A STUDY OF PERCOLATION THRESHOLD ON GRAPHENE CONDUCTIVE INK FILLED RESIN EPOXY

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DECLARATION

I declare that this project report entitled "A study of Percolation Threshold on Graphene Conductive Ink filled Resin Epoxy" is the result of my own work except as cited in the references.

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Date : JULY 2020

APPROVAL

I hereby declare that I have read this project report and in my opinion, this report is sufficient in term in terms of scope and the quality for the award of the degree of Bachelor of Mechanical Engineering.

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Name : TS. DR. MOHD AZLI BIN SALIM

Date : JULY 2020

ABSTRACT

Nowadays, there is a new technology that undergoes well development, which has the ability to conduct electricity by using the utility of the ink. This technology is called conductive ink and has been launch recently with the implementation of different materials. In most cases, materials such as silver, carbon, and other conductive materials are used as a filler in the composition of the conductive ink. Additionally, there is a material that still new in this conductive ink industry which is graphene. Graphene is a new treasure so-called “wonder material” that has excellent properties in conducting electricity. The use of graphene as a conductive material in the formulation of ink might give many advantages but some factors can be affecting the performance of the graphene during conducting electricity. One of the factors is the properties and behaviors of the graphene, which can be affecting the ink and its percolation threshold. These factors are a new thing that still undergoes a study by the researchers, as the use of graphene in conductive ink technology is still new. Based on the existing formulation of conductive ink, which is graphene as a filler, a study on the effect of properties and behavior on conductive ink and its percolation threshold has been conducted. The study is used graphene as a filler, epoxy resin works as binder and polytheramine as a hardener. These materials have been blend together by using the centrifugal mixer to form the samples of conductive ink. After that, the samples are undergone the printing and curing process at 100°C for thirty minutes. Samples of the ink have been patterned into four different types which are a straight line, zigzag, curve, and square pattern. Each pattern is print out with three different widths, which are 1mm, 2mm, and 3mm. In this study, to achieve the objectives, there are four tests have been conducted which are voltage and sheet resistivity test by using the four-point probe, hardness test by using nanoindentation, microstructure test on the top surface of ink by using image analyser and study on the existence of an electron by using a scanning electron microscope. This study is carried out both theoretically and experimentally to find out the results. Based on this study, it can be concluded that curve is the best pattern followed by straight line while vice versa with zigzag, and based on the width, 1mm samples has the less efficient in conducting electricity compared to a 3mm samples. Besides that, it is proved that the properties and behavior of graphene affecting the percolation of ink as it found out at the end of the study. These findings of this study will be used in the future for increases the performance of graphene in conductive ink technology.

ABSTRAK

Pada masa kini, terdapat satu teknologi baru yang melalui pengembangan yang pesat dengan mempunyai kemampuan untuk mengalirkan elektrik dengan menggunakan utiliti dakwat. Teknologi ini dipanggil dakwat konduktif dan telah dilancarkan baru-baru ini dengan pelaksanaan bahan yang berbeza. Dalam kebanyakan kes, bahan seperti perak, karbon, dan bahan konduktif lain digunakan sebagai pengisi dalam komposisi dakwat konduktif. Selain itu, terdapat bahan yang masih baru dalam industri dakwat konduktif ini iaitu graphene. Graphene adalah harta karun baru yang disebut sebagai "bahan ajaib" yang mempunyai sifat yang sangat baik dalam mengalirkan elektrik. Penggunaan graphene sebagai bahan konduktif dalam formulasi dakwat berkemungkinan memberikan banyak kelebihan tetapi beberapa faktor boleh mempengaruhi prestasi graphene semasa mengalirkan elektrik. Salah satu faktornya ialah sifat dan tingkah laku graphene yang boleh mempengaruhi dakwat dan ambang perkolasinya. Faktor-faktor ini adalah perkara baru yang masih menjalani kajian oleh para penyelidik, kerana penggunaan graphene dalam teknologi dakwat konduktif masih baru. Berdasarkan formulasi dakwat konduktif yang sedia ada, iaitu graphene sebagai pengisi, kajian mengenai pengaruh sifat dan tingkah laku pada dakwat konduktif dan ambang percolasinya telah dijalankan. Kajian ini menggunakan graphene sebagai pengisi, resin epoksi berfungsi sebagai pengikat dan polytheramine sebagai pengeras. Bahan-bahan ini telah dicampurkan bersama dengan menggunakan pengadun sentrifugal untuk membentuk sampel dakwat konduktif. Selepas itu, sampel menjalani proses pencetakan dan pengawetan pada suhu 100 ° C selama tiga puluh minit. Sampel dakwat telah dibentuk menjadi empat jenis yang berbeza iaitu garis lurus, zigzag, lengkung, dan corak segi empat. Setiap corak dicetak dengan tiga lebar yang berbeza iaitu 1mm, 2mm, dan 3mm. Dalam kajian ini, untuk mencapai objektif, terdapat empat ujian yang telah dilakukan iaitu ujian ketahanan voltan dan lembaran dengan menggunakan probe empat titik, ujian ketahanan dengan menggunakan nanoindentasi, ujian struktur mikro pada permukaan atas dakwat dengan menggunakan penganalisis gambar dan mengkaji kewujudan elektron dengan menggunakan mikroskop elektron imbasan. Kajian ini dijalankan secara teori dan eksperimen untuk mengetahui hasilnya. Berdasarkan kajian ini, dapat disimpulkan bahawa lengkung adalah corak terbaik yang diikuti dengan garis lurus dan sebaliknya untuk corak zigzag, dan berdasarkan lebarnya, sampel 1mm mempunyai tenaga yang kurang efisien dalam menjalankan elektrik berbanding dengan sampel 3mm. Selain itu, terbukti juga bahawa sifat dan tingkah laku graphene mempengaruhi perkolasi dakwat seperti yang dijumpai pada akhir kajian. Penemuan kajian ini akan digunakan pada masa akan datang untuk meningkatkan prestasi graphene dalam teknologi dakwat konduktif.

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LIST OF ABBREVIATIONS

FIB	-	Focused Iron
GnP	-	Graphene Nanoparticles
TPU	-	Thermoplastic Polyurethane
PC	-	Personal Computer
SEM	-	Scanning Electron Microscopy
EDX	-	Energy Dispersive X-Ray

LIST OF SYMBOLS

C	-	Carbon
O	-	Oxygen
Pt	-	Platinum
Bi	-	Bismuth
R	-	Resistance
V	-	Voltage
I	-	Current
ρ	-	Sheet resistivity
t	-	Thickness
C	-	Carbon
Ag	-	Silver
Cu	-	Copper
Au	-	Gold
C	-	Carbon
Tpa	-	Terapascal
Gpa	-	Gigapascal
mV	-	Millivolt
ηm	-	Nanometre
µm	-	Micrometre
mm	-	Millimetre
mN	-	Milli-newton

CHAPTER 1

INTRODUCTION

1.1 Overview

This chapter briefly explained on the background of this study on graphene based conductive ink filled epoxy resin. The primary objective highlighted in this study was to investigate on properties and behaviour graphene that affected the conductivity and percolation threshold ink. The study was done by used different pattern and width of samples.

1.2 Background

Electronics that produced by the printing technology is called printed electronics. This printed electronics is particularly is an electronic science and technology based on conventional printing techniques as the means to manufacture electronics devices and systems (Cui, 2016). Nowadays, these printed electronics things undergo fast development in order to replace the outmoded technology of electronics. In this printed electronics technology, conductive ink is one of the new well-developed products that be used widely in this world. Conductive ink is a blend of electricity conductivity materials with an ink utility to conduct electricity. According to Li, Lu, & Wong (2010), conductive ink is a technology that can pattern the conductive or semi-conductive materials in order to conduct electricity. In recent years, this conductive ink is hyped researchers and industries that related to doing research on it in order to replace the old circuit system of electricity. The supplier also is looking for some differentiation on the products based on conductive ink as it rapidly launching.

In most cases, the conductive ink is formulated together with micron-size particles which a conductive material to allow the ink to conduct electricity. The conductive polymers, carbon (C), or metallic particles such as silver (Ag), copper (Cu) and gold (Au) are the material that the most common selection to produce conductive ink (Cruz, Rocha, & Viana, 2018). Various materials have been undergoing research in order to find out the best materials to conduct electricity as the user demanding about low cost, flexible and smarter products. As the demand for the uses of conductive ink increases, the research on other conductive materials is such a race between the researchers. In the years 2010, graphene has been claimed to enable a new wave of innovation as a group of physicists successfully prepare a tiny sheet of graphene, which before this considered as theory only. This achievement is awarded a Nobel Prize in physics 2010, which awarded to Profs A. Geim and K. Novoselov of the University of Manchester for their ground-breaking experiments on graphene (Alvial-Palavicino & Konrad, 2019). This achievement makes researches hyped on these graphene materials as its properties are beyond expected.

This research on graphene is a hit in a conductive ink technology which many researchers have done research on it. Various type of research has been done on the graphene in conductive ink technology, which includes the properties of the graphene. The properties study on this graphene as been done in many terms of properties such as in electrical properties, mechanical properties, thermal properties and the application. (Papageorgiou, Kinloch, & Young, 2017). The electrical properties studies include the voltage and resistivity test while the mechanical properties studies are done on the hardness, strength, and stiffness of the ink. Thermal properties are studied on the thermal effect on the ink. Some researcher has done a study on the formulation of the ink and the pattern in order to conduct electricity effectively. In order to produce high-quality conductive ink, the graphene must be blended together with the other materials to produce ink. Some study on this graphene as conductive ink is still ongoing and takes quite some times as to figure out the best formulation and application on the good properties of the graphene.

This conductive ink has many advantages, as it will be widely used in various types of industries. This is because conductive ink is required a low cost for manufacturing as the conductive ink only takes a simple process to be turned into a form of ink. This conductive also can be cured at low temperature and the usage of the ink is gives low environmental effects. Besides that, this conductive ink needed a low maintenance cost it is very long-lasting products. It is also very flexible as it can go through a bending test, stretchable, flexible and dynamic condition.

This conductive ink also applies in many industries such as electronic packaging, healthcare, also the solar industries and many other applications. This technology will continue growing as it a very flexible technology that can be implemented in many applications in the industries.

1.3 Problem statement

For future use, many conductive materials been formulated such as carbon black, silver, carbon nanotube (CNT), copper and graphene. Many formulated been done in order to get a better result to find out the most suitable materials and formulated for conductive electricity. In this, research the material that is focused on conductive material, which is graphene. The graphene is been formulated with other materials to form a conductive ink. The incorporation of the graphene other material will produce good conductive ink but some factors might affect the performance of the graphene. Many studies have to go through the factors based on the properties of the graphene but the study on the thermal properties of the graphene is quite lesser than other properties. The graphene might produce good conductive ink but in order to produce an excellent product of ink, these thermal properties must be looking forward. This thermal property might affect the performance based on the percolation threshold of the graphene. In the term of the thermal effect, the concentration of the percolation threshold in the graphene will affect the performance in conducting electricity. In summary, to produce good conductive ink, many factors that might affect the performance of the ink must be considered. The percolation threshold on graphene is one of the factors that needed for a better understanding in order to make the graphene is the ideal materials for conductive electricity in the form of ink. For more specific, the following research questions are addressed as below;

1. There is no exact data on the properties and behavior of the graphene in the form of ink that conducts electricity?
2. There is no study on the effect of graphene properties and behavior on the percolation threshold.

Based on these two problem statements, a study is carried out in order to find out either the thermal effect gives a major effect on the conductive ink or not. Through some research on the existing conductive that used graphene as a material, these two problems in a factor of percolation threshold must be finding out in order to increases the performance of the conductive ink.

1.4 Objectives

A study is carried out in order to investigate and understand the problems related to the thermal properties of the graphene in the form of ink. The goal of the study is to understand the percolation threshold effect on the graphene conductive ink. Through this study, two aims that are related to the thermal effect on the percolation threshold of graphene conductive ink need to be achieved at the end of the study. Particularly, the study has the following objectives;

1. To identify the properties and behavior of the graphene conductive ink.
2. To investigate the effect of properties and behaviour on percolation threshold in graphene conductive ink.

This study will be done based on the ink properties and continue with the percolation threshold effect on the graphene. This study will be ended with a comparison between the simulation and experimentally results. After the results validate, the result will be used to improve the performance of graphene in conductive the electricity in the form of ink.

1.5 Scope

The conductive ink might have many factors that affect its performance however, this study will only covered up on the behavior and properties, focus and discuss the thermal effect which is percolation threshold on the graphene conductive ink due to four type of pattern which are straight line, zigzag, curve, and square with a width 1mm,2mm, and 3mm. This report is not covered up the other factors that affected the graphene. This study is used as an experimental approach to gain data.

1.6 Limitations

The limitation of this study is the material and the pattern of the samples. The results of this study can only be applied to graphene-based conductive ink. These results of the study did not valid for other materials. Pattern and the width of the samples also one of the limitations of this study as the pattern that be done on this study are a straight line, zigzag, curve, and square only and the width is limited to 1mm, 2mm, and 3mm. Next, during the printing process, it is hard to maintain the linear distribution of the samples as the process is done manually. The samples might have some defeat on the surface, as the distribution of the composite in the mold did not go well.

CHAPTER 2

LITERATURE REVIEW

2.1 Overview

This chapter briefly explains the necessity of graphene as conductive material in the conductive ink in study of effect properties and behaviour. The effect of percolation threshold in conductive ink is provided in this chapter. Other than that, the review of previous research in conductive ink field, which led to the justification for the need for this research to be conducted, is also discussed in this chapter.

2.2 Conductive ink

Many researchers have done a study on the conductive ink, as it is a new technology in the electronic and electrical industry. In recent years, all the research is been done on specific materials such as copper, silver, gold, carbon black and many other conductive materials. The presence of the free electron depends on the conductive materials (*Engineering Physics-II*, 2019). Conducting materials is a material that can conduct electricity due to electricity from free electrons when the difference in electrical potential is applied to it. The user of the conducting materials plays an important role in engineering and technology. As stated above, the conducting materials are widely used in conductive ink technology. This is because the conducting materials are good conductors of electricity and heat. In the conductive ink technology, the better the properties of the materials in conducting electricity the high the quality of the conductive ink been to produce.

In the line of this topic, during the process of turning the conductive materials into a form of ink, the materials will be blends together with other materials as a support to complete the composition of the ink (Alemour, Yaacob, Lim, & Hassan, 2018). The filler is terms that be used for the conductive materials in the composite such as carbon-based materials like graphene. When the filler combines together with the polymer resin or some hardener materials, the composite turn into a conductive composite. The filler in the composite plays a major responsibility in the composite to conduct electricity. As the filler contents in the composite increases, the particles of the filler will start to contact each other and make a path for particles freely moves and conduct the electricity. Usually, the conductive composite is in the form of a lightweight, resistance to corrosive, and can easily adapt to the needs of the specific applications. Therefore, these conductive composite might replace the use of metal in the industry for some applications.

2.3 Graphene

In 2010, by the ground-breaking experiment on the graphene, Professors Andre Geim and Konstantin Novoselov were awarded the 2010 Nobel Prize in Physics (Hancock, 2011). This achievement has rapidly increased the research on the graphene until this day. With the 2D microstructures, the C-C bonds and the good mechanical properties induced by its bond make the graphene more popular in the conductive materials. The band gap of the graphene is zero gaps which represent the superconductive property as well as the strong biocompatibility of graphene (Liu, Qing, Wang, & Chen, 2015).

If the graphene microstructure changed into a nanoparticle, different types of graphene-based materials such as graphene, graphene oxide, and metal-doped graphene can be obtained (Xiong et al., 2015). Graphene has remarkable qualities that come from the 2p orbitals that ultimately cause the p bands to pass over the carbon sheets that make up the graphene. Researchers also found that the graphene is extremely rigid from the past research on the graphene which displays thermal conductivity at a high level of it and clearly impermeable to gases (Sheehy, 2009). For commercial and applications, high-quality graphene is needed (Verma & Goh, 2019).

Graphene is also known as a wonder material. The research of graphene ha proven that how excellent the properties of the graphene. Some study on the graphene has exposed the functionality of the atomic crystal of graphene in 2-D. The functionality that is discussed is related to the graphene properties such as thermal conductivity is equally to 5000 $Wm^{-1}K^{-}$ with high electron mobility at room temperature. Besides that, the graphene also has a high property of the surface area per unit mass which is 2630 $m^2 g^{-1}$ (Zhu et al., 2010). Next, the graphene also has a high modulus of elasticity which reaches until 1 TPa (Terapascal) (Potts, Dreyer, Bielawski, & Ruoff, 2011). The graphene also has high electrical conductivity. However, some reach on the graphene as stated that the preparation and implementation of good quality of graphene in bulk quantities is really a difficult task. The dispersion of graphene will affects the properties of the graphene. Some study has done on the formulation for the graphene in order to build a strong composite that can conduct an electricity effectively. There some researches is undergoes a research on the pattern of the graphene-based composite as the pattern of the graphene might affecting the performance of the composite (Technologies et al., 2019). There are many studies have been regarding the excellent properties of the graphene.

2.4 Mechanical properties

Every material has physical properties that explain its behavior when loads are applied to it is a mechanical property. Mechanical properties for graphene have many aspects, which include the stiffness of the graphene, the strength and lastly the toughness. A study from the past on graphene has proven that the material has wonder properties (Verma & Goh, 2019).

2.4.1 Stiffness

Stiffness is one of the mechanical properties that indicate the strength of the structure of the material when resisting deformation or deflection by applying pressure or force. A flexible material usually has a low stiffness value. The reason why graphene stands out as an individual component and be a composite of reinforcement agent is the remarkable mechanical properties of graphene (Papageorgiou et al., 2017). This graphene lies in the stability of the sp^2 bonds that form a hexagonal lattice (refer to Figure 2.1) and oppose a variety of in-plane deformation. In a research of this stiffness of the graphene, the researchers have measured the elastic properties and intrinsic breaking strength of freestanding monolayer graphene membranes by nanoindentation in an atomic force microscope.

The intrinsic strength is the ability of the graphene to connect with other materials. The breaking strength of the graphene is be measured by some of the researchers which found that the breaking strength of the graphene is at $42Nm^{-1}$ and it is explained about the intrinsic strength of a defect-free sheet. These quantities also correspond to a Young's Modulus of E = 1.0 Tpa (Terapascals) and elastic stiffness of D = -2.0Tpa (Lee, 2012). This value shows that the graphene is the strongest material ever measured and directly shows that the graphene as the perfect nanoscale materials can be mechanically tested to deformation. Some other research has obtained different values of stiffness which probably the result of the intrinsic and unavoidable crumpling of graphene in a monolayer out-of-plane. This crumpling of graphene has found outcomes from the uneven stress at the boundary of the graphene and responsible for the degradation of the graphene in terms of mechanical properties. For this stiffness of the graphene, crumpling and wrinkling are play important roles on the nanomechanical systems as they might affecting the stiffness of the graphene as a composite (Cranford & Buehler, 2011).

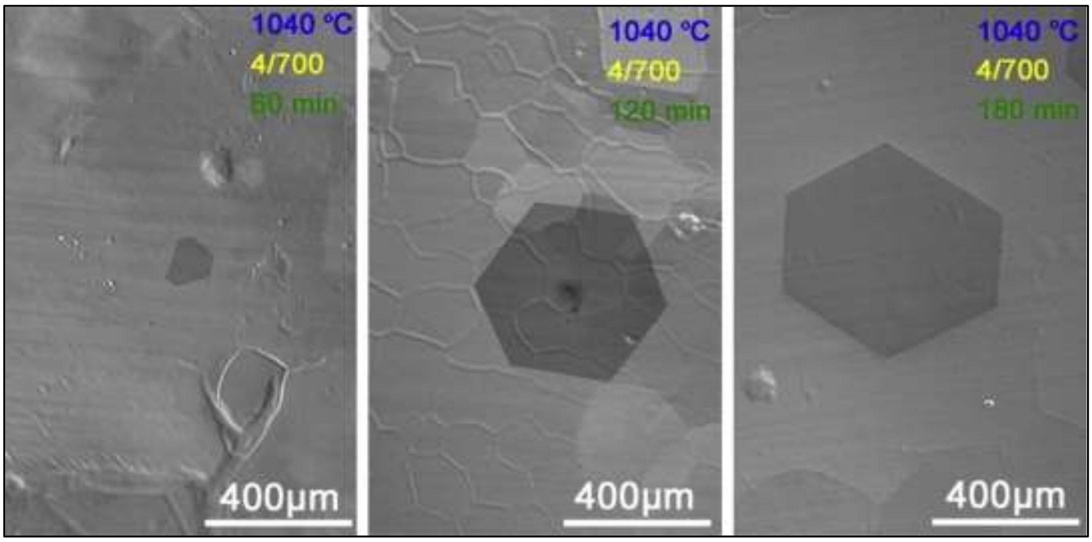


Figure 2.2.1: Synthesis of Hexagonal Single Crystal Graphene (Science, 2014)

2.4.2 Strength

As the intrinsic strength of the graphene is at $42Nm^{-1}$ and directly makes the graphene as the strongest materials with a high stiffness value. In the defect-free mode, graphene in a monolayer is considered as the strongest material ever tested. The intrinsic strength value of graphene at $42Nm^{-1}$ is equally to the intrinsic strength of 130GPa. It is quite interesting as the strength and stiffness of the graphene were maintain when the graphene even in higher densities in sp^3 bonds. When the graphene reaches the defect-regime, the strength of the graphene drops significantly.

2.4.3 Toughness

Fracture toughness of graphene is one of the most important mechanical properties of the graphene. This fracture toughness property is relevant to the engineering application. (P. Zhang et al., 2014). Some study has found that a central crack in the graphene membranes by using the focused ion beam (FIB) and the brittle fracture was determined by applying the load on the graphene. The fracture stress decreased with increasing the crack length and the critical strain of the graphene was found to be $15.9Jm^{-2.}$ The fracture toughness of graphene was measured as a critical stress intensity factor (KC) of 4.0 ± 0.6 MPa (B. Zhang et al., 2012). From the similar works that the durability of grphene relative to most membranes is heavily dependent on its weakest link.

2.5 Electrical properties

Every material that is used in any industry must have an electrical property either it is a good conductor or cannot even conducting electricity. The material usually is classified into three types depends on the characteristics of the materials such as conductor, semiconductor and nonconductor. Materials are divided into these three types of the group based on the capability of the material to conducting electricity. Graphene is a conductor material as it has the ability to conduct electricity. 2-D dimensional graphene has a unique electrical property that such as high mobility and ballistic transport at room temperature (Wang, Ma, Liang, & Sun, 2017). Graphene is a planar film composed of carbon atoms with a sp^2 hybrid orbital and has a honeycomb-like lattice structure. It is a two-dimensional (2D) material of only one atomic layer thickness. Fullerene (0-dimensional), carbon nanotube (1-dimensional), and graphite (3 dimensional) form the basic constituent elements, which can be regarded as infinite aromatic molecules" (Wang et al., 2017).

2.5.1 Percolation threshold

The percolation theory refers to the critical filler loading where electrical conductivity increases notably to several orders of magnitude because of the formation of continues electron and conducting paths (Ozkan, 2019). This study also stated that electrical conductivity experienced a saturation plateau when multiple electron paths exist above the percolation transition range. Electron paths will not exist below the percolation transition range. Some study has stated that the concentration of graphene must be above the percolation threshold so that the conducting network can be achieved in the composite

(Sadasivuni , Ponnamma, Kim, & Thomas, 2015). Many researchers have claimed that the percolation threshold affected the performance of the materials in conducting electricity.

In recent years, all the research is been done on specific materials such as a polymer and carbon black. Graphene is a new material that has been developed which good to conduct electricity in the form of ink. As it still new in the conductive ink industry, the review for the graphene in a factor of percolation threshold is still not expanded yet. There is some article related to properties and factors that affect the performance of the graphene. Graphene and carbon black have been dispersed in a high-performance thermoplastic polymer, the poly(ether ketone), to improve its electrical conductivity. The dispersion of graphene has a significant influence on the percolation threshold. A simple exfoliation protocol to obtain graphene monolayers has led to significant decreases in the percolation threshold. (C. Bessaguet, 2019). Some of the articles are discussed in the methods that be used to lower the percolation threshold in the graphene ink. Graphene nanocomposites with low percolation threshold and tunable negative permittivity can be prepared by coating and pressing methods. (H. Wu, 2017). These articles are discussed on the factors that make the percolation threshold low in the graphene.

There are many methods that be used in the study of the percolation threshold of the graphene either theoretically or experimentally. Some studies have used the extra-properties of graphene to another level. In the combination of the great mechanical properties, unique electrical properties, and thermal properties, the extraordinary of the electronic behavior of the graphene offers great potential for a variety of applications. This exaggerated the researchers around the world to study on this graphene in further. Some studies have conducted an experiment on the percolation of the graphene by combine the graphene with the polymer and produce a graphene-polymer hybrid composite (Mutlay & Tudoran, n.d.). In this study of the percolation behavior, graphene nanoplatelet filled electrically conductive nanocomposite of the polymer. Both did this study theoretically and experimentally. This study was found that the best values of the percolation threshold and maximum conductivity were found to be 0.3 vol.% and 10^{-1} s/cm, respectively. This study develops a simple modification of the power-law model for the percolation theory of electrically conductive. What is missing from the past studies is the approach to the percolation threshold effect on the graphene conductive ink. Therefore, in order to understand how the percolation threshold affected the performance of the graphene to conduct electricity, this research is the aim of it. It may be advantageous to study the percolation threshold in different patterns of graphene ink as it may give a different result (Verma & Goh, 2019).

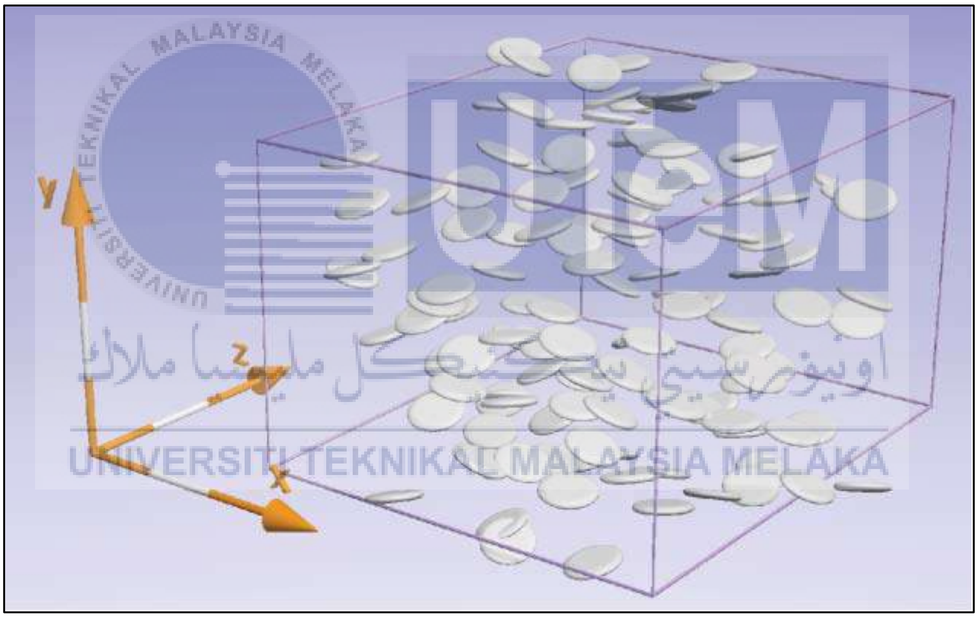


Figure 2.2.2: Schematically Simulated Visualization of Particle Distribution in a Graphene-Polymer (Agglomeration of the graphenes)

2.6 Thermal properties

Thermal properties are the properties that related to the conductivity of heats. This thermal property is important, as it is a property that how the materials react when it surround by the fluctuation heats. Some study has been done by the researches on the thermal properties of the graphene. Graphene has a very high thermal conductivity. The specific heat of the graphene is dominated by phonons and it is slightly higher than graphite and diamond below room temperature (Pop, Varshney, & Roy, 2012). If the graphene in a composite or 3D dimensional structures, the thermal properties could be highly tunable such tunability raises the interesting prospects of both ultrahigh thermal conductivity for heat-sinking applications and ultralow thermal conductivity for thermoelectric applications.

2.7 Application of the graphene

Graphene as nanofiller in the composite gives many advantages and will be implementing more in the electronic and electric industry. Electromagnetic interference shielding materials, coatings, sensors, batteries, and electrode are some of the examples of the application graphene as a composite. Graphene is used in the applications as a hybrid in a polymer matrix and it works more effectively in this graphene-polymer matrix.

CHAPTER 3

METHODOLOGY

3.1 Overview

The methodology section is an explanation of the method that be used to achieve the objectives of the study. There are many methods that can be implemented in a study depends on the field and the objective of the study. According to this study, the objectives are to identify the behavior and properties of the graphene and to investigate the concentration of the percolation threshold between the graphene (filler), binder and hardener. A few methods have been considered and used in order to achieve the objectives of the study. The first method that is used is the experimental approach and followed by data analysis.

3.2 Experimental approach

The experimental approach is the method used to identify the behavior and properties of the graphene conductive ink. For this experiment, the material is an important thing to be considered, as it is a major aim for this study. The experimental study starts with the formulation process that already standardized by the past study. For the formulation process, materials that be used in the experiment include the graphene, epoxy resin and hardener. These three materials are blended together in order to form a sample of conductive ink. After the formulation proses, the sample undergone a printing process and followed by curing proses with specific temperature and time. For this study, the printed sample that already solidifies is tested on its behavior and properties based on engineering standards to make sure the sample capable to conduct electricity. The test is conducted to look forward to the behavior and properties of graphene-based on the voltage, resistivity, hardness and the microstructure of the conductive ink.

3.2.1 Materials

In physically experimental, materials are important things to be considered even before the study start. This study combined three materials and turn into a form of composite. Generally, a relatively new family of materials is known as composites which distinguished by lighter weight and greater resistance than traditional material (Bayraktar, 2016). The materials used are graphene that works as a filler, epoxy resin as a binder, and hardener. This combination of materials is to build an application as it has structural materials that fulfill the criteria in engineering applications.

3.2.1.1 Filler

In this study, the filler is the most important material in this composition process. To be a filler as part of conductive ink, the ability to conduct and electricity is important things to be considered. This study is carried out with graphene as the filler and be added into the composite in order to increases the bulk, viscosity and firmness of the composite. Graphene is proved as one of the best conductor materials in this conductive ink technology as it has a high capability to conduct electricity and has good properties as a conductor (Wassei & Kaner, 2010).

As the graphene in any type of form, this study chose graphene nanoplatelets for this study. Graphene nanoplatelets (900439) are provided by Sigma Aldrich and were used as the main filler in this study. GnP or Graphene nanoplatelets have 500 m2/g surface area and 12.01 g/mol of molecular mass. Typically this material consists of submicron platelet aggregates having a particle smaller than 2 microns in diameter (Sheet, 2018).

