EFFECT OF HYBRIDIZATION ON THE FUNCTIONAL PROPERTIES OF NANOCARBON BASED ELECTRICALLY CONDUCTIVE ADHESIVE (ECA)

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In fulfilment of the requirement for the degree of

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DECLARATION

I declare that this project entitle “Effect of Hybridization On The Functional Properties of Nano Carbon-Based Electrically Conductive Adhesive” is the result of my own work except as cited in references.

Signature :……………………………..

Supervisor’s Name :………………………………

Date :………………………………

APPROVAL

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

Signature	: ....
Name of supervisor	: ....
Date	: ....

DEDICATION

First of all, I dedicate this project to God Almighty for giving me the strength to finish this project. I also like to dedicate this project to my beloved family for always supporting me throughout my life.

ABSTRACT

Due to its low processing temperature, Electrically Conductive Adhesive (ECA) is considered an alternative for interconnecting material that substitutes traditional material such as lead solder and lead-free solder. Because of its high conductivity and strength, silver is the most widely used metal fillers used in conductive adhesives. However, the cost of silver-filled conductive adhesives is much higher than the usual lead-free soldiers. Some of the limitations of silver are when blended with epoxy will produce low thermal conductivity, poor impact strength, and limited current carrying capabilities. Thus, the hybridization of the fillers in the ECA is aimed to enhance the functional properties of the single-filled ECA. This research is focused on the effect of the hybridization on the functional properties of ECA. The objective of this research is to evaluate the functional properties of hybrid ECA with varying silver flakes filler loading added to a constant amount of MWCNT conductive filler in an epoxy polymer binder. In this research, the hybrid ECA was formulated by adding 5 wt. % of MWCNT and silver flakes with filler loadings of 3wt.%, 4 wt.%, and 5 wt. % to the epoxy matrix in the Thinky mixer ARE-310 centrifugal planetary mixer machine. A JANDEL model, RM3000+ 4-point probe machine, was used to measure the resistivity of printed hybrid ECA, with reference to ASTM F390 standard guideline, in which each sample was tested 3 times to get a reliable set of data. The experimental result from the electrical characterization suggests that the sheet resistance and volume resistivity of the hybrid composites decrease with increasing filler loading, an indication that the electrical conductivity is enhanced for this range of Ag and MWCNT filler loadings in the hybrid ECA. Here, the percolation threshold was reached at approximately 10 wt.% of the fillers, in which 5 wt.% Ag + 5 wt. % MWCNT were added to the epoxy matrix. Based on the literature, with increasing fillers loading, the electrical properties of the hybrid ECA resulted in decreasing resistivity and became a better electrical conductor. Such observation could suggest that the percolation threshold is reached, creating a conductive path, therefore result in a decrease in the hybrid ECA's resistivity and increasing conductivity. As for the mechanical property, the results from lap shear test revealed that increasing the Ag filler from 3 to 4 wt.% in the hybrid ECA system, (which correspond to 8-9 wt.% total filler in the hybrid ECA), shows a gradual increase in the average maximum lap shear strength. However, beyond this filler loading, there is a gradual decrease, suggesting a saturation state for the hybrid ECA system, therefore suggesting that adding more filler does not further enhance the mechanical strength of the hybrid ECA.

