

**SCHOOL OF MATERIAL AND MINERAL RESOURCES**

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**STRETCHABLE CONDUCTIVE INK USING COPPER FILLERS**

By

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## DECLARATION

I hereby declare that I have conducted, completed the research work and written the dissertation entitled “**Stretchable Conductive Ink Using Copper Fillers**”. I also declare that it has not been previously submitted for the award of any degree or diploma or another similar title of this for any other examining body or university.

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## **LIST OF ABBREVIATIONS**

<b>UV</b>	Ultraviolet
<b>XRD</b>	X-Ray Diffraction
<b>SEM</b>	Scanning Electron Microscope
<b>DSC</b>	Differential Scanning Calorimetry
<b>TGA</b>	Thermogravimetric Analysis
<b>PDMS</b>	Polydimethylsiloxane
<b>PI</b>	Polyimide
<b>PDMS-OH</b>	Hydroxy Terminated Polydimethylsiloxane
<b>GPTMS</b>	3-Glycidyloxypropyl Trimethoxysilane
<b>VTMOS</b>	Vinyltrimethoxysilane
<b>D4</b>	Octamethylcyclotetrasiloxane
<b>DTG</b>	Differential Thermal Gravimetric
<b>PU</b>	Polyurethane

## LIST OF SYMBOLS

<b>S/m</b>	Siemens Per Meter
<b>%</b>	Percentage
<b>%T</b>	Percentage of Transmittance
<b>T<sub>g</sub></b>	Glass Transition Temperature
<b>T<sub>m</sub></b>	Melting Temperature
<b>T<sub>d</sub></b>	Degradation Temperature
<b>MPa</b>	Mega Pascal
<b>mPa.s</b>	Milli Pascal Second
<b>W /Mk</b>	Watts Per Meter-Kelvin
<b>1/K</b>	Per Kelvin
<b>Ωcm</b>	Ohm Centimetre
<b>Nm</b>	Nanometre
<b>Kn</b>	Kilo Newton
<b>Mg</b>	MilliGram
<b>°C</b>	Degree Celsius
<b>Min</b>	Minute
<b>ml</b>	Milli Litre
<b>G</b>	Gram
<b>μl</b>	Micro Litre
<b>wt%</b>	Weight Percent

# **DAKWAT KONDUKTIF MERENGGANG MENGGUNAKAN PENGISI**

## **TEMBAGA**

### **ABSTRAK**

Kajian ini membincangkan dakwat konduktif merenggang menggunakan pengisi tembaga. Pada masa kini, banyak aplikasi yang melibatkan dakwat konduktif merenggang menggunakan pengisi perak. Pengisi perak adalah salah satu daripada dakwat konduktif biasa dan popular. Walau bagaimanapun, disebabkan kos yang tinggi, ia menghadkan penggunaan dalam aplikasi yang lebih luas. Tembaga pengisi mempunyai kekonduksian yang setanding dengan pengisi perak. Justeru itu, pengisi tembaga telah dipilih sebagai pengisi konduktif dalam projek ini. Beberapa pengubahsuaian diperlukan kerana kecenderungan untuk tembaga teroksida dalam suasana persekitaran. Menggabungkan pengisi perak adalah satu kaedah untuk meningkatkan kekonduksian dakwat konduktif merenggang menggunakan pengisi tembaga. Dakwat konduktif merenggang menggunakan pengisi tembaga diuji ciri-ciri dengan UV ternampak pemindahan, XRD, SEM dan ujian tegangan. Dalam usaha untuk mengkaji sifat-sifat haba, DSC dan TGA dijalankan. Kekonduksian dakwat konduktif merenggang diukur dengan multimeter. Didapati bahawa kekonduksian dakwat konduktif merenggang meningkat apabila pengisi tembaga ditingkatkan tetapi kecenderungan yang lebih tinggi juga untuk pengisi tembaga untuk mengoksidakan seterusnya mengurangkan kekonduksian.

# **STRETCHABLE CONDUCTIVE INK USING COPPER FILLERS**

## **ABSTRACT**

This research focused on the stretchable conductive ink using copper fillers. Nowadays, a lot of application which involves stretchable conductive ink using silver fillers. The silver fillers are one of the common and popular conductive ink. However, due to its high cost, it limits the usage in the wider application. Copper fillers have the comparable conductivity with silver fillers. So, the copper fillers have been chosen as conductive fillers in this project. Some modification needed due to copper fillers tendency to oxidise in the ambient environment. Incorporation of silver fillers is a method to enhance the conductivity of stretchable conductive ink using copper fillers. The stretchable conductive ink using copper fillers are characterised by UV-visible transmittance, XRD, SEM and tensile testing. To study the thermal properties, DSC and TGA are conducted. The conductivity of stretchable conductive ink is measured by a multimeter. It found that conductivity of stretchable conductive ink increasing when increasing copper fillers but the higher tendency also for the copper filler to oxidise and reduce conductivity.