3.2.1.2 Binder

For the binder, it used to bind the particles of the composite together. In this study, the epoxy resin is the most suitable binder for this composite. The epoxy resin is a thermosetting group of polymer that usually used as a binder. This component also greatly influences the mechanical properties of the ECA. The epoxy resin used in this research was a bisphenol-A diglycidyl (DGEBA) ethers, that is made up of epichlorohydrin and bisphenol-A and have an average molecular density of ≤ 700 g/mol. The density of the resin is 1.168 g/ml, with a viscosity value in the range of 500-750 mPa.s at 25 °C. This colorless resin was purchased from Sigma Aldrich, with the trade name of Araldite 506 Epoxy Resin.

3.2.1.3 Hardener

Lastly, the used of hardener in this experiment is for solidifying the composite of the combination filler and binder. Since this type of polymer are two-component adhesives, therefore, a hardener was required to complete the curing process via the cross-linking process. The Hunstman polyetheramine D230 hardener was used, which is an amine group type of curing agent. The density of the curing agent is 0.947 g/ml with a viscosity of 9 mPa.s at 25 °C.

3.2.1.4 TPU substrate

Thermoplastic Polyurethane (TPU) substrate as shown in Figure 3.1 is the substrate that is used as a base for the ink during the printed process. These TPU materials are chosen as it has beneficial properties such as stretchable and can resist the high temperature as the ink was cured at a high temperature. This TPU was cut depends on the size of the ink which 15cm for height and 4cm for the width. Each sample has the same dimensions, as each size of the patterns is the same.

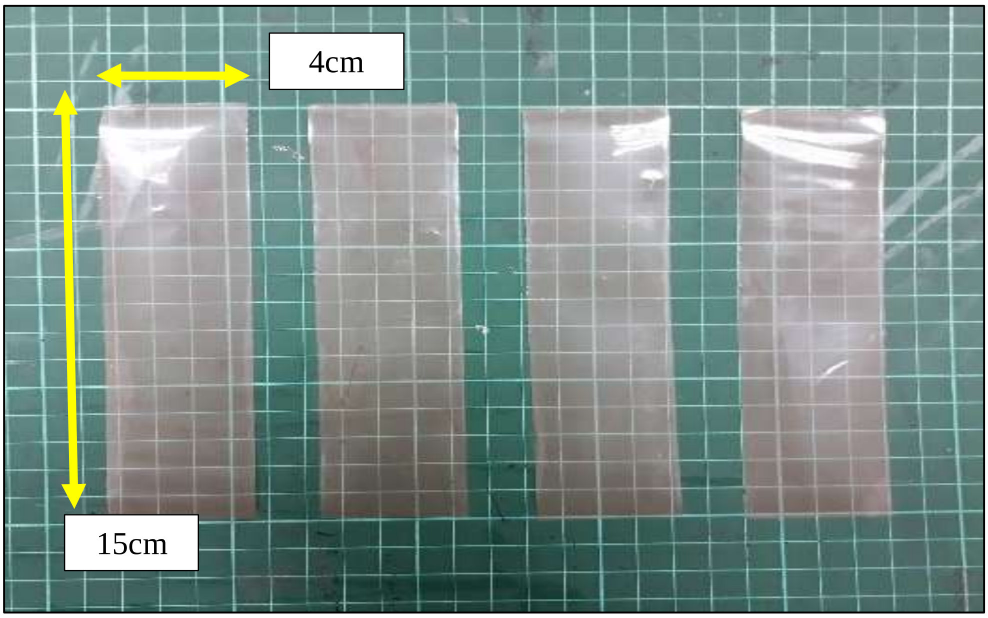


Figure 3.1: Thermoplastic Polyurethane (TPU)

3.2.2 Equipment

Equipment is an apparatus or machine that be used in this experimental approach. For this study, the equipment that is used is a weight scale, a centrifugal mixer, which is used in the mixing process, continued with oven for curing proses. For the test part, the equipment that is used is a 4-point probe, image analyzer and lastly the nanoindenter. All this equipment will be explained more in this subsection.

3.2.2.1 Analytical balance

The analytical balance as shown in Figure 3.2 is used in this experimental approach for weighting all the materials used in the composite of conductive ink. Mettler Toledo is the supplier of this analytical balance and the model that be supplied is me204E. This machine has come with an AC adapter and a few replaceable components. The replaceable components are been install based on the range of the weight that will be weighting as this machine has functionality that emphasizes the balancing with the readability of 0.1mg, 1mg, 10mg or 100mg. For this experiment, balancing with the readability of 0.1mg is chosen as the materials be used is a lightweight type. This machine has a precision feature, which suitable for the composite as the ratio of the materials is important. This machine was used during the formulation of proses.

Figure 3.2: Weight Scale

3.2.2.2 Centrifugal mixer

In the mixing process, the materials will be mix together by using the centrifugal mixer. The centrifugal mixer used is a Thinky mixer (ARE-310 model) which is a non-vacuum mixer. Figure 3.3 shows that the thinky mixer, which is a new technology of mixer as it is, has a feature of the untouchable system and did not use the blade to mixing the materials. Based on (*Pioneering planetary centrifugal mixers*, n.d.), this machine can uniformly be mixing materials with different viscosities or specific gravities. It is also can dispersion of high-density material with no sedimentation. This thinky mixer is can dispersion of nanoparticles such as graphene with no aggregation and it been made for nano-level pulverizing and dispersing of insoluble compounds. In this study of the graphene, this thinky mixer is an important machine as the graphene has high density and in nanoparticles form.

This Thinky mixer comes with complete components such as container and container holder as shown in Figure 3.4. The container is come with a different size and was used depends on the weight of the material. This because to avoid an overflow material during the mixing process. In addition, the container is used to combine the materials together. The container holder is used to hold the container during the mixing process. The holder is placed in the mixer during the mixing in a 45° of angles.

Figure 3.3: Centrifugal Mixer

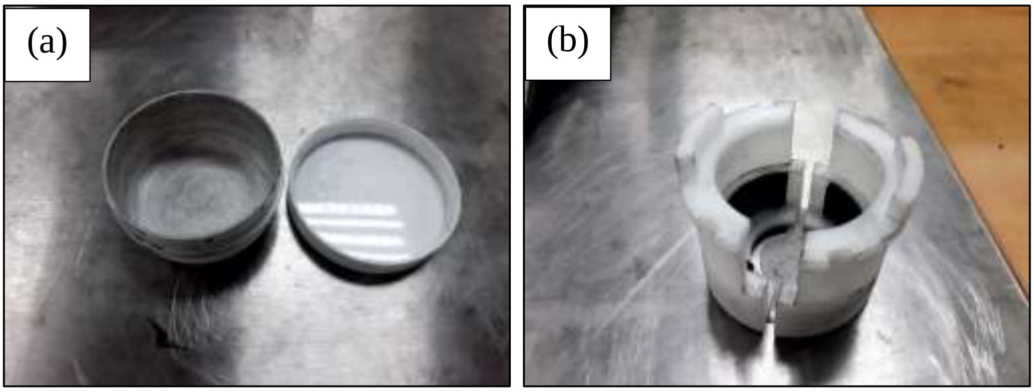


Figure 3.4: (a) Container (b) Container holder

3.2.2.3 Mold

For a piece of extra information, the mold was used in this experimental study only to pattern the samples and not include the curing process. The material of the mold is mild steel, the laser cutter on the mild steel carves the patterns for this experiment, and the thickness of the mild steel is 1mm. For this experiment, there are four types of pattern, which are a straight line, zigzag, curve, and square, and three different types of width, which is 1mm, 2mm, and 3mm. All the samples have the same thickness as the mold. Figure 3.5 shows that the patterns and width of the samples.

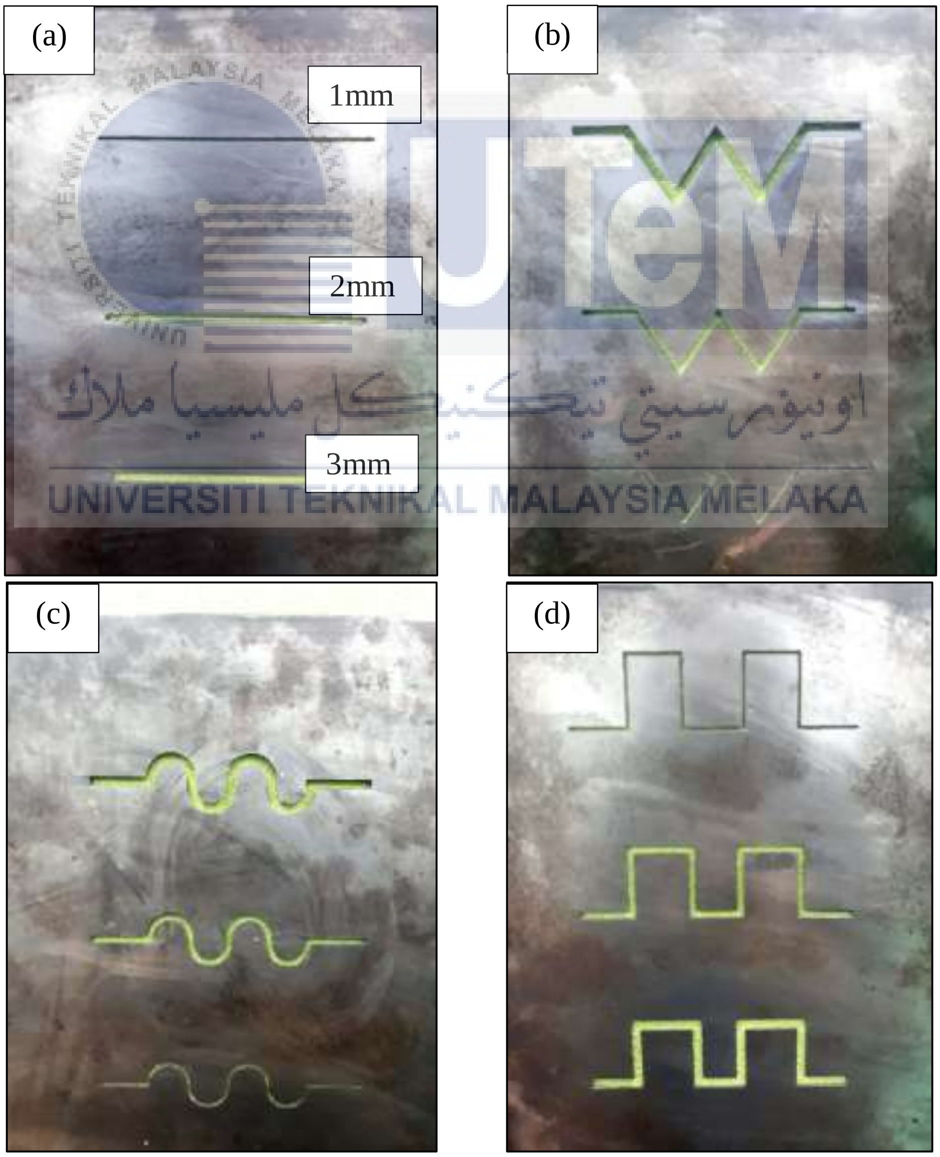


Figure 3.5: The mold; (a) straight line (b) zigzag (c) curve (d) square

3.2.2.4 Oven

This study also involved an oven to complete the conductive ink samples in the curing process. An oven that shows in Figure 3.6 is used to cure the samples after been printed on the substrate. This oven comes with the tray and the touchable display. The time and temperature can be set up to 300°c. As this oven was used in high heat, the precaution steps must be followed in order to avoid any damage to the user of the oven. Precautions that need to be followed were used Personal Protection Equipment such as a glove. This glove is worn during to put in and out the tray into the oven.

Figure 3.6: Oven

3.2.2.5 4-Point probe

The printed samples have undergone a test that gives data on the voltage and resistivity of the samples by using a 4-point probe machine. This machine was used as it is the most convenient machine to read the resistivity and voltage value of the samples. The Jandel supplies this machine and the model for the machine is RM3000. This 4 –point probe is consists of a cylindrical probe and the digital voltmeter. The voltmeter is connected to the personal computer (PC) to logging the data into the Jandel RM3000 software. This machine is supplied with a constant current and displays the resultant voltage, sheet resistance or volume resistivity depending on which function has been chosen. For this experiment, the digital voltmeter will display the reading of the voltage in a unit millivolt (mV).

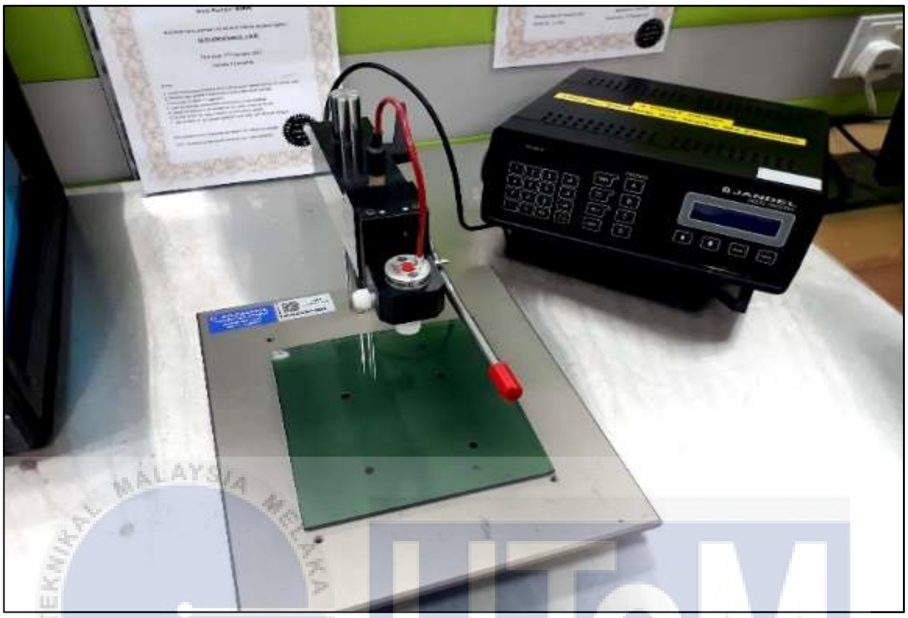


Figure 3.7: 4-Point Probe

3.2.2.6 Image analyser

Image analyser is an equipment that used to recognizing, differentiating and quantifying the diverse type of image including greyscale or colour images (Park & Lu, 2015). This image analyser is required in this study of the behaviour of the composite. The image analyser that is used is an Axioskop 2 Mat model (refer to Figure 3.8). This model offers an excellent feature, which comes with a variety and high-performance microscopes. This image analyser is easy to use, has accessible controller, reliable and fast focusing and lastly has many stages of the microscope. The machine has many small components that very functional during the experiment.

Figure 3.8: Image Analyser

3.2.2.7 Nano indentation

Nano indentation is a machine used to test the hardness of the samples. Hardness is a measure of the material resistance to a surface by an indenter with a force applied to the sample. Figure 3.9 shows the Nano indenter that is used in the study. This machine can be controlled by both manuals and automatically in adjusting the lens and the position of the sample. If used manually, there is the controller for focus and position while for automatically method, the software called DUH must be connected to the personal computer (PC) and the adjusting can be done by controlling through the software as shown in Figure 3.10.

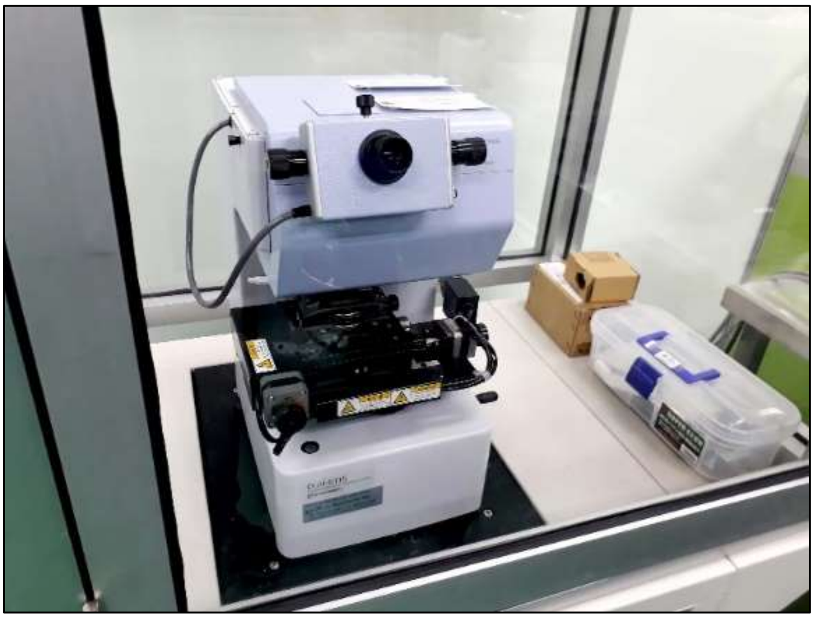


Figure 3.9: Nano Indenter

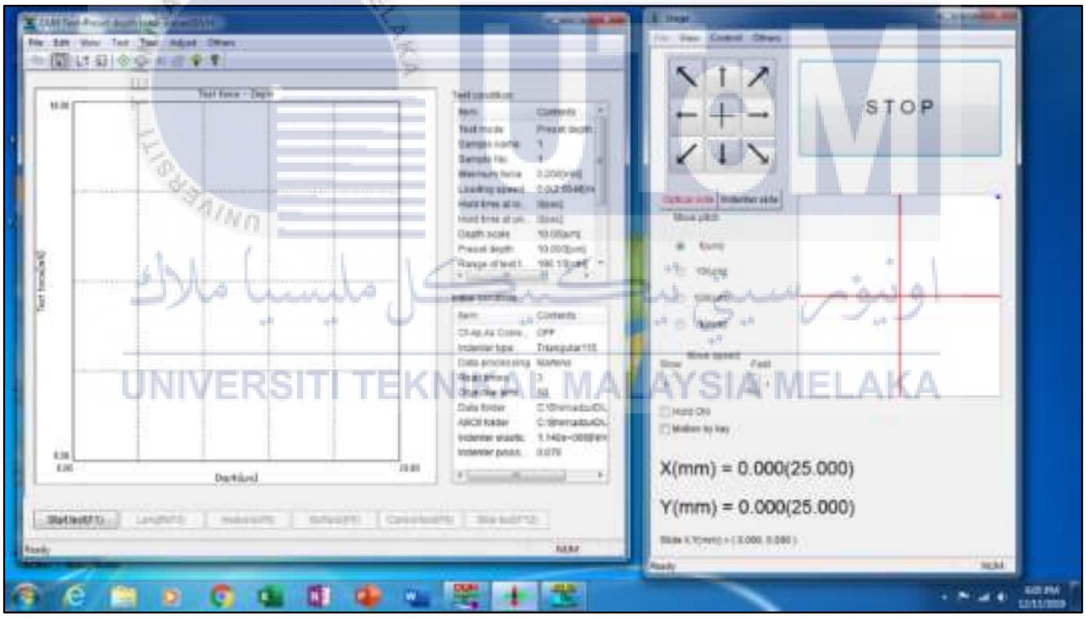


Figure 3.10: Duh Software

3.2.3 Procedural steps

This section is discussed about the procedural steps that be taken from the start of the materials been formulated until the test that been done. This study on graphene behaviour and properties as a conductive ink take a few procedural steps in order to form good and quality samples. These procedural steps include the formulation and mixing process, printing process, curing proses, testing the samples based on the objectives of the study. All the experimental is done in the laboratory.

3.2.3.1 Material preparation

The experimental approach is started with the study on the formulation of the materials used as this approach is focused on the materials used. As the test was done on the printed samples based on the behavior and properties of the composite, the composition process is important. The past study has done research on the formulation of the conductive ink in the form of composite (Technologies et al., 2019). This study also concludes that from the percentage of all the filler shows in Table 3.1, only 30% and 40% used of the filler can detect a resistivity during the test.

Table 3.1: The Composition of Filler Loading

Sample	Filler		Binder		Hardener (g)	Total (g)
	(%)	(g)	(%)	(g)		
1	10	0.2	90	1.8	0.54	2.54
2	20	0.4	80	1.6	0.48	2.48
3	30	0.6	70	1.4	0.42	2.42
4	40	0.8	60	1.2	0.36	2.36
5	50	1.0	50	1.0	0.30	2.30
6	60	1.2	40	0.8	0.24	2.24
7	70	1.4	30	0.6	0.18	2.18
8	80	1.6	20	0.4	0.12	2.12

Based on the past study, the formulation that is used for this experiment is 35% of filler (graphene), 65% of the binder (polymer resin) and hardener 30% from the value of binder (refer to Table 3.2). The total weight of the composite is 2.39g which equally 3 samples.

Table 3.2: Formulation of Composition

Sample	Filler		Binder		Hardener (g)	Total (g)
	(%)	(g)	(%)	(g)		
1	35	0.7	65	1.3	0.39	2.39

This experiment starts with the formulation process. The formulation process was started by taking out the materials from the chemical storage cabinet. Figure 3.11 shows that the materials which are graphene (filler), epoxy resin (binder) and the hardener. Then switch on the analytical balance by press the “on” button at the adapter. Before weighting the materials, the analytical machine must be calibrated first. Open the door of the machine and put the containers inside the machine (refer to Figure 3.12). Then close the door and pressed the “tare” button. The machine will read zero on the container after a few seconds.

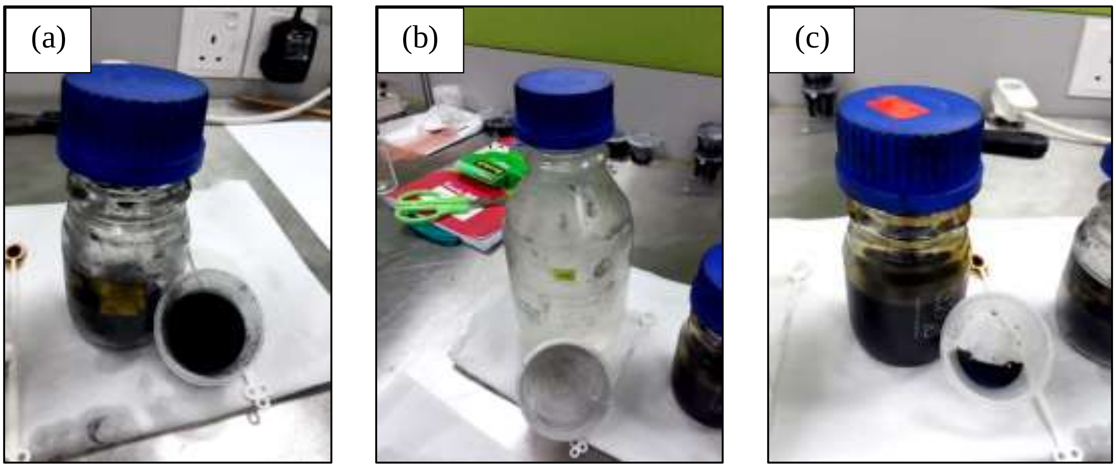


Figure 3.3.11: The materials: (a) Graphene (b) Epoxy Resin (c) Hardener

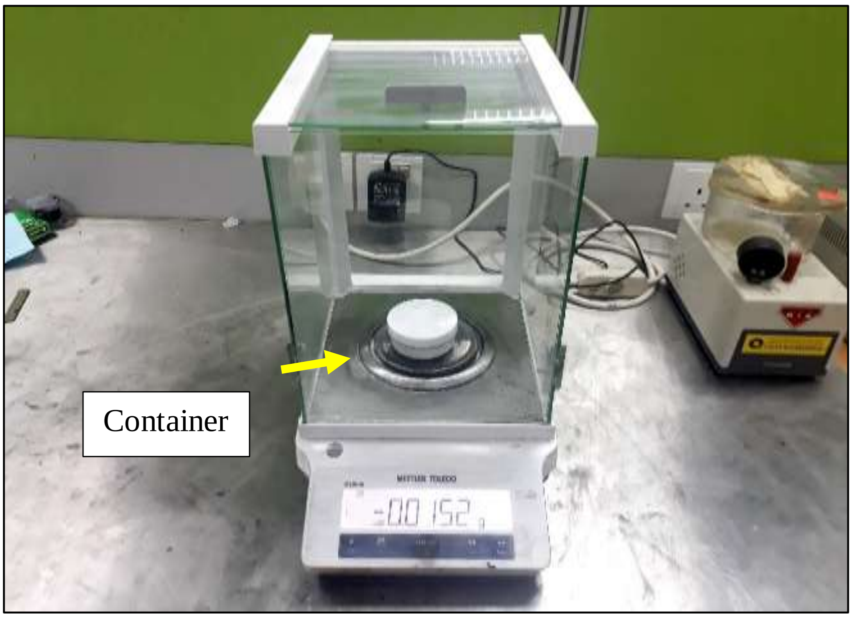


Figure 3.12: Container inside the Balance

After calibrating the container, open the door of the balance again and put the material in it depends on the formulation. The balance will read the weight of the material and stop put the material when balance displayed the reading that needed. This process was repeated for other materials. After finished weighting the materials, the formulation process was taken over by the mixing process. The materials were blends together and stirred until form a rough composite. After that, combine the container that fills with composite with container holder and put it together in the balance machine. The balance read the weight of the container and the container holder. The weight was applied in the calibration for Thinky mixer. By referring to Figure 3.13 (a), open the mixer put inside container holder into the mixer in 45° of tilt angle. The mixer must be calibrated before starting the mixing process by setting the weight balance based on the weight of the container and container holder that has been weighted before as shown in Figure 3.13 (b). This process continued by setting the time and speed at the display of the mixer at 3 minutes and 2000rpm. Close the mixer and press the ”start” button. After 3 minutes, take off the container holder. As shown in Figure 3.14, the different before and after the user of the Thinky mixer.

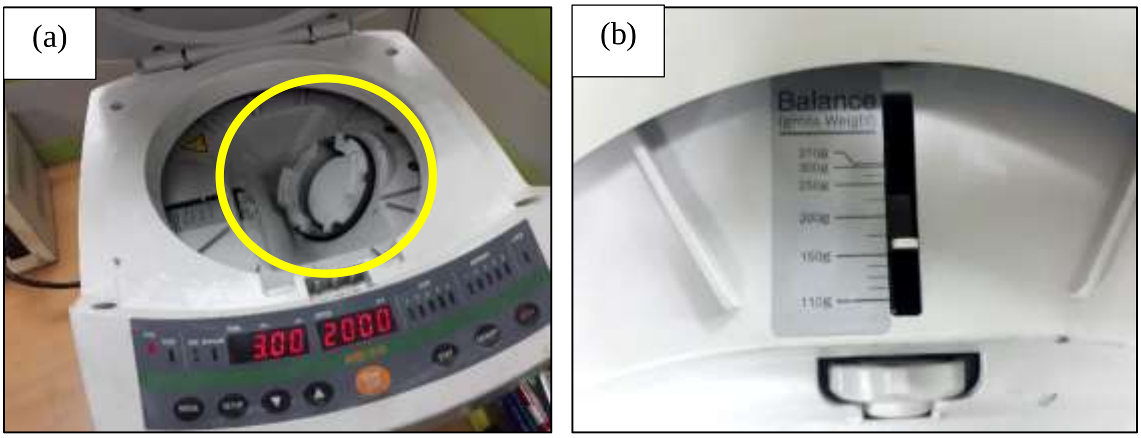


Figure 3.13: Calibration; (a) Container inside Mixer (b) Weight Balance

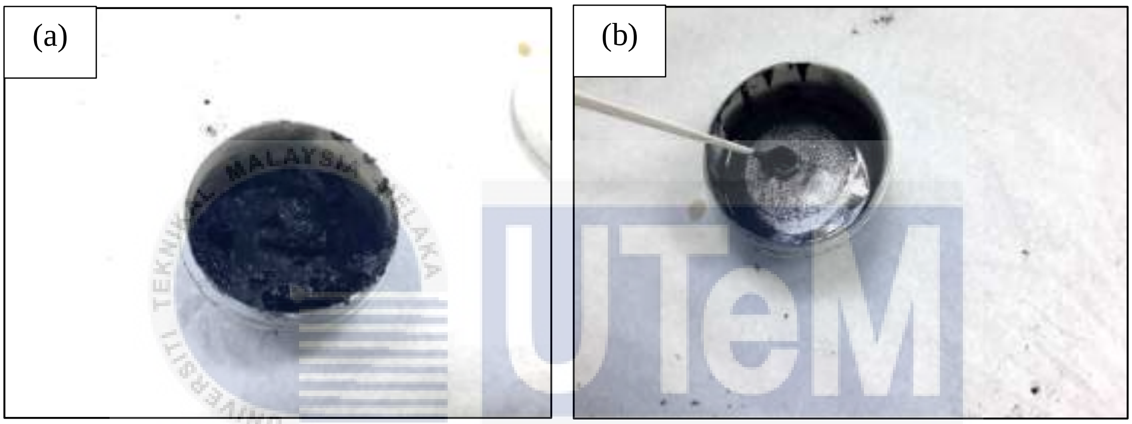


Figure 3.14: (a) Before Mixed (b) After Mixed

3.2.3.2 Printing process

Following the material preparation, the procedural steps are continued with the printing process. For this study, the printing process is carried out in order to various the width and pattern of the samples. There is a method that is implemented the concept in this printing process which is a casting method with the help of a doctor-blade technique. The casting process is usually used with a liquid composite that been put into the mold that followed the desired shape needed while the doctor-blade technique is the used of the blade to place the wet composite into the mold properly. The doctor blade is a technique that widely used in producing a thin film on the substrates (Technique, 2017). In this study of the behavior and properties, after the material preparation was done, the composite that has been mixed together with the filler, binder and the hardener is gone through a printing process. The printing process is started with glue together with the TPU substrate at the lower surface of the mold with the tape.

The TPU substrate must be clean first with the acetone in order to avoid any small particles that can damage the surface of the ink after the curing process. After the TPU substrate been glue together with the mold, the mold position been change by upside down it. The wet composite then been put on the top of the mold and here how the doctor-blade technique works. Based on Figure 3.15, it shows how the doctor-blade technique works. When the wet composite been put on the top of the mold surface, the blade starts to move constantly towards the direction shown in Figure 3.15. This blade is been controlled manually by human power. This doctor-blade has been used in order to give a lot of pressure on the wet composite for its spread in the mold by the shaped. This technique also lesser the space of air or bubbles in the wet composite. After the printing process is done, the TPU substrates been take off from the bottom of the mold. This process needs to be done slowly in order to avoid the ink stick on the mold. The sample is considered to fail if the wet composite did not take off from the mold. This printing process was repeated two times for each pattern and widths.