ABSTRAK

Oleh kerana suhu pemprosesan yang rendah, Perekat Konduktif Elektrik (ECA) dianggap sebagai alternatif untuk saling menghubungkan bahan untuk menggantikan bahan tradisional seperti solder plumbum dan solder bebas plumbum. Disebabkan kekonduksian dan kekuatannya yang tinggi, perak adalah logam yang paling banyak digunakan dalam pelekat konduktif. Walau bagaimanapun, kos pelekat konduktif jenis perak jauh lebih tinggi daripada pelekat konduktif tanpa plumbum biasa. Antara kelemahan perak adalah apabila dicampurkan dengan epoksi akan menghasilkan kekonduksian termal yang rendah, kekuatan hentaman yang lemah, dan keupayaan mengalirkan arus yang terhad. Oleh itu, hibridisasi pengisian dalam ECA bertujuan untuk meningkatkan fungsi ECA yang diisi secara tunggal. Kajian ini akan memfokuskan pada kesan hibridisasi terhadap sifat fungsi ECA. Objektif kajian ini adalah untuk menilai sifat fungsi ECA hibrid dengan campuran serpihan perak yang berbeza dan ditambah dengan jumlah pengisi konduktif MWCNT yang tetap bersama pengikat polimer epoksi. Dalam kajian ini, ECA hibrid dirumuskan dengan menambahkan 5 wt. % kepingan MWCNT serta sebanyak 3.wt %, 4wt. % dan 5wt. % serpihan perak bersama matriks epoksi ke dalam Thinker mixer ARE-310. Model JANDEL, mesin probe 4-point RM3000, digunakan untuk mengukur ketahanan ECA hibrid, dengan merujuk kepada garis panduan standard ASTM F390, di mana setiap sampel diuji 3 kali untuk mendapatkan satu set data yang baik. Hasil eksperimen dari pencirian elektrik menunjukkan bahawa rintangan lembaran komposit hibrid berkurang dengan peningkatan pengisian, ini menunjukkan bahawa kekonduksian elektrik dapat ditingkatkan dalam julat muatan pengisi Ag dan MWCNT ini dalam ECA hibrid. Dalam kajian ini, ambang perkolasi dicapai sekitar 10% berat pengisi, di mana 5%Ag + 5 wt. % MWCNT ditambahkan ke matriks epoksi. Berdasarkan literatur, dengan peningkatan pemuatan pengisi, sifat elektrik ECA hibrid mengakibatkan penurunan rintangan dan menjadi konduktor elektrik yang lebih baik. Pemerhatian ini dapat menunjukkan bahawa ambang perkolasi tercapai, mewujudkan jalan konduktif, oleh itu mengakibatkan penurunan rintanganECA hibrida dan peningkatan kekonduksian. Bagi sifat mekanik, hasil ujian ricih menunjukkan bahawa peningkatan pengisi Ag dari 3 hingga 4% berat dalam sistem ECA hibrid, (yang sesuai dengan jumlah pengisi 8-9% berat dalam ECA hibrida), menunjukkan peningkatan dalam kekuatan ricih pusingan maksimum. Walau bagaimanapun, di luar pengisian ini, terdapat penurunan secara beransur-ansur, menunjukkan keadaan tepu untuk sistem ECA hibrid, oleh itu menunjukkan bahawa menambahkan lebih banyak pengisi tidak meningkatkan lagi kekuatan mekanikal ECA hibrid.

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LIST OF ABBREVIATIONS

ECA	Electrically Conductive Adhesive
ICA	Isotropic Conductive Adhesive
ACA	Anisotropic Conductive Adhesive
CNT	Carbon Nano-Tube
SWCNT	Single-Walled Carbon Nano-Tube
MWCNT	Multi-Walled Carbon Nano-Tube
Ag	Silver
ASTM	American Society for Testing Material

CHAPTER 1

INTRODUCTION

1.1 Introduction

This chapter provides the background of the research study, the problem statement of the research, objective and scope of research study, and also the planning and executions of the research study.

1.2 Background

An electrically conductive adhesive is an adhesive that is mainly used for electronics. Electrically Conductive Adhesives (ECAs) have been used for high-reliability applications such as automotive, medical, and telecommunication products. Due to its low processing temperature, Electrically Conductive Adhesive (ECA) is considered an alternative for interconnecting material that substitutes traditional material such as lead solder and lead-free solder. ECA consists of a matrix of polymers that acts as a binder for the conductive fillers. For standard ECA, due to improved electrical conductivity, metallic materials such as silver are used as conductive fillers.

Two major classifications of electrically conductive adhesive (ECA) are isotropic conductive adhesive (ICA) and anisotropic conductive adhesive (ACA). (Yi Li Daniel Lu C.P. Wong, 2010.). Isotropic conductive adhesives are composites of polymer resin and conductive fillers, also known as "polymer solder." Through touching the conductive particles, the conductive fillers provide the material with electrical conductivity. For increasing concentrations of the filler, the electrical properties of the ICA turn it from an insulator to a conductor. Anisotropic conductive adhesives (ACAs) are a group of materials that usually combine epoxy or acrylic adhesives with conductive particles to allow electrical connection over what would otherwise be a regular mechanical adhesive assembly. These vary from isotropic conductive adhesives such as silver epoxy in that the conductive particles are charged and distributed in such a way that they do not work in the bulk of the adhesive, but when they are stuck between the electrodes on the top and bottom substrates. Similar to isotropic adhesives or other solder techniques, it helps them to deliver several unique advantages. Such advantages are primarily related to its low temperature, and high interconnect density capabilities in the case of touch panels, although assembly cost and speed may also be considerations (Jain 2016).

