

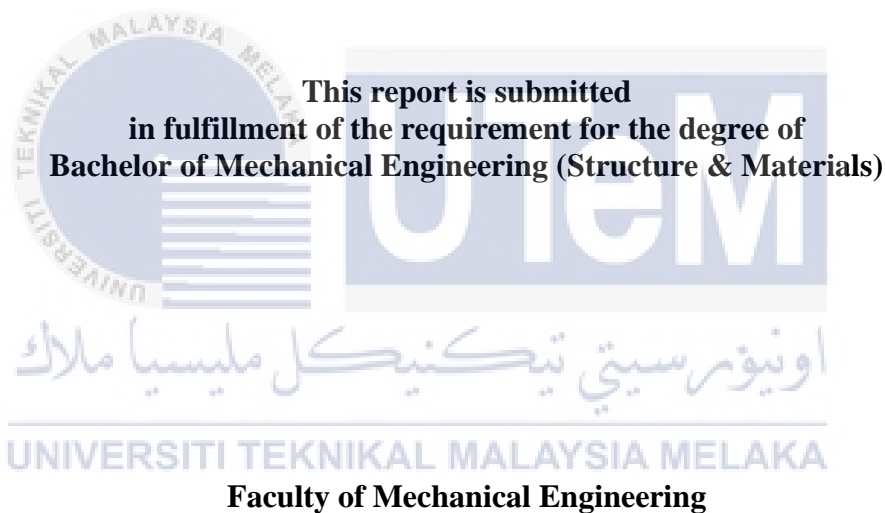
**THE PERFORMANCE OF STRETCHABLE CONDUCTIVE INK (SCI) UNDER  
MECHANICAL FACTOR**



**UNIVERSITI TEKNIKAL MALAYSIA MELAKA**

**THE PERFORMANCE OF STRETCHABLE CONDUCTIVE INK (SCI)  
UNDER MECHANICAL FACTOR**

**MUHAMMAD IZWAN BIN ABDUL RAHIM**



**UNIVERSITI TEKNIKAL MALAYSIA MELAKA**

**JANUARY 2022**

## DECLARATION

I declare that this project report entitled “The Performance of Stretchable Conductive Ink (SCI) Under Mechanical Factor” is the result of my own work except as cited in the references

Signature : .....

Name : .....

Date : .....



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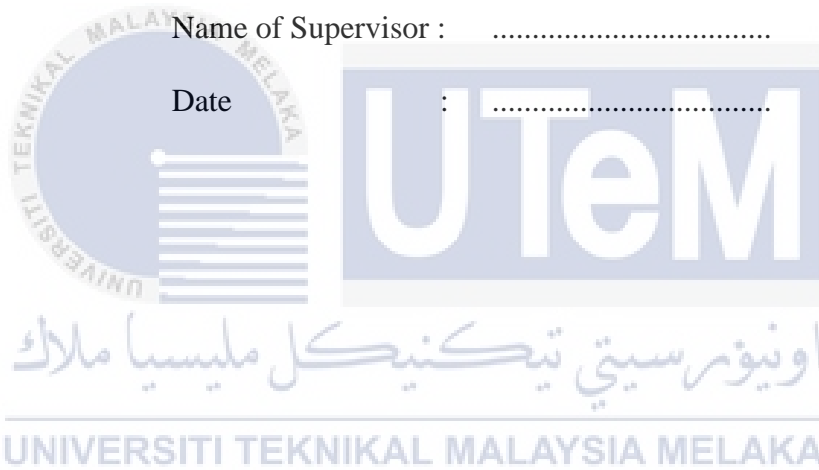
## APPROVAL

I hereby declare that I have read this project report and in my opinion this report is sufficient in terms of scope and quality for the award of the degree of Bachelor of Mechanical Engineering (Structure & Materials).

Signature : .....

Name of Supervisor : .....

Date : .....



## DEDICATION

To my beloved mother and father



## ABSTRACT

Graphene Nanoplatelets (GNPs) are excellent electrical and mechanical fillers for conductive polymers. However, the GNP size might affect the conductive polymer's conductivity and reliability, especially when it is subjected to various types of loading. This study shows the effect of GNP particle size on conductivity and reliability of conductive polymer composites when subjected to mechanical fatigue stress through the stretch test. In this work, two types of GNP filler sizes are considered which is the 5 $\mu$ m and 15 $\mu$ m with a mixed of surfactant. Following the manual cyclic stretch test, the results show that the resistivity increases as the number of cycles increases due to cracks' formation. The zero reading of bulk resistivity of 5 $\mu$ m and 15 $\mu$ m particles size was obtained when the stretch reached approximately 13 cm to 14 cm in length, which is 225% to 250% of the strain percentage. The applicable elongation of both particle sizes to withstand longer with better bulk resistivity is 125% with an elongation of 0.5 cm from the initial length of 4.0 cm. 15 $\mu$ m. Other than that, it was found that the peel strength of 15 $\mu$ m GNP which is 0.00419 N/mm is better compared to 5 $\mu$ m GNP which is 0.00328 N/mm. The surface morphology of each particle was described as a result of its bulk resistivity after being stretched through the cycles.