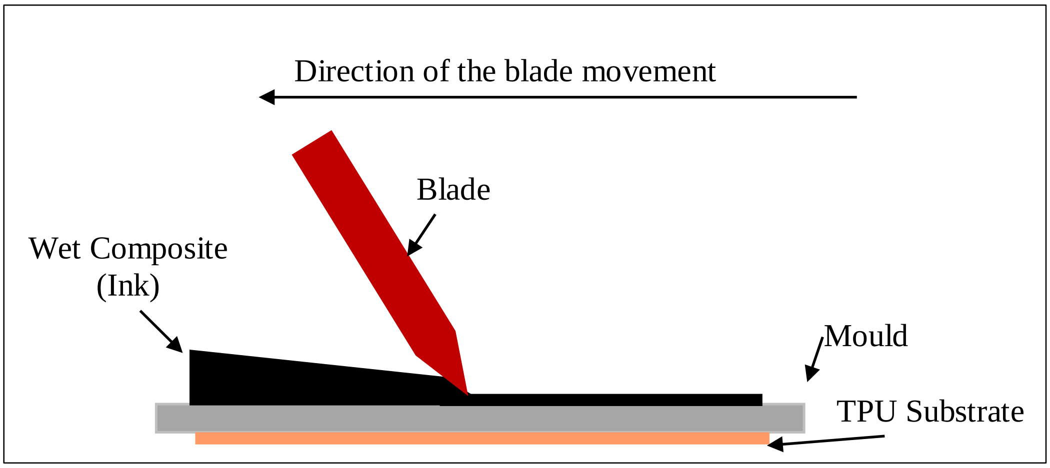


Figure 3.15: Doctor-blade Technique

Figure 3.16 (a) shows that the printing process during the experimental at the Amchal's laboratory. As there are four patterns (straight line, zigzag, curve, and square) and three different types of width, the samples that we produce during the printing process is around 24 samples as each width has 2 samples. This is because to find out the average reading during the properties test. Figure 3.16 (b) shows that the wet composite after the printing process.

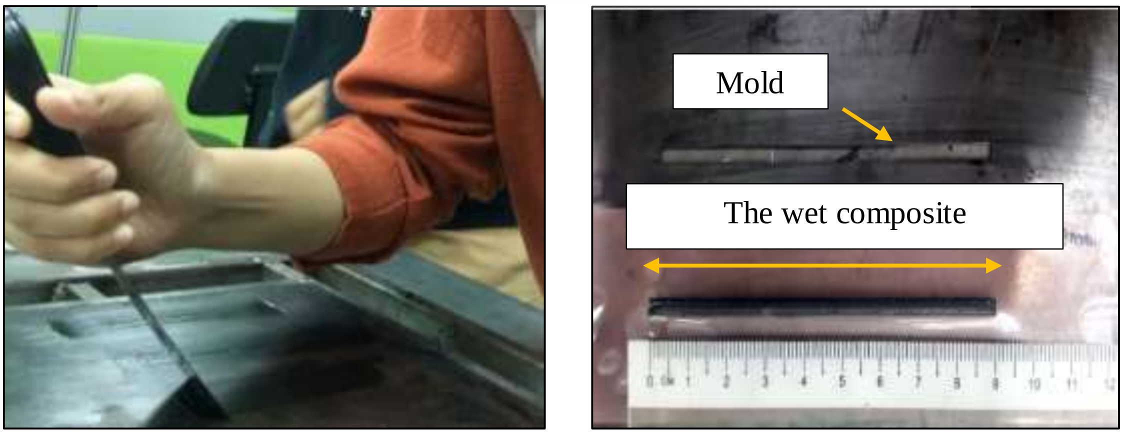


Figure 3.16: Printing process; (a) Printing the Composite (b) The Wet Composite

3.2.3.3 Curing process

The curing process is follow up after the printing process was done. The aim of this curing process is to cure the wet composite and turn it into a composite that has high strength bonds. After the printing process, the samples then have undergone a curing process by putting the samples on the tray and put the samples into the oven. This curing process is been set up for 30 minutes with 100°C of the temperature. This process also makes the wet composite growth in the high-temperature environment. After 30 minutes, the tray is put outside the oven and the samples been cooling process by using the natural air. This sample was then marked to five-point for the test as shown in Figure 3.17.

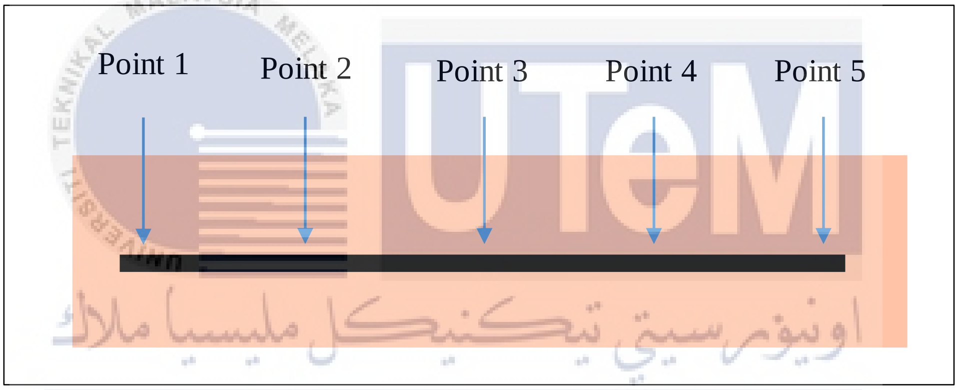


Figure 3.17: The Point for the Data Taken

3.2.3.4 Voltage and resistivity test

In this study, to obtain the reading of the voltage and resistivity on the conductive ink is by using the 4-point probe machine. The test is started by the switch on the machine and set up the voltmeter of the machine. This square meter must be clear from the previous data to avoid any continue data on the in the software. To clear the previous data, the button 'Enter' must be press for a quite sometimes around 5 seconds then the previous data will be erased from the voltmeter memory. The personal computer (PC) must be connected to this Jandel machine software, as the data will be logging into this software automatically. In this study, the current that been put is on 10μA and this current been applied for all samples. After the setup process is done, the sample is been put on the base under the probe as shown in Figure 3.18.

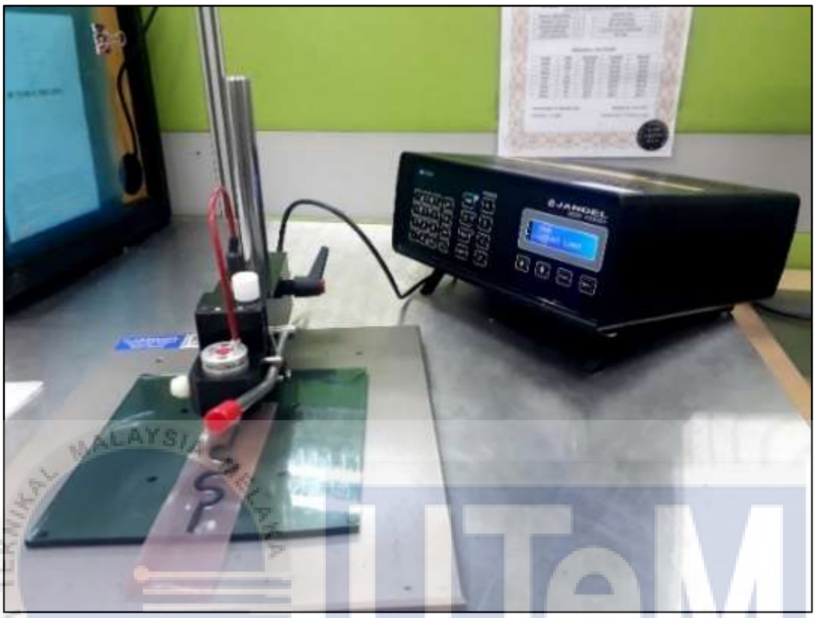


Figure 3.18: 4-Point Probe Machine

This test is been done by adjusting the position of the cylindrical probe near the surface of the sample by using the twirl lever beside the cylindrical probe. Before the cylindrical probe been move (refer to Figure 3.19 (a)), there is a gap between the probe and the sample while after the probe been move, the probe is touch the sample surface as shown in Figure 3.19 (b). When the twirl lever goes down, the cylindrical probe goes down and touches the surface of the sample. When the probe touches the surface of the sample, the voltmeter will read the voltage of the sample in the millivolt (mV) and resistivity in ohm/sq (Ω/sq). The voltmeter displayed the reading of the voltage. In order to log the data into the software automatically, the data at the voltmeter must be saved. By entering the 'enter' button, data is logging into the software. After that, the data need to be export into a folder.

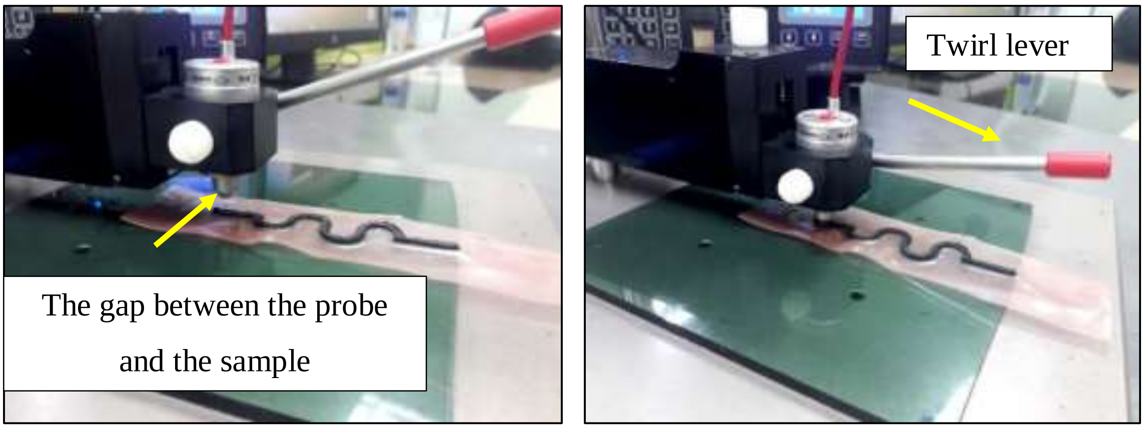


Figure 3.19: 4-Point Probe Test; (a) The Probe has a Gap Between Sample (b) The Probe Touch the Sample Surface

3.2.3.5 Microstructure analysis

After the sample undergoes voltage and resistivity tests, the sample proceeds with the microstructure test by using an image analyzer test. The aim of this test is to analysis the microstructure of the sample. This study of microstructure analysis will be using a digital image processing technique. (Sarojadevi, Shetty, & Murthy, 2013). This technique will be processing a clear image. The test of this microstructure analysis is been done by the switch on the personal computer (PC) and press the 'on' button at the image analyzer which places at the side of the image analyzer. Then, click the Isolution Lite software in the PC and when the software pop up at the screen, click the image 'camera'. The new window will pop out at the right corner of the screen. The sample then is put on the mechanical stage as shown in Figure 3.20 where the objective lens on the top of the sample surface. Then click the 'live' on the software in order to view the microstructure of the sample. In order to find a clear image, the fine focus must be adjusted until the clear image appears in the software. After the clear image appears in the software, the image must be captured and saved in a folder. Besides that, the sample undergone the SEM and EDX test in order to identify the effect of the properties graphene on conductive ink.

Figure 3.20: Sample on the Image Analyser

3.2.3.6 Hardness test

The samples also have undergone a hardness test by using the nanoindenter machine. Based on this test, the data on the hardness of the composite can be determined. This hardness test is important has the conductive ink must be flexible enough. To start the hardness test, the sample is placed on the mechanical stage of the nanoindenter. For the adjustment, it can be done manually as shown in Figure 3.21. In order to make sure the data can be read clearly, the samples must be on focus and in the right position.

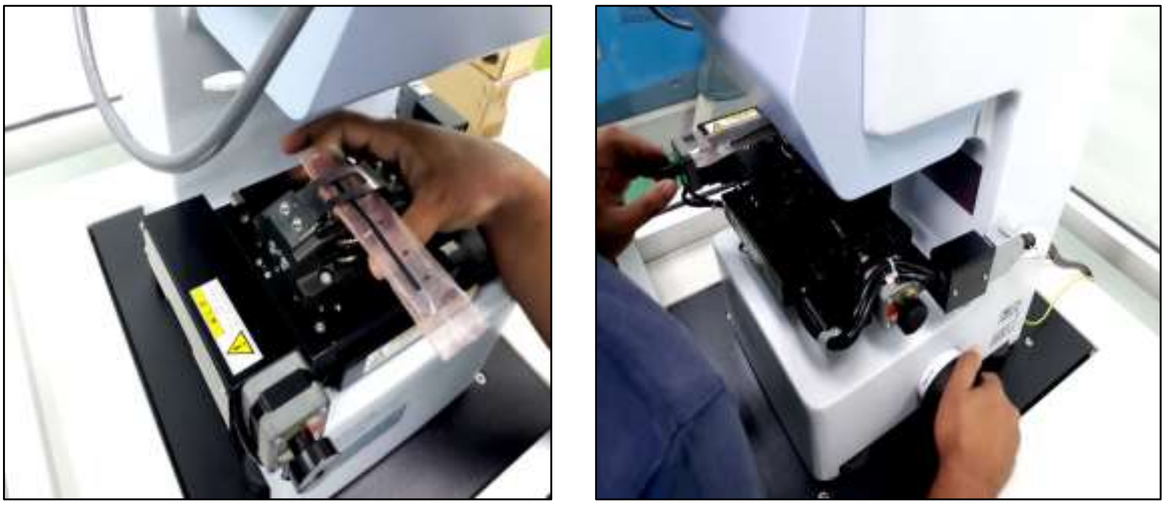


Figure 3.21: Hardness Test; (a) Sample on the Machine (b) Reposition sample

This hardness test is started from the windows “Start” menu and open the DUH software. The focus of the lens can be adjusted and the process repositions of the sample by using this software automatically. When the test is started, the reading of the hardness will be shown in terms of the graph. After the graph is finished, the data need to be saved in a folder.

CHAPTER 4

RESULTS AND DISCUSSIONS

4.1 Overview

This chapter is divided into five sub-chapter on the test results and three on discussion. For the five first sub-chapter, it explained on the test that has been done while for the three sub-chapter, it emphasises the analysis on all the results and the relationship between properties and behaviour on the performance of conductive ink

4.2 Properties of voltage

In this study, all graphene-based conductive ink samples undertaken in voltage test by using the 4-point probe. This voltage test was done to figure out the effect of the voltage on the current flow in the samples of ink. Voltage is an electrical pressure, which forces the current flow in the specific material which conductive material (Patrick & Fardo, 2008). Under these circumstances, it shows that the greater the voltage the greater the current flow in the material. To make sure that the samples have an electric force and can drive the electric current along with the samples, all the data on the voltage test has been taken and be analysed to figure out which pattern and width have the greatest voltage.

4.2.1 Voltage test

Table 4.1 shown that the graph on data taken of voltage in straight-line samples while Table 4.2 displayed the graph on zigzag samples. Table 4.3 and Table 4.4 are curve and square patterns graphs that have been analysed. All the graphs is based on the data taken during running experimental tests on each sample. The data taken is been tabulated as attached in APPENDIX A.

Table 4.1: The Graph on Data of Voltage for Straight Line Pattern

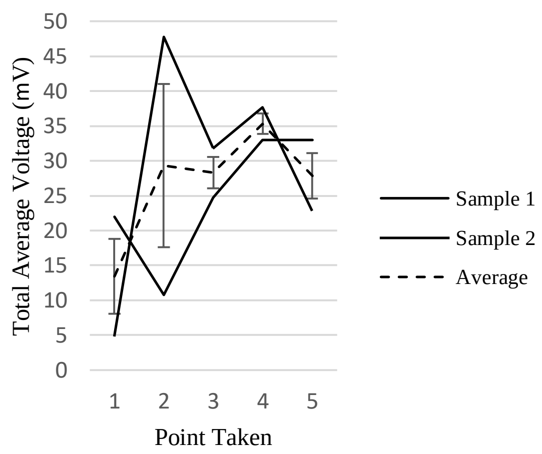
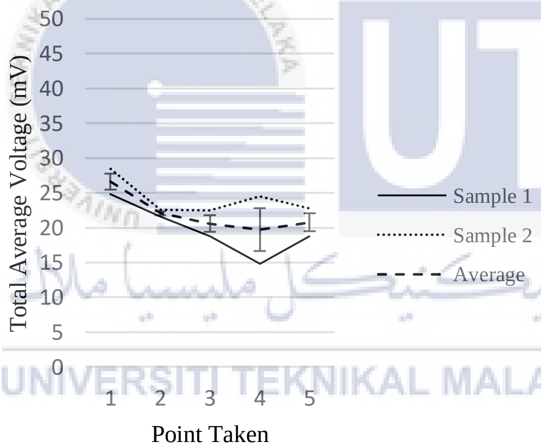
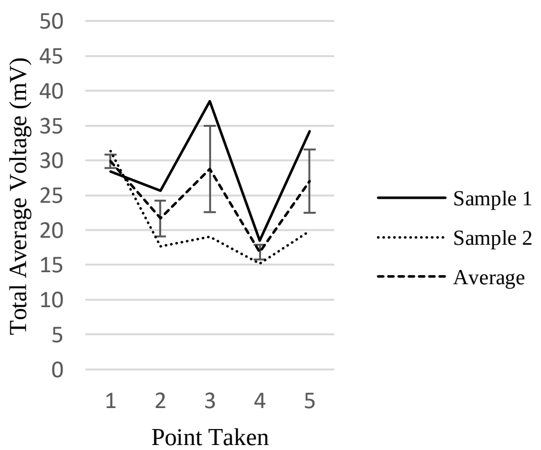
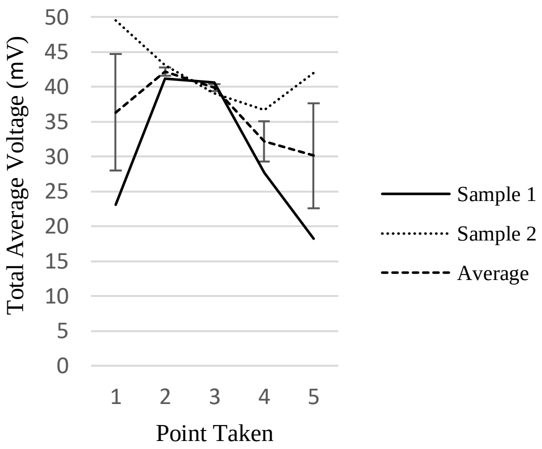
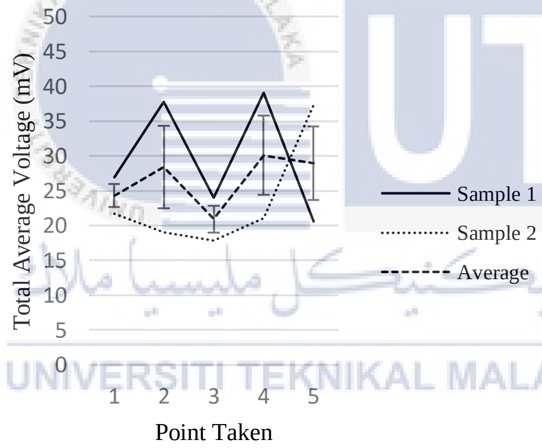
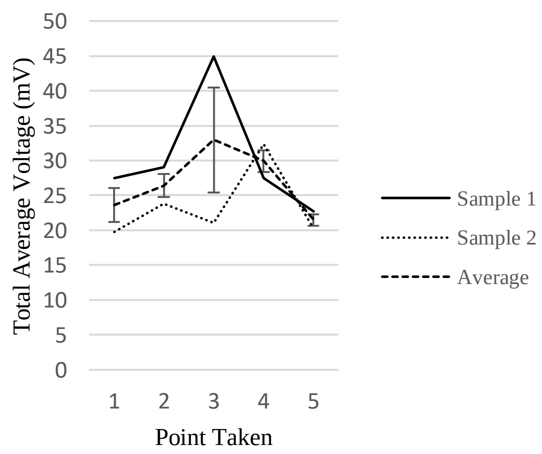
Width (mm)	Graph on Data of Voltage	Analysis
1		As shown in the graph, point 4 has the highest average voltage reading while point 1 has the lowest reading. Besides that, Point 2 has the biggest gap for error bar between sample 1 and sample 2, which contrast with point 4. Hence, point 2 has the lowest reliability.
2		This data showed a slightly difference in data reading between sample 1 and sample 2. Point 1 has the highest reading while point 4 has the lowest reading of voltage.
3		Graph of 3mm width sample show the highest average reading of voltage compared to 2mm and 1mm samples. On the other hand, the highest average reading of voltage is at point 1 and vice versa with point 4.

Table 4.2: The Graph on Data of Voltage for Zigzag Pattern

<table>
<tr><th>Width (mm)</th><th>Graph on Data of Voltage</th><th>Analysis</th></tr>
<tr><td>1</td><td>


</td><td>The highest average value between sample 1 and sample 2 is at point 2 which 42.16mV while contrast with point 5 that has 30.14mV of voltage reading. Both point 1 and point 5 has low reliability in data taken at the same time both point 2, and point 3 has high reliability.</td></tr>
<tr><td>2</td><td>


</td><td>Through the graph, the average reading taken has the highest value at point 4 and lowest value at point 3. The reliability for data for both sample 1 and sample 2 is low in each point. The average reading can be affected by the reliability of data between these two samples.</td></tr>
<tr><td>3</td><td>


</td><td>The graph showed that reading started to gradually increase at point 3 even though there is biggest gap between reading on sample 1 and sample 2. Point 5 showed the lowest average reading of voltage as both samples have lowest reading of voltage.</td></tr>
</table>

Table 4.3: The Graph on Data of Voltage for Curve Pattern

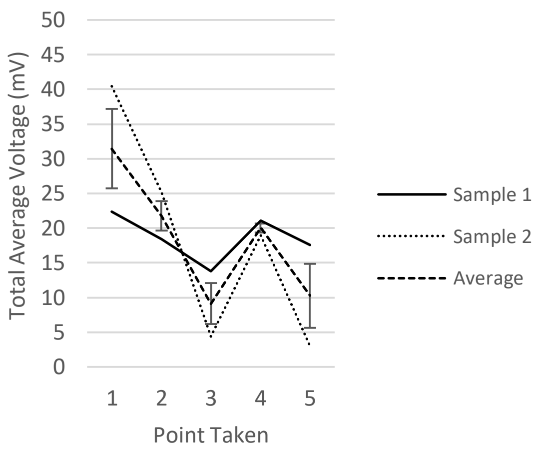
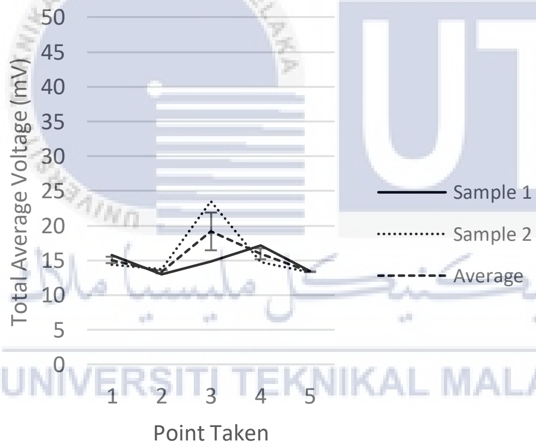
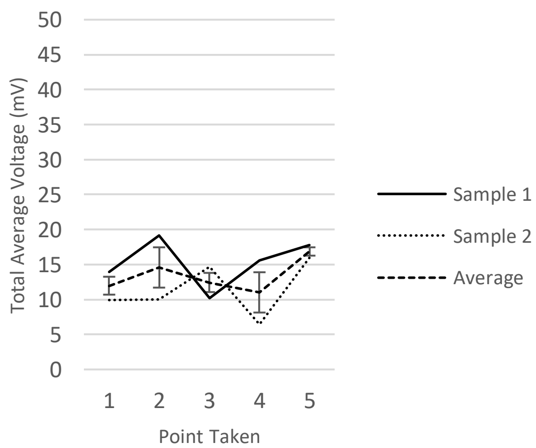
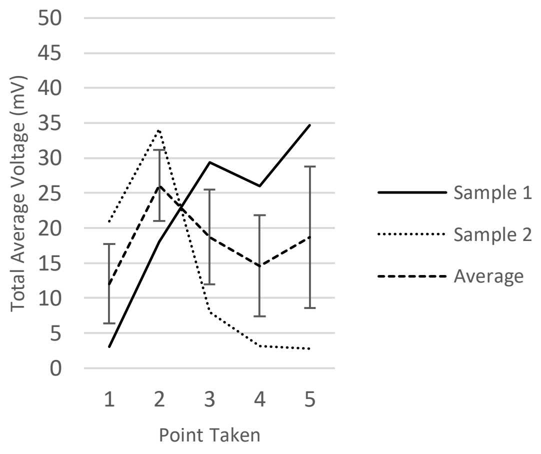
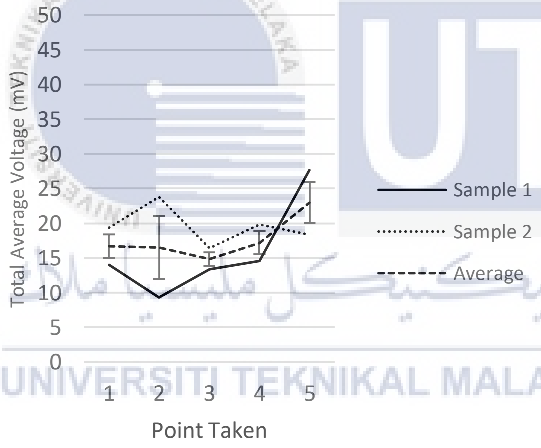
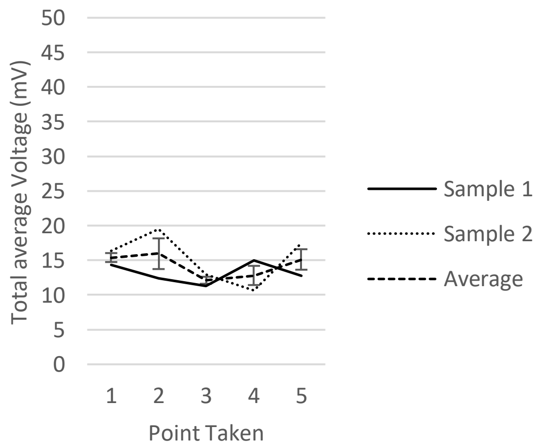
Width (mm)	Graph on Data of Voltage	Analysis
1		Based on the graph, the data taken is in nano measurement, which make 1mm sample, has the least reading of voltage compared to 2mm and 3mm samples. As shown, the reading is decreased rapidly from point 1 until point 3 and increased slightly at point 4.
2		2mm sample shows a little bit different in average voltage reading which in the range from 13mV to 20mV. The reliability of the data on sample 1 and sample 2 also did not show much different.
3		From the graph, the average voltage value is higher at point 5 while lower value at point 4. Both point 2 and point 4 show a bigger gap between sample 1 and sample 2 in voltage reading.

Table 4.4: The Graph on Data of Voltage for Square Pattern

Width (mm)	Graph on Data of Voltage	Analysis
1		The average value for voltage at point 2 is the highest while the lowest value is at point 5. As shown in graph, this reading have the biggest gap between sample 1 and sample 2 for all point.
2		The graph illustrates high average voltage at point 5 and low value at point 3. The average reading is in a range between 14mV to 23mV.
3		Point 2 has the highest value of average reading of voltage while contrast with point 3 which has the lowest value. This graph shows a little difference in voltage for all point except point 2.

4.3 Properties of sheet resistivity

Resistivity is described by how material resists the flow of electricity in the electrical circuit (Pickover, 2008). The unit for resistivity is in ohm per square (Ω/sq) which in order to distinguish between sheet resistivity and resistance (Shahat, 2017). Sheet resistivity is performed using a four-point probe machine due to its independent of the square and low resistivity of the thin film (Serway, Jewett & Peroomian, 2019). Past study has stated that material has a low resistivity when it good and has the ability to conduct electricity easily (Taherian & Kausar, 2019). In this study, the material that been used is graphene which is a conductive material while graphene has low resistivity with ($10x^{-6}$ Ωcm). This sheet resistivity test is conducted to figure out the ability of graphene in assisting binder and hardener as a composition to conduct electricity.

4.3.1 Sheet resistivity test

Sheet resistivity is conducted by using a four-point probe machine where all the samples are taken part to figure out the characterization samples in resistivity. Table 4.5, Table 4.6, Table 4.7, and Table 4.8 are represented data of sheet resistivity reading that taken and transforms into the form of a graph. All the data taken during experimentally can be referred in APPENDIX B. Generally, based on findings, 1mm has the highest reading of resistivity compared to other widths. The data will be compared by pattern and width to figure out which pattern and width have the lowest sheet resistivity.

Table 4.5: The Graph on Data of Resistivity for Straight Line Pattern

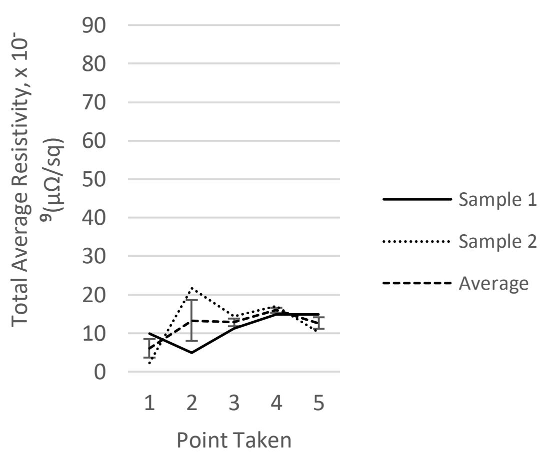
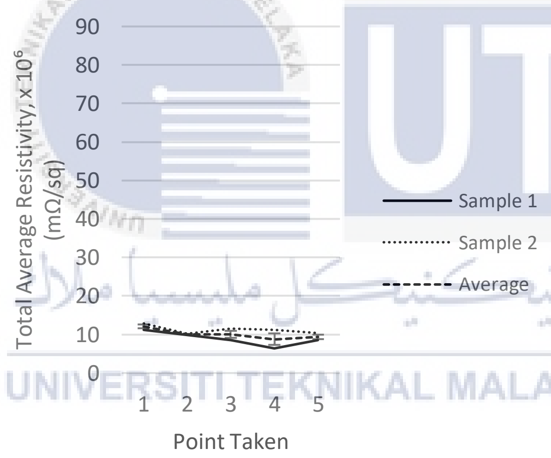
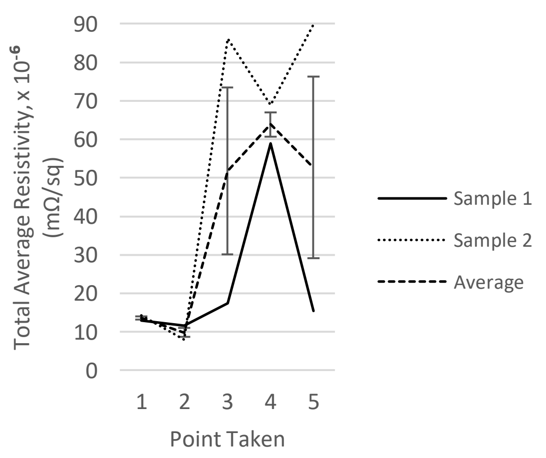
Width (mm)	Graph on Data of Resistivity	Analysis
1		It has low resistivity that is in range 2.252 µΩ/sq to 21.662 µΩ/sq, which compared to sample 2mm and 3mm. The highest value of resistivity is at point 4 while contrast with point 1. The graph shows that the average resistivity is increases and slightly decrease at point 4.
2		This area has moderate average resistivity compare to previous sample and show a little gap between other points. The average resistivity has the lowest value at point 4 and vice versa with point 1. Point 4 also has the biggest gap between sample 1 and sample 2.
3		The graph illustrates high value of resistivity with slightly bigger gap between sample 1 and sample 2 which at point 4 while point 2 has the lowest value. This sample has the highest value of average resistivity compared to others samples because it has big width of sample.