Carbon nanotubes (CNTs) are cylindrical clusters of single-layer carbon atoms (graphene) rolled-up plates. These can be single-walled (SWCNT) with a diameter of less than 1 nanometer (nm) or multi-walled (MWCNT), consisting of several dense nanotubes reaching more than 100 nm in diameter. Their length can exceed many or even millimeters of micrometers. CNTs are chemically bonded with sp^2 bonds, like their building block graphene, an extremely strong type of molecular interaction. Combined with the natural inclination of carbon nanotubes to rope together via van der Waals forces, this feature provides the opportunity to produce high-strength, low-weight materials with highly conductive thermal and electrical properties. The rolling-up path of the graphene layers determines the nanotubes' electrical properties.

On the other hand, multi-walled carbon nanotube (MWCNT)-filled adhesives are resistant to oxidation and metal migration, high in strength and lightweight compared to metal-filled conductive adhesives. Because of the resistivity of the underlying material, the carbon-based filler can never equal the electrical quality of metallic materials like gold, silver, or even copper. The resistivity of MWCNTs is 1x 10^{-4}.cm, while 6 x 10^{-6}.cm is the resistivity of silver (Nasaruddin et al., 2019). A good gain in mechanical properties, however, is the added value of using carbon nanotube as a compared filler.

1.3 Problem Statement

While the tin/lead soldering technique has been widely used to create electrical connections and packaging for electronic components, for various reasons, it is replaced by lead-free alternatives. Toxicity is the first and foremost concern with lead soldiers. The electronic industry is replacing lead soldering at a rapid pace due to toxicity and environmental impact issues. The other problem when using the lead soldering is the high-temperature problem. The assembly is subject to very high temperatures during the soldering process. Because of this high-temperature exposure, some heat-sensitive components in the near vicinity may be harmed. Tin/lead solder is also able to dissolve gold and form some inter brittle metallic compounds. In these situations, the joint's mechanical power is significantly decreased. The use of electrically conductive adhesives (ECAs) is an alternative to lead-free solder. These are a polymer binder and conductive filler used for chip-to-die connections in the semiconductor industry and are considered environmentally friendly (Lewis and Coughlan 2008). Advantage of using an electrically conductive adhesive (ECAs) are serviceability at high and low temperature, low stress, and peel strength, and also resistance in thermal cycling. The composition of an electrically conductive adhesive is made up of a binding material and a conductive filler, the combination of which defines the adhesive's strength and electrical properties. (Lewis and Coughlan 2008).

Unlike other types of adhesives, there are two critical roles for electrically conductive adhesives. First, conductive adhesives form joints with sufficient strength to link two surfaces, and second, the two bonded surfaces form an electrical interconnection. The typically used conductive fillers include carbon black, graphite dust, and metal particles like gold, nickel, copper, and aluminum that are micron or Nano-sized. Epoxy, silicone, polyamide, and polyurethane are typical polymer matrices. High and stable electrical conductivity is the primary properties of these adhesives. (Sancaktar and Bai 2011).

The use of electrically conductive adhesive to replace the lead soldering offers many advantages. Electrically conductive adhesives (ECA) have very low volume resistivity (< 0.001 ohm-cm) in combination with high-temperature resistance. These compounds are relatively low epoxy outgassing, and many electrically conductive epoxies also meet the low outgassing standards of NASA. Tough compositions can improve the serviceability for thermal cycling without becoming brittle. There is an excellent mechanical strength and durability.

1.4 Objective

The objective of this research is to formulate and characterize the functional properties of Ag-MWCNT hybrid electrically conductive adhesive with varying Ag flakes filler loading.