# CHAPTER 1

## INTRODUCTION

### 1.1. Background of Study

Numerous researches have been done towards the future generation of high degree flexibility, cheap, and easy to be manufactured printed conductive electronics devices. Thus, advanced materials have been explored to fulfil criteria for printed conductive electronic devices. The effectiveness of printed conductive depend on conductive fillers added into the mixture, which forms the electrically conductive composites that are the key material for printed conductive to be used in the fields of printed conductive electronics devices as shown in figure 1.1 (Kenneth, 2016).

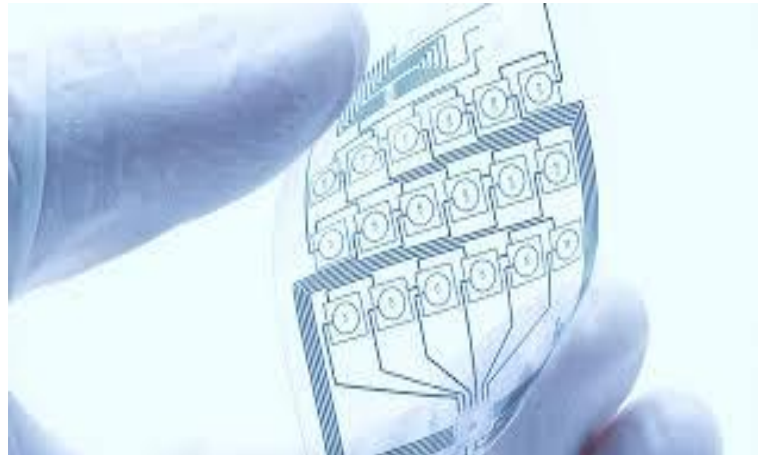


Figure 1.1: Flexible printed conductive circuit (Dupont, 2017)

Electrically conductive composite formulations were created in the efforts to develop printed conductive electrical device with the desired specifications of electrical and mechanical properties. The studies toward this application are highly unique and valuable to forming the printed conductive electronic devices, which able to alter the function of materials to form a high-quality product.



Nowadays, the electrically conductive composite has benefited the development of the applications of the strain sensor, health monitor, antenna etc. In the future, the fully wearable sensor be used in our daily lives and routines with the well-develop future electronic devices as in figure 1.2. To do so, the electronic devices must meet advance requirements and specification that involve wearability of electronic devices.

The wearable electronic device will be excellence in its quality and their ability to recognise the activity of their user as well as the situation of the user thus interprets this info to modify its system.



Figure 1.2: Wearable electronic device (Textile Industries Media Group, LLC, 2016)

On the other hand, despite their various advantages compared to the previous device, the material and technologies itself have its own weakness such as low electrical conductivity have been considered as the obstacles (Venkata, 2015).

## **1.2 Problem Statement**

The challenge of this research is how to use the copper fillers in the stretchable conductive ink to be used in the various electronic devices application. Conductive fillers, such as silver, have been studied as active fillers for the conductive fillers (Kimberly et al., 2004).

This silver filler now has been widely used as the stretchable conductive ink in a variety of applications. Silver has slightly higher conductivity than copper. However, we can say that both of their conductivity is comparable. According to Douglas (1995), the conductivity of copper is  $5.95 \times 10^{-7}$  S/m conductivity while  $6.29 \times 10^{-7}$  S/m are the conductivity for silver. Copper price also very low compared to silver. Therefore, by using copper we can cut the cost of material and still produce the stretchable conductive ink.

To use copper fillers in the stretchable conductive ink, we need to study about their structure, properties, conductivity and its morphology. We also need to study the amount and chemicals to be used in the formulation of stretchable conductive ink.

The formulation of ink needs to ensure their conductivity under the strain condition. The ink also needs to be compatible with the substrate so that the ink remains on the substrate under strain.

The outcome of this research is higher wt% of copper fillers having higher conductivity. This research will bring a lot of benefit and contribution toward many fields especially based on electrical technology application. This research also will increase the use and demand of the polymeric material which can help in our daily life application and yet to widen the option of the material available for a lot of application in the future.

### **1.3 Objectives**

The objective of this research is list below:

- To fabricate the stretchable conductive ink using copper fillers
- To study the properties of PDMS substrate with copper fillers
- To study effect of mixing copper fillers with silver fillers in stretchable conductive ink

### **1.4 Scope of Work**

This study focused on the formulation of stretchable conductive ink using copper fillers. The right and suitable formulation and method needed to make sure conductivity of ink to produce the sample that can conduct electricity using copper fillers by incorporating silver fillers. To do so, we need to perform the UV-visible spectrometer, XRD, SEM, tensile test, DSC and TGA to characterise the material while using a multimeter to characterise the conductivity of the material.

### **1.5 Dissertation Outline**

Chapter 1 discusses the printed conductive electronic and the introduction of using copper fillers in conductive ink and its application. Chapter 2 contains literature review and related research associated with this study. Chapter 3 include the methodology used for data collection for analysis. Chapter 4 provides the presentation of data collected and analysed for this study. Chapter 5 covers the conclusions for this study and recommendations for future study.

## **CHAPTER 2**

### **LITERATURE REVIEW**

#### **2.1 Overview**

In this high technology era, there are great interests and research toward flexible electronic devices especially in sports, entertainments, and medical care's application. This technology helps to improve our daily life quality (Shi Bai et al., 2016).