Table 4.6: The Graph on Data of Resistivity for Zigzag Pattern

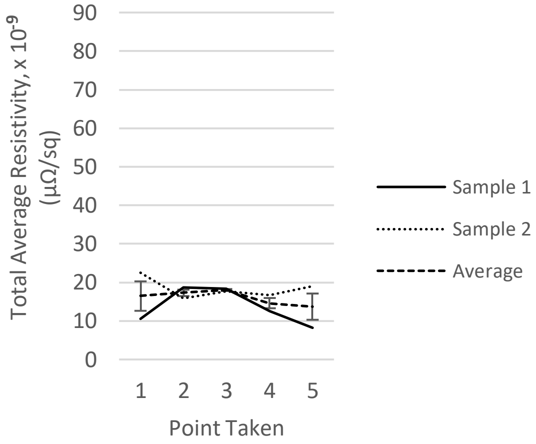
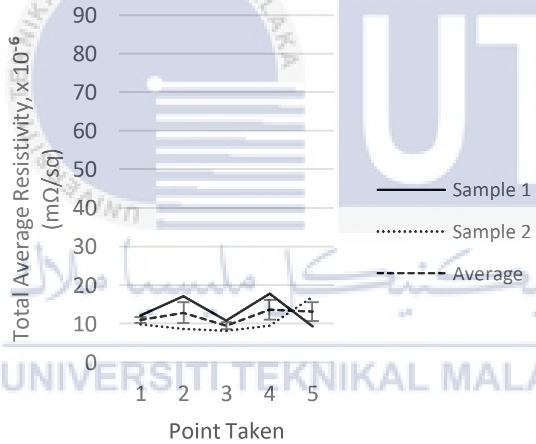
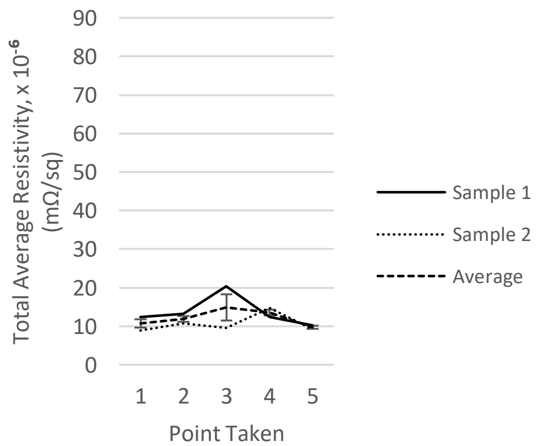
<table>
<tr><th>Width (mm)</th><th>Graph on Data of Resistivity</th><th>Analysis</th></tr>
<tr><td>1</td><td>
</td><td>The value of average resistivity is increase from point 1 until point 3 and decrease. The error bar can barely be seen at each points except at point 3. This is show that the average value of resistivity has a low certainty.</td></tr>
<tr><td>2</td><td>
</td><td>Through the graph, resistivity has shown fluctuate pattern for all points. The highest value of resistivity is at point 4 while point 3 has the lowest value. The error bar is in big dimension for almost each points, which make sample, has low reliability.</td></tr>
<tr><td>3</td><td>
</td><td>The data displays up to not much difference among all of the points except point 3. Error bar can barely be seen at point 3 since it has the highest and lowest value of resistivity for sample 1 and sample 2.</td></tr>
</table>

Table 4.7: The Graph on Data of Resistivity for Curve Pattern

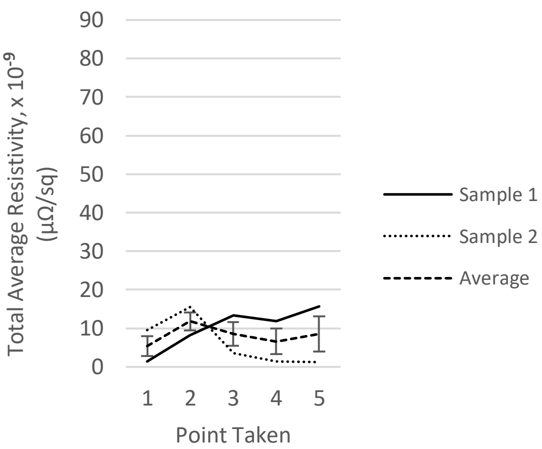
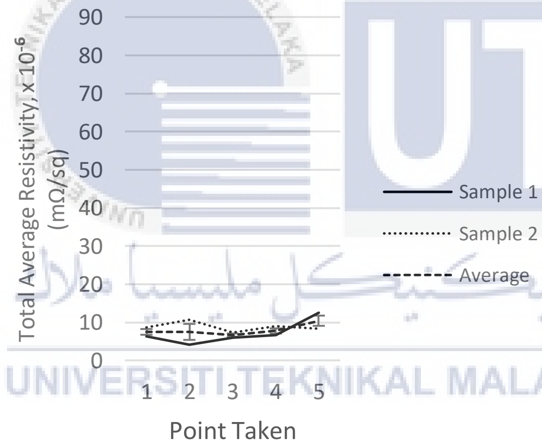
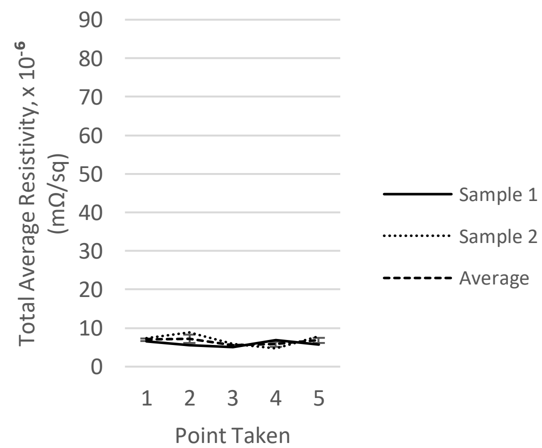
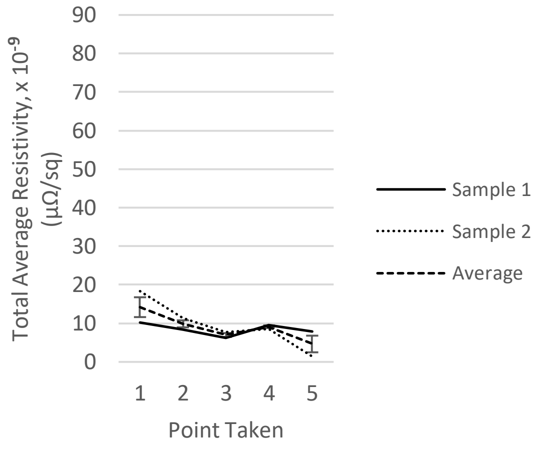
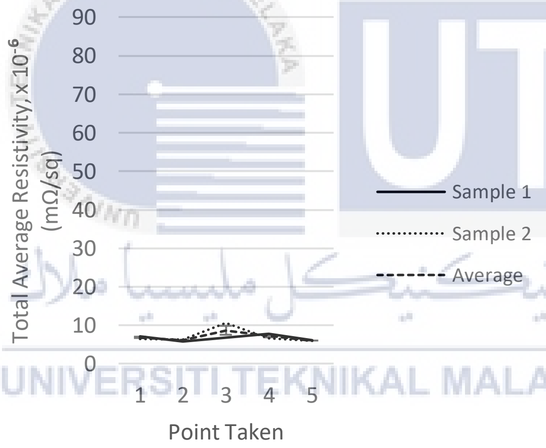
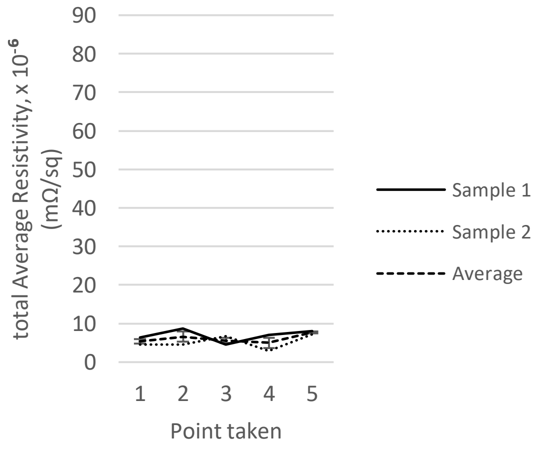
Width (mm)	Graph on Data of Resistivity	Analysis
1		The average value for resistivity at point 2 is the highest while the lowest value is at point 1. As shown in graph, this reading have the biggest gap between sample 1 and sample 2 for all points, which make it, has low certainty.
2		The graph illustrates high average resistivity at point 5 and low value at point 3. The average reading is in a range between 7 mΩ/sq to 10 mΩ/sq .
3		Point 2 has the highest value of average resistivity while contrast with point 3 which has the lowest value. This graph shows a little difference in resistivity value for all point except point 2.

Table 4.8: The Graph on Data of Resistivity for Square Pattern

Width (mm)	Graph on Data of Resistivity	Analysis
1		Based on the graph, the data taken is in nano measurement, which make 1mm sample, has the lowest reading of resistivity compared to 2mm and 3mm samples. As shown, the reading is decreased rapidly from point 1 until point 3 and increased slightly at point 4.
2		2mm sample shows a little bit different in average resistivity reading which in the range from 6 mΩ/sq to 9 mΩ/sq. The reliability of the data on sample 1 and sample 2 also did not show much different except at point which the error bar has bigger gap.
3		From the graph, the average resistivity value is higher at point 5 while lower value at point 4. Both point 2 and point 4 show a bigger gap of error bar between sample 1 and sample 2 in resistivity reading.

4.4 Properties of hardness

Hardness Vickers is the measurement of hardness value in micro size on the specimen. The graphene-based conductive ink samples undergone the micro-hardness test to investigate the ability of the material to resist local plastic deformation by standardized loading. The hardness of each sample was measured and constant load at 6mN load was applied during the test. The findings in this test is been tabulated and to simplified the results of this micro-hardness test the data that has been tabulated is been transfer in the form of a graph. From the observation of the hardness test, there were variations in hardness values due to different contents of hardener in the ink. Table 4.9, Table 4.10, Table 4.11 and Table 4.12 show the hardness results for each samples based on each pattern and width. All the data taken is attached in APPENDIX C.

Table 4.9: The Graph of Hardness for Straight Line Pattern

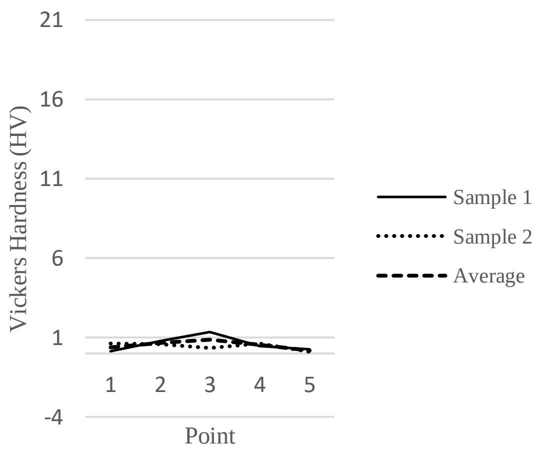
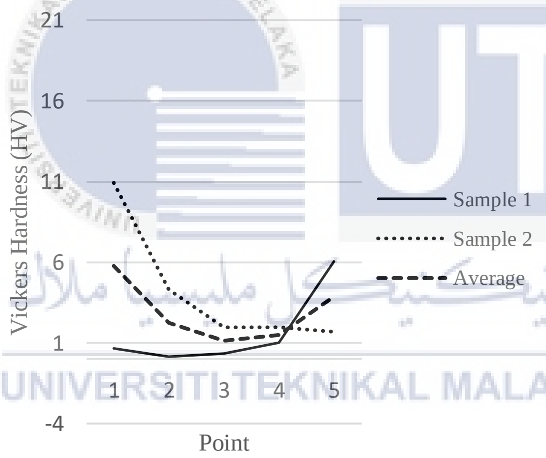
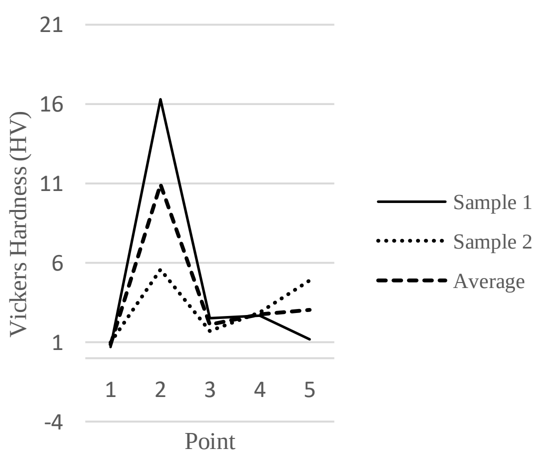
Width (mm)	Graph on Data of Resistivity	Analysis
1		From the graph, point 3 has the highest average hardness value, which is 0.868 HV while point 5 has the lowest average value with 0.191 HV.
2		Through the graph, point 1 has shown the higher value of hardness while point 3 has the lowest value. The graph show that the average value dropping start at point 2 and increase at point 4.
3		As shown in graph, point 2 has the highest average value of hardness while point 1 has the lowest value. The range of the hardness for this sample is from 10.946 HV to 0.859 HV.

Table 4.10: The Graph of Hardness for Zigzag Pattern

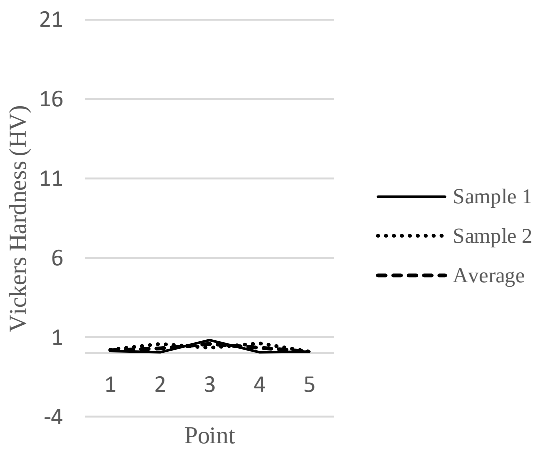
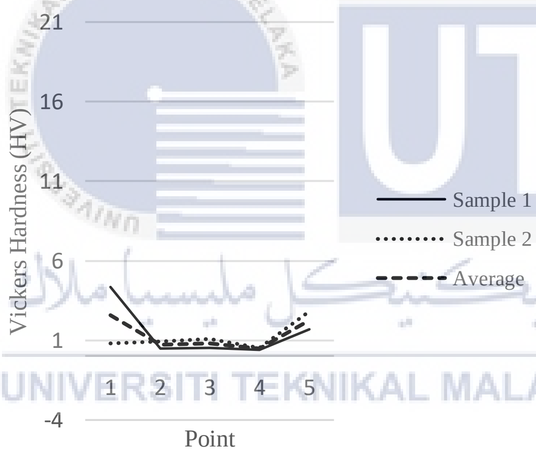
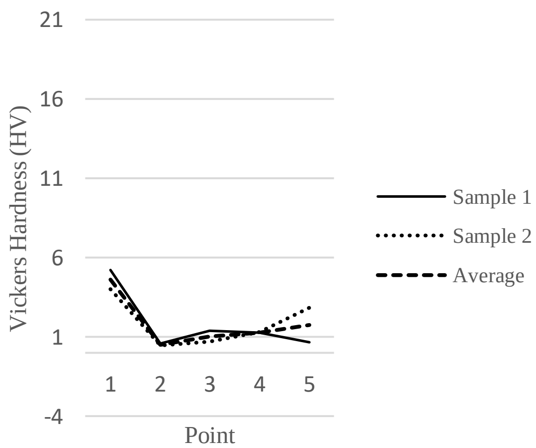
Width (mm)	Graph on Data of Resistivity	Analysis
1		The graph illustrates that the fluctuate pattern of graph. As shown, the highest average value of hardness is at point 3 and the lowest value at point 5.
2		The average value of hardness is high at point 1 and point 5, which contrast with other points.
3		The graph illustrates a fluctuate pattern of hardness data. The pattern described as the average value of hardness decreases at point 2 and continuously increases starting at point 3.

Table 4.11: The Graph of Hardness for Curve Pattern

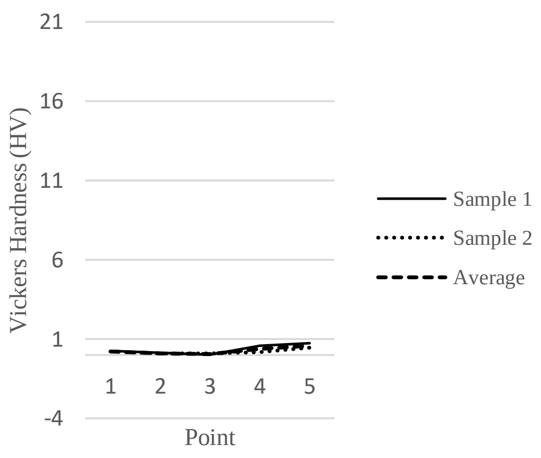
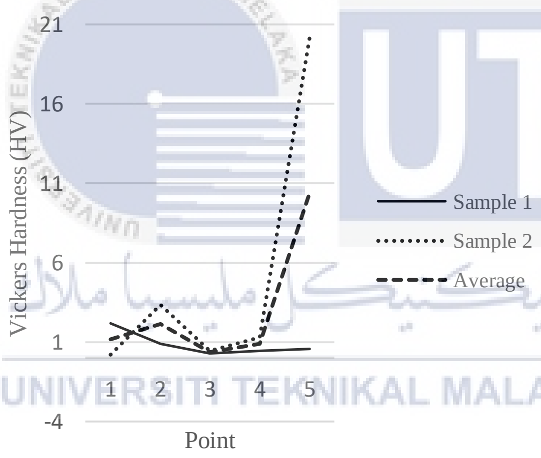
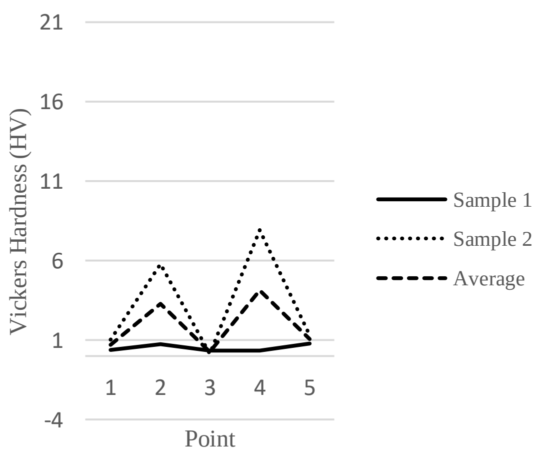
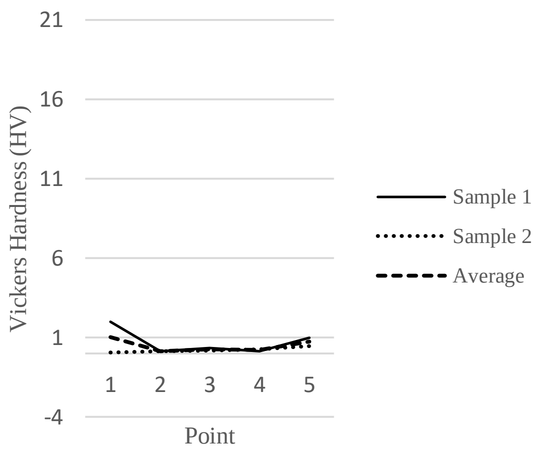
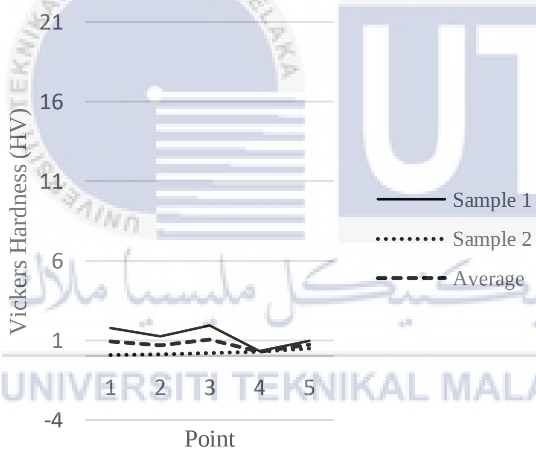
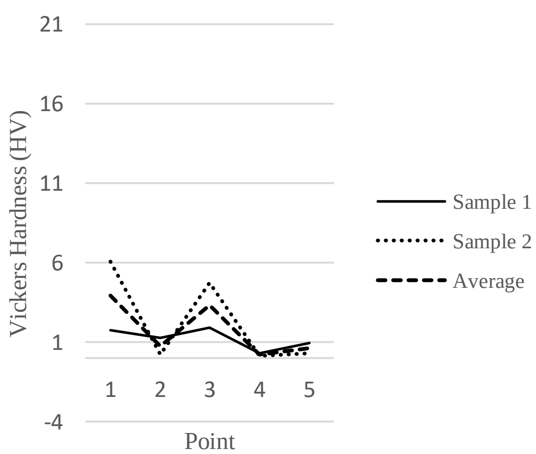
Width (mm)	Graph on Data of Resistivity	Analysis
1		This sample has average value in the range between 0.065 HV to 0.597 HV. The graph is shows that the average value started to drop at point 2 and has the lowest value at point 3. The graph start to increase at point 4 and reach the maximum value at point 5.
2		It has average hardness value between 0.185 HV to 20.183 HV. This sample has the highest value of hardness compared to 1mm and 3mm samples.
3		The graph shown fluctuate pattern of data where the average value of hardness keep increases and decreases continuously. The highest value is at point 4, which contrast with point 3.

Table 4.12: The Graph of Hardness for Square Pattern

Width (mm)	Graph on Data of Resistivity	Analysis
1		From the graph, point 1 has the highest average hardness value, which is 1.029 HV while point 2 has the lowest average value with 0.130 HV.
2		Through the graph, point 3 has shown the higher value of hardness while point 4 has the lowest value. The graph show that the fluctuate pattern of data as the data increase and decrease continuously.
3		As shown in graph, point 1 has the highest average value of hardness while point 4 has the lowest value. The range of the hardness for this sample is from 3.916 HV to 0.221 HV.

4.5 Characterization of microstructure

In this graphene-based conductive ink study, microstructure analysis has been done to determine the microscopic condition of the composite based on the ink surface. This analysis is one of the methods that used to find out the factors that might affect the physical properties of the ink. Physical properties is necessary for this study as structure might affect the performance of the ink during conducting electricity. Therefore, by did a microstructure analysis, some behavior of the ink composition can be recognized, and most importantly, prediction on the failure of the ink can be made, and the factors that caused the failure can be determined.

Besides that, there might be agglomeration effect that occurs in the composition of the ink. Agglomeration effect is where particles of the materials tend to agglomerate together and caused resistance. Particle agglomeration tends to reduce the strength of material even though the agglomerate may be strong enough to increases the initial modulus. Within the material, agglomerates are weak points and break easily when stress is applied. A broken agglomerate then acts as a strong concentrator of stress. Besides, since agglomerates are larger than the primary filler particles, it can produce weaker materials than composites containing the dispersed particles (Feng, Zhu, Liu & Wang, 2019).

Many factors might affect the performance of ink thus; microstructure analysis was done on each samples of the conductive ink. This microstructure analysis was done by used two different analyses, which are morphological analysis by using image analyzer and by images from Scanning Electron Microscopy (SEM) with Energy Dispersive X-Ray (EDX) analyzer. The results of both analyses will be discussed based on the images captured.

4.5.1 Morphological test

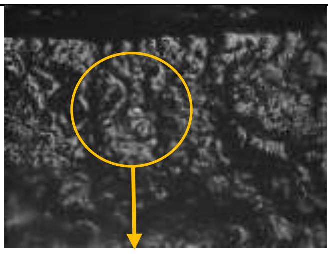
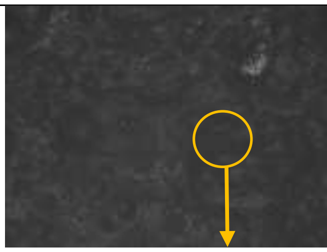
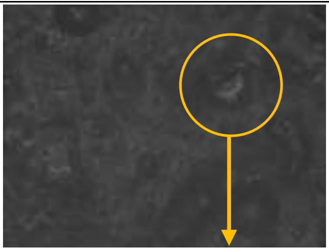
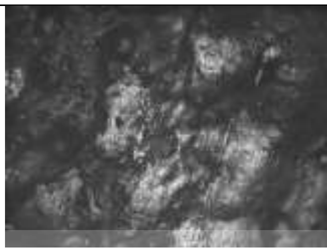
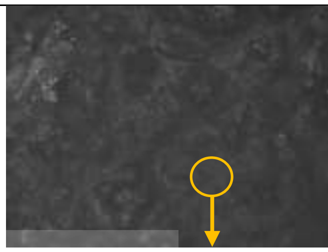
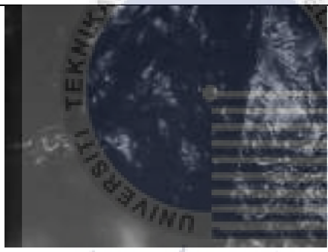
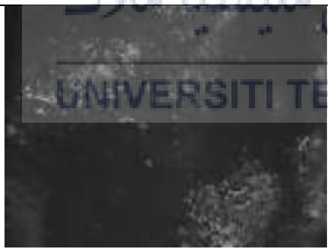
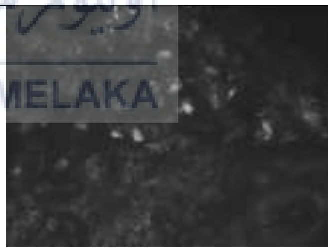
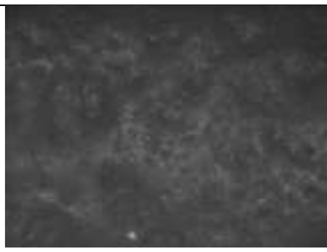
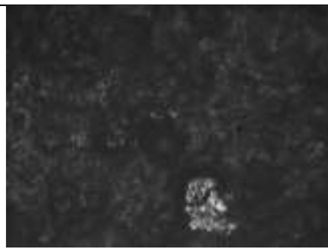
In this study, morphological analysis was done on the surface by using images captured by the image analyzer. The scales of the magnification on the microstructure images are 10x as shown in the table in the next section. This study was done to figure out the relationship between morphological surfaces with electrical conductivity of the graphene-based conductive ink samples.

4.5.1.1 Morphological analysis on straight line pattern

Based on Table 4.13, it shows that the optical images that captured by image analyzer on straight-line samples. By referring to the table, all images show surfaces that have similarities for each point but the contrast with different widths. Roughly, the optical images show a wrinkle surface and microspores on top of surface structure.

By referring images in the table, the sample of graphene-based conductive ink illustrates the similarity form of the surface but slightly different on the amount of presence. Based on the observation of the 1mm sample, each point shows a crude surface with many wrinkles. These wrinkles can be seen under this 10x scales of a magnification image analyzer. Next, the 2mm and 3mm samples show that smooth surface but more microspores on the top of the surface. These pores can be easily on each point for both samples. The difference between these two samples is the number of pores where samples with 2mm width have more pores compared to 3mm samples. This porous surface was present due to oxidation during the printing process. This also shows that the biggest the width the lesser the microspores on the top surface and have the lower the resistance. Besides that, the light area is bigger than the dark area as the amount of binder is 65% compared to 35% of filler. The dark area has high conductivity, as graphene is conductive materials.

Table 4.13: Optical Images of Straight Line Samples

Point	Width (mm)		
	1	2	3
1	 Wrinkles	 Dark area	 Pores
2			 Light area
3			
4			
5			

4.5.1.2 Morphological analysis on a zigzag pattern

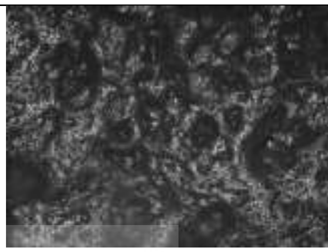
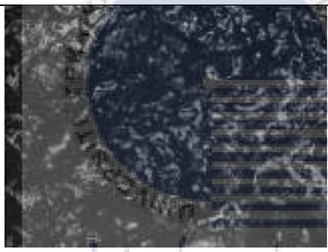
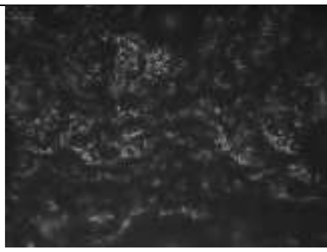
Based on the observation in Table 4.14, the table shows that the optical images captured by the image analyzer on a sample of the zigzag pattern. Five points were marked on each sample of 1mm, 2mm, and 3mm width. Each point undergoes image analysis as shown in the table below. Generally, for each width, these optical images illustrate similarity on all of the points except for 1mm sample, as there is a difference for some points.

In the first place, the sample of zigzag pattern with 1mm width shows inconsistency in the optical images. As shown, point 1 and point 5 has similarity as it has less crude on the top surface compared to other points. Even though there is a difference in some points, these images show similarities by having many microspores, especially in the middle area. In line with this, microspores can be seen especially at point 3.

For samples with 2mm width, the optical images show sameness of top surface structure for each point. Image of point 2 shows the most presence of pores compared to other points. Based on the observation, point 3 and point 4 shows the most smooth surface and fewer pores. This shows that the porous and crude surface is different for each point even in the same sample. Besides that, the structure of the top surface of point 2 illustrates the cruder surface while point 3 shows the less crude of the surface. This crude and pores surface can be related to the resistance in which the cruder surface the higher the resistance for the current flow. It goes to the porous surface; it can be one of the physical resistance to the current flow.

Lastly, the table also shows the optical images of the 3mm width sample of the zigzag pattern. Based on the images, point 3 shows the fewer pores on the surface and more smooth compared to others. This is an interesting study as the wider the width of the sample the less the pores on top of the surface.