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## ABSTRAK

*Graphene nanoplatelets (GNPs) adalah pengisi elektrik dan mekanikal yang sangat baik untuk polimer konduktif. Walau bagaimanapun, saiz GNP mungkin menjejaskan kekonduksian dan kebolehpercayaan polimer konduktif, terutamanya apabila ia tertakluk kepada pelbagai jenis beban. Kajian ini menunjukkan kesan saiz zarah GNP pada kekonduksian dan kebolehpercayaan komposit polimer konduktif apabila tertakluk kepada tekanan keletihan mekanikal melalui ujian regangan. Dalam kajian ini, dua jenis saiz pengisi GNP dipertimbangkan iaitu 5 $\mu$ m dan 15 $\mu$ m dengan surfaktan. Berikutan ujian regangan kitaran manual, keputusan menunjukkan bahawa rintangan meningkat apabila bilangan kitaran meningkat disebabkan oleh pembentukan retak. Bacaan sifar rintangan pukal 5 $\mu$ m dan 15 $\mu$ m zarah saiz diperolehi apabila regangan mencapai kira-kira 13 cm hingga 14 cm panjang, iaitu 225% hingga 250% daripada peratusan terikan. Pemanjangan yang dibenarkan bagi kedua-dua saiz zarah untuk bertahan lebih lama dengan rintangan yang lebih baik adalah 125% dengan pemanjangan 0.5 cm dari panjang awal 4.0 cm. Selain itu, didapati bahawa kekuatan lekatan 15 $\mu$ m GNP iaitu 0.00419 N/mm adalah lebih baik berbanding 5 $\mu$ m GNP iaitu 0.00328 N/mm. Morfologi permukaan setiap zarah digambarkan sebagai hasil daripada rintangan pukal selepas diregangkan melalui kitaran.*

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## ACKNOWLEDGEMENT

Alhamdulillah, all praises to Allah for His blessing for me to complete this Projek Sarjana Muda (PSM) 2. First and foremost, I would like to express my sincere appreciation and sincere gratitude to my supervisor, Dr. Nadlene Binti Razali for essential supervision, support, and encouragement towards the completion of this experimental and report writing.

I would also like to express my deepest gratitude to postgraduate students in Advance Materials Characteristic Laboratory (AMCHAL), Andee Faeldza Bin Dziaudin and the other members in AMCHAL LAB for their assistance, time spent and effort in lab and work analysis. My sincere appreciation also extends to all AMCHAL's staff for their support and invaluable experience and memorable research atmosphere during my last year.

My sincere gratitude to my parents and my friends for their unconditional love, continuous support, encouragement and prayers throughout this study. Lastly, thank you to everyone who had been to the crucial parts of the realization part of this project directly or indirectly. Thank you.



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## LIST OF ABBREVIATIONS

PCB	Printed Circuit Board
FPCB	Flexible Printed Circuit Board
SCI	Stretchable Conductive Ink
CNT	Carbon Nanotube
CF	Carbon Fibre
CB	Carbon Black
SWCNT	Single-Wall Nanotubes
MWNT	Multi-Walled Carbon Nanotube
PFC	Printed Flexible Circuits
PET	Polyethene Terephthalate
TPU	Thermoplastic Polyurethane
GNP	Graphene Nanoplatelets
PEDOT:PSS	poly(3,4-ethylenedioxythiophene) polystyrene sulfonate
EG	Mono Ethylene Glycol
DMSO	Dimethyl sulfoxide
TX	Triton X-100

# CHAPTER 1

## INTRODUCTION

### 1.1 Background

Recently, the stretchable conductivity ink of electronic applications has gained significant attention from electronic industries. However, a conventional printed circuit board (PCB) has reached its limitation due to a rigid structure applied at a flexible texture. Therefore, the researcher actively explores the replacement. It might be flexible printed circuit boards (FPCB) made of polymer materials resistant to corrosion, moisture, lubricants, temperature, and impact.

