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

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

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**in fulfillment of the requirements for the degree of**
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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.

Signature : .....

Name : AINA NATASHA BINTI HOSNIE

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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## 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).