Table 4.14: Optical Images of Zigzag Sample

Point	Width (mm)		
	1	2	3
1			
2			
3			
4			
5			

4.5.1.3 Morphological analysis of curve pattern

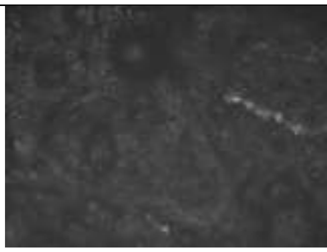
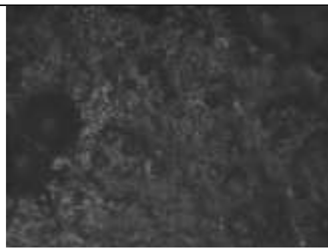
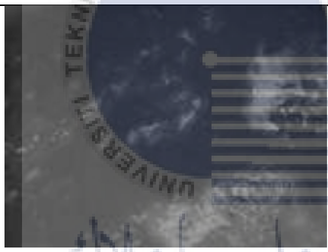
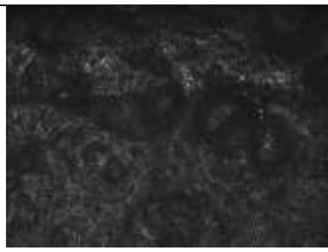
As can be seen in Table 4.15, the microstructure of the curve pattern for all three different width shows some differences regarding the surface structure and the porosity. Optical images for 1mm sample show some similarity for each point as it can be seen each image has a lot of porosity which makes this sample has high resistance and low conductivity. Besides that, the sample for 1mm has a cruder top surface compared to 2mm and 3mm samples. This crude surface might affect the flow of electricity, which makes the resistance increases.

For optical images for a 2mm sample, all the images for each point show some consistency regarding the top surface. The porous on the top surface can be seen in the images and it less than 1mm sample. The amount of porosity is consistent for each point. In this case, a 2mm sample has consistency regarding the flow of electricity and has high conductivity compared to a 1mm sample.

3mm sample has the highest conductivity compared to others sample has it is the biggest width compared to others. Besides that, this sample optical image shows that the less porous on the top of the surface compared to 1mm and 2mm samples. This makes the sample has the least resistance compared to others.

Lastly, for this curve pattern, it can be seen that a 3mm sample has the highest conductivity because it has a better microstructure compare to 1mm and 2mm samples which vice versa with 1mm, which has the lowest conductivity.

Table 4.15: Optical Images of Curve Pattern

Point	Width (mm)		
	1	2	3
1			
2			
3			
4			
5			

4.5.1.4 Morphological analysis on a square pattern

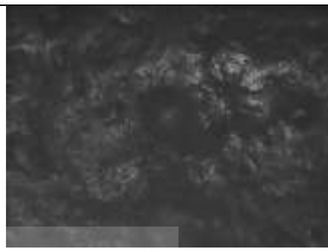
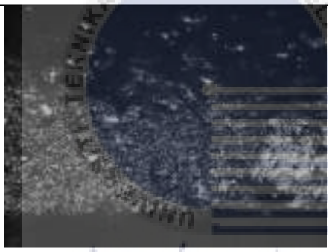
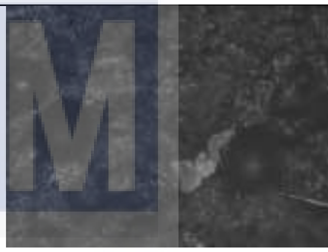
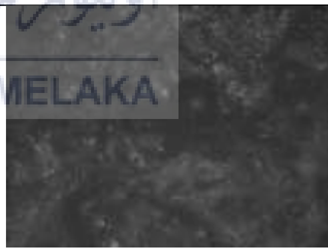
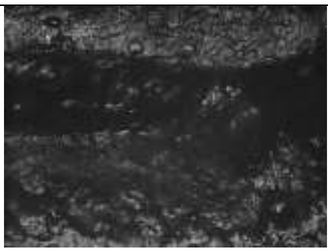
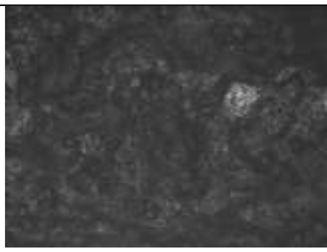
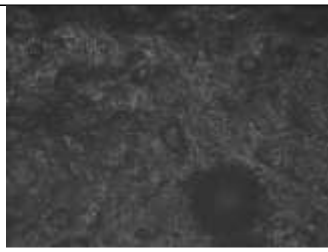
For this square pattern, all three different widths have been analysed through a microscopic analyser. All the optical images were captured and shown in Table 4.16. Based on this table, it can be seen that all the optical images shows some differences, especially at a 1mm sample. Sample 2mm and 3mm show some similarities regarding the porosity and the microstructure of the top surface.

For a 1mm sample, as it has the smallest width and top surface compared to other samples, it shows a lot of porosity, which might affect the flow of electricity. This is because this sample has the highest resistance compared to others. The top surface also has the crudest surface compared to others. Sample 1mm has many wrinkles on top of the surface compared to others. This might occur during the printing process.

As for sample 2mm, there is no much difference between this sample and sample for 3mm. Even though, these two sample shows slightly different still 2mm has more porosity on the top surface compared to 3mm. Based on this, it can be seen that the size of the sample plays an important role in conducting electricity. Besides that, the presence of filler, binder n hardener can be seen based on the dark and light area. The dark area is represented as filler, which is conductive materials while the light area is binder n hardener, which is non-conductive materials. The continuous dark area shows that electricity can be flow over this surface as there is the presence of conductive materials. Based on these 2mm and 3mm samples, 3mm shows darker area compared to 2mm and make 3mm has more an ability to conduct electricity.

In conclusion, as same as other samples, the 3mm width of sample has the less porosity compared to others because it is has the biggest size of samples. The 3mm sample also has the highest conductivity compared to others.

Table 4.16: Optical Images of Square Pattern

Point	Width (mm)		
	1	2	3
1			
2			
3			
4			
5			

4.5.2 Scanning Electron Microscopy (SEM) with energy dispersive X-Ray (EDX)

In this SEM and EDX analysis, the percolation threshold can be observed through the presence of the high contrast area, which represents the high number of electron and low contrast as less number of electron. The percolation threshold will be better if the high contrast area connected for each point and formed a large area. If the high contrast area is bigger, the conductivity in the ink is better as the percolation in the composition connected well and can conduct electricity. The top surface of each sample also was analyzed through the SEM images. EDX analysis is important to identify the present elements in the ink to figure out the effect of the element to the ink properties. The results as shown in Table 4.16 to Table 4.26 is interpreted and been concluded in the next sub-topic.

Table 4.17: SEM and EDX Images of Straight Line for 1mm Sample

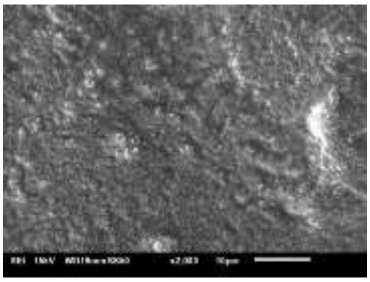
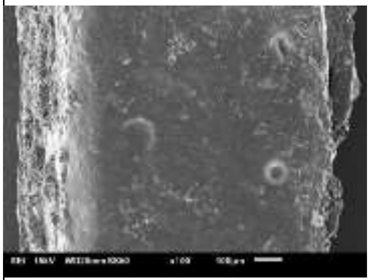
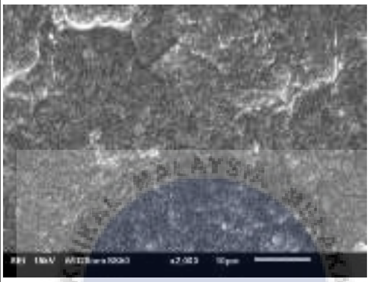
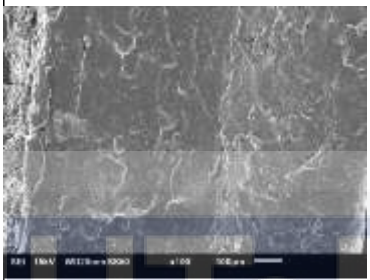
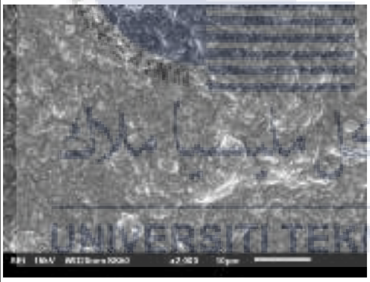
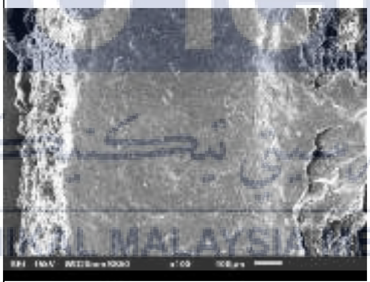
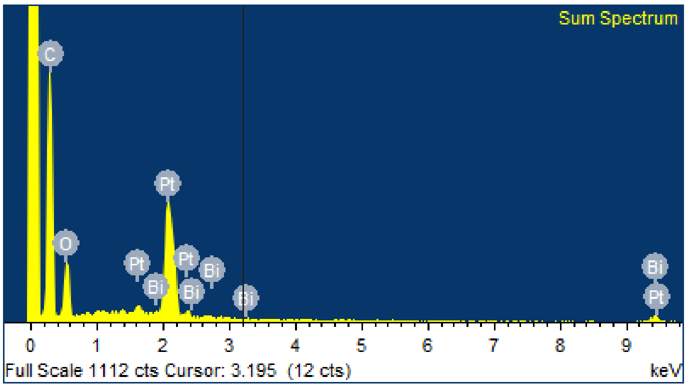
Point	SEM Image (10 µm)	SEM Image (100 µm)	Analysis
1			SEM image at point 1 shows that the presence of connected filler in the dark spot form.
3			The image displays the smoother surface compared to point 1 and points 5 and the present of filler which represented by high contrast area.
5			This image illustrates the top surface of point 3 that cruder compared to point 1. The present of the filler also can be seen through this image.
	EDX Image		
			EDX image shows that the presence of carbon (C) with 78.17% atomic and oxygen (O) with 19.49% atomic in the sample.

Table 4.18: SEM and EDX Images of Straight Line for 2mm Sample

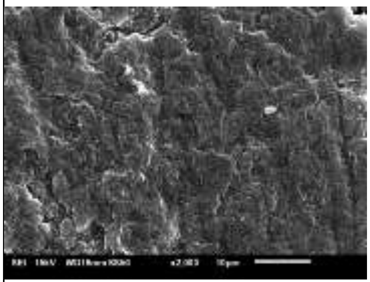
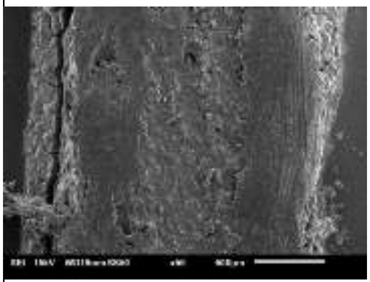
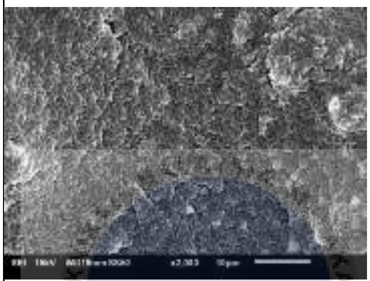
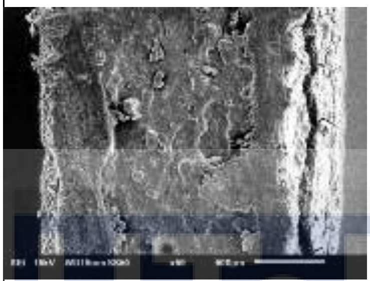
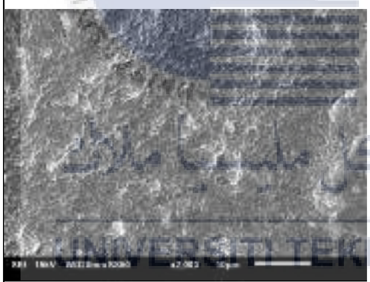
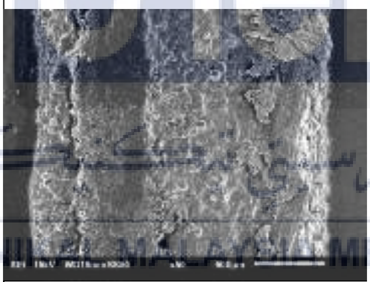
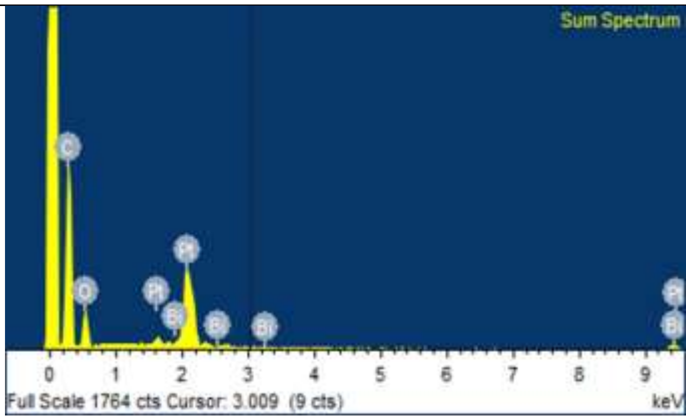
Point	SEM Image (10 µm)	SEM Image (500 µm)	Analysis
1			Based on SEM image, point 1 shows the cruder surface compared to others and the present of filler as high contrast area.
3			This image shows the area of low contrast and high contrast area is equally.
5			The image displays that the largest area of low contrast which represented as the non-conductive materials
	EDX Image		
			The image shows that the sample is consists of 81.48% atomic carbon (C), 17.04% of atomic oxygen (O) and 1.48% of platinum (Pt).

Table 4.19: SEM and EDX Images of Straight Line for 3mm Sample

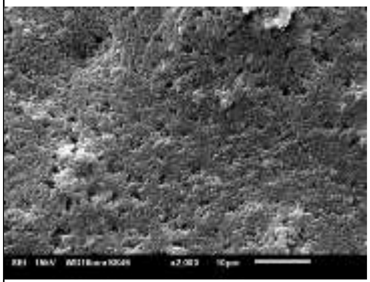
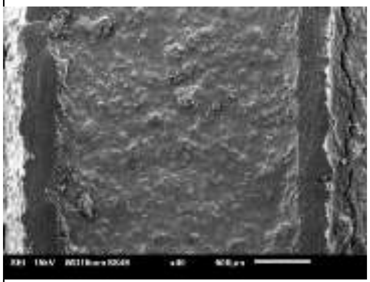
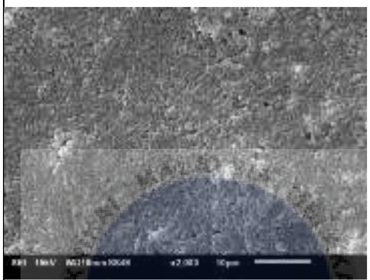
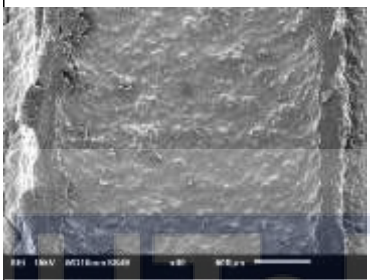
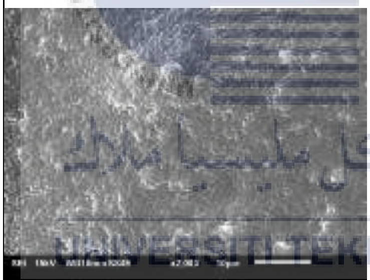
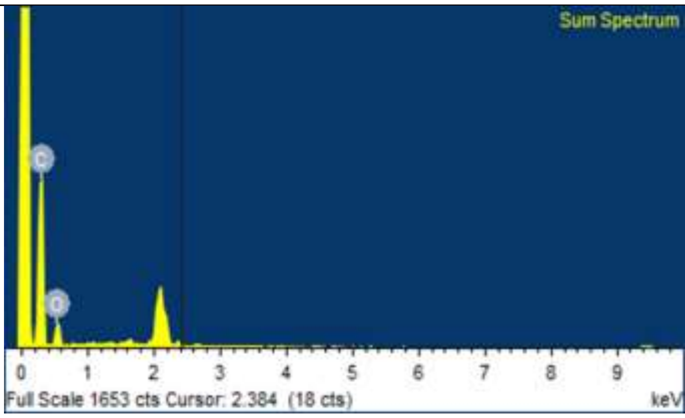
Point	SEM Image (10 µm)	SEM Image (500 µm)	Analysis
1			As can be seen in the SEM image, top surface at point 1 shows a lot of porousity and connected high contrast area.
3			This image shows the smoother area and has the highest area of high contrast, which represented the filler.
5			Point 5 shows that the connected area of high contrast and low contrast equally.
	EDX Image		
			The atomic percentage is 84.92% of carbon (C) which is the highest amount others samples. The rest of elements is oxygen (O) which consists of 13.92% and platinum (Pt) 1.17%.

Table 4.20: SEM and EDX Images of Zigzag for 1mm Sample

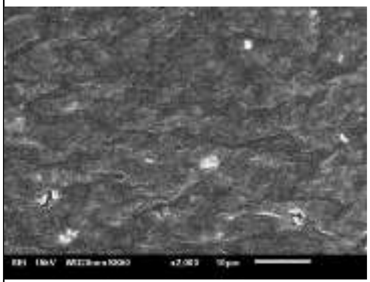
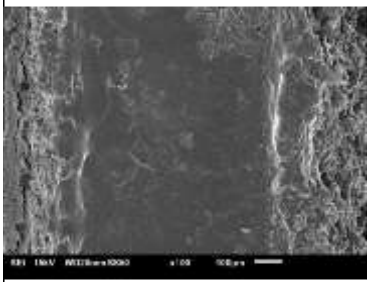
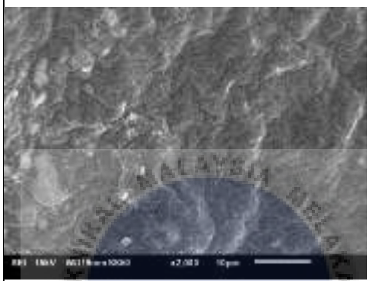
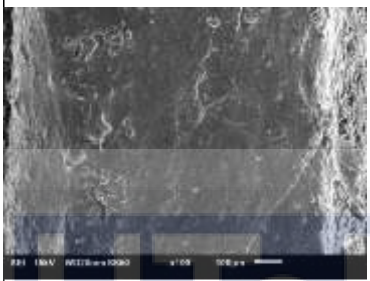
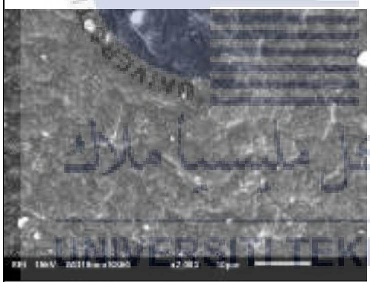
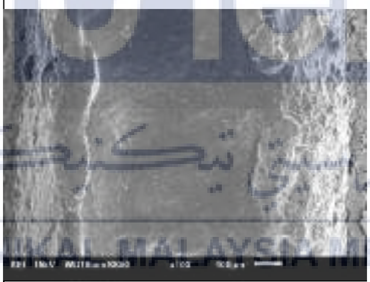
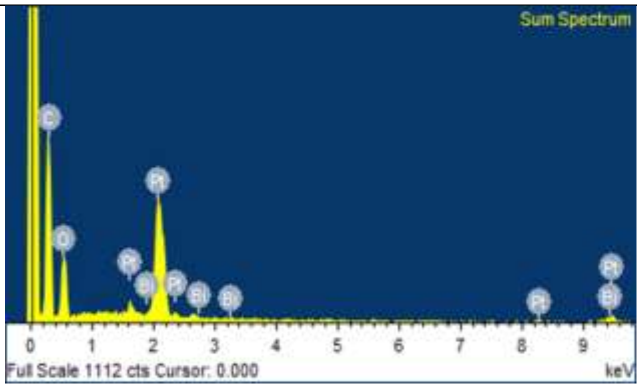
Point	SEM Image (10 µm)	SEM Image (100 µm)	Analysis
1			The image displays that point 1 has the largest high contrast area which high conductivity compared to other points.
3			Point 3 image illustrates the largest area of low contrast, which has the lowest conductivity compared to other points.
5			This image shows that the moderately of low and high contrast area.
	EDX Image		
			From the EDX image, the sample consists of 75.74 % of atomic carbon (C), 22.33% of oxygen (O), and 1.9% of platinum (Pt). This sample has the lowest atomic percentage of carbon (C) compared to 2mm and 3mm samples.

Table 4.21: SEM and EDX Images of Zigzag for 2mm Sample

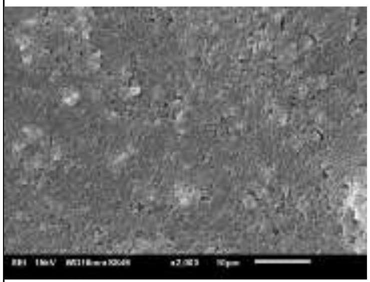
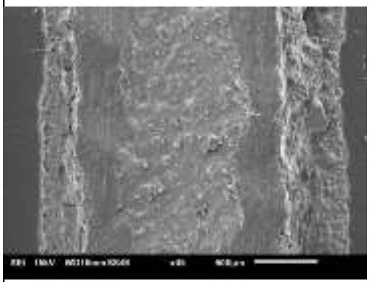
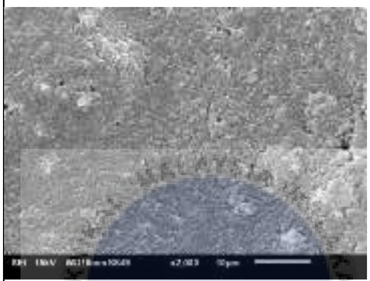
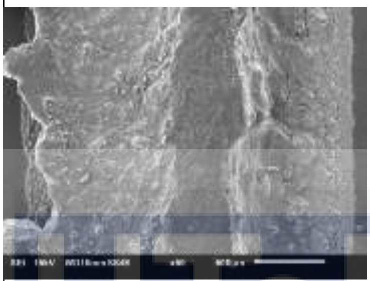
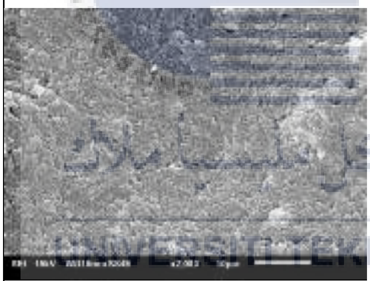
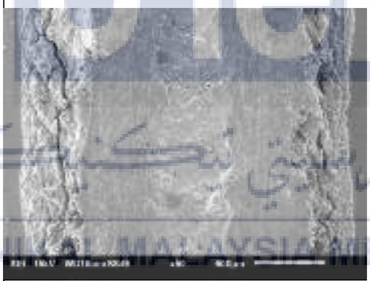
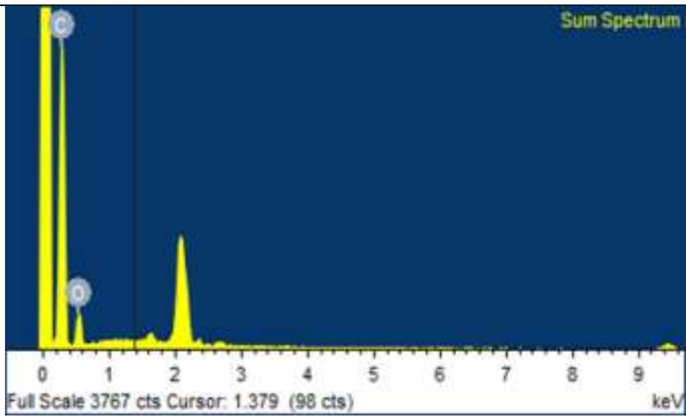
Point	SEM Image (10 µm)	SEM Image (500 µm)	Analysis
1			Based on SEM image for point 1, the top surface shows the smoother compared to other points. The area for high contrast also large as same to other points.
3			Point 3 shows some similarity to point regarding the area of filler in the sample.
5			This point 5 also shows some similarity as same as points before and make this sample has consistently in connected area of high contrast.
	EDX Image		
			As be seen in EDX image, this sample has 87.40% atomic of carbon (C), 11.30% of oxygen (O), and 1.30% of platinum (Pt). This sample has the highest percentage of carbon compared to 1mm and 3mm width.

Table 4.22: SEM and EDX Images of Zigzag for3mm Sample

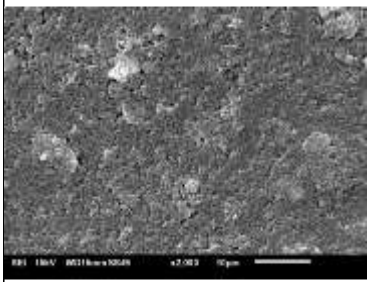
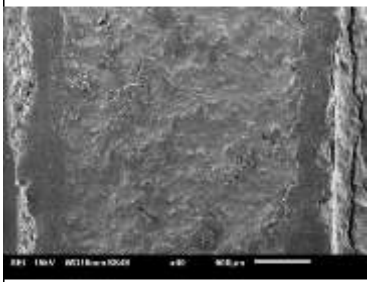
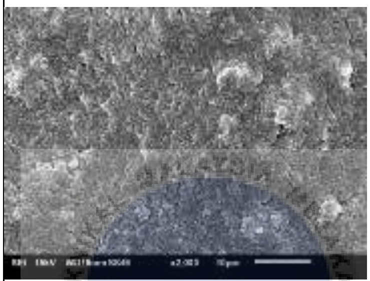
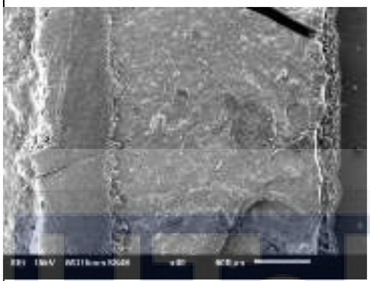
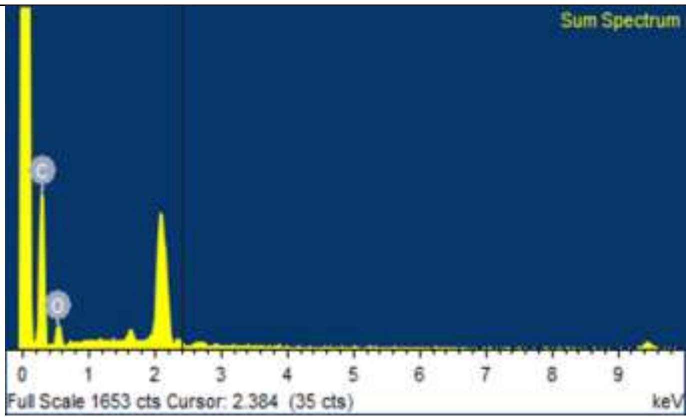
Point	SEM Image (10 µm)	SEM Image (100 µm)	Analysis
1			Based on the image, point 1 shows the largest high contrast area which it has the high conductivity compared to other points.
3			Point 3 illustrates that the largest low contrast area, which make point 3, has the lowest conductivity compared to other points.
5			SEM image shows the smoother surface and has more high contrast area compared to low contrast area.
	EDX Image		
			This EDX image shows that present of element in 3mm sample. This sample only consists of carbon (C) with 86.13% atomic, 11.13% oxygen (O).

Table 4.23: SEM and EDX Images of Curve for 1mm Sample

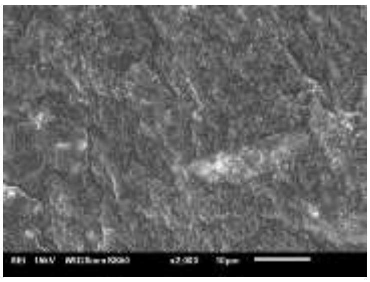
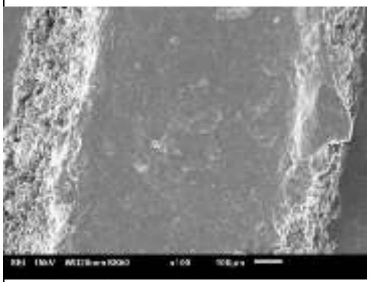
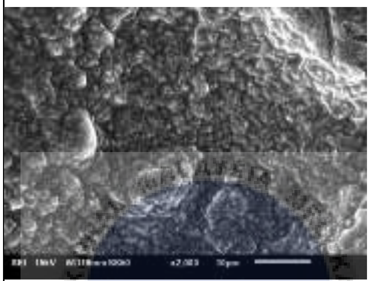
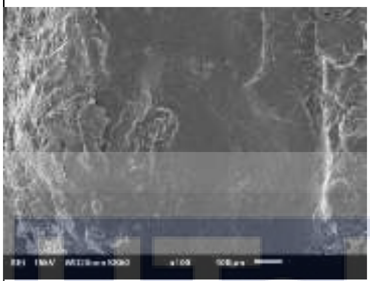
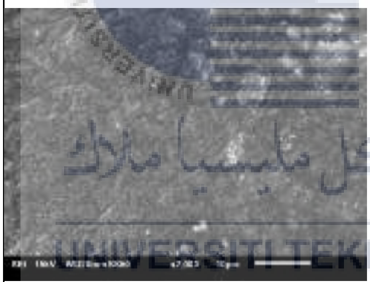
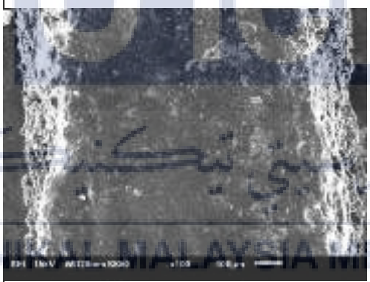
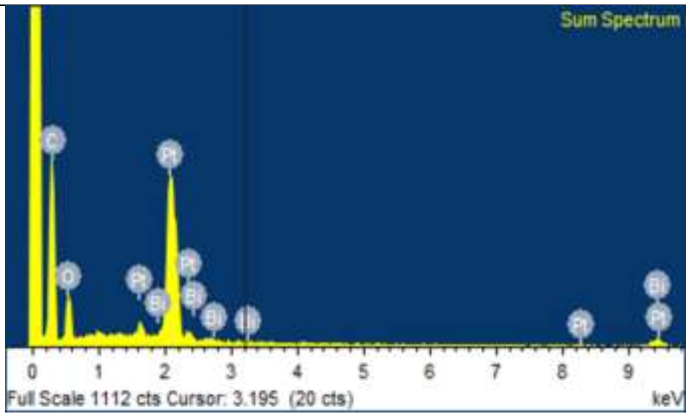
Point	SEM Image (10 µm)	SEM Image (100 µm)	Analysis
1			SEM image shows that the present of the high contrast area in this point 1.
3			As can be seen in this image, point 3 has the cruder surface compared to other points. The high contrast area shows the least in this image.
5			The image displays that present of connected high contrast area which, make it has conductivity.
	EDX Image		
			EDX image shows that the present of carbon (C) with 77.12% atomic and oxygen (O) with 18.87% atomic in the sample.