At present, customers will purchase a limited range of stretchable electronics products in the market. Their prices remain incredibly high but not fully stretchable and have a standard hard module. Furthermore, these devices have a few issues on water resistance and product costing. However, FPCB technology provided designers with unprecedented levels of flexibility, but it is still a plastic foil that can only adhere to basic surface topographies and reaches its limitations when stretched. (Vieroth et al., 2009)

In this project, stretchable conductive ink is developed subjected to different particle size. The samples will then undergo a stretch test to assess the behaviour of the mechanical, physical, and electrical properties of SCI. A great achievement from these initiatives has the ability to alter our perception of electronics, transforming it from rigid, planar chips to flexible, curvilinear sheets. (Rogers et al., 2010).

## 1.2 Problem Statement

Conductive inks are composed of conductive fillers, binders, and solvents that give electrical conductivity. The inks are produced with a specially formulated rheology to provide the best performance for a specific printing process. Conductive ink is a versatile material that may be used in various applications, including printed and flexible electronics. Therefore, they may be used to manufacture low-cost and high-performance electronic devices. Conductive ink's components are non-toxic and environmentally safe.

Stretchable Conductive Ink is important for the development of stretchable electronics. The ink should be capable of being strained to at least 20% of its original length for at least 500 cycles without increasing its resistance by more than 30 times its original value while retaining electrical and mechanical integrity. (Mohammed & Pecht, 2016). This is a good approach for this innovative technology because 20% stretchability can suit many current requirements.

In future, the conventional PCB will face the problem due to the flexibility and stretchability of their rigid components. Thus, the SCI development is a great replacement as the alternative way to overcome their flexibility and stretchability. However, to achieve the better performance of SCI, a few problems are still discovered such as conductivity, hydrophobicity and hardness that may be affected during making it stretchable. In addition, the SCI will undergo a few tests to evaluate its reliability on the mechanical factor to endure the mechanical deformation. The printed screen is used along this experimental to reduce the cost of making SCI in large quantity.

### 1.3 Objective

1. To evaluate the effect of the stretch test on the mechanical and electrical performance of Graphene nanoplatelets (GNPs) on TPU substrate.
2. To investigate the effect of adhesion properties of Graphene nanoplatelets (GNPs) on the substrates subjected to different particle size

### 1.4 Scope of Project

This project focused more on the manual stretching test experiment to determine the effect of resistivity of SCI for the maximum strain and the applicable elongation stretch with varying particle GNPs size between 5 $\mu$ m and 15 $\mu$ m. The surface morphology of both particle sizes is analysed by using the optical microscope before and after undergoing the multiple manual cyclic stretch tests. The peel test is also performed to evaluate the effect of adhesion properties that may be affected the SCI performance.

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## CHAPTER 2

### LITERATURE REVIEW

#### 2.1 Electronic Packaging

The semiconductor electronics industries work very hard to improve electronic packaging production and assembly. However, during the early stages of the electronic packaging industry, most connection materials used were made of lead (Pb), which is a highly hazardous substance. (Chew et al., 2014). An electronic packaging refers to an electronic device that protects the electronic and electrical system components from each other and the environment. Thus, the stretchable interconnects enable the overall electronic system to withstand large deformation in order to help improve the performance of printed circuit board. In addition, electronic packaging gives protection to mechanical, chemical or electromagnetic components and interconnections (Li, Y. 2007).

#### 2.2 Conductive filler

Conductive filler is a material used to make conductive ink, which is electrically conductive to SCI. In general, the electrical conductivity of a conductive filler is determined by the type of filler and aspect ratio of the conductive filler materials. Thus, a highly conductive filler is added to the composite matrix to form a three-dimensional network of filler particles throughout the component. This critical state is known as the percolation threshold. (Sandler et al., 2003). Percolation theories

are widely used to describe the transitions from insulator to conductor in a continuous conductive network via a composite which is consisting of a conductive filler and an insulating matrix. It has been demonstrated that increasing the aspect ratio of the conductive filler reduces the value of the percolation threshold. (Untereker et al., 2009). Therefore, to obtain high conductivity, the filler concentration must be equal to or higher than the percolation critical concentration.

There are two types of the metal filler used in SCI which is metal and non-metals based. Non-metals based consist of carbon based and conducting polymer. Both types of non-metal can conduct electricity. However, Carbon-based materials are widely used for SCI due to better electrical and mechanical properties compared to polymer-based materials. Table 2.1 shows the conductivity of metal and carbon fillers. Carbon filler consists of carbon fibre (CF), carbon black (CB) and carbon nanotube (CNT) while metallic filler consist of metal flakes, metal powder, metal nanowire and metal coated fibre.