This electronic field has been undergoing the tremendous development by producing an intelligent electronic device that can be used in a lot of application such as heart rate monitoring, health and fitness activity tracking and so on.

According to Venkata (2015), many advantages of this technology compares to the past and present technologies for the future applications. Even though there is research being conducted in this field, much more must be done to implement them in real-time applications.

Printed electronics give huge advantages for electronic devices such as sensor by reducing material waste and production costs. This technology not only gives an alternative to reduce cost but also to the development of materials that can perform various advanced functions (Matsuhisa, et al, 2015).

Due to these benefits and advantages, electronic printing market more favourable thus increase the investment toward this market. The electronics printing market was worth USD 9.8 billion in 2012 and are predicted to grow to a value of USD 63.2 billion by 2022.

Furthermore, around 33% of all electronic devices are coming from conductive inks. This field seems to attract more company to try this market, which requires the printing of a variety of conductive patterns (Jaewon et al., 2014).

## 2.2 Stretchable substrate

A stretchable conductor with both high conductivity and stretchability is one of the key features of this stretchable electronics device. The stretchable conductor must also include two components; conductive components and stretchable substrates component as shown in figure 2.1.

Realisation of their wide applications for bending, stretching, twisting, and other complex deformation, flexible materials could be a suitable alternative as electrical conductive components (Kimberly et al., 2004).

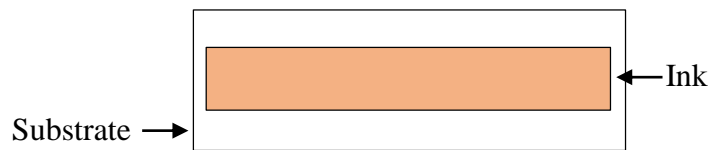


Figure 2.1: The stretchable conductive ink sample

According to Oxford (2017), flexible is defined as bending capability without deforming while stretchable is the capability to pull wider and longer without deform.

Joshua (2011) reported that many materials have been explored to maximise the performance of the applications involved and the result found that thermosetting polymer is ideal for most applications. The thermoset materials were used either as the substrate and/or matrix use to incorporate the conductive filler.

Thermosetting polymers are cured by forming a three-dimensional network that forms one large molecule. These include epoxy, polyimide, silicones, polyurethanes, etc. The advantages and disadvantages of the thermosetting polymer are shown in table 2.1.

Table 2.1: The advantages and disadvantages of thermosetting polymer (Joshua, 2011)

Material	Advantages	Disadvantages
Epoxies	Thermally stable	Long cure cycles with
	Good moisture and	anhydride hardeners
	chemical resistance	Large exothermal when
	High purity	amine-cured
Silicones	Highest purity	Low surface energy
	High and low-temperature	Low Tg
	stability	Large CTE
	Low-temperature cure	
Polyurethanes	Stretchable and flexible	
	Good flexibility at low	Lower thermal stability
	temperatures	and service temperature
	Highly versatile chemistry	(150-163 °C)
Polyimides		Moderate bond strength
	Higher temperature	Requires multi-step curing
	stability	to volatilize solvent
		May absorb moisture in
		cured condition

Takao (2012) reported that there are specific features had to be introduced to obtain a stretchable circuit as shown in figure 2.5. Firstly, instead of using the normal rigid epoxy material, an elastic material is used, allowing deformation of the circuit without damage the circuit.

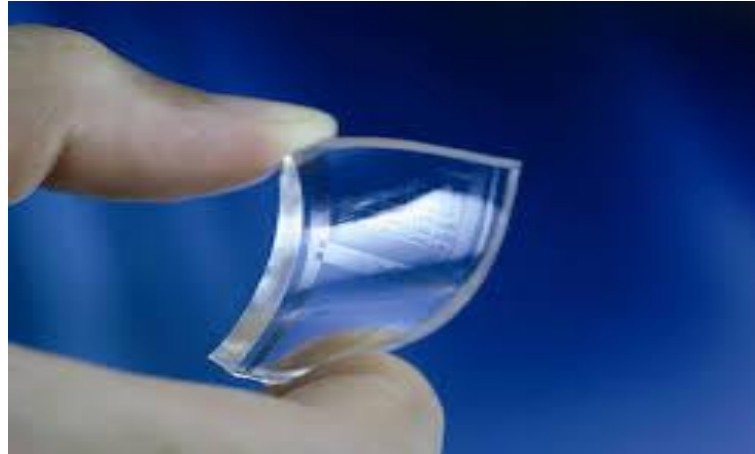


Figure 2.2: Flexible, stretchable, transparency PDMS

The main classes of substrate used are PDMS (Polysiloxane) and PUs (Polyurethanes) in figure 2.3 and 2.4 respectively.

Another important flexible substrate is PI (polyimide) which can use in this electronic device (Shi Bai et al., 2016) as shown in figure 2.5. PDMS and PU are popular to be used as substrate or matrix compare to polyimide and epoxy due to its flexibility and stretchability.