Table 4.24: SEM and EDX Images of Curve for 2mm Sample

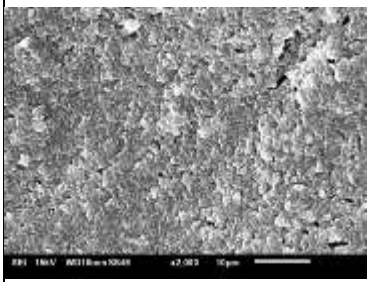
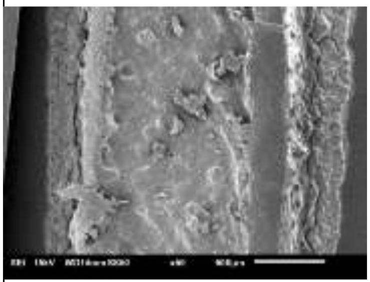
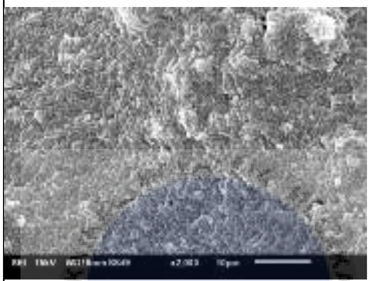
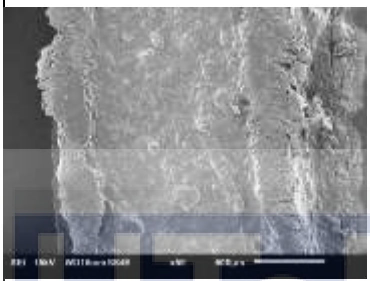
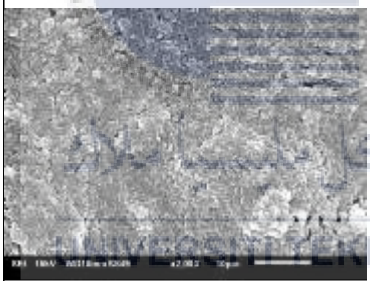
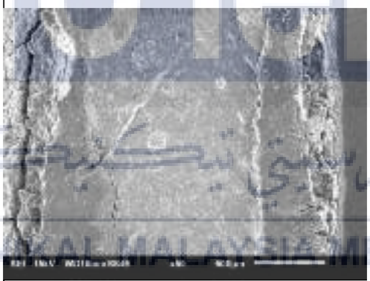
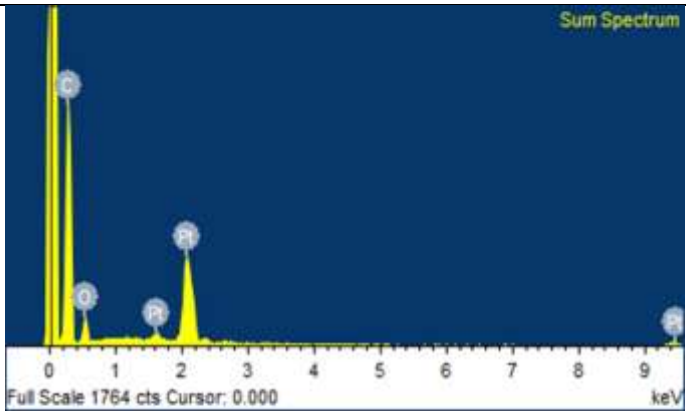
Point	SEM Image (10 µm)	SEM Image (100 µm)	Analysis
1			Based on SEM image, point 1 shows the cruder surface compared to others and the present of filler as high contrast area can be seen in the image.
3			This image shows the largest area of high contrast. This is make that point 3 has the highest conductivity.
5			The image displays that the smallest area of low contrast, which, make point 5, has the lowest conductivity.
	EDX Image		
			The image shows that the sample is consists of 88.05% atomic carbon (C), 10.53% of atomic oxygen (O) and 1.42% of platinum (Pt).

Table 4.25: SEM and EDX Images of Curve for 3mm Sample

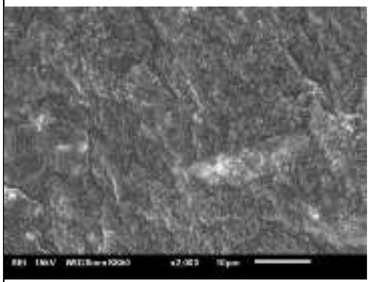
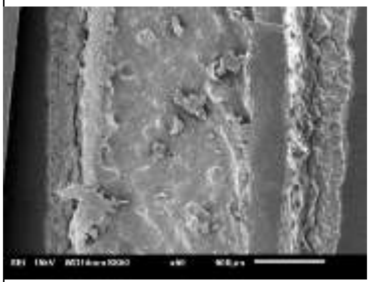
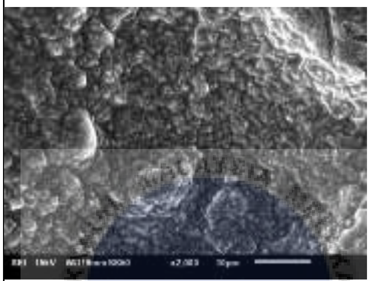
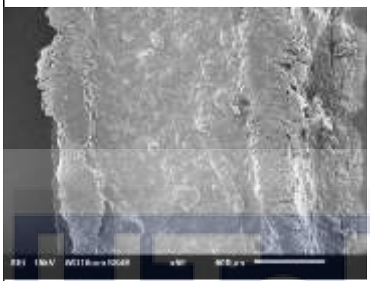
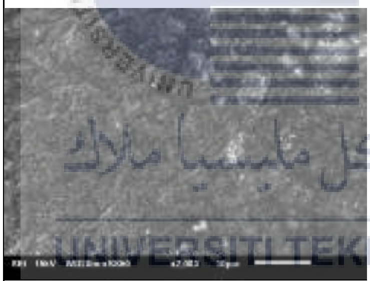
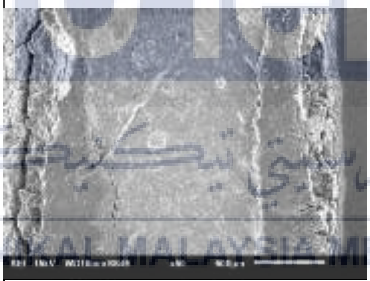
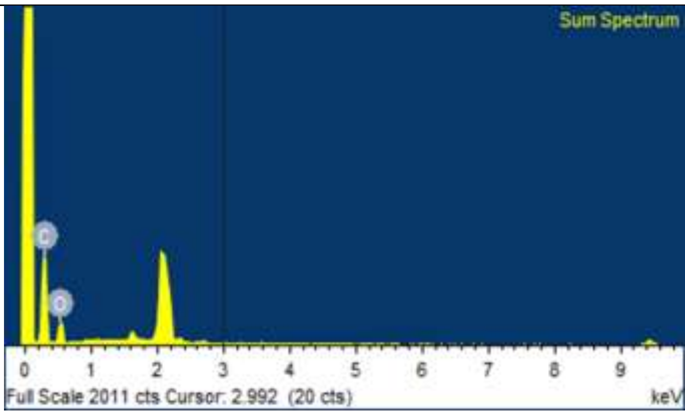
Point	SEM Image (10 µm)	SEM Image (100 µm)	Analysis
1			This image shows that the moderately of low and high contrast area
3			Point 3 image illustrates the largest area of low contrast, which has the lowest conductivity compared to other points.
5			The image displays that point 5 has the largest high contrast area which make it has high conductivity compared to other points.
	EDX Image		
			From the EDX image, the sample consists of 80.94 % of atomic carbon (C), 16.07% of oxygen (O), and 2.99% of platinum (Pt).

Table 4.26: SEM and EDX Images of Square for 1mm Sample

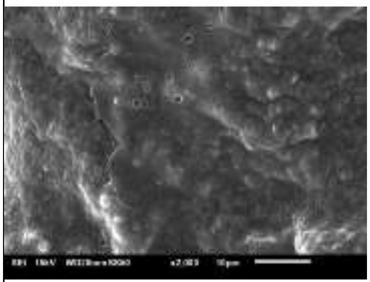
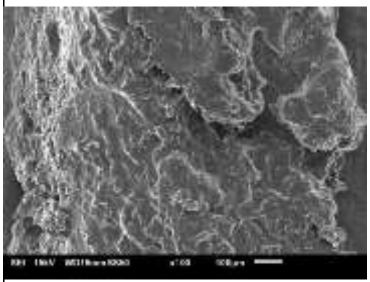
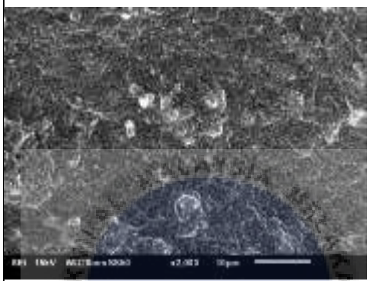
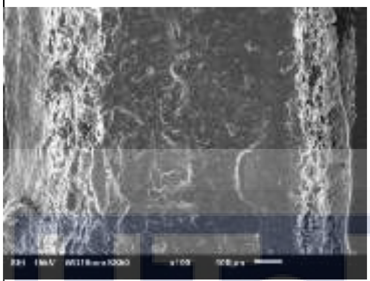
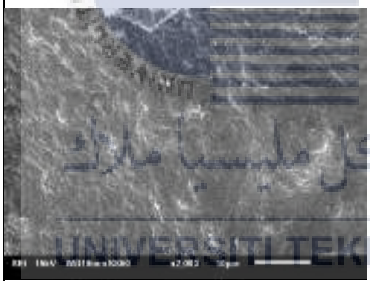
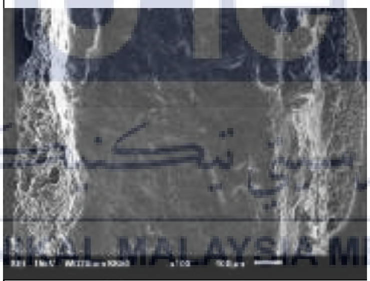
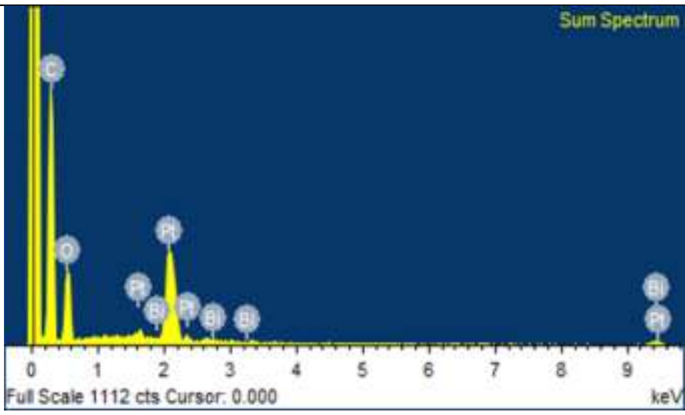
Point	SEM Image (10 µm)	SEM Image (100 µm)	Analysis
1			This point 1 shows that the area, which is consistently, connected area of high contrast.
3			Point 3 shows some similarity to point 5 regarding the area of filler in the sample.
5			Based on SEM image for point 5, the top surface shows the smoother compared to other points. The area for high contrast also large as same to point 3.
	EDX Image		
			As be seen in EDX image, this sample has 76.37% atomic of carbon (C), 22.51% of oxygen (O), and 1.12% of platinum (Pt). This sample has the lowest percentage of carbon compared to 2mm and 5mm width

Table 4.27: SEM and EDX Images of Square for 2mm Sample

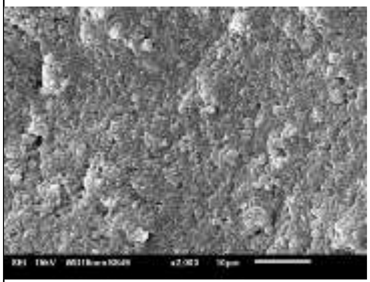
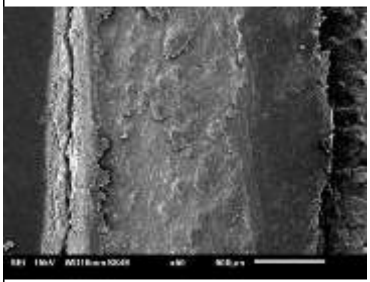
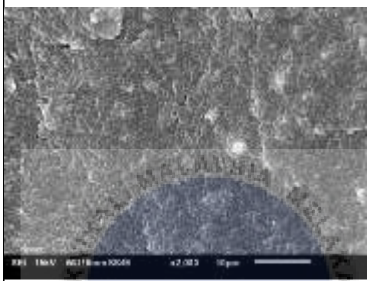
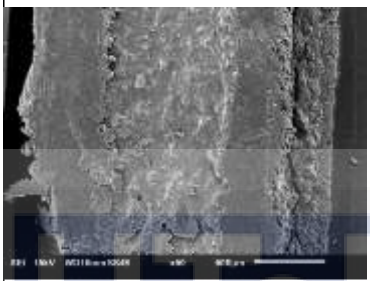
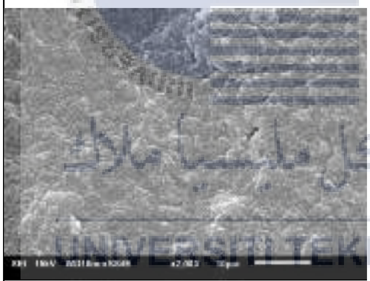
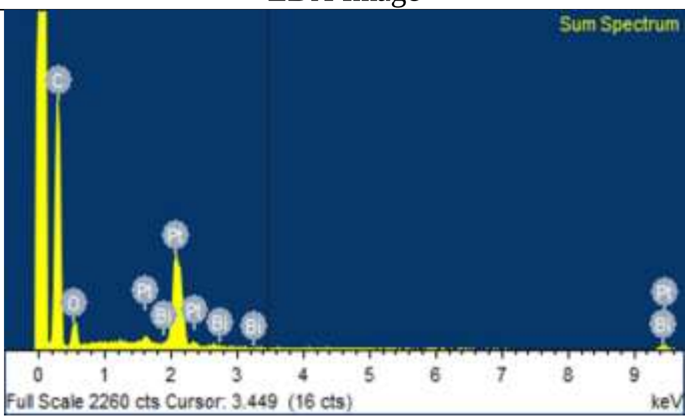
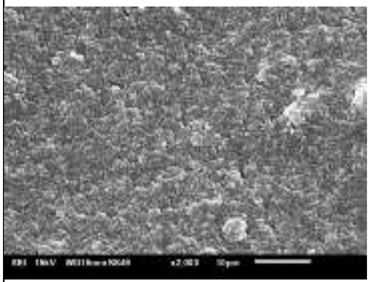
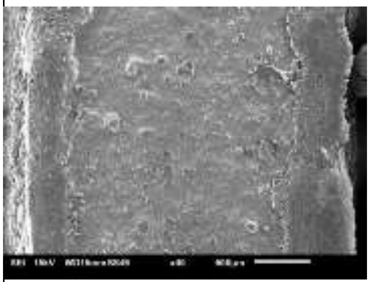
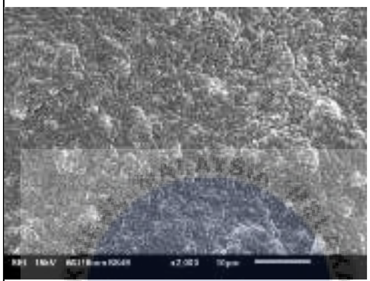
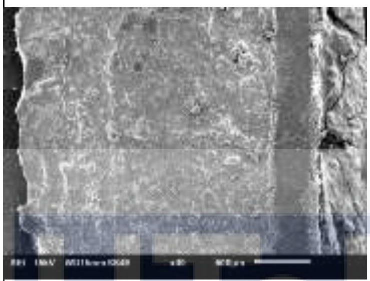
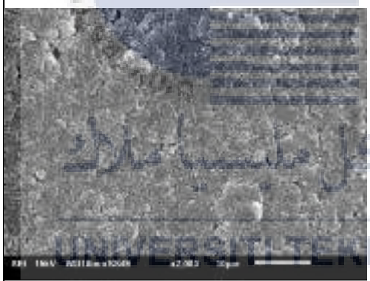
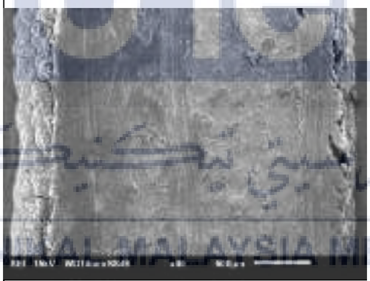
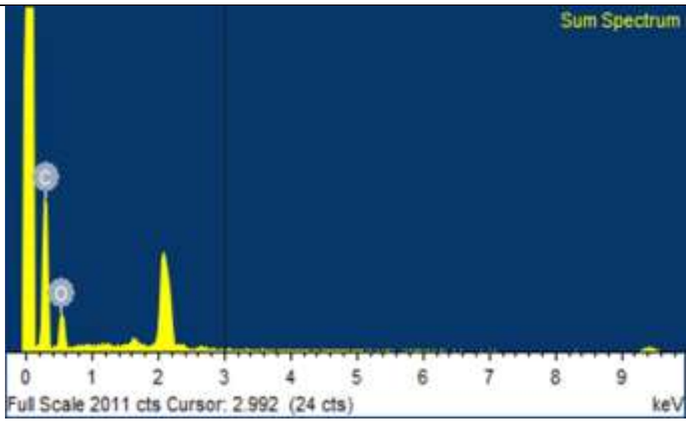
Point	SEM Image (10 µm)	SEM Image (100 µm)	Analysis
1			Based on the image, point 1 shows the smallest high contrast area which it has the lowest conductivity compared to other points.
3			Point 3 illustrates that the largest high contrast area, which make point 3, has the highest conductivity compared to other points.
5			SEM image shows the smoother surface and has more high contrast area compared to low contrast area.
	EDX Image		
			This EDX image shows that present of element in 2mm sample. This sample only consists of carbon (C) with 88.49% atomic, 10.08% oxygen (O). This sample has the highest percentage of carbon compared to 1mm and 3mm width

Table 4.28: SEM and EDX Images of Square for 3mm Sample

Point	SEM Image (10 µm)	SEM Image (100 µm)	Analysis
1			SEM image at point 1 shows that the present of connected filler in the dark spot form which has large area of it.
3			The image displays the present of filler, which represented by high contrast area and make it has high conductivity.
5			This image illustrates the present of the connected filler can be seen through this image.
	EDX Image		
			As be seen in EDX image, this sample has 80.21% atomic of carbon (C), 17.86% of oxygen (O), and 1.92% of platinum (Pt).

4.6 Analysis on properties and characterization of the graphene conductive ink

In this study, there are two properties was figured out to ensure the ability of the graphene-based conductive ink to conduct an electricity and measure the hardness of the ink. Clearly, electrical properties was determined based on the voltage and sheet resistivity as both of it is relatable. Mechanical properties is equally important, as the analysis on this property will answer the measurement of hardness on conductive ink.

Generally, electrical properties is explained based on Ohm's law where shown in Eq. (4.1) (Pickover, 2008). Ohm's law can found out the relationship of resistance R, voltage V, and current I. In order to produce an effective ink that can conduct an electricity efficiently, the ability to current flow in the ink must increases and vice versa with resistance. In this experiment, resistance is directly proportional to voltage, V that measure in (mV) and inversely with current, I (μA) as refer to Eq. (4.2);

$$V = IR \tag{4.1}$$

$$R = \frac{V}{I} \tag{4.2}$$

Sheet resistivity is the capability of the ink materials in resists the flow of electricity in the form of thin films (Taherian & Kausar, 2019). As an illustration for the relationship of voltage and sheet resistivity, an equation of sheet resistivity is used where shown in Eq. (4.3) (Bhore, 2013).

$$R_S = R\frac{l}{W} \tag{4.3}$$

Where R_S is the symbol for sheet resistivity while l is the length (mm), *w* is the width of the sample (mm) and *R* represented as resistance (Ω). When the resistance from the Ohm's law is substitute in the sheet resistivity equation, the final equation is generally define as in Eq. (4.4):

$$R_S = \frac{Vl}{IW} \tag{4.4}$$

Back to main objective of the study, which is to identify the properties of the graphene-based conductive ink, the sheet resistivity equation is used to point out the relationship between the voltage, current and sheet resistivity. Based on the equation, the sheet resistivity is directly proportionally to voltage and length where contrast with width and current. Theoretically, the higher the sheet resistivity value, the lower the current flow in the sample and the wider the width. Based on this theory, the sample of conductive ink should has lower sheet resistivity in order to increases the current flow.

In this study of conductive ink, the results from the experimental was analysed based on two different characteristics, which are pattern and width. As stated in methodology, the conductive ink is patterned in four different type (straight line, zigzag, curve and square) and three different width (3mm, 2mm and 1mm). In order to figure out which pattern and width have the most efficient in conducting an electricity, the analysis was done as following discussion in the next sub-chapter.

4.6.1 Characterization: pattern

Table 4.29 displays that the graphs for analysis of pattern on 1mm samples. Each patterns is presented in the graph of voltage, sheet resistivity and hardness. The plotted graph of voltage showed that zigzag pattern has the higher average value of voltage from approximately 30mV to 43mV. This graph of zigzag is vice versa with curve pattern as it has the lowest value of voltage which approximately from 31mV to 9mV. For the sheet resistivity graph, zigzag pattern is plotted in the graph as the highest average value among all these patterns with approximately value which ranging between 13-19 μΩ/sq. On the other hand, curve has the lowest average value with approximately between 4-14 μΩ/sq. Based on the average results; zigzag has less an ability to conduct an electricity compared to other patterns. Thus, the highest sheet resistivity as stated from previous study might affected the performance of the ink. This of results is continuous with the present of hardness Vickers results where straight-line pattern come out with the highest value of hardness while zigzag has the lowest value.

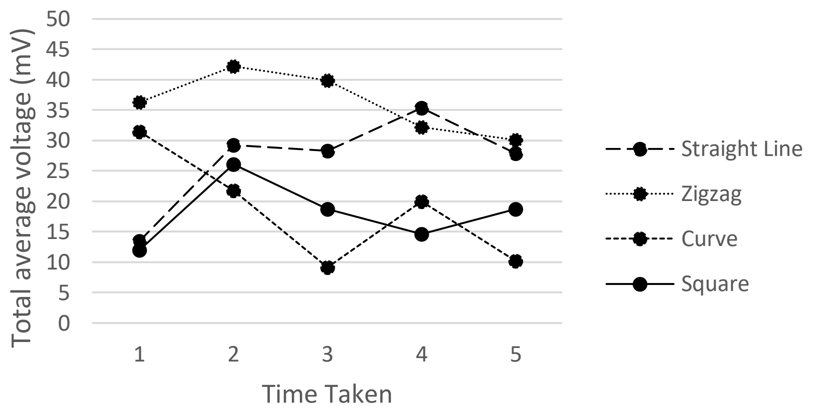
To continue investigate the effect of properties on the conductive ink samples by patterns, Table 4.30 shows the results of 2mm samples. By referring this table, zigzag pattern has the highest value of voltage within approximately 21-30mV while curve has a lower value which approximately from 13mV to 19mV. For a sheet resistivity results, as same as 1mm results zigzag recorded as it has the highest value from 9-13 μΩ/sq. Based on the plotted graph, curve pattern shows the lowest value of sheet resistance between 4 μΩ/sq to 14 μΩ/sq. Results on mechanical properties shows that straight line patterns has the highest hardness value.

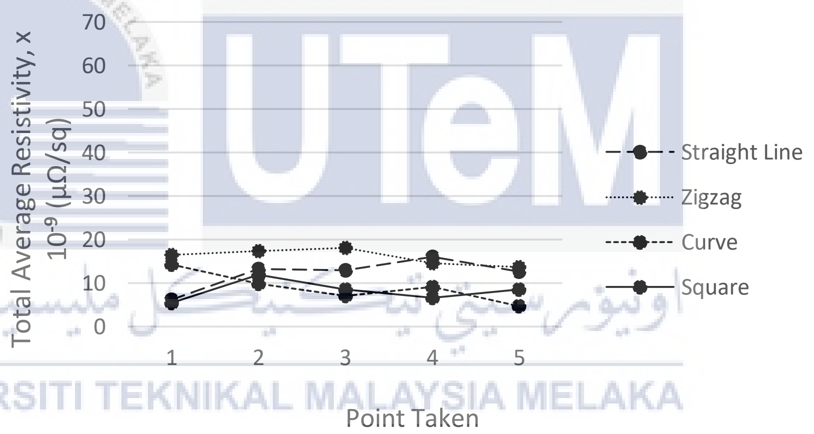
As shown in Table 4.31, results of 3mm samples is plotted in line graph with different pattern of samples. Based on this table, it clearly shows that zigzag pattern has the highest value of voltage between 21mV to 34mV while the lowest value from 10mV to 17mV is curve pattern. Next, accordingly to sheet resistivity graph, straight line plotted the highest average value from 10 μΩ/sq to 60 μΩ/sq. Following this results, curve pattern shows the lowest graph line with value in ranging from 5-9 μΩ/sq. Next, the hardness Vickers graph shows some similarity with the results of 1mm and 2mm samples as straight line shows the highest value between 0.1 HV to 11 HV while square shows the lowest value within 0.1 Hv to 4 HV.

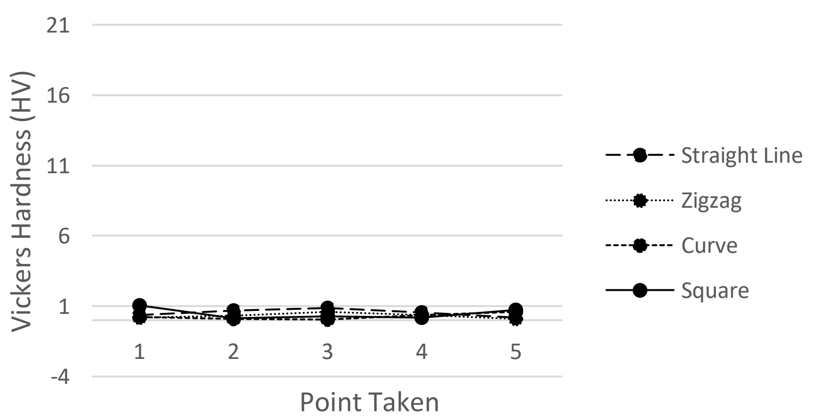
As can be seen in the final analysis for properties study by patterns, zigzag shows the highest value of voltage for each width (1mm, 2mm and 3mm) where vice versa with curve which has the lowest value of voltage for each width. For sheet resistivity, the results shows the similarity with voltage results as zigzag has the highest average value of sheet resistance, which contrast with curve pattern. Theoretically, voltage has relationship with sheet resistivity as these two things might affect the performance of current flow in the conductive ink. This shows that the higher the value of voltage the higher the value of sheet resistivity and it based on experimentally.

Based on these results, it can be conclude that the composite that consists of graphene which a conductor materials and non-conductive materials (epoxy resin and hardener) can allow current flow. This is because as stated in previous study that graphene has high mobility. Besides that, some inconsistency of results might cause during printing process. In the study of conductive ink, there is study stated that printing process is one of the factor that can affect the ink properties.

Table 4.29: The Analysis of Pattern on 1mm Samples

<table>
<tr><th>Width (mm)</th><th>Voltage (mV)</th></tr>
<tr><td rowspan="5">1</td><td>


</td></tr>
<tr><td>Sheet Resistivityx10^{-3} (μΩ/sq)</td></tr>
<tr><td>


</td></tr>
<tr><td>Hardness Vickers (HV)</td></tr>
<tr><td>


</td></tr>
</table>

Table 4.30: The Analysis of Pattern on 2mm Samples

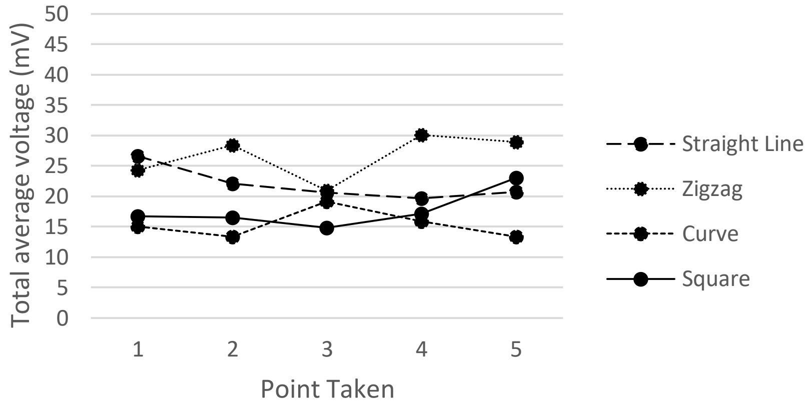
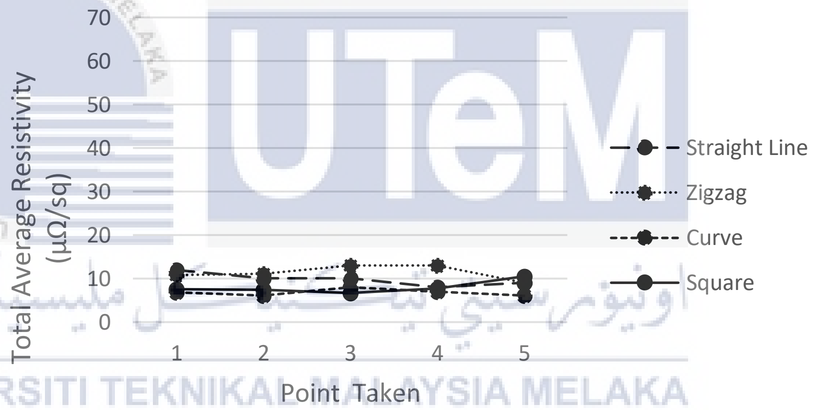
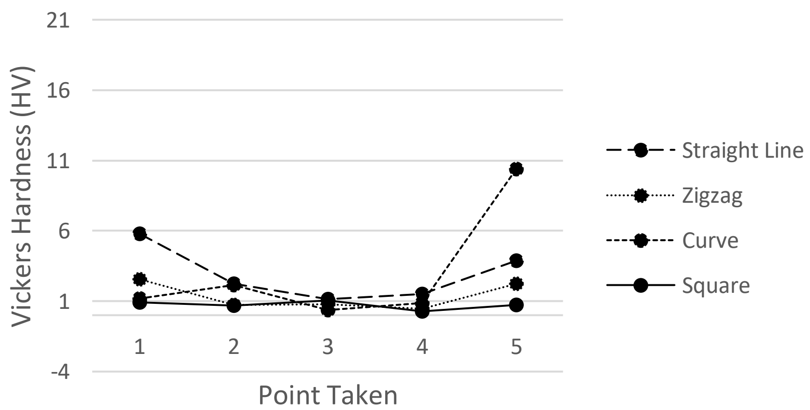
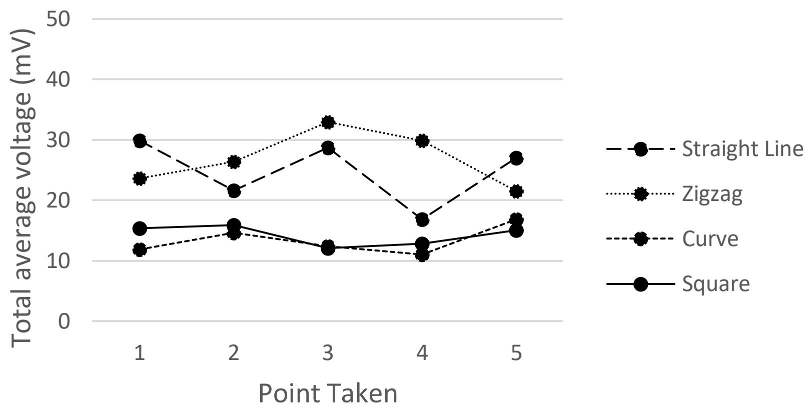
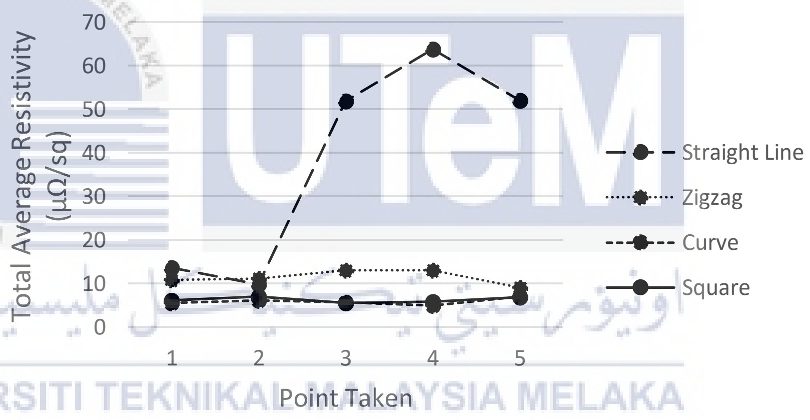
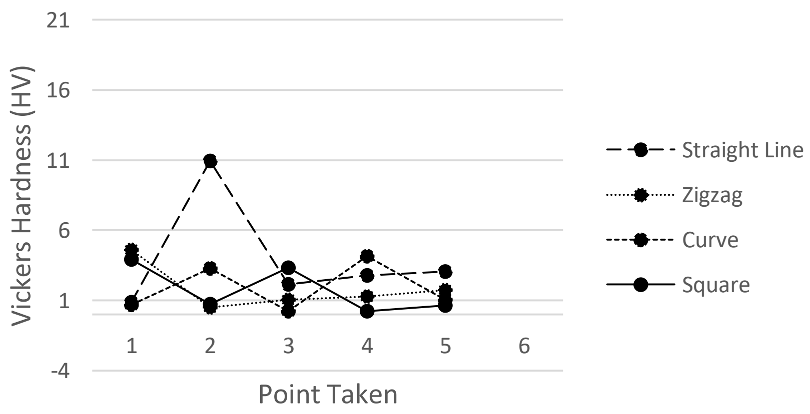
<table>
<tr><th>Width (mm)</th><th>Voltage (mV)</th></tr>
<tr><td rowspan="5">2</td><td>
</td></tr>
<tr><td>Sheet Resistivity (μΩ/sq)</td></tr>
<tr><td>
</td></tr>
<tr><td>Hardness Vickers (HV)</td></tr>
<tr><td>
</td></tr>
</table>

Table 4.31: The Analysis of Pattern on 3mm Samples

<table>
<tr><th>Width (mm)</th><th>Voltage (mV)</th></tr>
<tr><td rowspan="5">3</td><td>
</td></tr>
<tr><td>Sheet Resistivity (μΩ/sq)</td></tr>
<tr><td>
</td></tr>
<tr><td>Hardness Vickers (HV)</td></tr>
<tr><td>
</td></tr>
</table>

4.6.2 Characterization: width

Table 4.32 shows that the end results of analysis of width on straight-line samples. From the observation of the voltage graph, clearly 1mm sample shows the highest graph. Followed by plotted graph on 3mm samples with fluctuate pattern line graph as it has the value of voltage within 16mV to 30mV. 2mm samples has the lowest value compared to 3mm and 1mm graph with a value from 20mV to 25mV. Besides this voltage results, sheet resistivity shows that 3mm has the highest value in the range between 10-65 μΩ/sq while 1mm shows the lowest plotted line graph. 2mm maintain in the middle between other samples with value from 10 μΩ/sq to 15 μΩ/sq. For the hardness test, based on the observation of the graph, there is no differences from the results of voltage and sheet resistivity as 3mm sample consists the highest value of hardness and followed by the 2mm and 1mm.

