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UNIVERSITI TEKNIKAL MALAYSIA MELAKA

Faculty of Mechanical Engineering

EFFECT OF LOW TEMPERATURE ON THE PERFORMANCE OF CARBON-BASED CONDUCTIVE INK FOR FLEXIBLE PRINTED CIRCUIT

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Master of Mechanical Engineering (Energy Engineering)

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EFFECT OF LOW TEMPERATURE ON THE PERFORMANCE OF CARBON-BASED CONDUCTIVE INK FOR FLEXIBLE PRINTED CIRCUIT

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**A thesis submitted
in fulfilment of the requirement for the degree of Master of
Mechanical Engineering (Energy Engineering)**

Faculty of Mechanical Engineering

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2019

DECLARATION

I declare that this thesis entitle "Effect of Low Temperature on the Performance of Carbon-Based Conductive Ink for Flexible Printed Circuit" the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature :

Name : NORSHEILA BINTI BUYAMIN

Date : 15/3/2019

APPROVAL

I hereby declare that i have read this thesis and in my opinion this thesis is sufficient in terms of scope and quality for the Master of Mechanical Engineering (Energy Engineering)

Signature :

Supervisor Name : DR. MOHD NUR AZMI BIN NORDIN

Date : 15/3/2019

DEDICATION

To my beloved husband, children, mother, father, mother in law and all family member

ABSTRACT

Nowadays, it is known that printing methodologies can be used to print electrically functional devices on a variety substrates such as organic and inorganic substrate's materials. Currently, printing technologies have been an attractive alternative printing method to fabricate flexible electronic devices, keeping to its advantages including easy handling, wide use and low cost. However, using flexible substrate open up new possibilities for printed electronic (PE), where certain applications expose the flexible substrate to a mechanical bending which might decreased the performance or becomes a cause to a functionality. Moreover, developing of an appropriate ink at an extreme surrounding temperature with high conductivity and good dispersion of the ink-jet printing is one of the critical problems that need to be solved. Thus, the objectives of this study are determine the effects of cyclic loading on the elasticity and the conductivity of carbon-based conductive ink after exposing to low temperature. The sample of conductive ink has been tested by resistivity test, nanoindentation test and microstructure analysis after doing cyclic test. This study is focused on the conductivity behaviour of printed conductive ink after being exposed to two different temperature; low temperature at -6°C and room temperature at 26°C. After being exposed to each of the temperature set, the cyclic loading test of 1000 and 5000 cycles each was carried out where non-cyclic sample was prepare as the bench-marking sample. From the cyclic test result, it is understood that the resistivity of printed ink at low temperature is lower than that of at room temperature. Besides, the lowest resistivity is recorded as 105.71Ω/sq for sample that being tested at 5000 cyclic after exposed to low temperature. This is due to the deformation of the elasticity properties of the ink when exposed to the low temperature after the printing process. On the other hand, the hardness of low temperature sample is larger than room temperature. However, the highest resistivity in this study is 5000 cyclic sample at room temperature almost 206.52Ω/sq. Moreover, in cyclic test, the printed conductive ink will be bend and stretched at the maximum limit. Therefore, the surface structure become scratch and crack onto conductive ink. Finally, the 5000 cyclic sample at room temperature shows more defect on the surface structure than others.

