Faculty of Mechanical Engineering

INTERFACIAL ADHESION BETWEEN SILVER INK AND THERMOPLASTIC POLYURETHANE AND ELECTROMECHANICAL RELIABILITY OF FLEXIBLE PRINTED CIRCUIT

Afiqah binti Mohd Yunos

Master of Science in Mechanical Engineering

2021

INTERFACIAL ADHESION BETWEEN SILVER INK AND THERMOPLASTIC POLYURETHANE AND ELECTROMECHANICAL RELIABILITY OF FLEXIBLE PRINTED CIRCUIT

AFIQAH BINTI MOHD YUNOS

A thesis submitted
in fulfillment of the requirements for the degree of Master of Science
in Mechanical Engineering

Faculty of Mechanical Engineering

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

2021

DECLARATION

I declare that this thesis entitled "Interfacial Adhesion Between Silver Ink and Thermoplastic Polyurethane and Electromechanical Reliability of Flexible Printed Circuit" is 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 : AFIQAH BINTI MOHD YUNOS

Date : 24/09/2021

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 award of Master of Science in Mechanical Engineering.

Signature : ....

Name : PROFESOR IR. DR. GHAZALI BIN OMAR
Timbalan Naib Canselor (Penyelidikan & Inovasi)
Universiti Teknikal Malaysia Melaka

Date : 24/09/2021

DEDICATION

To my beloved mother, father, siblings and friends

ABSTRACT

Flexible printed circuit (FPC) is one of the promising components in the electronic industries. The advantages of FPC are that its fabrication process is environmentally friendly, low cost, and efficient, which makes it a favourable choice for applications in industrial and medical. The conductive ink and substrate are the main components of FPC and they need to perform with good flexibility as that indicates that it is able to withstand a degree of deformation before occurring loss in conductivity. Nonetheless, issues that arise are interfacial adhesion strength between conductive ink and substrate, and reliability of conductive ink upon exposure to a stretchable type of deformation. Therefore, this study aims to investigate two different adhesion-enhancing techniques, which are thermal control and self-assembled monolayer of adhesion promoter. In addition, reliability of the conductive ink when exposed to stretchable type of deformation is also investigated. The silver conductive ink and thermoplastic polyurethane (TPU) were used in this study. The thermal control technique involved curing printed silver conductive ink at selected temperatures: room temperature, 60^{o}C, 80^{o}C, 100^{o}C, 120^{o}C, 130^{o}C, and 140^{o}C. The TPU underwent thermal analysis by using Differential Scanning Calorimetry (DSC) to study thermal properties of TPU. Meanwhile, self-assembled monolayer technique involved the construction of adhesion promoter layer onto the surface of TPU by dipping it in the adhesion promoter solution. Adhesion promoter used in this study was 3-aminopropyltrimethoxy silane (APTS). The success of the APTS construction was evaluated through Fourier Transform Infrared Spectroscopy (FTIR) and water contact angle (WCA) analysis. The evaluation of adhesion performance was assessed according to the cross-cut test (ASTM D3359) and 180^o peel-test. The changes in electrical, mechanical, and surface energy characteristics were carried out within this research to investigate whether these techniques would cause changes in the mentioned properties. The experimental results showed an improvement in adhesion when both methods were executed. The thermal control has 4B/0.22 N/mm to 5B/0.55 N/mm rating when curing started at 100^{o}C to 140^{o}C. APTS-treated TPU showed adhesion was improved to 3B/0.17 N/mm. An increase in conductivity of printed silver with lower hardness was observed when the temperature was elevated. However, insignificant changes in conductivity and hardness were observed for APTS-treated TPU. The surface energy of TPU changed when it was exposed to thermal, showing an insignificant effect in promoting the adhesion. The improvement of adhesion was described as influenced by the changes in thermal properties of TPU. Meanwhile, surface energy of APTS-treated TPU showed polar properties due to the presence of polar head functional groups that allowed affinity bond with the silver particles. Reliability of silver ink was tested by printing with different geometrical patterns (straight, square, sinusoidal, and zig-zag), and different widths (1 mm, 2 mm, and 3 mm). The electromechanical measurement was carried out by manually stretching the pattern until it lost its conductivity. Zig-zag with 3 mm width showed excellent electromechanical performance with 7.78% maximum strain.