Results of zigzag pattern (refer to Table 4.33) shows some differences in voltage and sheet resistivity results compared straight-line pattern. In voltage and sheet resistivity graph, 1mm stay has the highest average value while 2mm shows the moderate plotted line with a average value from 20mV to 30mV and 10 μΩ/sq to 15 μΩ/sq. The 3mm sample replaced the 2mm results in straight-line pattern and stay in middle between 2mm sample and 1mm sample. Hardness results shows some similarity with straight-line pattern as 3mm maintain in having the highest value of hardness and followed by 2mm and 1mm.

Based on the results of curve pattern (refer to Table 4.34), 2mm sample has the moderate value for both voltage and sheet resistivity. It is just the same with zigzag results as the value is from 13mV to 20mV and 6 μΩ/sq to 9 μΩ/sq. 3mm stay as the last with a value of voltage and sheet resistivity from 10mv to 16mV and 5 μΩ/sq to 8 μΩ/sq and 1mm as the highest. The hardness Vickers shows differences between results from other pattern where 2mm has the highest hardness with value from 0.3HV to 10HV and followed by 3mm.

Table 4.35 shows that the results of square pattern. Based on the results, as same as results in zigzag and curve, 2mm is main as the sample that has moderate voltage and sheet resistivity value with a value from 16mV to 23mV and 7 μΩ/sq to 11 μΩ/sq. This result is followed by 3mm with a value of voltage between 11mV to 16mV and sheet resistivity value from 5 μΩ/sq to 7 μΩ/sq approximately while 1mm stay as the highest value. The hardness results shows some similarity with curve results which 2mm can endurance more force compared to 3mm samples. The value of hardness Vickers for this square pattern is between 0.1 HV to 4 HV.

Based on the discussion, it can conclude that width results shows some inconsistency regarding the voltage, sheet resistivity and hardness. Graphene based conductive ink shows that 1mm width has higher resistivity compared to 3mm width. This shows that the wider the size of samples the lower the value of resistivity. This because of the wider the samples width, it has low resistance which make the flow of current in the ink more efficient compared to small width.

Table 4.32: The Analysis of Width on Straight Line Samples

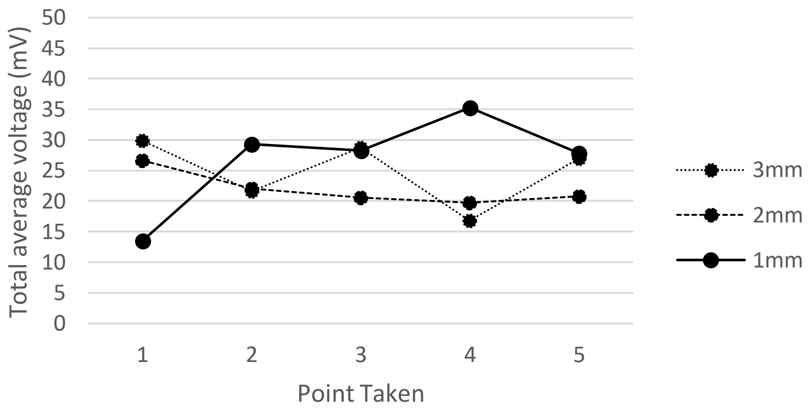
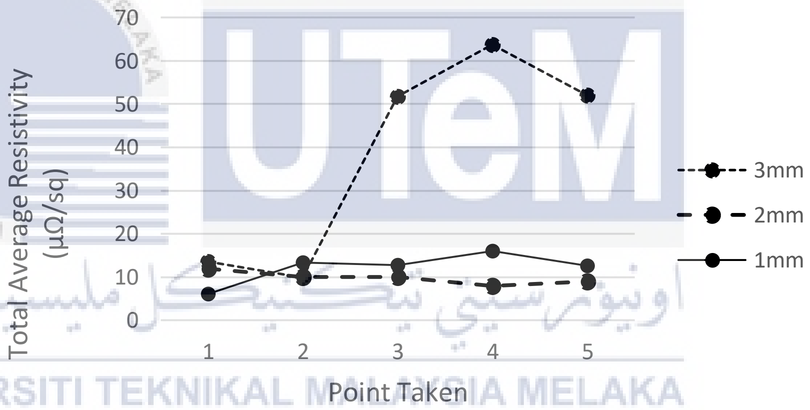
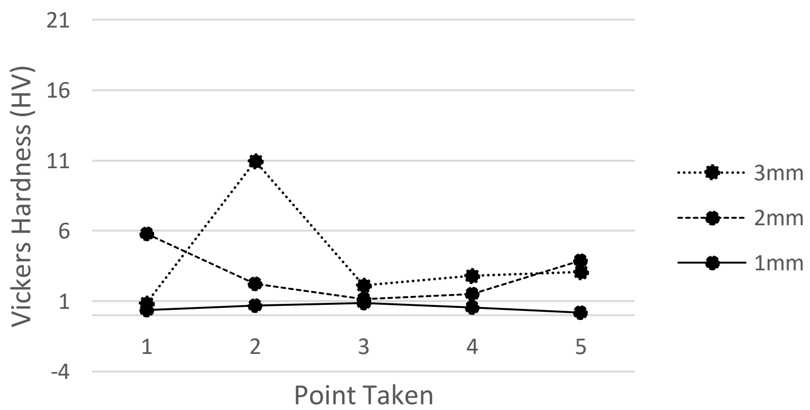
<table>
<tr><th>Pattern</th><th>Voltage (mV)</th></tr>
<tr><td rowspan="5">Straight Line</td><td>
</td></tr>
<tr><td>Sheet Resistivity (µΩ/sq)</td></tr>
<tr><td>
</td></tr>
<tr><td>Hardness Vickers (HV)</td></tr>
<tr><td>
</td></tr>
</table>

Table 4.33: The Analysis of Width on Zigzag Samples

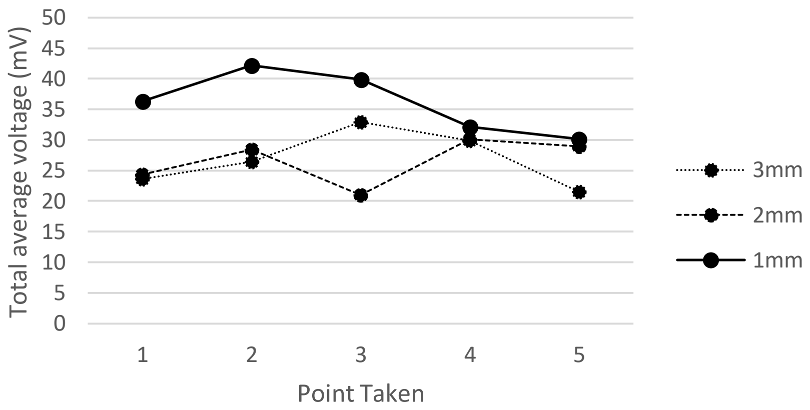
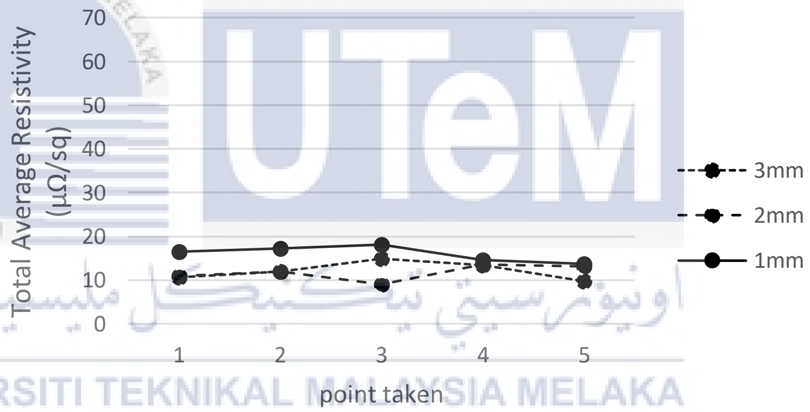
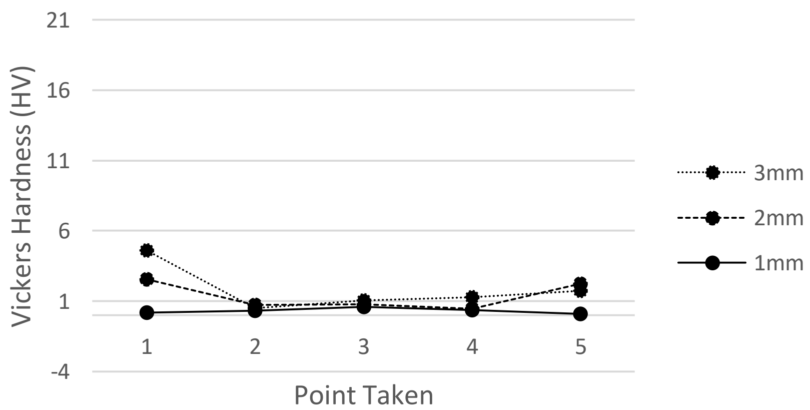
<table>
<tr><th>Pattern</th><th>Voltage (mV)</th></tr>
<tr><td rowspan="5">Zigzag</td><td>
</td></tr>
<tr><td>Sheet Resistivity (μΩ/sq)</td></tr>
<tr><td>
</td></tr>
<tr><td>Hardness Vickers (HV)</td></tr>
<tr><td>
</td></tr>
</table>

Table 4.34: The Analysis of Width on Curve Samples

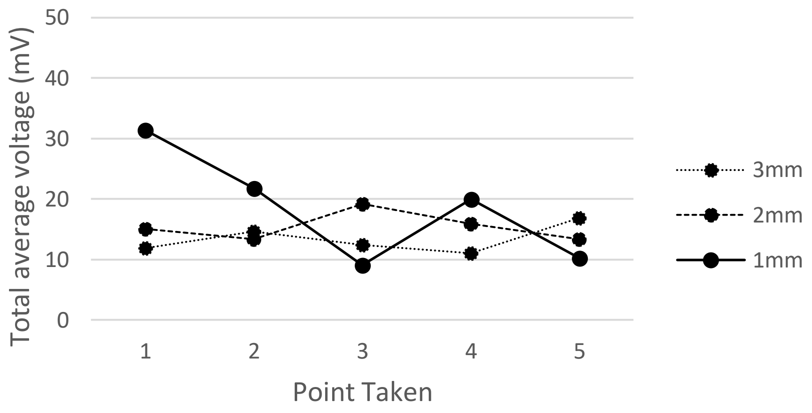
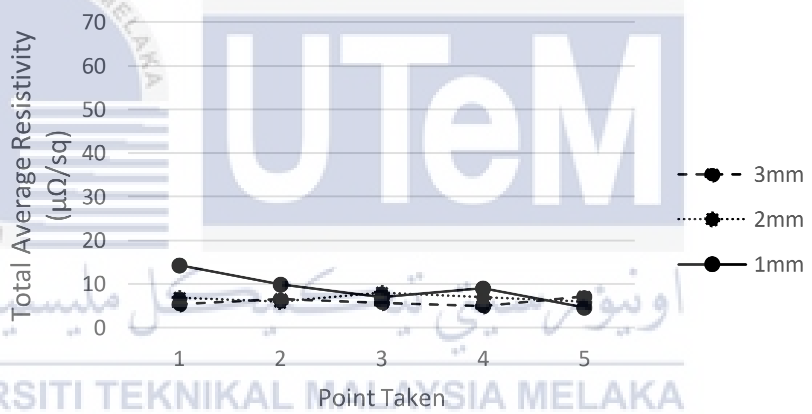
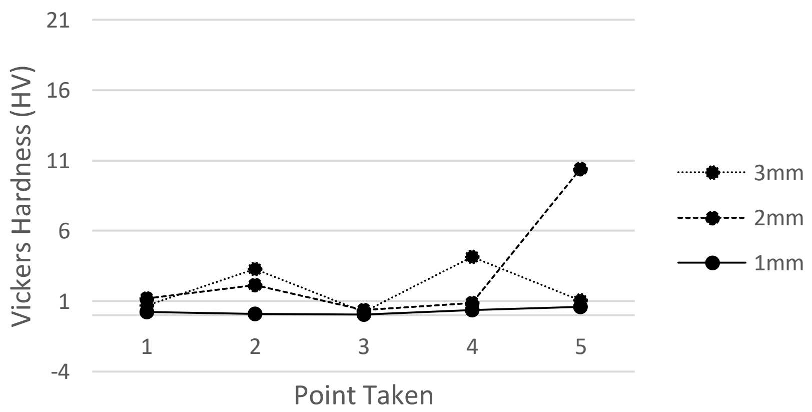
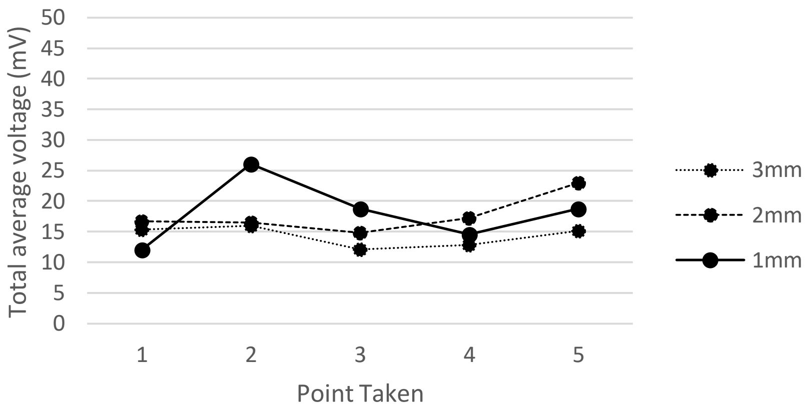
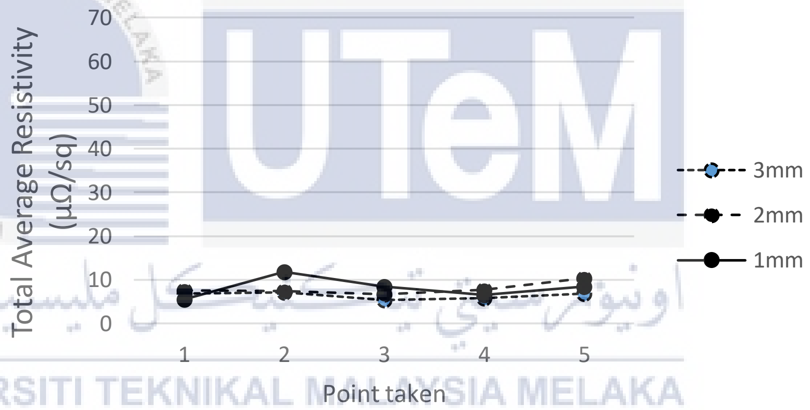
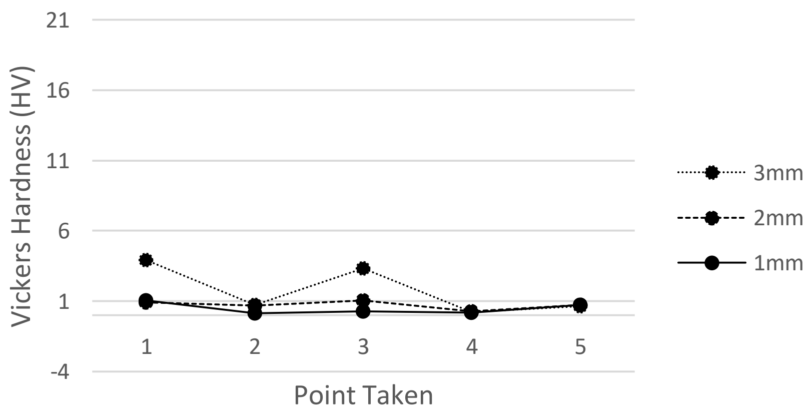
Pattern	Voltage (mV)
Curve	
	Sheet Resistivity (μΩ/sq)
	
	Hardness Vickers (HV)
	

Table 4.35: The Analysis of Width on Square Samples

<table>
<tr><th>Pattern</th><th>Voltage (mV)</th></tr>
<tr><td rowspan="5">Square</td><td>
</td></tr>
<tr><td>Sheet Resistivity (μΩ/sq)</td></tr>
<tr><td>
</td></tr>
<tr><td>Hardness Vickers (HV)</td></tr>
<tr><td>
</td></tr>
</table>

4.7 Relationship between voltage, sheet resistivity, hardness and microstructure conductive ink

Based on the observation of the results in Table 4.30, it can concluded that zigzag pattern is the efficient in conducting electricity. This is because zigzag has the highest reading in voltage, sheet resistivity which contrast with curve pattern. By referring on the Eq. (4.4), it shows that the highest the resistivity, the lowest the current flow (Bhore, 2013). .This results regarding on zigzag pattern is due to the complexity of the design. Curve which followed nearly by the straight line has the most efficient in flowing the current. This because both pattern have good properties and behaviour if the conductive ink.

For the width, 3mm shows the most efficient compared to other widths. Theoretically, the lowest the resistivity the wider the width and it shown through Eq. (4.4). This theory is proven as 3mm stay as the better width. Figure 4.1, Figure 4.2 and Figure 4.3 shows the overall results regarding the voltage reading, sheet resistivity and hardness test. For hardness test, straight line have the better structure in endurance force and contrast with zigzag. This is because the structure of the straight-line pattern simpler compare to others.

Table 4.36: The overall results

Properties	Pattern		Width	
	Highest	Lowest	Highest	Lowest
Voltage, mV	zigzag	Curve	1mm	3mm
Sheet Resistivity, ohm/square	zigzag	Curve	1mm	3mm
Hardness Vickers, HV	zigzag	Curve	3mm	1mm
Behaviour	Cruder	Less cruder	Cruder	Less cruder
Image analyser	zigzag	Curve/straight line	1mm	3mm
	High contrast	Low contrast	High contrast	Low contrast
SEM image	Curve/ straight line	zigzag	3mm	1mm

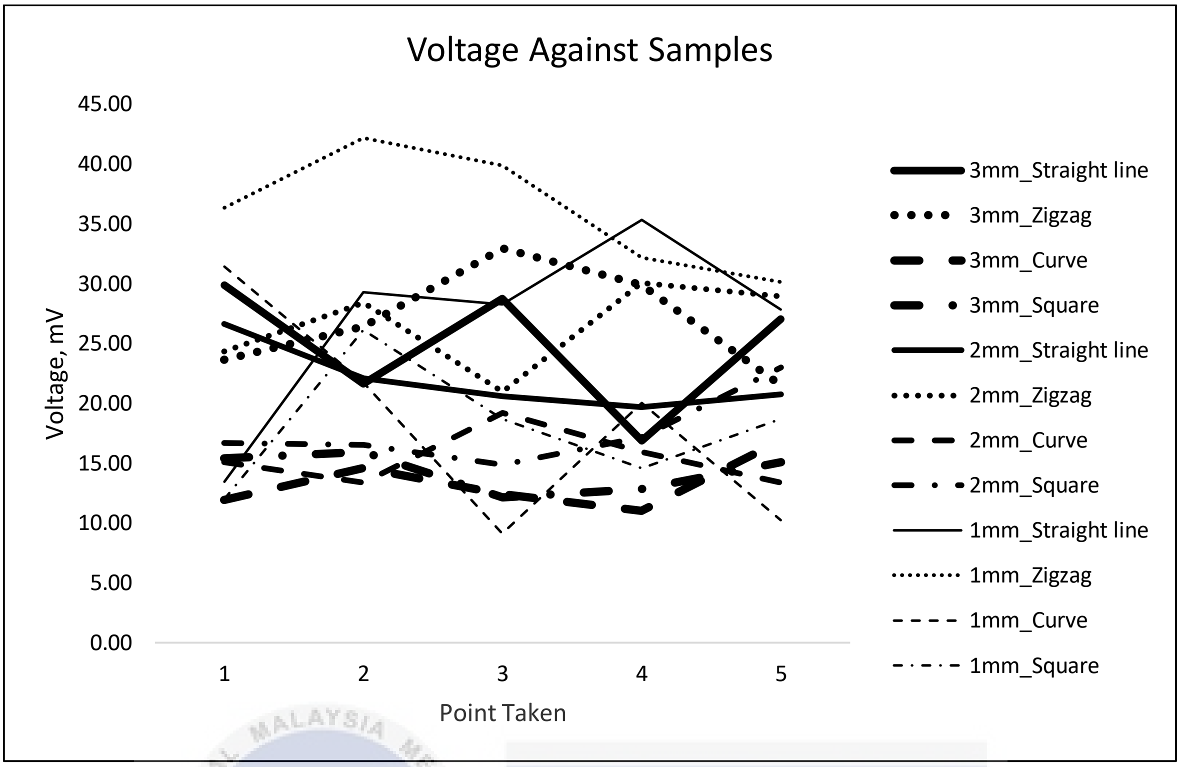


Figure 4.1: Graph voltage against samples

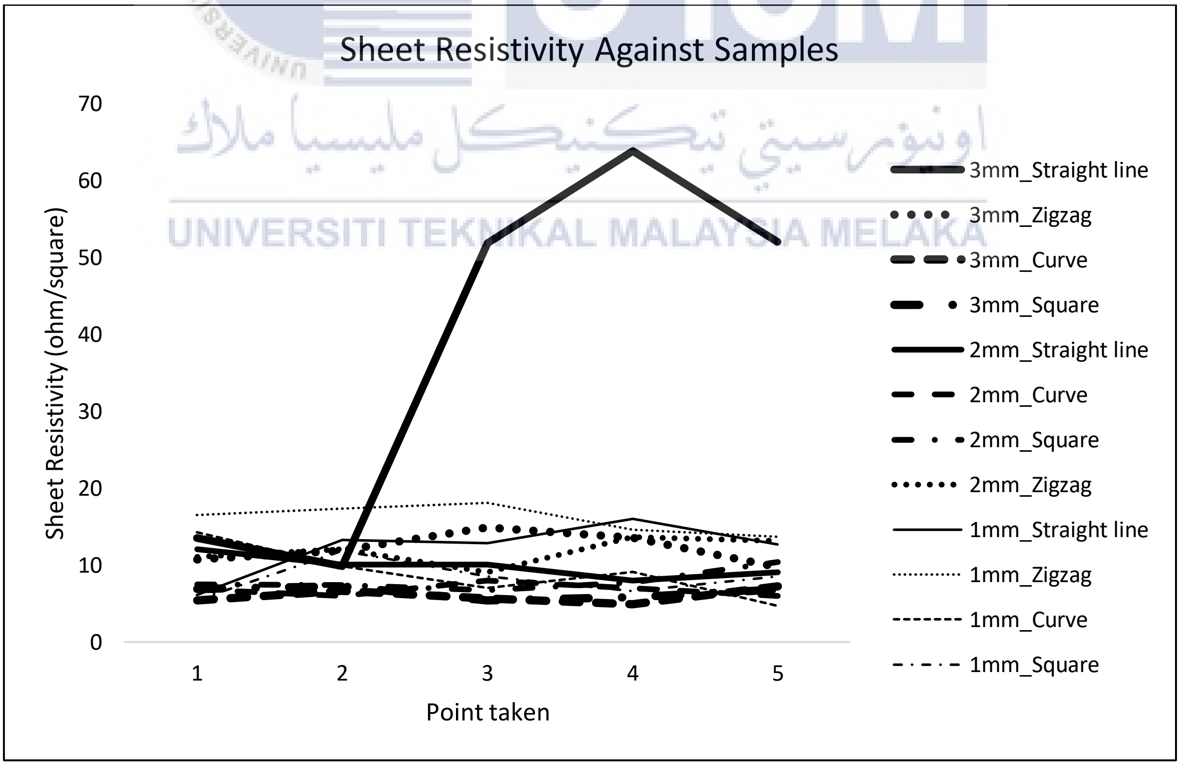


Figure 4.2: Graph sheet resistivity against samples

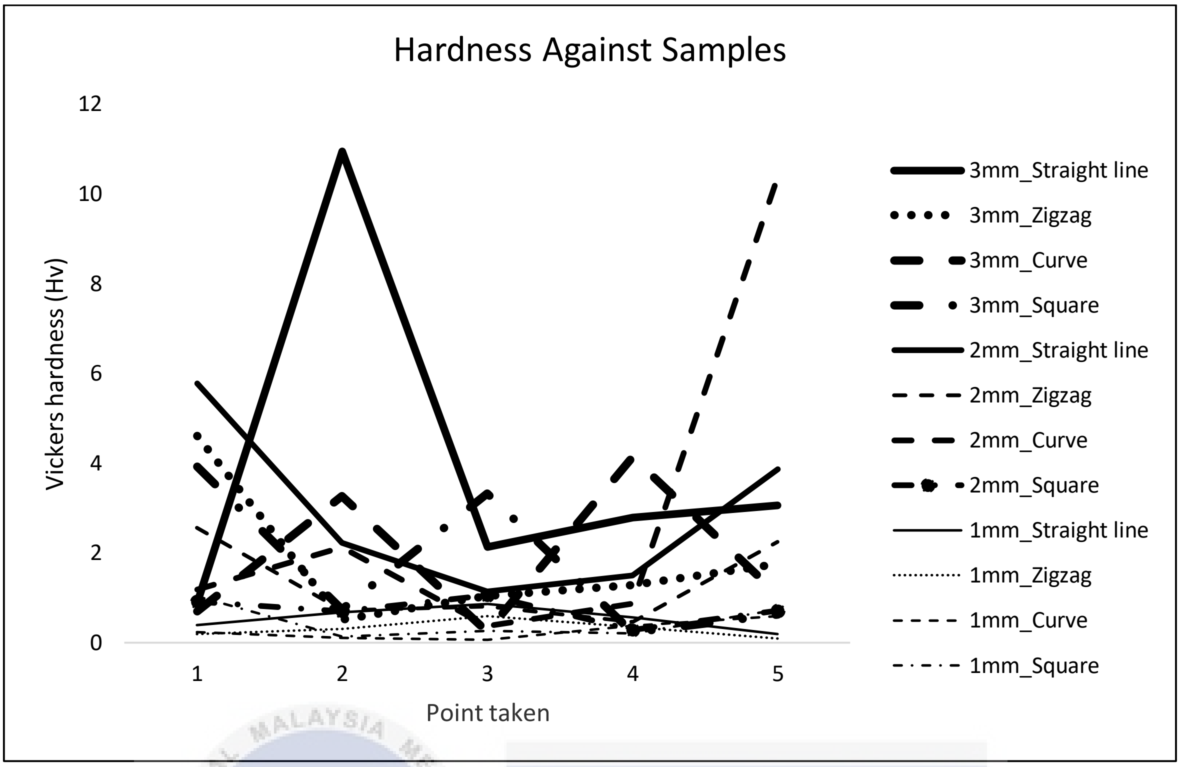


Figure 4.3: Graph hardness against samples

4.8 Percolation threshold effect in graphene conductive ink

Percolation threshold is the lowest concentration of filler at which non-conductive materials is converted to conductive materials (Wypych, 2016). Based on the past study regarding this percolation threshold, 30% wt to 40% wt of filer is the lowest concentration value to filler converting the binder and hardener to conductive materials. In this study, the filler used is 35%, 65% of binder and 30% from binder is hardener concentration. As the suitable value is determined in past study, this study is come out to identify on how the percolation threshold can effect in graphene conductive ink.

