In the solution mixing method, the MWCNT and polymer is dispersed in the solvent agent to de-bundle the MWCNT. This also allows the linkage interaction between the conductive filler, MWCNT with polymer matrix. Polydimethylsiloxane, PDMS is the most popular and flexible polymer matrix used in manufacturing composite electrode due to its hydrophobic and easy to manipulate during polymer composite manufacturing.

However, poor dispersion may affect the electrical properties of the polymer composite. Thus, the physical mixing method such as shear mixer, sonication or mechanical stirring can enhance the proper solution mixing method to achieve a conductive polymer composite. The dispersion of MWCNT/PDMS composite is investigated using electrical characterisation to produce conductive MWCNT/PDMS composite.

1.3 Objectives

The research objectives are:

(a) To fabricate the MWCNT/PDMS composite electrode by using solution mixing method.

(b) To optimize the concentration of MWCNT inside PDMS to produce conductive electrode.

(c) To characterise electrical properties of the fabricated MWCNT/PDMS composite.

1.4 Scopes

This research is primarily concerned with the operation of dry electrode fabrication and characterisation. The research scope is limited to fabricating the conductive polymer composite using the Multi-Walled Carbon Nanotube (MWCNT) as conductive filler and polydimethylsiloxane (PDMS) polymer matrix. The facile solution mixing technique disperses MWCNT in PDMS using toluene as a solvent agent to obtain a better composite mixture as an electrode. This technique consists of a sonication process and a mechanical stirring process together to fabricate conductive polymer composite.

The concentration of MWCNT varies from 2 wt% to 10 wt% of MWCNT/PDMS composite. The MWCNT/PDMS composite was fabricated in three different diameters; 1 cm, 2 cm and 3 cm. The thickness of the electrode is 0.1 cm similar to the conventional electrode thickness. Raman Spectroscopy analysis is used to characterise the dispersion quality of MWCNT in PDMS. The electrical properties of the fabricated MWCNT/PDMS composite electrodes are studied through I-V measurement.

1.5 Thesis Outline

This subsection of introduction provides the work frame of this research project. The research project is divided into five chapters.

Chapter 1 explains theoretically about electrical system occurred in the heart and clinical tools used. The motivation, problem, aim of the research and scope also stated in this chapter.

Chapter 2 briefly describes the literature review of past research related to the research project. All subtopics will be discussed and reviewed in detail.

Chapter 3 holds the methodology part where the chemical preparation and fabrication process of the fabricated electrode are explained. The measurement analysis also included in this chapter.

Chapter 4 is about result and discussion of the fabricated electrode. The results obtained are then discussed in this chapter thoroughly.

Lastly, Chapter 5 is about summary of the project regarding the research conducted and recommendation of this study for future work.

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LIST OF PUBLICATIONS

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