Table 2.1: The Conductivity of Metal and Carbon Fillers (Huang et al., 2019)

Filler type	Electrical conductivity (S/cm)	Thermal conductivity (W/mK)	Density (g/cm <sup>3</sup> )
Aluminium	$3.538 \times 10^5$	234	2.7
Copper	$5.977 \times 10^5$	386-400	8.9
Silver	$6.305 \times 10^5$	417-427	10.53
Nickel	$1.43 \times 10^5$	88.5	8.9
CNTs	$3.8 \times 10^5$	2000-6000	2.1
CF	$10^2-10^5$	10-1000	1.5-2.0
Graphene	6000	4000-7000	1.06
Graphite	$10^4$	100-500	2.25
Aluminium nitride	$<10^{-13}$	100-319	3.235
Boron nitride	$10^{-14}$	185-400	2.27

Table 2.2: Classified of Metal Based and Carbon Based

Based Material	Metal	Carbon
Filler Type	Silver Aluminium Copper	Graphene CNTs Graphite

### 2.2.1 Metal Based Materials

Conductive ink that made up of metal-based material known as metal fillers. Commonly, the fillers are combined together with polymer binders, and solvents, which typically include volatile solvents and non-volatile organic polymers. Conductive fillers that offer conductivity and polymer binders give physical and mechanical properties for conductive ink such as adhesion, stretchability, and influence strength.

#### 2.2.1.1 Silver

Silver-based conductive ink is a metal filler that is commonly used in printed electronic applications to generate a conductive electronic line. Silver is considered a potential replacement of conductive filler due to its strong electrical ( $6.3 \times 10^7 \text{ S m}^{-1}$ ) and thermal conductivity ( $429 \text{ W m}^{-1} \text{ K}^{-1}$ ). (Gao et al., 2011). Besides from excellent charge transport properties, conductive ink for ink-jet and printing applications must also meet general ink requirements such as high conductivity, low resistivity, low viscosity, high chemical stability, low temperature process ability, and surface tension. (D. Y. Wang et al., 2015). Furthermore, the electrical and mechanical properties of silver particles ink are highly dependent on their size, shape, and chemical treatment. Silver nanoparticles with spherical, cubic, and flake-like morphologies exhibit unique characteristics. In comparison to silver nanoparticles with a spherical form, provides greater advantages such as a bigger contact area and superior electrical properties. (Sun et al., 2003)

### 2.2.1.2 Copper

Copper is a reddish element that has a shiny metallic. It also ductile and malleable materials. Copper base alloys with high electrical or thermal conductivity have gained popularity in a wide variety of electrical applications. In term of thermal conductivity, copper has good thermal conductivity as high of  $\lambda=380W/m.K$ . However, it has poor mechanical properties which the tensile strength is below 225MPa. (Coddet et al., 2016). Besides, copper has its disadvantages due to instability against oxidation under ambient temperature and it tends to form an insulating layer of copper oxide. Furthermore, the formation of copper oxidation generates poor conductivity in sintered copper films and interferes with the sintering of copper particles. (Coddet et al., 2016).

### 2.2.2 Carbon Based Material

Carbon-based conductive inks have acquired significant appeal in printed and electronic packaging applications during the last few years due to their low cost, environmental compliance, and lower assembly temperature. The carbon-based materials include carbon black (CB), carbon fibres (CF), graphite, carbon nanotubes (CNTs), and graphene.

#### 2.2.2.1 Carbon Nanotube (CNT)

Carbon nanotube (CNT) is a nano particle conductive filler material with excellent electrical conductivity, lightweight, high strength modulus, and free oxidation. CNTs have a basic structure that can be referred to as single-wall nanotubes (SWCNT) with a single layer rolled tube. Besides, another form of common CNT is multi-walled nanotube (MWCNT), which is rolled into a multi-layer tube. SWCNT

has better electrical properties than MWCNT. Due to its excellent electrical properties, it is suited for use as connecting materials, which have formed the basis for producing modern electronics. (Ayatollahi et al., 2011) Stretchable conductors like CNT were embedded within elastomers making them stretchable. A Carbon Nano Tube CNT tube on polyurethane can handle up to 400% strain (Lee et al., 2012).

#### **2.2.2.2 Graphene**

Graphene is a hexagonal crystalline single layer of graphite. Graphene exists in various forms such as graphene nanoplatelets, nano-sheets and 3D graphene. Among these, graphene nanoplatelets are widely used in electronics applications due to their pure graphitic composition which is excellent in electrical and thermal conductors. Graphene nanoplatelets come in black or grey powder.