Scope of the project

The scope of this project is as follows:

1. Formulation of the electrically conductive adhesive (ECA) hybrid nanocomposites using Ag filler loading of 3,4 and 5 wt.% with a constant MWCNT filler loading of 5 wt.% in the epoxy binder.
2. Fabrication of the hybrid electrically conductive adhesive (ECA) using a planetary centrifugal mixer.
3. Electrical properties of electrically conductive adhesive (ECA) using a four-point probe test.
4. Mechanical characterization of electrically conductive adhesive (ECA) using a lap shear test.

1.5 Planning and Execution

The research planning and activities for PSM I and PSM II are shown below. The research planning will include title selection, literature review, and submission of progress report, experimental design, formulation and fabrication of ECA, test for ECA, and the analysis of the experiment. For both PSM I and II, the completion of all research will be followed by the data analysis, report submission and the presentation

Table 1.1 PSM I Gantt chart

WEEK / ACTIVITIES	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Research Title Selection														
Background Study														
Literature Review														
Lab Visit														
Submission Progress Report														
Formulation and fabrication of ECA														
ECA Electrical conductivity test														
Data Analysis														
Report Writing														
Report Submission														
PSM 1 Seminar														

Table 2.2 PSM II Gantt chart

WEEK / ACTIVITIES	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Literature Review														
Methodology														
Electrical Characterization														
Mechanical characterization														
Data Analysis														
Progress Report Submission														
PSM II Report Writing														
Draft Report Submission														
PSM II Report Submission														
PSM II Seminar														

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter presents a review of the electrically conductive adhesive (ECA), which will include the type of polymer matrix, type of filler, type of carbon nanotube, and functional properties of ECA, as reported in the literature.

2.2 Electrically Conductive Adhesive (ECA)

The use of electrically conductive adhesives (ECAs) is an alternative to lea d-free solder. These consist of a polymer binder, and conductive liner has been used for chip-to-die connections in the semiconductor industry and is considered environmentally friendly. Although conductive adhesives have become commercially available and have a lower processing temperature advantage, they have not been used extensively as a general solder replacement. Such observation is due to weaknesses such as high joint strength and low mechanical strength, especially when exposed to different environmental conditions. (Lewis & Coughlan, 2008). Some advantages offered by ECA will include the reduced cost through reduction in several processing steps, low curing temperature, and also ECA has better fatigue resistance compared to solder joints.

The composition of an electrically conductive adhesive is made up of a binding material and a conductive filler, the combination of which defines the adhesive's strength and electrical properties. Unlike other adhesive types, two primary functions are performed by electrically conductive adhesives. First, conductive adhesives form joints with sufficient strength to link two surfaces, and second, the two bonded surfaces form an electrical interconnection. Normally this dual functionality is accomplished in a composite form by particle dispersion in an isolating adhesive matrix. The widely used conductive fillers include carbon black, graphite dust, and metal particles like silver, nickel, copper, or aluminum that are micron or Nano-sized. Epoxy, silicone, polyamide, or polyurethane are common polymer matrices. The primary properties of these adhesives are high and stable electrical conductivity. The metal fillers applied to the epoxy resin typically boost their other properties such as strength, thermal conductivity, etc. and can, therefore, also be used to express different composite properties such as thermal expansion, thermal conductivity, and shrinkage and heat resistance. (Sancaktar & Bai, 2011).

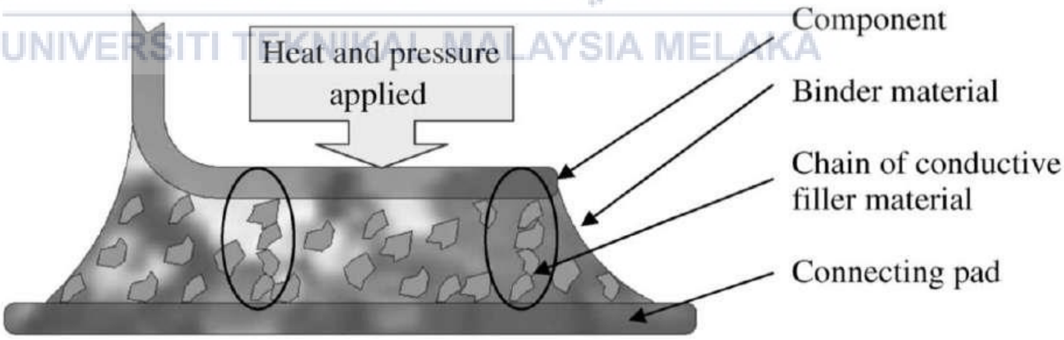

Figure 2.1: Schematic example of an electrically conductive adhesive bonded to an electrical component and a connecting pad. (Lewis & Coughlan, 2008).