In this study, graphene (filler) is combined with non-conductive materials (binder and hardener) which turn into composite of ink. The effect of properties and behaviour of graphene on percolation threshold in this composite is identified through microstructure test. Based on the microstructure result and the properties results, it can be conclude that zigzag pattern shows that the least effective pattern compared to others. This is because it electrical properties shows that it has least an ability to conduct and electricity. By the support of microstructure results, zigzag also has the cruder surface compared to others and has the least connected of high contrast area, which shows that it has lowest number of electron. Theoretically, the low percolation shows the composite has lowest number of electron (Rafiee, 2017).

On the other hand, curve is the best pattern in conducting an electricity as it has low resistivity and voltage which current can flow efficiently in this pattern. Morphological structure of curve pattern also is the smoother, which allow current flow with less resistance. Besides that, SEM images also shows that curve has the biggest high contrast area, which connected together.

As can be seen, it can be conclude that properties and behaviour of the graphene does affected the percolation threshold in conductive ink. This show as the highest contrast are shown in the SEM images, the better the percolation threshold. In addition, graphene has the best properties in conducting electricity, which help more in figured out the percolation threshold effect.

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusions

Overall, the present work had successfully fabricated graphene incorporated with Epoxy resin and Polyetheramine, which acted as composite for conductive ink. In order to produce the ink, material preparation, printing process and curing process was done. All the test was done on the conductive ink samples that have been fabricated. The results from present work of testing had achieved all the objectives as described and drawn below.

Electrical properties shows that the curve which followed by straight line has the best properties condition that allow current efficiently flow while contrast with zigzag pattern. These results is supported by microstructure results regarding on the surface structure and the percolation threshold in the ink. This is also at once prove that the properties and behavior of graphene does effect the percolation in the conductive ink. Besides that, this study also has conclude that 3mm width for the samples is the more suitable width for conductive ink has it shows better properties compared to 2mm and 1mm. For mechanical properties, straight-line pattern is the best in endurance force.

Some results have inconsistently data, which due to the printing process as the surface of the samples might defect during pulling out the samples from the mold. This also can be because of the mixing process as some of materials did not blend enough has all the materials have different density. The dispersion of graphene in the conductive ink is inconsistent. Further research needs to be carried out in order to prove that graphene is the best conductive materials as the conductive ink filler and to produce a low-cost conductive ink with best properties of conductive materials.

5.2 Recommendations

In spite of successful, identify the properties of graphene based conductive ink and the best characteristic for samples there is some recommendations for future works. The recommendations are listed as follows;

1. As the results from experimentally shows some inconsistency, printing process can be one of the factors that might be affect the properties of ink. This is because the mold for the printing ink need to be improve as it might defect the ink during the pulled out process of the printed ink from the mold. The mold can be change with an automatic technology.
2. Next, the materials for the mold also can be change into anti-rust materials such as stainless steel as the present of platinum is detected during the EDX analysis. It might resist the current to flow.
3. Finally, next study should be conducting regards on thickness of samples as it one of factor that might affecting the performance of conductive ink.

REFERENCES

Cui, Z. (2016). Printed Electronics: Materials, Technologies, and Applications. Wiley.

Li, Y., Lu , D., & Wong, C. (2010). *Electrical Conductive Adhesives with Nanotechnologies.* New York Dordrecht Heidelberg London: Springer .

Cruz, S. M. F., Rocha, L. A., & Viana, J. C. (2018). Printing Technologies on Flexible Substrates for Printed Electronics. *Flexible Electronics*.

Alvial-Palavicino, C., & Konrad, K. (2019). The rise of graphene expectations: Anticipatory practices in emergent nanotechnologies. *Futures*, *109*(September 2018), 192–202.

Papageorgiou, D. G., Kinloch, I. A., & Young, R. J. (2017). Progress in Materials Science Mechanical properties of graphene and graphene-based nanocomposites. *Progress in Materials Science*, *90*, 75–127.

Alemour, B., Yaacob, M. H., Lim, H. N., & Hassan, M. R. (2018). *Review of Electrical Properties of Graphene Conductive Composites*. *11*(4), 371–398.

Bayraktar, E. (2016). Section 12 Composites Materials and Technologies. *Reference Module in Materials Science and Materials Engineering*, 1–2.

Cranford, S. W., & Buehler, M. J. (2011). *Packing efficiency and accessible surface area of crumpled graphene*. *205451*, 1–7.

Hancock, Y. (2011). The 2010 Nobel Prize in physics - Ground-breaking experiments on graphene. *Journal of Physics D: Applied Physics*, *44*(47). Lee, C. (2012). *No Title*. *385*(2008).

Liu, L., Qing, M., Wang, Y., & Chen, S. (2015). Defects in Graphene: Generation, Healing, and Their Effects on the Properties of Graphene: A Review. *Journal of Materials Science and Technology*, *31*(6), 599–606.

Mutley, İ., & Tudoran, L. B. (n.d.). *Fullerenes , Nanotubes and Carbon Nanostructures Percolation Behavior of Electrically Conductive Graphene Nanoplatelets / Polymer Nanocomposites : Theory and Experiment*. (February 2014), 37–41.

Park, B., & Lu, R. (2015). Hyperspectral imaging technology in food and agriculture. In *Hyperspectral Imaging Technology in Food and Agriculture*.

Pioneering planetary centrifugal mixers. (n.d.).

Pop, E., Varshney, V., & Roy, A. K. (2012). *Thermal properties of graphene : Fundamentals and applications*. *37*(December), 1273–1281.

polymer nanocomposites. *Polymer*, *52*(1), 5–25. htPotts, J. R., Dreyer, D. R., Bielawski, C. W., & Ruoff, R. S. (2011). Graphene-based

Sarojadevi, H., Shetty, A. B., & Murthy, A. K. (2013). *Digital Image Processing Technique for Microstructure Analysis of Spheroidal Graphite Iron*. 1–5.

Science, C. M. (2014). *Graphene: Synthesis and Preparation 3*. *2*.

Sheehy, D. E. (2009). *Optical transparency of graphene as determined by the fine-structure constant*. 2–5.

Sheet, S. D. (2018). *SIGMA-ALDRICH*. 1–7.

Technique, I. (2017). *Instrumental Technique : Electrode coater (DOCTOR BLADE)*. Technologies, S., Potential, S., Technique, B., Glycol, E., Nanofluids, B., Parameters, C., … Helicopters, L. F. (2019). *SCIENCE & TECHNOLOGY RESEARCH INSTITUTE FOR DEFENCE (STRIDE)*. *12*.

Verma, D., & Goh, K. L. (2019). Functionalized Graphene-Based Nanocomposites for Energy Applications. In *Functionalized Graphene Nanocomposites and their Derivatives*.

Wang, J., Ma, F., Liang, W., & Sun, M. (2017). Electrical properties and applications of graphene , hexagonal boron nitride (h-BN), and graphene / h-BN heterostructures. *Materials Today Physics*, *2*, 6–34.

Wassei, J. K., & Kaner, R. B. (2010). Graphene, a promising transparent conductor. *Materials Today*, *13*(3), 52–59.

Xiong, D., Li, X., Shan, H., Zhao, Y., Dong, L., Xu, H., … Sun, X. (2015). Oxygen-containing Functional Groups Enhancing Electrochemical Performance of Porous Reduced Graphene Oxide Cathode in Lithium Ion Batteries. *Electrochimica Acta*, *174*, 762–769.

Zhang, B., Mei, L., Xiao, H., Zhang, B., Mei, L., & Xiao, H. (2012). *Nanofracture in graphene under complex mechanical stresses Nanofracture in graphene under complex mechanical stresses*. *121915*, 1–6.

Zhang, P., Ma, L., Fan, F., Zeng, Z., Peng, C., Loya, P. E., … Lou, J. (2014). Fracture toughness of graphene. *Nature Communications*, 1–7.

Zhu, B. Y., Murali, S., Cai, W., Li, X., Suk, J. W., Potts, J. R., & Ruoff, R. S. (2010). *Graphene and Graphene Oxide : Synthesis , Properties , and Applications*. 3906–3924.

D. R. Patrick and S. W. Fardo, *Electricity and electronics fundamentals*. Lilburn, GA: Fairmont Press, 2008.

C. A. Pickover, *Archimedes to Hawking: Laws of Science and the Great Minds Behind Them.* Oxford University Press, 2008

R. A. Serway, J. W. Jewett, and V. Peroomian, *Physics for scientists and engineers with modern physics*. Boston, MA: Cengage, 2019.
R. Taherian and A. Kausar, *Electrical conductivity in polymer-based composites: experiments, modelling and applications*. Amsterdam: Elsevier/WA, William Andrew, 2019.

A. E. Shahat, *Electrical resistivity and conductivity*. Croatia, Rijeka: InTech, 2017.

Y. Feng, L. Zhu, Z. Liu, and X. Wang, “Design and Application of Intelligent Plate Making Process for Box Packaging and Printing,” *Advances in Graphic Communication, Printing and Packaging Lecture Notes in Electrical Engineering*, pp. 551–558, 2019.

G. Wypych, “Physical Properties of Fillers and Filled Materials,” *Handb. Fill.*, pp. 303–371, 2016

Rafiee, R. (n.d.). *Carbon nanotube-reinforced polymers from nanoscale to macroscale*. Amsterdam: Elsevier.

LIST OF APPENDICES

APPENDIX A1

Results of voltage for straight line

Width (mm)	Point Taken	Voltage Reading, Sample 1 (mV)			Average (mV)	Voltage Reading, Sample 2 (mV)			Average (mV)	Total Average Voltage (mV)	Standard Deviation	Standard Error
		1	2	3		1	2	3				
1	1	16.29	11.65	37.91	21.95	5.91	2.55	6.46	4.97	13.46	12.01	5.37
	2	9.13	11.99	11.18	10.77	55.19	35.45	52.74	47.79	29.28	26.18	11.71
	3	20.72	28.41	25.06	24.73	25.06	37.38	33.06	31.83	28.28	5.02	2.25
	4	39.93	19.21	39.83	32.99	63.61	40.45	8.84	37.63	35.31	3.28	1.47
	5	32.00	30.02	36.96	32.99	35.55	10.37	22.15	22.69	27.84	7.28	3.26
2	1	23.32	25.59	25.59	24.83	26.15	28.55	30.63	28.44	26.64	2.55	1.14
	2	18.77	23.94	22.04	21.58	21.40	22.93	23.38	22.57	22.08	0.70	0.31
	3	17.49	19.28	19.43	18.73	24.46	25.83	26.06	25.45	20.59	2.63	1.18
	4	14.45	14.76	15.28	14.83	24.79	24.27	24.56	24.54	19.69	6.87	3.07
	5	18.43	17.66	20.16	18.75	21.79	23.31	23.24	22.78	20.77	2.85	1.27
3	1	27.57	27.94	29.70	28.41	29.92	31.73	32.38	31.34	29.88	2.07	0.93
	2	27.51	24.33	25.18	25.67	14.98	18.56	19.37	17.64	21.66	5.68	2.54
	3	37.80	38.21	39.52	38.51	18.52	19.11	19.40	19.01	28.76	13.79	6.17
	4	18.33	19.14	17.98	18.49	14.53	15.48	15.60	15.20	16.85	2.33	1.04
	5	32.39	34.54	35.61	34.18	19.44	20.00	20.06	19.83	27.01	10.15	4.54

APPENDIX A2

Results of voltage for zigzag

Width (mm)	Point Taken	Voltage Reading, Sample 1 (mV)			Average (mV)	Voltage Reading, Sample 2 (mV)			Average (mV)	Total Average Voltage (mV)	Standard Deviation	Standard Error
		1	**2**	**3**		**1**	**2**	**3**				
1	1	41.87	4.43	23.06	23.12	45.16	76.69	26.83	49.56	36.34	18.70	8.36
	2	43.70	32.54	47.39	41.21	26.65	58.30	44.40	43.12	42.16	1.35	0.60
	3	56.16	42.98	22.77	40.64	41.34	32.45	43.48	39.09	39.86	1.09	0.49
	4	19.08	22.94	41.01	27.68	43.82	42.03	24.18	36.68	32.18	6.37	2.85
	5	6.40	26.31	22.03	18.24	40.38	61.55	24.16	42.03	30.14	16.82	7.52
2	1	27.24	26.67	26.78	26.90	20.86	21.57	22.70	21.71	24.31	3.67	1.64
	2	35.52	38.07	39.65	37.75	17.85	19.41	19.85	19.04	28.40	13.23	5.92
	3	21.79	25.19	25.03	24.01	17.45	16.38	19.82	17.88	20.95	4.33	1.94
	4	38.84	38.96	39.38	39.06	20.87	21.83	20.54	21.08	30.07	12.71	5.69
	5	19.99	20.79	20.94	20.57	32.79	37.30	41.74	37.28	28.93	11.82	5.28
3	1	26.10	27.56	28.78	27.48	18.80	20.17	20.35	19.77	23.63	5.45	2.44
	2	27.16	30.21	29.69	29.02	25.90	23.12	22.37	23.80	26.41	3.69	1.65
	3	46.07	45.63	42.95	44.88	21.35	21.17	20.57	21.03	32.96	16.86	7.54
	4	25.84	28.30	28.17	27.44	34.11	30.88	32.16	32.38	29.91	3.49	1.56
	5	25.17	21.95	21.06	22.73	18.11	20.52	22.02	20.22	21.48	1.77	0.79

APPENDIX A3

Results of voltage for curve

Width (mm)	Point Taken	Voltage Reading, Sample 1 (mV)			Average (mV)	Voltage Reading, Sample 2 (mV)			Average (mV)	Total Average Voltage (mV)	Standard Deviation	Standard Error
		1	2	3		1	2	3				
1	1	1.41	35.21	30.50	22.37	59.70	17.91	43.78	40.46	31.42	12.79	5.72
	2	11.65	17.45	26.13	18.41	8.13	63.40	3.81	25.11	21.76	4.74	2.12
	3	22.66	2.53	16.28	13.82	4.23	4.49	4.49	4.41	9.11	6.66	2.98
	4	18.18	30.68	14.30	21.06	18.75	28.11	10.13	19.00	20.03	1.46	0.65
	5	20.72	15.99	15.87	17.53	2.40	3.12	3.33	2.95	10.24	10.30	4.61
2	1	15.57	15.93	15.78	15.76	14.09	14.41	14.65	14.38	15.07	0.98	0.44
	2	12.38	13.57	12.96	12.97	13.14	13.80	14.37	13.77	13.37	0.57	0.25
	3	13.63	15.48	15.48	14.86	22.28	23.88	24.31	23.49	19.18	6.10	2.73
	4	16.90	17.29	17.24	17.15	14.06	14.87	15.20	14.71	15.93	1.73	0.77
	5	13.46	13.57	13.27	13.43	12.78	13.35	13.65	13.26	13.35	0.12	0.05
3	1	13.29	13.89	14.75	13.98	9.60	10.01	10.08	9.90	11.94	2.88	1.29
	2	17.18	21.74	18.57	19.16	9.67	10.02	10.46	10.05	14.61	6.44	2.88
	3	10.10	10.17	10.33	10.20	13.99	14.65	15.35	14.66	12.43	3.15	1.41
	4	15.38	15.72	15.63	15.58	6.37	6.48	6.39	6.41	11.00	6.48	2.90
	5	17.11	16.72	19.59	17.81	15.35	16.00	16.49	15.95	16.88	1.32	0.59

APPENDIX A4

Results of voltage for square

Width (mm)	Point Taken	Voltage Reading, Sample 1 (mV)			Average (mV)	Voltage Reading, Sample 2 (mV)			Average (mV)	Total Average Voltage (mV)	Standard Deviation	Standard Error
		1	**2**	**3**		**1**	**2**	**3**				
1	1	2.86	3.09	3.23	3.06	2.12	40.60	20.25	20.99	12.02	12.68	5.67
	2	3.11	20.43	30.71	18.08	40.79	30.88	30.71	34.13	26.10	11.35	5.07
	3	2.49	66.24	19.51	29.41	1.25	1.47	21.26	7.99	18.70	15.15	6.77
	4	19.21	39.81	18.91	25.98	4.34	2.57	2.64	3.18	14.58	16.12	7.21
	5	39.34	40.71	23.87	34.64	3.09	2.65	2.48	2.74	18.69	22.55	10.09
2	1	13.12	14.44	14.49	14.02	19.01	19.30	19.76	19.36	16.69	3.78	1.69
	2	9.33	9.25	9.19	9.26	21.33	25.48	24.45	23.75	16.51	10.25	4.58
	3	12.83	14.13	13.04	13.33	15.68	16.63	16.87	16.39	14.86	2.16	0.97
	4	14.17	14.73	14.83	14.58	19.50	19.89	19.92	19.77	17.18	3.67	1.64
	5	25.34	27.13	30.44	27.63	17.98	18.50	18.60	18.36	23.00	6.55	2.93
3	1	13.55	14.97	14.58	14.37	16.06	16.41	16.67	16.38	15.38	1.42	0.64
	2	12.46	12.40	12.37	12.41	18.58	19.81	20.05	11.31	15.95	5.00	2.24
	3	11.27	11.16	11.50	11.31	12.94	12.57	13.20	12.91	12.11	1.13	0.51
	4	14.42	15.07	15.55	15.01	10.19	10.68	10.93	10.60	12.81	3.12	1.39
	5	13.52	12.69	12.00	12.74	16.63	17.35	18.34	17.44	15.09	3.32	1.49

APPENDIX B1

Results of resistivity for straight line

Width (mm)	Point Taken	Reading Resistivity for Sample 1 (µΩ/sq)			Average (µΩ/sq)	Reading Resistivity for Sample 2 (µΩ/sq)			Average (µΩ/sq)	Total Average Resistivity (µΩ/sq)	Standard Deviation	Standard Error
		1	**2**	**3**		**1**	**2**	**3**				
1	1	7.38	5.27	17.18	9.97	2.67	1.15	2.92	2.25	6.10	5.44	2.43
	2	4.13	5.43	5.06	4.88	25.01	16.06	23.90	21.66	13.27	11.86	5.30
	3	9.39	12.87	11.35	11.20	11.35	16.94	14.98	14.42	12.81	2.27	1.01
	4	18.09	8.70	18.05	14.95	28.83	18.33	4.00	17.05	16.00	1.48	0.66
	5	14.50	13.60	16.75	14.95	16.11	4.69	10.03	10.28	12.61	3.30	1.47
2	1	10.57	11.59	11.60	11.25	11.85	12.94	13.88	12.89	12.07	1.15	0.51
	2	8.50	10.84	9.99	9.78	9.70	10.39	10.59	10.22	10.00	0.31	0.14
	3	7.92	8.73	8.80	8.49	11.08	11.70	11.81	11.53	10.01	2.15	0.96
	4	5.98	6.54	6.69	6.40	11.23	10.99	11.13	11.12	8.76	3.33	1.49
	5	8.35	8.00	9.13	8.49	9.87	10.56	10.53	10.32	9.42	1.29	0.57
3	1	12.49	12.66	13.46	12.87	13.56	14.37	14.67	14.20	13.54	0.94	0.42
	2	12.47	11.02	11.41	11.63	6.79	8.41	8.77	7.99	9.81	2.57	1.15
	3	17.13	17.31	17.91	17.45	83.93	86.62	87.93	86.16	51.80	48.58	21.72
	4	8.30	86.76	81.50	58.86	65.83	70.16	70.72	68.90	63.88	7.10	3.17
	5	14.68	15.65	16.13	15.49	88.08	90.64	90.90	89.88	52.68	52.60	23.52

APPENDIX B2

Results of resistivity for zigzag

Width (mm)	Point Taken	Reading Resistivity for Sample 1 (µΩ/sq)			Average (µΩ/sq)	Reading Resistivity for Sample 2 (µΩ/sq)			Average (µΩ/sq)	Total Average Resistivity (µΩ/sq)	Standard Deviation	Standard Error
		1	**2**	**3**		**1**	**2**	**3**				
1	1	18.97	2.00	10.44	10.47	18.97	2.00	10.44	10.47	16.47	8.47	3.79
	2	19.80	14.74	21.47	18.67	19.80	14.74	21.47	18.67	17.29	1.95	0.87
	3	25.45	19.47	10.32	18.41	25.45	19.47	10.32	18.41	18.06	0.49	0.22
	4	8.64	10.39	18.58	12.53	8.64	10.39	18.58	12.53	14.58	2.89	1.29
	5	2.89	11.92	9.98	8.26	2.89	11.92	9.98	8.26	13.65	7.62	3.40
2	1	12.34	12.08	12.13	12.19	9.45	9.76	10.28	9.84	11.01	1.66	0.74
	2	16.10	17.25	17.96	17.10	8.09	8.79	8.99	8.62	12.86	5.99	2.68
	3	9.87	11.41	11.34	10.88	7.91	7.42	8.98	8.10	9.49	1.96	0.87
	4	17.60	17.65	17.84	17.70	9.45	9.89	9.30	9.55	13.62	5.76	2.57
	5	9.06	9.42	9.49	9.32	14.86	16.90	18.91	16.89	13.11	5.35	2.39
3	1	11.82	12.49	13.04	12.45	8.52	9.14	9.22	8.96	10.70	2.46	1.10
	2	12.30	13.69	13.45	13.15	11.74	10.47	10.13	10.78	11.96	1.67	0.74
	3	20.87	20.68	19.46	20.34	9.67	9.59	9.32	9.53	14.93	7.64	3.41
	4	11.71	12.82	12.76	12.43	15.46	13.99	14.57	14.67	13.55	1.58	0.70
	5	11.40	9.95	9.54	10.30	8.21	9.30	9.98	9.16	9.73	0.80	0.360

APPENDIX B3

Results of resistivity for curve

Width (mm)	Point Taken	Reading Resistivity for Sample 1 (μΩ/sq)			Average (μΩ/sq)	Reading Resistivity for Sample 2 (μΩ/sq)			Average (μΩ/sq)	Total Average Resistivity (μΩ/sq)	Standard Deviation	Standard Error
		1	**2**	**3**		**1**	**2**	**3**				
1	1	0.63	15.95	13.82	10.13	27.05	8.11	19.84	18.33	14.23	5.79	2.59
	2	5.27	7.09	11.84	8.34	3.68	28.73	1.72	11.38	9.86	2.14	0.96
	3	10.26	1.47	7.37	6.26	19.19	2.03	2.03	7.75	7.09	1.05	0.47
	4	8.24	13.06	6.48	9.54	8.50	12.73	4.58	8.60	9.07	0.66	0.29
	5	9.39	7.26	7.19	7.94	1.08	1.41	1.51	1.33	4.64	4.67	2.08
2	1	7.09	7.28	7.15	7.14	6.38	6.53	6.64	6.51	6.83	0.44	0.19
	2	5.61	6.10	5.87	5.87	5.95	6.25	6.51	6.24	6.06	0.25	0.11
	3	6.17	7.06	7.05	6.75	10.09	10.82	11.06	10.64	8.69	2.75	1.23
	4	7.66	7.88	7.81	7.77	6.37	6.74	6.88	6.66	7.21	0.78	0.35
	5	6.10	6.10	6.01	6.08	5.79	6.05	6.18	6.01	6.04	0.05	0.02
3	1	6.02	6.27	6.68	6.33	4.35	4.53	4.56	4.48	5.41	1.30	0.58
	2	7.78	9.81	8.41	8.68	4.38	4.54	4.74	4.55	6.62	2.92	1.30
	3	4.57	4.69	4.68	4.62	6.33	6.64	6.95	6.64	5.63	1.43	0.64
	4	6.97	7.16	7.08	7.06	2.88	2.93	2.89	2.90	4.98	2.93	1.31
	5	7.75	7.57	8.87	8.07	6.95	7.25	7.47	7.22	7.64	0.59	0.26

APPENDIX B4

Results of resistivity for square

Width (mm)	Point Taken	Reading Resistivity for Sample 1 (μΩ/sq)			Average (μΩ/sq)	Reading Resistivity for Sample 2 (μΩ/sq)			Average (μΩ/sq)	Total Average Resistivity (μΩ/sq)	Standard Deviatio n	Standard Error
		1	**2**	**3**		**1**	**2**	**3**				
1	1	1.29	1.39	1.46	1.38	0.95	18.40	9.17	9.51	5.44	5.74	2.56
	2	1.40	9.25	13.91	8.19	18.48	13.99	13.91	15.46	11.83	5.14	2.30
	3	1.12	30.02	8.84	13.33	0.56	0.66	9.63	3.62	8.47	6.86	3.07
	4	8.70	18.04	8.57	11.77	1.96	1.16	1.19	1.44	6.68	7.30	3.26
	5	17.88	18.45	10.81	15.69	1.40	1.20	1.12	1.24	8.47	10.22	4.57
2	1	5.94	6.54	6.56	6.35	8.61	8.74	8.95	8.77	7.56	1.71	0.76
	2	4.23	4.19	4.16	4.19	9.66	11.54	11.08	10.76	7.48	4.64	2.07
	3	5.81	6.40	5.90	6.04	7.10	7.53	7.64	7.43	6.73	0.98	0.43
	4	6.42	6.67	6.72	6.60	8.83	9.01	9.02	8.96	7.78	1.66	0.74
	5	11.43	12.29	13.77	12.55	8.14	8.38	8.42	8.32	10.43	2.97	1.32
3	1	6.12	6.78	6.61	6.51	7.28	7.43	7.55	7.42	6.96	0.64	0.28
	2	5.68	5.61	5.60	5.62	8.41	8.98	9.08	8.82	7.22	2.26	1.01
	3	5.10	5.06	5.21	5.12	5.86	5.69	5.98	5.84	5.48	0.51	0.22
	4	6.53	6.83	7.04	6.80	4.61	4.82	4.95	4.80	5.80	1.41	0.63
	5	6.12	5.75	5.43	5.77	7.53	7.86	8.31	7.90	6.83	1.50	0.67

APPENDIX C1

Results of hardness for straight line

Width (mm)	Point	Force (mN)	Sample 1		Sample 2		Average Depth (µm)	Average Vickers Hardness (HV)
			Depth (µm)	Vickers Hardness (HV)	Depth (µm)	Vickers Hardness (HV)		
1	1	6	0.42	0.13	1.10	0.63	0.76	0.38
	2		0.28	0.79	0.04	0.57	0.16	0.68
	3		0.00	1.37	0.64	0.34	0.32	0.86
	4		0.01	0.48	4.36	0.63	2.19	0.55
	5		0.25	0.27	0.63	0.10	0.44	0.19
2	1	6	0.25	0.64	0.00	10.91	0.12	5.78
	2		0.89	0.15	0.00	4.29	0.44	2.22
	3		0.40	0.32	0.00	1.94	0.20	1.13
	4		0.18	1.02	0.00	1.97	0.09	1.49
	5		0.00	6.04	0.01	1.67	0.00	3.86
3	1	6	0.18	0.71	0.51	1.00	0.34	0.85
	2		0.00	16.29	0.00	5.59	0.00	10.96
	3		0.00	2.54	0.00	1.71	0.00	2.12
	4		0.00	2.68	0.90	2.88	0.45	2.78
	5		0.20	1.20	0.00	4.91	0.10	3.05

APPENDIX C2

Results of hardness for zigzag

Width (mm)	Point	Force (mN)	Sample 1		Sample 2		Average Depth (µm)	Average Vickers Hardness (HV)
			Depth (µm)	Vickers Hardness (HV)	Depth (µm)	Vickers Hardness (HV)		
1	1	6	0.31	0.14	1.12	0.22	0.72	0.18
	2		1.22	0.05	0.00	0.56	0.61	0.30
	3		0.32	0.82	0.64	0.34	0.48	0.58
	4		4.98	0.05	4.36	0.63	4.67	0.34
	5		0.80	0.08	0.43	0.10	0.61	0.09
2	1	6	0.00	4.33	0.38	0.78	0.19	2.55
	2		0.26	0.49	0.01	0.93	0.14	0.71
	3		0.24	0.50	0.01	1.09	0.13	0.79
	4		0.40	0.39	0.01	0.53	0.20	0.46
	5		0.00	1.68	0.00	2.80	0.00	2.24
3	1	6	0.00	5.19	0.00	4.00	0.00	4.60
	2		0.33	0.56	0.01	0.46	0.17	0.51
	3		0.11	1.38	1.56	0.68	0.83	1.03
	4		0.18	1.25	0.00	1.30	0.09	1.28
	5		0.00	0.66	0.00	2.83	0.00	1.74

APPENDIX C3

Results of hardness for curve

Width (mm)	Point	Force (mN)	Sample 1		Sample 2		Average Depth (µm)	Average Vickers Hardness (HV)
			Depth (µm)	Vickers Hardness (HV)	Depth (µm)	Vickers Hardness (HV)		
1	1	6	0.24	0.24	5.56	0.21	2.90	0.23
	2		1.01	0.10	1.14	0.10	1.07	0.11
	3		2.15	0.03	0.28	0.09	1.21	0.06
	4		0.21	0.57	0.66	0.17	0.43	0.37
	5		0.25	0.74	10.40	0.45	5.32	0.59
2	1	6	0.00	2.18	0.52	0.18	0.26	1.18
	2		0.02	0.89	0.00	3.36	0.01	2.12
	3		1.54	0.29	0.28	0.44	0.91	0.37
	4		0.33	0.44	0.00	1.29	0.16	0.86
	5		4.03	0.57	0.00	20.18	2.01	10.38
3	1	6	1.54	0.38	0.70	1.01	1.12	0.70
	2		0.19	0.74	0.00	5.79	0.09	3.27
	3		3.08	0.32	4.55	0.15	3.81	0.24
	4		0.55	0.34	0.00	7.92	0.27	4.13
	5		0.18	0.80	0.16	1.32	0.17	1.06

APPENDIX C4

Results of hardness for square

Width (mm)	Point	Force (mN)	Sample 1		Sample 2		Average Depth (µm)	Average Vickers Hardness (HV)
			Depth (µm)	Vickers Hardness (HV)	Depth (µm)	Vickers Hardness (HV)		
1	1	6	0.20	1.99	8.19	0.04	4.20	1.02
	2		3.54	0.12	0.75	0.16	2.15	0.13
	3		0.27	0.33	0.59	0.19	0.43	0.26
	4		0.76	0.13	6.46	0.26	3.61	0.19
	5		0.29	1.00	0.01	0.46	0.15	0.73
2	1	6	0.01	1.77	8.19	0.06	4.10	0.91
	2		0.00	1.25	0.75	0.13	0.37	0.69
	3		0.00	1.92	0.59	0.19	0.29	1.06
	4		0.37	0.30	6.46	0.26	3.42	0.28
	5		0.00	0.95	0.01	0.46	0.01	0.70
3	1	6	0.01	1.77	0.00	6.06	0.00	3.91
	2		0.00	1.25	1.85	0.23	0.92	0.74
	3		0.00	1.92	0.00	4.72	0.00	3.32
	4		0.37	0.30	0.57	0.14	0.47	0.22
	5		0.00	0.95	1.17	0.30	0.59	0.62