PELEKATAN ANTARAMUKA ANTARA DAKWAT PERAK DENGAN POLIURETENA TERMOPLASTIK DAN KEBOLEHHARAPAN ELEKTROMEKANIKAL LITAR CETAK FLEKSIBEL

ABSTRAK

Litar bercetak fleksibel (FPC) adalah salah satu komponen yang diyakini dalam industri elektronik. Kelebihan seperti proses mesra alam, murah dan prosess pembuatan yang efisien menjadikannya digemari dalam sektor industri dan perubatan. Dakwat konduktif dan substrat merupakan komponen utama dalam FPC perlu bersifat fleksibel agar mampu menahan tahap ubah bentuk sebelum kehilangan kekonduksiannya. Namun, terdapat masalah timbul iaitu kekuatan lekatan antara dakwat dan substrat serta kebolehpercayaan dakwat konduktif apabila perubahan bentuk jenis regangan dikenakan. Oleh itu, kajian dijalankan bagi menyiasat dua teknik peningkatan lekatan iaitu kesan haba dan monolayer dipasang sendiri. Selain itu, kebolehpercayaan dakwat konduktif apabila terdedah kepada jenis ubah bentuk yang boleh diregangkan juga disiasat. Dakwat konduktif perak dan termoplastik poliuretana (TPU) digunakan di dalam kajian. Teknik kesan haba melibatkan pengawetan dakwat konduktif perak pada suhu yang terpilih (suhu bilik, 60^{o}C, 80^{o}C, 100^{o}C, 120^{o}C, 130^{o}C dan 140^{o}C). Analisi haba terhadap TPU dijalankan menggunakan Calorimetri Pengimbasan Berbeza (DSC). Sementara itu, teknik monolayer dipasang sendiri melibatkan pembinaan lapisan penggalak lekatan. Penggalak lekatan digunakan dalam kajian ini adalah 3-aminopropyltrimethoxy silane (APTS). Kejayaan pembinaan APTS dinilai menggunakan Spektroskopi Inframerah Transformasi Fourier (FTIR) dan sudut sentuhan air (WCA). Penilaian prestasi lekatan dinilai berdasarkan ujian rentas (ASTM D3359) dan ujian lucutan 180^{o}. Perubahan elektrik, mekanikal dan tenaga permukaan dianalisis bagi menyiasat kedua-dua teknik boleh menyebabkan perubahan pada sifat-sifat tersebut. Hasil eksperimen menunjukkan lekatan yang lebih baik oleh kedua-dua kaedah. Kesan haba mempunyai kekuatan lekatan 4B/0.22 N/mm hingga 5B/0.55 N/mm ketika pengawetan dilakukan pada suhu 100^{o}C hingga 140^{o}C. TPU yang mempunyai lapisan APTS menunjukkan lekatan meningkat kepada 3B/0.17 N/mm. Kekonduksian dakwat konduktif perak meningkat dan kekerasan yang lebih rendah apabila suhu meningkat. Manakala perubahan kekonduksian dan kekerasan tiada perubahan ketara oleh dakwat konduktif dicetak diatas TPU yang mempunyai lapisan APTS. Perubahan tenaga permukaan TPU disebabkan kesan haba tidak memainkan peranan penting dalam meningkatkan lekatan. Peningkatan lekatan dipengaruhi oleh perubahan sifat terma TPU. Sementara itu, TPU yang mempunyai lapisan APTS menunjukkan sifat polar kerana wujudnya kumpulan fungsian kepala kutub yang membenarkan ikatan pertalian dengan zarah perak. Kebolehpercayaan dakwat perak dilaksanakan dengan mencetak pelbagai corak geometri (lurus, persegi, sinusoidal dan zig-zag) yang dibezakan dengan lebar (1 mm, 2 mm dan 3 mm). Pengukuran elektromekanikal dilakukan dengan meregangkan corak secara manual sehingga hilang kekonduksian. Corak zig-zag dengan lebar 3 mm menunjukkan prestasi elektromekanikal yang sangat baik dengan regangan maksimum 7.78%.

ACKNOWLEDGEMENTS

In the name of Allah, the Most Gracious and the Most Merciful

Alhamdulillah, all the praises to Allah for the strengths and His blessing in helping me through all the difficulties in completing this research degree. For this golden opportunity, I would like to thank my supervisor, Prof. Dr. Ghazali bin Omar for his supervision, guidance, encouragement and kindness throughout my study. I am also very thankful for my co-supervisor, Dr. Nor Azmmi bin Masripan for his guidance and motivation throughout my study.

Special thanks to Universiti Teknikal Malaysia Melaka (UTeM) for short term funding of Zamalah 2.0 scheme. Also, special thanks to Advanced Material Characterization Lab (AMCHAL) from Faculty of Mechanical Engineering (FKM) and Composite Lab from Faculty of Manufacturing Engineering (FKP) as well as all staffs for all technical support and assistance in completing this project. I would like to extend my gratitude to all my friends who have been directly and indirectly involved in the successful completion of this thesis.