2.3 Polymer Matrix Composite

Carbon fibre polymer-composites have been known excellent for their mechanical properties, electrical conductivity, thermal conductivity, and low thermal expansion. In general, composites of polymer matrix are much easier to manufacture than composites of the metal matrix, carbon matrix or ceramic matrix, whether the material is a thermoset or a thermoplastic polymer. The advantage of polymer matrix composites is they are lower in manufacturing cost and also give a better performance of properties.

Carbon fibers are brittle (low ductile), although they are high in tensile strength and unit. Consequently, a carbon fiber polymer matrix composite, particularly one with continuous carbon fibres, tends not to be tough enough. The polymer matrix is stronger and more ductile than carbon fiber. (Deborah Chung 2017). Various polymers, however, vary in their hardness. The durability of a carbon fiber reinforced polymer matrix, therefore, depends heavily on the choice of the polymer matrix. Table 2.1 shows some material properties for some commonly used material and fibre composites.

During its operation, the downside of polymer matrix composite may be harmful to humans by emitting more hazardous products throughout material processing, dangerous toxicity, and worse manufacturing of products. Content does have an improvement in applicability and economics, but it is harmful to humans, restricting use and growth (Ru-Min Wang, 2011).

Table 2.1: Specific strength and specific modulus of some commonly used materials and fibre composites (Ru-Min Wang, 2011).

Materials	Density (G/cm^3)	Tensile Strength (GPa)	Elastic Modulus (10^2GPa)	Specific Strength (10^6Cm)	Specific Modulus (10^8Cm)
Steel	7.8	1.03	2.1	1.3	2.7
Aluminium alloy	28	0.47	0.75	1.7	2.6
Titanium alloy	4.5	0.96	1.14	2.1	2.5
Glass fibre composite materials	2.0	1.06	0.4	5.3	2.0
Carbon fibre ii/epoxy composite materials	1.45	1.50	1.4	10.3	9.7
Carbon fibre i/epoxy composite materials	1.6	1.07	2.4	6.7	15.0
Organic fibre/epoxy composites	1.4	1.40	0.8	1.0	5.7
Boron fibre/epoxy composites	2.1	1.38	2.1	6.6	10.0
Boron fibre/aluminium matrix composites	2.65	1.0	2.0	3.8	7.5

2.4 Thermoplastic

Thermoplastics are defined as polymers that can be almost indefinitely melted and recast. When heated, they are molten and harden when cooled. A thermoplastic, however, becomes glass-like and subject to fracture when frozen. These features, which give the material its name, are reversible, so that the material can be repeatedly heated, reshaped, and frozen. As a result, thermoplastics can be reused mechanically. Polypropylene, polyethylene, polyvinylchloride, polystyrene, polyethylene, and polycarbonate are some of the most common types of thermoplastic. Thermoplastics have a simple structure of molecules consisting of chemically independent macromolecules. They are softened or melted after heating, then shaped, formed, welded, and, when cooled, solidified. It is possible to repeat multiple heating and cooling cycles, allowing reprocessing and recycling. Figure 2.2 shows show the example of the thermoplastic resin structure.

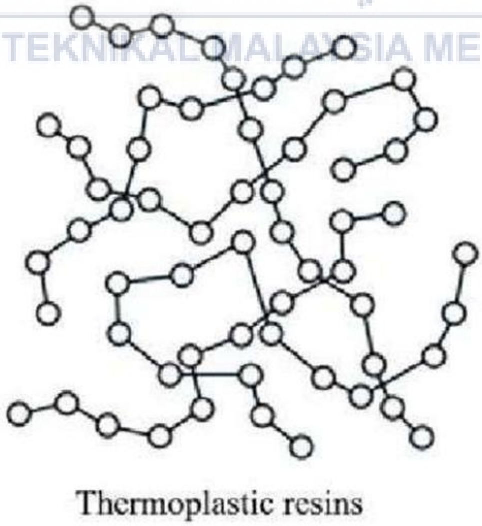

Figure2.2: Thermoplastic chemical structure (Liu, Zwingmann and Schlaich, 2015)

2.5 Thermoset Plastic

Thermoset materials are those materials made of polymers joined by chemical bonds that develop a strongly interlinked polymer structure. The highly interlinked structure formed by chemical bonds in thermoset materials is directly responsible for the high mechanical and physical strength compared to thermoplastics or elastomer materials.