ABSTRAK

Pada masa kini menunjukkan *kaedah-kaedah mencetak boleh digunakan di dalam fungsi peranti eletrikal yang dicetak di atas pelbagai permukaan substrat samada dalam permukaan organik atau bukan organik. Pada masa ini juga, teknologi percetakan mempunyai alternatif kaedah percetakan yang menarik untuk mereka bentuk fleksibel peranti elektronik di mana antara kelebihan-kelebihannya adalah senang diurus, penggunaan yang meluas dan harga yang lebih murah. Walaubagaimanapun, penggunaan fleksibel substrat membuka satu kemungkinan kapada percetakan elektronik di mana aplikasi lenturan percetakan elektronik ini mendedahkan ia kepada mekanikal lenturan yang berkemungkinan menyebabkan kecekapan berkurangan atau menjadikannya gagal. Lebih-lebih lagi adalah dengan membangunkan dakwat aliran yang sesuai pada suhu persekitaran yang terlampau dengan kekonduksian yang tinggi dan penyebaran yang bagus dalam percetakan dakwat jet merupakan salah satu masalah kritikal yang perlu diselesaikan. Oleh itu, objektif kajian ini adalah menentukan kesan-kesan beban kitaran ke atas keanjalan dan kekonduksian dakwat konduksi berasaskan karbon selepas didedahkan kepada suhu yang rendah. Selepas melakukan ujian kitaran, sampel dakwat konduksi karbon telah diuji dengan ujian kerintangan, ujian nanopelekukan dan analisis mikrostruktur. Kajian ini difokuskan kepada dua suhu yang berbeza iaitu pada suhu rendah, -6°C dan pada suhu bilik, 26°C. Selepas didedahkan kepada suhu yang ditetapkan, ujian 1000 dan 5000 kitaran beban telah dijalankan di mana sampel yang tiada kitaran dijadikan sebagai sampel rujukan. Daripada keputusan ujian kitaran, ia difahamkan bahawa kerintangan dakwat percetakan pada suhu rendah adalah lebih rendah daripada suhu bilik. Selain itu, kerintanagn paling rendah adalah dicatatkan sebagai 105.71Ω/sq bagi sampel pada 5000 kitaran selepas didedahkan kepada suhu yang rendah. Ini juga disebabkan oleh sifat pemanjangan elastic dakwat apabila didedahkan kepada suhu rendah selepas proses percetakan. Di antara lain adalah kekerasan pada sampel suhu rendah adalah lebih besar daripada suhu bilik. Walau bagaimanapun, kerintangan yang tertinggi adalah sampel 5000 kitaran pada suhu bilik dengan nilainya adalah 206.52Ω/sq. Tambahan lagi, dalam ujian kitaran, percetakann dakwat, konduksi akan melentur dan menegang pada tahap maksimum. Oleh itu, permukaan struktur menjadi calar dan retak ke atas dakwat konduksi tersebut. Akhiryer, sampel 5000 kitaran pada suhu bilik menunjukkan terdapat lebih kecacatan pada permukaan struktur berbanding sampel yang lain.*

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LIST OF ABBREVIATIONS

3-MPTMS	Mercaptopropyltrimethoxysilane
ASTM	Amarican Socity for Testing and Materials
CB	Carbon Black
CNT	Carbon Nanotubes
DC	Direct Current
LT	Low Temperature
FE	Antennas on a Flexible
PCB	Printed Circuit Board
PE	Printed Electronic
PET	Flexible Polyethylene Terephthalate
PTF	Polymer Thick Film
RT	Room Temperature
SEM	Scanning Electron Microscopy
TPU	Polyurethane

CHAPTER 1

INTRODUCTION

1.1 Background

Printed electronics refers to the application of printing technologies as a fabrication method to meet industrial requirements for low cost and high volume demand of electronic circuits and devices at low processing temperature. Besides the recognized technological advantages, printed electronic (PE) is estimated to grow into a multibillion business in the near future (Happonen, 2016). A potential high volume of production method for printed electronics is roll-to-roll (R2R) printing (Kololuoma *et al.*, 2004; Noh *et al.*, 2010; Kim *et al.*, 2012) enabling a non-stop electrical structure production with a low cost of substrate than printed circuit board (PCB). A few researcher stated that this fabrication method also facilities of manufacturing devices with organic semiconductor, such as solar cell(Søndergaard *et al.*, 2012; Krebs *et al.*, 2013). Moreover, the market of printed application is estimated to exceed $300 billion, i.e. requires the methods be more efficient, faster, cheaper and user friendly (Kamyshny and Magdassi, 2014).