My deepest gratitude to my parents Mohd Yunos bin Mohd Shariff and Rohana binti Atan and family members for the continuous support, unconditional love, and prayers throughout my life. All the sacrifices that they made on my behalf for me was what sustained me thus far. They are ultimate role models.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

APTS	-	3-aminopropyltrimethoxysilane
Au	-	Gold
Ag	-	Silver
BFR	-	Brominated Flame Retardants
CNT	-	Carbon Nanotube
CTE	-	Coefficient of Thermal Expansion
Cu	-	Copper
DSC	-	Differential Scanning Calorimetry
EDX	-	Energy-Dispersive X-Ray
EEE	-	Electrical and Electronic Equipment
FEA	-	Finite Element Analysis
FPC	-	Flexible Printed Circuit
FTIR	-	Fourier Transform Infrared Spectroscopy
OECD	-	Organization for Economic Co-operation and Development
PC	-	Polycarbonate
PCB	-	Printed Circuit Board
PEN	-	Polyethylene Naphthalate
PDMS	-	Polydimethylsilane
PEG	-	Polyethylene glycol
PET	-	Polyethyleneterephthalate

PI	-	Polyimide
PMMA	-	Poly (methyl methacrylate)
PVP	-	Poly (4-vinyl phenol)
RFID	-	Radio Frequency Identification
SAM	-	Self-Assembled Monolayer
SEM	-	Scanning Electron Microscopy
TPU	-	Thermoplastic Polyurethane
TV	-	Television
WEEE	-	Waste of Electrical and Electronic Equipment

LIST OF SYMBOLS

I	-	Current.
R	-	Resistance per unit length
Ri	-	Calculated initial resistance,
T_g	-	Glass transition temperature
T_m	-	Melting point temperature
T_r.	-	Crystallinity temperature
V	-	Voltage
γ_{LV}	-	Liquid-gas surface tension
γ_{SV}	-	Solid-gas surface tension
γ_{SL}	-	Solid-liquid surface tension
γ^p	-	Surface free energy of polar component
γ^d	-	Surface energy of dispersive component
θ_{eq}	-	Equilibrium contact angle

LIST OF PUBLICATIONS

The research papers produced and published during the course of this research is as follows:

1. Yunos, A.M., Omar, G., Salim, M.A., Masripan, N.A. and Al-Mola, M.H.A., 2020. The Effect of Temperature on the Electrical Conductivity and Microstructure Behaviour of Silver Particles. *International Journal of Nanoelectronics and Materials*, 13, pp. 431-438.

CHAPTER 1

INTRODUCTION

1.1 Research background

Nowadays, improper electronic waste (e-waste) management has been drawing environmental and social concerns globally as it is reported to create threat towards public health and the environment (Baibergenova et al., 2003; Grant et al., 2013; Fu et al., 2018; Esenduran et al., 2019). According to the Global E-waste Monitor, the continued growth of waste amount has resulted in it being exceeded the recycled one by 44.7 million metric tonnes and is expected to gradually increase to 52.2 million metric tonnes by 2021 (Baldé et al., 2017). Besides that, estimation on the global level made by UN Environment Programme indicates that the amount of e-waste dramatically rises three times faster than other forms of municipal waste by generating 20-50 million tonnes per annum (Burke, 2007). This serious upstream of e-waste is because of the large consumption of electrical and electronic equipment (EEE) in daily life due to the urbanisation and industrialisation events. (Huang et al., 2009; Grant et al., 2013).

Generally, printed circuit boards (PCBs) are core integral components in almost all EEE with their percentage usage varying from 3% to 6% (Zhou and Qiu, 2010). According to IPC's World PCB Production Report, PCB industry achieved an estimated real growth of 13.9% in 2017 and China reportedly dominated more than half of the world's production value (52.7%) (IPC, 2018). Huang et al. (2009) reviewed that the presence of advanced technology and intense marketing by China caused a rapid update rate of EEE and shortened its average lifespan, and lead to a dramatic increase in the amount of e-waste. Besides that, several components exist in PCB wastes (brominated flame retardants (BFR), PVC plastic, and heavy metals) of which improper discarded activities can contribute to the generation of hazardous by-products, such as dioxins, furans, polybrominated organic pollutants, and polycyclic aromatic hydrocarbons, and the effects can get worse if these by-products leach into groundwater or soil (Huang et al., 2009).