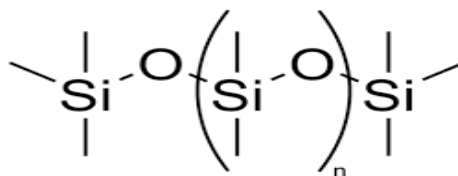


Figure 2.3: Polydimethylsiloxane chemical structure

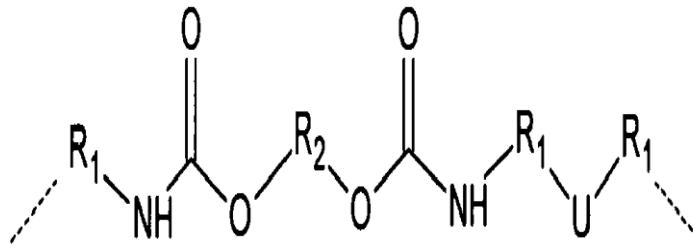


Figure 2.4: Polyurethane chemical structure

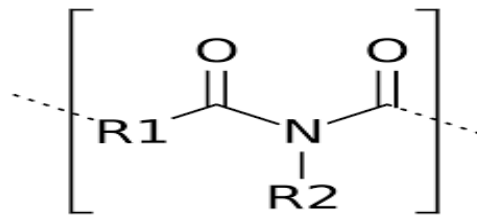


Figure 2.5: Polyimide chemical structure

In general, PDMS having the flexible Si-O bond while PU is produced from diisocyanates and polyols (hydroxyl groups). This hydroxyl group contribute to its flexibility and stretchability. The diisocyanates part contribute to its good mechanical properties. The polyols part contributes to its elastic and flexible (Kenneth, 2016).

The main advantage of stretchable material electronics is the forming of flexible, lightweight and cost-effective material. Moreover, the key attraction of flexible electronics is its ability to undergo varies deformation without affecting the performance and properties while maintaining its function (Deepalekshmi et al., 2016). In this way, electronics devices could be upgraded into clothes as an example where the deformations involved.



### **2.2.1 PDMS stretchable substrate**

PDMS is the mostly used as an elastomeric substrate for the formation of a stretchable substrate due to its unique characteristics, such as high stretchability and biocompatibility (Kimberly et al., 2004). It possesses not only high stretchability and durability but also good thermal stability, compatibility, and chemical resistance (Kimberly et al., 2004).

These interesting properties of PDMS are from its chemical structure, intermolecular and intramolecular forces. High temperature and chemical stability coming from the high bond energy of Si-O bond which is more than a C-C bond energy. Moreover, the Si-O backbone is highly flexible compared to the hydrocarbons. As the result, PDMS is one of the lowest known polymer glass transition temperatures, between -123°C to -54°C (Joshua, 2011).

Additionally, transparency and low-cost manufacturing process could also be the reason for PDMS chosen as a substrate (Kimberly et al., 2004). PDMS has low Young's Modulus, 140 to 600 kPa at the thickness of 0.6mm. Low modulus assures the PDMS film has high stretchability (Edward and Michael, 2014). The common properties of commercial PDMS elastomers are shown in Table 2.2.

Table 2.2: The common properties of commercial PDMS (Joshua, 2011)

Colour	Transparent
Resin viscosity (mPas)	3900
Shore-A hardness	50
Tensile Strength (MPa)	-
Elastic Modulus (MPa)	1.8
Thermal Conductivity (W /mK)	0.18
Thermal Expansion Coefficient (1/K)	310
Dielectric Constant	2.65
Resistivity ( $\Omega\text{cm}$ )	1.2x10 <sup>14</sup>

Generally, PDMS substrate can be obtained by adding a pre-polymer (vinyl terminated PDMS) with cross-linker such as dimethyl and methyl hydrogen siloxane in specific amount and condition. The mechanical properties of PDMS can be varied by changing its degree of the crosslink. The high the degree of crosslinking produced the stiffer material (Deepalekshmi et al., 2016).

Joshua (2011) explained in his report that cross-linked PDMS can be obtained by three typical routes: condensation reaction, addition reaction or radical reaction.

Condensation of silanol groups to form siloxanes. Cross-linkers containing silane and silicone hydrides to crosslink as shown in figure 2.6. To fasten the rate of reaction by using Tin as catalyst (Joshua, 2011). This reaction forming crosslink PDMS with higher thermally stability. However, requires a longer curing time.

Condensation curing process is cheaper and better for most applications. Condensation curing process is easy to mix and have long curing time that allowed slow and even crosslinking process (Joshua, 2011).