Another advantage in PE is the possibility of using various type of flexible substrates on the electrical structure. This includes polymers, papers and also fabrics. Recently, most of the conductive circuits use the flexible and stretchable polymer substrate in order to vary the applications, besides to enhance the flexibility of the circuit for reduction critical problem purpose. In this study, the flexible polyethylene terephthalate (PET) and polyurethane (TPU) thin film is employed as a substrate for carbon-based conductive ink sample. Basically, a manufacturing process and materials used such as devices and circuit line printed on flexible (bendable polymer or paper) substrate is an important thing to be concerned. Traditionally, electronics devices were produced by photolithography, vacuum deposition and electroless plating processes. All these methods are multistage and required high cost equipment and used an undesirable chemical which become large amounts of waste. Table 1.1 shows the comparison between the traditional and electronic printing process.

Table 1.1 Comparison traditional printing and printed electronics (Joshi, 2011)

Requirements	Traditional	Electronics
Resolution	15 – 100micron	<<20 micron
Registration	Low	High
Edge Sharpness	High	Very High
Uniformity of Layers	Not Really Important	Very Important
Adhesion of Layers to Substrate	Important	Important
Adhesion of Layers to Other layers	Less Important	Very Important
Solvents in Ink	Cost Issues	Functional Issues
Purity of Solution	Not Really Important	Very Important
Visual Properties	Very Important	Not Important
Electrical Properties	Not Important	Very Important

Currently, ink-jet printing technology has received growing attention as a method to produce a low cost of electronics products, sensors and antennas on a flexible (FE) and printed electronics (PE) devices. While, products of the FE and PE requiring to cover all the electronic application, however they have certain elements that are in common. One of the important elements is the need for conductivity. Furthermore, any technology exposes commercially when it has the potential to sustained, long-term significance in various applications.

All ink-jet printing technologies are based on the controlled operation and injected into the nozzle to drops of liquids ink with diameter range between 50-80μm onto a substrate. After reaching on the substrate, the droplet ink spread and blends with the previously deposited droplets to form a line or pattern. The printed line is then dried and sintered or cured by another process if necessary, to form a solid track (Sze Kee Tam *et al*, 2016). This process takes a long time to ensure the ink is fully attached onto the substrate.

1.2 Problem Statement

Currently, the ink-jet printing technologies have been an attractive alternative printing method to fabricate flexible electronic devices, keeping to its advantages including easy handling, wide use and low cost (Zhang *et al.*, 2016). But, using flexible substrate open up new possibilities for PE, where the application needs to be used in a curved form or requires repeated cyclic loading. Additionally, bending a PE application exposes it to a mechanical bending which might cause lower performance or becomes a failure (Happonen, 2016). The other issue is developing of an appropriate ink at an extreme surrounding temperature with high conductivity and good dispersion of the ink-jet printing are the challenges for the manufactures. In fact, when conductive ink exposes to the low temperature the grain of the material will become smaller from the original size and the conductivity ink will be increased. Thus, this study aims to determine the effect of low temperature on the mechanical properties and the performance of the carbon-based conductive ink. Several issues are listed below to address the problem statement throughout this research project: -

i. What the performance effect of carbon-based conductive ink after exposing at low temperature?

ii. How does carbon-based conductive ink will react at low temperature?

iii. How does cyclic loading will give an effect on the elasticity of carbon-based conductive ink at low temperature?

iv. What the different performance effect of modulus and resistivity carbon-based conductivity ink between expose at room temperature and low temperature?