On the other hand, this highly interconnected structure provides poor elasticity or elongation of these materials. The gel point, which refers to the time when the material changes from an irreversible way-viscous liquid state to a solid-state during the curing process, is one of the characteristic parameters of thermosets materials. Once the gel point is moved, the fluid will stop flowing, and it will not be possible to mold or process the grid. The properties of thermoset material are they cannot be melt, insoluble, and high resistance to creep. One of the negative aspects of thermosets is their ability to recycle because it is impossible to return to liquid phase material once they are crosslinked or cured. Thermoset materials have the property of not melting or deforming before moving into a liquid state in the presence of temperature or heat. Figure 2.3 shows an example of a thermoset resin structure.

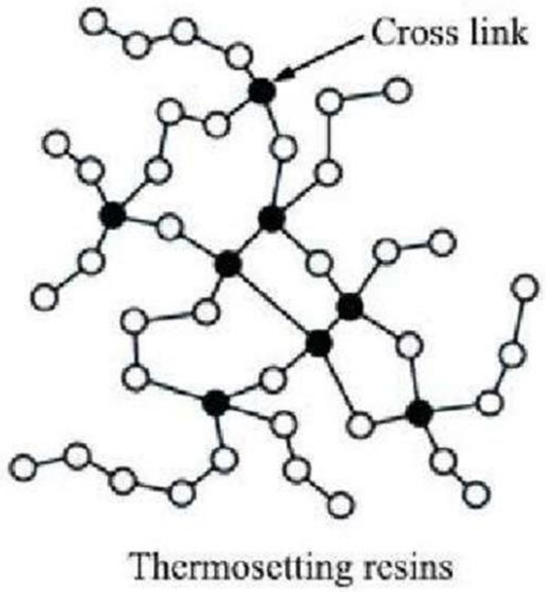

Figure2.3: Thermoset chemical structure (Liu, Zwingmann, and Schlaich, 2015).

2.6 Epoxy

Epoxy is by far the most widely used polymer matrix for carbon fibers. Epoxy has excellent mechanical properties and resistance to oxidation, is dimensionally stable, has good adhesion, and is relatively cheap. Besides, the low molecular weight in the liquid state of uncured epoxy resins results in exceptionally high molecular mobility during processing. Epoxy resins have two or more groups of epoxy per molecule.

An epoxy group's chemical structure is shown in Fig 2.4. Epoxy is a cyclic ether with a ring of three atoms. The bonds are strained as this ring is triangular, allowing the epoxy to be highly reactive. Due to the high adhesiveness of epoxy, epoxy resins are most widely used to shape the matrix of composites.

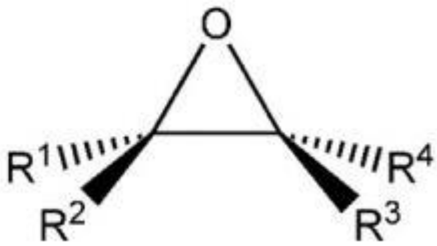

Figure 2.4: The structure of a generic epoxide, R1, R2, R3, and R4 are four atoms or functional groups that are covalently bonded to the two carbons atoms at two corners of the triangle. (Deborah Chung 2017).