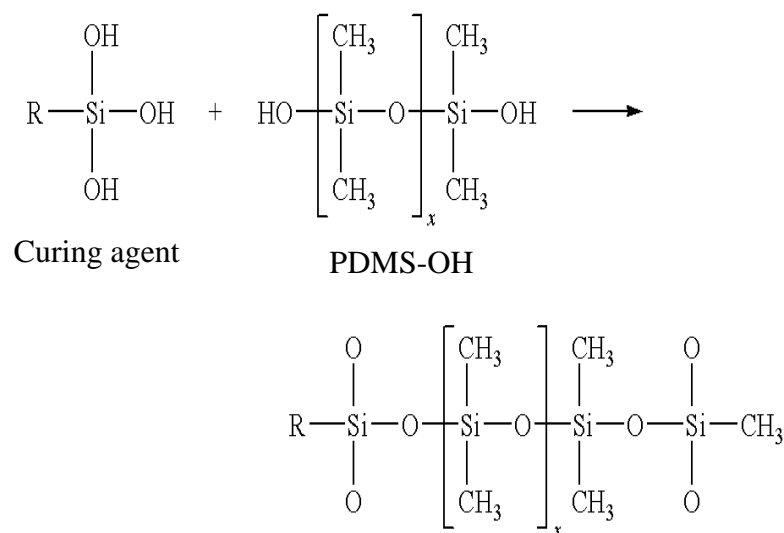


Figure 2.6: Condensation Process

Addition reaction form by combining a silicone hydride and an olefin, creating an alkylenes linkage. The mostly used olefins are vinyl silicones. The three-dimensional crosslink can occur when silicones hydrides and vinyl silicones are reacted as shown in figure 2.7. Hydrosilylation reactions are usually catalysed by platinum complexes like platinum divinyltetramethyldisiloxane as a quick reaction and need at the very low amount (Joshua, 2011).

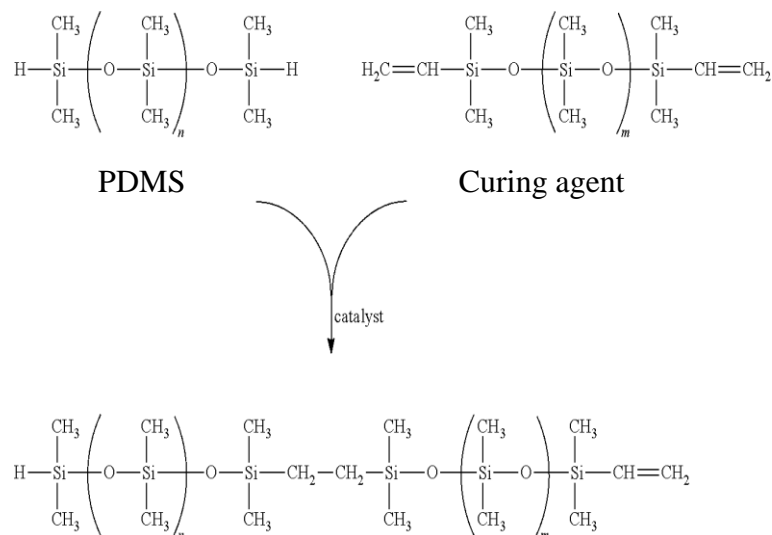


Figure 2.7: Addition Process

The major benefit of addition reaction crosslinking is no by-products produced and provides a more stable material at elevated temperatures and was developed for rapid processing and fast curing rate. This reaction of PDMS requires less mixing but requires an elevated curing temperature (Joshua, 2011).

PDMS also can be cured by peroxide catalyst as shown in figure 2.8. Curing initiates when a peroxide catalyst undergoes homolytic cleavage producing free radicals. These radicals will abstract hydrogen from a methyl group on the PDMS and undergo the reaction to form ethylenic linkages between siloxanes or react with vinyl groups on PDMS. Once the free radical is formed on or adjacent to the vinyl groups the radical can attack another vinyl or methyl group forming crosslinks (Joshua, 2011).

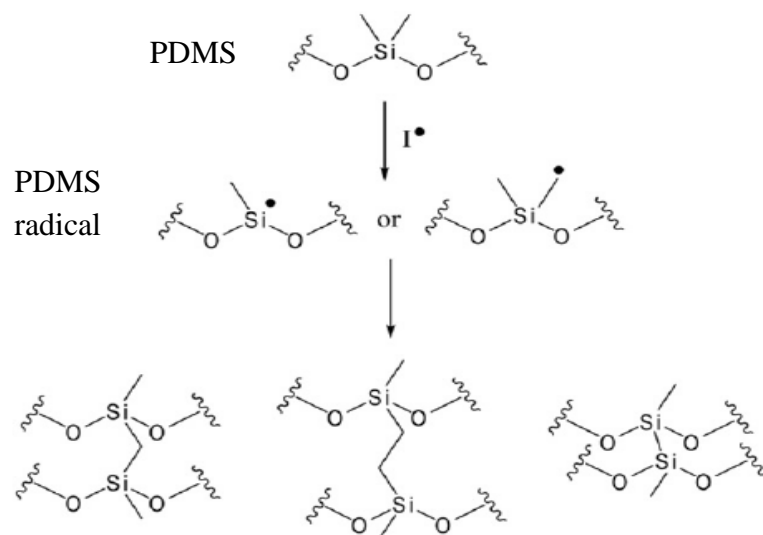


Figure 2.8: Radical Process

Termination occurs by joining of two radicals or through hydrogen abstraction from a peroxide molecule. The radical reaction can react at a broad range of temperatures depending on the decomposition temperature of the peroxide catalyst used.