1.3 Objectives

The purposes of this research projects are: -

i. To investigate the conductivity of carbon-based conductive ink after exposing to low and room temperature.

ii. To determine the effect of cyclic loading on the elasticity of carbon-based conductive ink at low temperature

1.4 Project Scope

The scopes of this research are focused on:-

i. Fabrication of carbon-based conductive ink sample by a manual hand-printing technique using a mesh-screen stencil on flexible printed circuit; polyethylene terephthalate (PET) and polyurethane (TPU) substrate.

ii. No new composition is developed during the work, however, conductive ink is purchasable from dedicated suppliers.

iii. Experimental works on carbon-based conductive ink sample to find out the effect of different low temperatures on the mechanical properties performance of the carbon-based conductive ink.

iv. Comparison of the sheet resistance, hardness and young's modulus readings between room temperature and low temperature.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter reviewed some of the related topics towards in this research based on the current and previous research. The main topics that have been reviewed are flexible printed circuit, conductive ink, electrical and mechanical properties of conductive ink, effect of low temperature and cyclic loading on printed conductive ink. Printed circuit is a wide electrical application usage in the current industries. Currently, many studies have begun to focus on the investigation of highly performance and flexible usage of printed circuit.

2.2 Flexible Printed Circuit

Nowadays, the stretchable and flexible substrate is commonly used on electronic circuits to replace a rigid printed circuit board (PCB) for electronic applications. The printed electronic belong to an effectively functional conductive ink that can be used high precision conducting circuits on flexible substrates such as paper, polymide, polyethylene terephthalate (PET) and polyurethane (TPU), using simple and low cost techniques (Liu *et al.*, 2016).

The proper substrate used in electronic circuits have many requirement. Hence, the specific adhesion of the inks onto the substrate is needed in a proper range of substrate energy surface. In fact, substrate should be non-conductive so that it is not related with electrical properties of conductive ink., Moreover, it should be flexible and have a sufficient of smoothness and porosity that will be printed conductive ink thick film (Kattumenu, 2008).

Therefore, this study is focusing on the flexible substrate which are PET and TPU to investigate which one is suitable with carbon-based conductive ink when exposed process at room temperature and low temperature.

PET is an excellent commercial thermoplastic polymer resin of the polyester family. PET substrate has attracted interest all the researcher because it is low cost, good thermal stability, space inertness, good spin ability and excellent moisture resistance. The most important that many researcher are using this substrate because of its optical properties with optical transmission higher than 85% in the visible range and also it have mechanically flexible under bending or buckling conditions (Faraj *et al.*, 2011). However, TPU is a unique category of plastic where it has high elongation and tensile strength; elasticity; ability to resist oil, grease, solvents, chemicals and abrasion. These characteristic make TPU extremely popular among market industries as a substrate (Huntsman, 2006).

In recent years, research interests of printed electronics on polymer flexible substrate are increasing due to growing demand for flexible display, radio frequency identification tags, x-ray; electronic paper; electronic solar cell arrays, wearable electronics and biomedical devices as shown in Figure 2.1 (Choi *et al.*, 2008; Wang *et al.*, 2016). The printed circuit is one of the major components that acknowledge the development of conductive ink especially in terms of types and formulating ink where both of substances; ink and substrate, will react with each other. Various factors have been considered in formulating ink such as the ratio of ingredients, method, temperature and many others. In addition, the conductive ink must be well-matched with the substrate due to compatible and adhesion of both substances. Apart from that, an additional process of heat treatments such as sintering or curing are carried out to ensure the adhesion of thin film ink on their substrate whole improving its mechanical and electrical properties.