Graphene inks recently have dramatically improved flexible print electronics since they are cheap, easy to manufacture, more flexible and have a greater conductivity. Graphene is essentially a single atomic layer of graphite, a common material that is an allotrope of carbon composed of extremely closely linked carbon atoms organised in a hexagonal lattice. Graphene is so special in its sp<sup>2</sup> hybridisation and very thin atomic thickness of 0.345nm. (Mevold et al., 2015). This allowed graphene to surpass so many records for strength, electricity, and heat conduction.

### 2.3 Polymer Binder

The polymer binder phase is essential for the stretchability of the stretchable conductive inks. In general, the polymer binder in SCI serves to give strong adhesive strength between conductive ink and substrates while also providing stretchability in the sintered form.(Hsu et al., 2013). Furthermore, the adhesive connection must be able to resist the device's whole operational temperature range due to exposed environmental factors during operation.(Hsu et al., 2013).

Thermosetting and thermoplastic resins were frequently utilised as matrix in polymer conductive composites. Polypropylene (PP), polyethylene (PE), and polystyrene are examples of thermoplastic resins (PS). Polypropylene is widely available due to its superior mechanical qualities, great resistance to heat, low cost, ease of processing, and fully recyclability, meanwhile thermosetting resins include epoxy resin, vinyl-ester, and polyester. (Alemour et al., 2018).

Both thermosetting plastics and thermoplastics react differently when heated. Thermoplastics can melt when exposed to heat after curing, whereas thermoset plastics retain their shape and remain solid when exposed to heat once cured. (AlMaadeed et al., 2020). Thermoplastic and thermosetting resins are electrically insulating, with very low electrical conductivity values. (Alemour et al., 2018).

### 2.3.1 Epoxy Resin

Epoxy that acts as a binder consists of two parts: resin and hardener mixed to cause it to cure. Epoxy resin is the most suitable polymer binder for conductive inks due to its viscosity properties. It is commonly used in demanding applications because of its strong chemical and corrosion resistance, good adhesive qualities, low shrinkage, and inexpensive cost. (Chatterjee et al., 2012)

In this study, the Araldite 506 Epoxy Resin (Bisphenol A-epichlorohydrin) has a density of 1.168g/ml and a viscosity at 500-750 mPa.s 25°C was used as a polymer binder. Epoxy resin is manually mixed up with the Hardener JeffAmine D-230 with 63.75% and 21.25 %, respectively for about 1 minute. A hardener is a solvent applied to the ink mixture to cause it to harden and provide a more durable ink and a curing agent for epoxy. The hardener has a vital role in epoxy water absorption. (Wu et al., 2006).

### 2.3.2 PEDOT: PSS

PEDOT: PSS is an advanced conducting polymer with high conductivity, excellent electrochemical properties, and low redox potential. Although this polymer is fully organic and non-metal, it could conduct electricity very well. In addition, the other advantage of PEDOT: PSS is used in a wide variety of electronic applications because of its water dispersibility, excellent visible-light transparency, high stability in ambient temperature, mechanical flexibility, and good solubility. (Netnapa et al., 2017). However, Netnapa also states that the more layer of conductive ink, the lower sheet resistivity. The composite contains 1.6 wt.% PEDOT: PSS has a high stretchability of up to 340% but an extremely poor conductivity. (Yang et al., 2020).

When PEDOT: PSS is mixed with filler such as graphene or CNT, it could optimise the electrical, mechanical, thermoelectric and other properties. (Yang et al., 2020)

#### **2.4 Factor Affecting Mechanical and Electrical Properties**

Some of the most important factors that can affect the sheet resistance of conductive ink are the curing conditions of the ink, the viscosity of the ink, and the filler content of the ink. (S Merilampi & Ruuskanen, 2009). The other filler properties, such as particle size, might affect the electrical conductivity. It has been demonstrated that smaller particle sizes reduce the percolation threshold for spherical particles. (Zaremba & Smoleński, 2000). Besides, it has been proven that having a greater aspect ratio (length to diameter ratio) and a wider range of aspect ratios reduces the percolation threshold. (Zaremba & Smoleński, 2000). Electrical properties are affected by the composition and amount of polymer matrix, as well as the particles (size, amount, shape, distribution, and orientation).

In addition, the surface properties of the filler and polymer also have a significant effect on the composite's conductivity and percolation threshold. Both the filler and the matrix have free surface energies, and the difference between their surface energies determines how well a polymer wets a filler surface. The polymer wets the filler better when the surface energies are closer together. (E. P. Mamunya, 1997). Therefore, improving the filler's wetting can improve its dispersion inside the matrix material. Thus, the composite's percolation threshold may increase, improving the composite's overall conductivity. For better electrical conductivity, a lower surface energy difference between the filler and the polymer is preferable.