This reaction is favoured by industry due to the longer shelf life and lower cost than others method. However, there is a drawback of radical reaction. Radical reaction rubbers tend to become discolour and become yellow after curing, smell bad and leave peroxide residues in the rubber (Joshua, 2011).

To produce PDMS with excellence mechanical properties, incorporation of fillers is needed. Commonly reinforcing fillers can be used to mechanical properties are silica. Studies have found that fumed silica, produced by flame annealing droplets of amorphous silica, are highly efficient to reinforce silicones than amorphous silica.

The improved reinforcing of fumed silica was closely related to its surface chemistry. In fumed silica, there are present of randomly distributed isolated hydroxyl groups that covalently bond to silicone while amorphous silica containing short polysialic acid, which cannot covalently bond to silicone (Joshua, 2011). The reinforcing capability of silica is very dependent on the surface area and bonding between fillers and polymer.

Although PDMS possesses so many beneficial properties, however, the weak adhesion between the PDMS substrate and the electrically conductive components has a severe problem to process the good and stable performance of the printed electronic device. This problem coming from its highly hydrophobic surface (Kimberly et al., 2004).

## **2.3 Stretchable conductive ink**

Printed electronics industry with applications including antennas, flexible electronics, and displays highly demanded conductive ink. To do so, expensive noble metals have been used in this conductive ink, which surely increases the cost (Ruvini et al., 2013).

Many studies have been conducted to explore a variety of materials as electrical conductive components such as conducting polymers, carbon tubes, graphite, and other metal materials (Kimberly et al., 2004).

### **2.3.1 Silver fillers**

Currently, silver fillers have been mostly developed to enable outstanding conductivity and excellent printability. Silver fillers are very popular among others metal in this application. Because silver fillers have high electrical conductivity.

The silver fillers form a silver oxide layer which good electrical conductivity, unlike many other metal oxide layers. Due to the unique characteristic, the ageing of silver fillers will not result in significantly reduced conductivity. Other benefits are it being the most conductive element that their thermal conductivity can withstand extreme temperature, anti-bacterial, etc.

A disadvantage of silver fillers is its tendency to migrate and it is relatively expensive (Xingcun, 2011). Apart from that silver is one of the most expensive elements silver also limits wide industrial application. Prices of noble metal materials, such as silver, gold and platinum, have increased in years (Venkata et al., 2015).

### **2.3.2 Copper fillers**

Researcher found low-cost processing by using copper-based conductive ink have been developed for both glass and flexible substrates (Ruvini et al., 2013). Copper has been considered as a potential alternative material for other expensive noble metals (Shi Bai et al., 2016). Copper fillers utilise the low-temperature sintering processes will reduce the costs involved with conductive patterns (Ruvini et al., 2013).

According to Venkata et al. (2015), copper is the second element with high conductivity after silver based on figure 2.9. Electrical conductive fillers are added to the polymer matrix to make the composite electrically conductive. Thus, copper fillers are a good replacement material for silver fillers because of its high electrical conductivity and low price (Jaewon et al., 2014).

Kinds of Materials	Electric Conductivity (1/ $\Omega\text{m}$ )
Aluminum	$35.3 \times 10^6$
Copper	$58.0 \times 10^6$
Gold	$41.0 \times 10^6$
Iron	$10.3 \times 10^6$
Silver	$62.9 \times 10^6$
Epoxy	$10^{-12} \sim 10^{-13}$

Figure 2.9: Electrical conductivity of elements (Lee et al., 2006)

Interestingly, copper fillers are can be used in writing. We can use a pen filled with copper fillers to design a series of copper patterns, such as lines and electrodes. This is an easy and interesting way of the fabrication process to make conductive patterns for portable applications (Chai-Yang et al., 2015).

However, this low-cost conductive filler, copper fillers easily oxidise. Once oxidised the conductivity is drastically reduced and become a poor conductor. Even if corrosion inhibitors are used, copper fillers electrical conductivity is not stable during accelerated ageing (Joshua, 2011).

This poor stability during ageing affects its thermal and electrical properties. Even with antioxidants, copper fillers will show an increase in resistivity on ageing especially under humidity condition (Xingcun, 2011). The copper oxides formed on the surface of have two negative effect which is it will increase the required sintering temperature and reduces the electrical conductivity.

Moreover, few researched reports that the thickness of the oxide layer is the factor which affects the electrical conductivity of the copper fillers obtained after sintering (Chia-Yang et al., 2015). If this oxide could be removed or changed to a conductive form, copper fillers would be very good conductors (David, 2000).



Only a few of reports have tried to explain the oxidation problem, which basically by reducing the exposure of the copper to oxygen, by a protective layer. There have been several reports presenting various approaches which help copper fillers resistance toward oxygen under ambient conditions, by coating the obtained copper fillers with a layer of capping agents to prevent aggregation and agglomeration in dispersions.

Coated copper fillers help improve conductivities by direct printing. Another way of removing the oxide is to add reducing agents such as amines. These approaches open new possibilities in printed electronics due to the very expensive cost of the silver fillers limits development application.