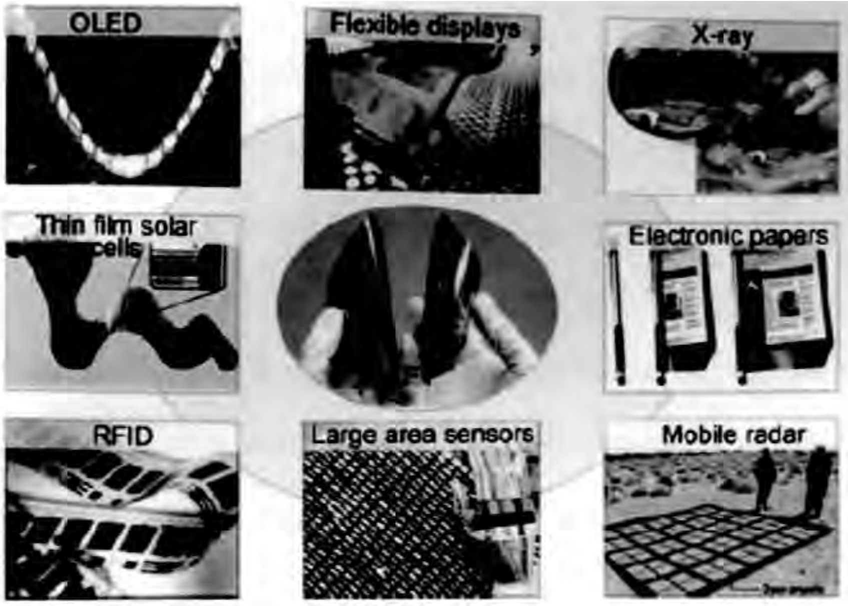

Figure 2.1 Flexible substrate enabling technologies for a whole range of novel applications (Zhouping *et al.*, 2010)

2.3 Conductive Ink

Conductive ink for printed electronics applications is commonly made of metal particles filler in a polymer matrix (Salam *et al.*, 2011). There are various types of conductive materials use as the fillers for conductive ink such as copper, silver, conductive polymer (Xiong and Liu, 2012), grapheme (Huang *et al.*, 2011), carbon (Pidcock and In Het Panhuis, 2012) and metal nanoparticles. However, conductive polymer, graphene and carbon fillers have lower conductivity as compared to the metal filler. Meanwhile, copper-based conductive ink is naturally ten times less conductive as compared to the silver-based conductive ink but with more cost-effectively (Sze Kee Tam *et al.*, 2016).

Normally, the quality of printed ink performance is based on the formulation of the conductive ink including the amount of conductive fillers, binder, dispersing medium, resin, dispersant and other additives.(Sze Kee Tam *et al.*, 2016). Recently, many researchers are focusing on the development of conductive ink properties that influencing the functionality of the circuit. P. Liu et al. (2016) mentioned that silver filler that is incorporated in the conductive ink is a highly conductive metal with a good oxidation resistance, has large contact area and also has been used widely as a conductive material (Liu *et al.*, 2016). In addition, B. Salam et al (2011). stated that copper based conductive ink inherently has lower material cost and higher electrical resistivity compared with silver-based conductive ink (Salam *et al.*, 2011). Moreover, the improvement of conductive ink in recent years involve the development of conductive ink directly at the ink-jet printing method, the enhancement of filler-substrate contact area, and also focusing on its mechanical and electrical properties.

2.4 Electrical and Mechanical Properties of Conductive Ink

2.4.1 Silver-Based Conductive Ink

In the technology era, silver-based conductive ink is one of the metal fillers that is widely used to form a conductive electronic line for PE applications. In addition to the excellent charge transport properties, the conductive ink for ink-jet and printing applications must meet other requirements as a general ink such as high conductivity, low resistivity, low viscosity, high chemical stability, low temperature process ability and surface tension (Wang *et al.*, 2015). Moreover, the electrical and mechanical properties of silver particles ink is highly depended on its size, shape and applied chemical's treatment. Nanoparticles silver materials with shape spherical, cubic and flake-like morphologies show special properties. As compared to the spherical-shape silver nanoparticle, has more advantages such as larger contact area and better electrical properties (Sun *et al.*, 2003; Gilbertson *et al.*, 2015).

Metal nanoparticles act as an important element in conductive ink. Recently, many researchers have focused on the development of the conductive ink directly at the ink-jet printing process on the substrate, the nanoparticles conductive ink (Lee *et al.*, 2009) is available in the market with nanoparticles of silver (30-50wt.%) are blended in a solvent