Consequently, environmental policies like Directive 2002/96/EC of the European Union (EU) for all EU member states, were established to ensure end-of-life (EOL) recovery system operated for e-waste recovery, and similar legislations have been adopted in non-EU member states as well (Directive, 2002). Moreover, the end destination of all e-wastes is controlled according to The Basel Convention of 1989, which was designed to minimise the globalised transboundary movement of hazardous waste and ensure environmentally sound management through reduction of generated waste by all parties (Kummer, 2017; Zoeteman et al., 2010). All the mentioned environmental policies on the waste of electrical and electronic equipment (WEEE) had been imposed globally to restrict disposal practices to non-Organisation for Economic Co-operation and Development (OECD) countries, yet the e-waste streams ended up in cheap waste disposable sites like China, India, and West Africa, and this has caused difficulty in achieving the sustainable objective (Zoeteman et al., 2010). Since a few years, recycling and recovery movement have been part of the solution in the management of e-waste at a global level. However, reusing and recycling printed circuit boards (PCBs) offered challenges in the separation of components and materials, due to the variety of attached components to serve their functions in the appliances. Electronic components need to be separated from the solder, which involved a complex process, and the application of temperature during dissembling makes components impossible to be reused. An improvement was introduced trough recycling process that was more efficient and had a less complex operation. However, the implementation of this recycling process still depended on awareness from various parties (such as industrial and consumer), which made the implementation of this effort difficult. Therefore, a continuous push for a better solution requires a way that can prevent any continuous adverse effects on the environment, and this can be achieved by substituting the traditional PCB with a new alternative (Esfandyari et al., 2015).

Therefore, the effective alternative to replace PCB became centre attention among the researchers, and the most promising solution is the introduction of a flexible printed circuit (FPC) (Esfandyari et al., 2015; Leong et al., 2012; Tsai, 2014). Thermoplastics, as a part of flexible printed circuit components, has become an attractive material due to its advantageous features; it is non-toxic, lightweight, easy to process, has low production cost, and most importantly, recyclable (which is an important factor that resolves the environmental concern). The conductive ink that can directly cure on the substrate gives an advantage as it can eliminate the soldering processes that cause harmful effects on the environment. The general environmental aspects of printable electronics have been reviewed by Kunnari et al. (2009). They concluded that the material exhibits good efficiency during the production as no extra material is involved, which eventually leads to less produced waste. Furthermore, its development stage promises minimum involvement of hazardous substances compared to the traditional electronic production as all material used ends up on the surface of the substrate. In addition, recyclability is possible as the fabricated component is a printed electronic, which is more environmentally friendly compared to the conventional one (Kunnari et al., 2009).

Flexible printed circuit (FPC) is a component that comprises the patterned arrangement of printed circuits and components on the flexible base substrate, which is layered up by optional flexible coverlay (IPC, 1996). FPC allows new prospects to be introduced in the field in which future electronics will be tolerant under large deformation of bending. Furthermore, the research and development (R&D) efforts within this field will ultimately create potential in facilitating extreme device applications, which will pave the way for medical applications such as conformable healthcare applications and conventional consumer electronics such as sensor and displays. The flexibility properties displayed by FPC are mainly gained from polymer substrate as an underlayer, for example, polyimide (PI) (Bouhamed et al., 2017), thermoplastic polyurethane (TPU) (Cruz et al., 2016), and polydimethylsilane (PDMS) (Chun-Yi and Ying-Chih, 2016), which has recently gained interest among researchers. Their properties, which are good tensile strength, low moisture absorption, low cost, and excellent dimensional stability, make them the best candidates for the designation of microelectronic devices (Bennet and Kim, 2014; Cardoso et al., 2001; Couty et al., 2012; Inagaki et al., 1996). Besides that, the conductive ink that will be printed on FPC, should have a certain extent property that shows the capability to withstand several degrees of deformation. Stretchable conductive ink is the ideal conductor material that is used to fabricate flexible printed circuit, with metallic-type filler being one of the best electrical conductors (Harris et al., 2015; S. Park et al., 2013).

In the electronic industry, the conductive circuit is an essential part of the printing circuit board (PCB) (Kim et al., 2007). Therefore, the fabrication of conductive patterns on flexible substrate is necessary for the manufacture of flexible printed electronic devices. Advances in flexible materials and electronics have resulted in the printing technology starting to take over the conventional manufacturing processes, such as photolithographic and electroless deposition, of conductive circuits. The processes which involved complex, high-cost processing, and generation of a large quantity of chemical waste are causing drawback in industry line. On the contrary, the rise of printing technology in electronic industries simplify the processes by enabling fast and cost-saving electronic fabrication (Wade et al., 2018). The success of this technology is reflected through the development of