However, studies found it is only effective in for short-term protection against oxidation and affect the cured properties of the resin system. Even with this reducing the conductivity will reduce over time. Since copper fillers is a potentially low-cost metal compared with silver or gold, a driving force toward the development of copper fillers surface treatment for resulting in high conductivity (David, 2000).

However, replacement of silver fillers to copper fillers is not easy. So, a method to produce and handling is important to form high-quality copper fillers ink for the printed electronics application (Fan et al., 2013).

#### **2.4.3 Copper-Silver fillers**

Coated copper fillers by silver fillers show a better resistance toward oxidation process by having an electric resistivity of  $5.7 \times 10^{-3} \Omega \cdot \text{cm}$ . This can be applied in electronic industry. Higher silver fillers content coated copper fillers increase adhesives stability and reduce the resistance between metal fillers (Zhou, 2017).

Hiroshi (2010), found the electrical resistivity of conductive ink using a silver-coated copper fillers having lower and much stable compared to uncoated copper fillers. The resistivity of conductive ink of pure copper fillers was highly influenced by the copper oxide layer on the ink surface. The silver helps to form great resistance toward oxygen for copper.

The researchers tried to investigate the use of silver coated copper fillers as the way to reduce cost while maintaining its properties (Siyuan et al., 2012). Moreover, the characterizations of the copper fillers on the flexible substrate have not been well reported.

Thus, we need to understand the microstructure of the copper fillers ink on a flexible substrate to fully develop the potential in copper fillers and further technological advancements in flexible printed electronics (Jaewon et al., 2014).

#### **2.3.4 Factor affecting good conductivity**

Conductivity is usually determined by measuring resistivity which is given by

$$\text{Conductivity} = L/RA \quad (1)$$

Where

R is the measured resistance between two points,

A is the cross-sectional area through which the current flows

L is the distance between the two points (David, 2000).

The shape and size of the conductive fillers play a crucial role in determining the conductivity of ink itself. As the size of the fillers decrease it becomes highly more difficult to prevent aggregation and agglomeration of the conductive fillers. The benefits using the high aspect ratio fillers are not able to solve this processing challenges which will cause these particles to melt and agglomerate (Tengyuan, 2013).

To reduce this problem in the dispersion of the conductive fillers, surfactants which typically fatty acids will act as compatibilizers between the conductive filler and the polymeric matrix (Joshua, 2011).

Next, the concentration of fillers which the polymer composite becomes conductive is referred to as the percolation threshold. Percolation is defined as the fillers loading when the conductivity of the composite dramatically increases until eventually reaching a plateau (Tengyuan, 2013).

At this point, conductive fillers network is formed. This allowed the mobility of charge of the fillers through the matrix and able form certain degree of electrical conductivity (Teh et al., 2011). So, to make materials with high conductivity, high loading level of conductive fillers needed.

The high fillers loading, however, not only impacts deformability, manufacturing, surface finish, and limit the ability to maintain desired conductivity at under deformation but also reduce the mechanical properties of the materials (Vikas, 2010).

Walter et al. (2009) found that the higher fillers loading has increased the viscosity of ink which will influence the processing and mechanical properties. The conductive fillers will improve the conductivity and incorporate of conductive fillers at high concentration will affect adhesion by forming aggregation (Chorbadjiev and Kotzev, 1988).

The conductive fillers must be able to conduct electrical conductivity by increasing crosslinking density of matrix (Siyuan et al., 2012). High crosslink density will reduce the inter fillers distance and enhance the conductivity. At higher fillers loading, the mobility of molecular is restricted due to the interaction between polymer and fillers and reduce stretchability (Mansour, 2008).

Under strain, the polymer matrix composite break the three-dimensional crosslink formed and causing the loss of the contact between fillers and the increase in the inter fillers distance (Sari et al., 2010).

## **CHAPTER 3**

### **METHODOLOGY**

#### **3.1 Overview**

This chapter will be discussing the steps involved in the research study which explained the flow of this research. This chapter also presents the preparation of the stretchable substrate and stretchable conductive ink. The procedures of the research stretchable conductive ink are well explained in this chapter.

#### **3.2 Materials**

All the materials were used directly as supplied without any modification or treatment mode.

Poly (dimethylsiloxane) hydroxyterminated (PDMS-OH), fume silica, dibutyltin dilaurate, 3-Glycidyloxypropyl trimethoxysilane (GPTMS), silver filler, vinyltrimethoxysilane (VTMOS) used in this researched is supplied by Sigma-Aldrich Sdn. Bhd.

Toluene is supplied by J.T. Baker Chemicals. Octamethylcyclotetrasiloxane (D4) used in this researched is supplied by Penchem Technologies Sdn. Bhd. The copper filler used in this researched is supplied by Merck Sdn. Bhd. and acetic acid used in this researched is supplied by Qrec (Asia) Sdn. Bhd.

#### **3.3 Research Methodology Flow Chart**

This section will highlight all the work done during the study and the experiment conducted in the laboratory, School of Material and Mineral Resources Engineering, USM. The experimental work flowchart is illustrated in figure 3.1.

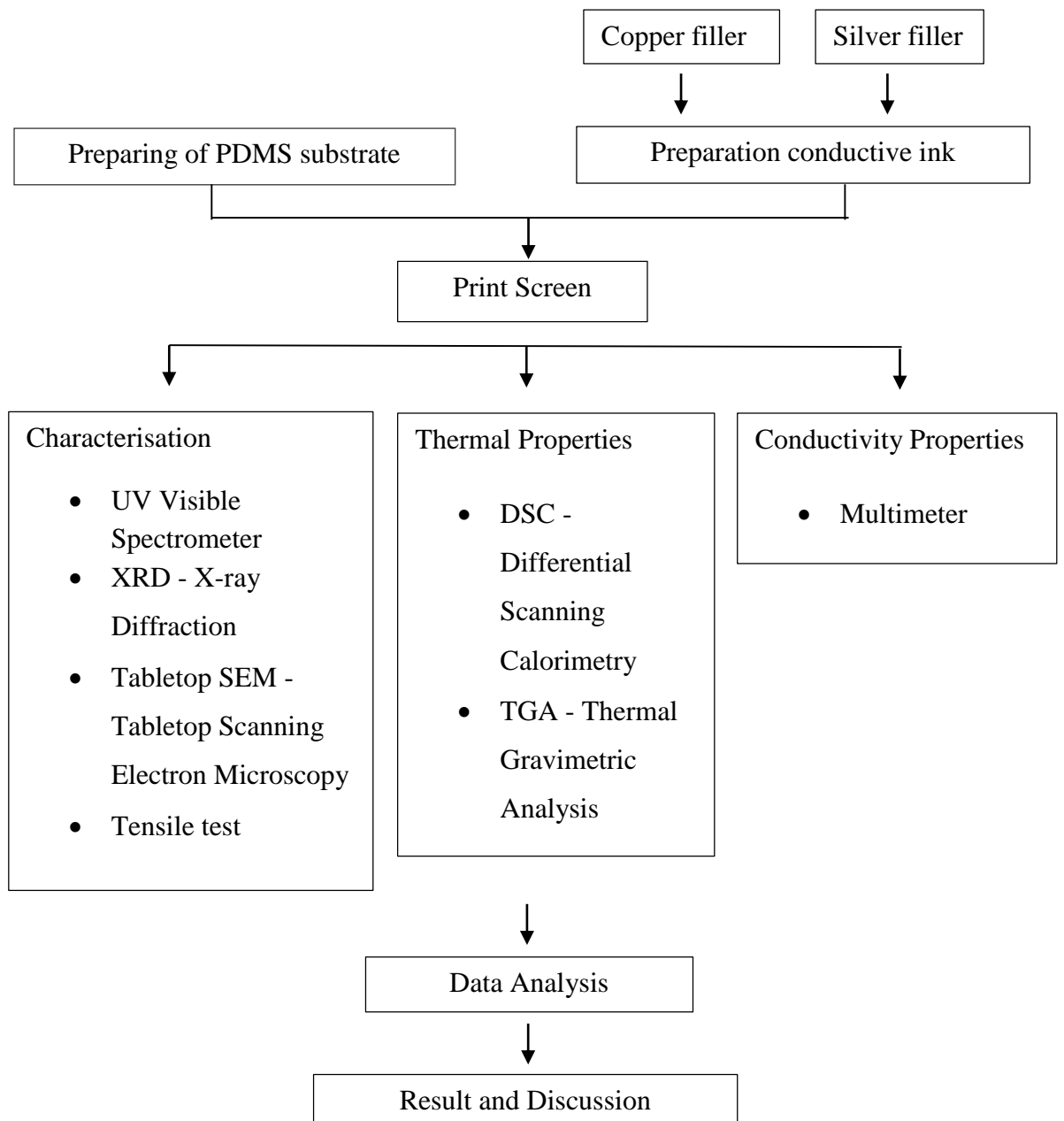


Figure 3.1: Research methodology flowchart

### 3.4 Preparation stretchable substrate

#### 3.4.1 Materials and Apparatus

PDMS-OH terminated, fume silica (2%), toluene, dibutyltin dilaurate, GPTMS, plastic cup, weight balance, mould and stick.

Table 3.1: Composition of stretchable substrate

Composition	Weight	Description
PDMS-OH terminated	7g	Matrix
Fume Silica (2%)	0.14g	Reinforcing agent
Toluene	13.86ml	Solvent
Dibutyltin dilaurate	0.07ml	Catalyst
GPTMS	0.20ml	Crosslinking agent

### 3.4.2 Procedures

Based on composition in table 3.1, PDMS-OH terminated and fume silica (dissolve in Toluene) is weighted respectively using weight balance and poured into the plastic cup. The mixture is stirred manually using stick until homogenous. Then, GPTMS is poured and stirred until homogenous. Then, the mixture is degassed for 5 minutes and then dibutyltin dilaurate is added and mixed well. After that, the mixture is poured into the mould shown in figure 3.2 and cured at the room temperature for 24 hours. Then the substrate been cut with dimension (5cm x 2cm x 0.1cm).



Figure 3.2: The mould to produce PDMS substrate (11cm x 11cm x 0.1cm)