2.7 Filler For ECA Composites

Conductive fillers should be applied to the ECAs ' polymer matrix to make an insulating polymer electrically conductive; when the critical filler concentration (percolation threshold) is reached, the insulating polymer will become a conductive one. Various materials such as metallic fillers (e.g., copper, nickel, gold, or silver particles) or carbon-based materials (e.g., carbon black, carbon nanotubes, and graphene) can be used as conductive fillers. Among all these fillers, silver was the first and foremost conductive filler used in industrial ECAs due to its strongest electrical and thermal conductivity at room temperature, ease of processing, and the conductive quality of its oxide. (Amoli 2015)

2.7.1 Metal

Metal has high thermal and electrical conductivity, ductility, and high light reflectivity. Metals such as copper, gold, and silver are found in ores (mineral-bearing substances) that do not react with other elements. Aluminum atoms have in the outermost shell less than half the full electron complement, and they prefer not to form compounds with each other. Silver, gold, and platinum are less reactive metals, but lithium, potassium, and radium are highly reactive metals. Metal's mechanical properties are resistance, hardness, the strength of fatigue, ductility, and malevolence (BELL, 2019).

2.7.2 Silver (Ag)

Because of its high conductivity and strength, silver is the most widely used metal fillers used in conductive adhesives. However, the cost of silver-filled conductive adhesives is much higher than the usual lead-free soldiers. Some of the limitations of silver are when blended with epoxy will produce low thermal conductivity, poor impact strength, and limited current carrying capabilities. (Qiao et al., 2014). Moreover, although silver has high conductivity, it comes with a high cost. (Wu et al., 2007).

2.7.3 Silver Flakes (Ag)

Silver flakes have the highest electrical conductivity compared to other metals; silver flakes were produced using mechanical milling from the silver powder. Silver (Ag) flakes for electrically conductive adhesives (ECAs) are widely used as fillers. To ensure proper rheology of the ECAs, the Ag flakes must be pretreated with organic lubricants. Lubricants on the Ag flakes have an impact on ECAs ' rheology, conductivity, and other properties. The thin layer is produced on the surface of silver flakes as the organic lubricant is added during the milling process. The thin layer is nonconductive. Once the layer is removed, the ECA will become conducive to electricity. (Lu, Tong & Wong, 1999).

2.7.4 Carbon Black (CB)

Carbon black has been used for a decade as conductive filler added into a polymer-based composite (Hou, Zhou and Wang, 2018). Carbon black is integrated into polymers that need resistivity between 1 and 10^6 Ω cm for permanent electrostatic discharge protection, explosion prevention, and polymer applications. Conductive carbon black fillers give polymers with lower critical volume fractions electrical conductivity than conventional carbon blacks and therefore influence the mechanical properties of the resulting polymer compound to a lower degree. The fundamental feature of this family of exceptional carbon black grades is a high carbon black structure, for example, a high void volume. High-structured carbon black materials are preferred fillers to make polymers conductive as they allow high concentrations of polymer to be retained while the conductive network is formed. Carbon black is used in composite plastic for electromagnetic interference shielding, conductive adhesive, electrostatic discharge shielding for sensing applications as well as conductive photoresists as electrically conductive micro-components (Hauptman et al., 2012).

2.7.5 Non-Metal Fillers

The non-metal filler is generally incapable of conducting electricity and heat. Non-metal are brittle and impossible to be roll or pounded. They are usually are in solid and gas state. (Bentor, 2018). Non-metal atoms were generally small, containing relatively large numbers of electrons in the outermost shell, almost filled shells of electrons, thus requiring some additional electron to balance out. It has a pronounced tendency to draw electron to itself due to low electronegativity from other electrons (Physician et al., 2017).

2.7.6 Carbon Nano-Tubes (CNT)

Carbon nanotubes (CNTs) are cylindrical clusters of single-layer carbon atoms (graphene) rolled-up plates. Carbon Nano-Tube (CNT) were categorized into Single-Walled Carbon Nano-Tubes (SWCNT) and Multi-Walled Carbon Nano-Tubes (MWCNT) (Hirsch, 2002). Their length can reach several or even millimeters of micrometers. CNT offers the opportunity to develop high-strength, low-weight materials with high-level electrical and thermal properties. For many applications, it makes them highly attractive. Carbon Nano-tube has been introduced in the manufacturing process using screen printing technology because it is a low cost, small size, and easy to prepare (Pan, Zhu and Gao, 2008). Carbon Nano-Tubes has a high aspect ratio, can be ideal materials for connecting metal flakes in the electrically conductive adhesive (ECA) to create conductive networks and minimize metal content, thus reducing the cost of electrically conductive adhesive.

2.7.7 Single-Walled Carbon Nano-Tubes (SWCNT)

Single Walled Carbon Nanotubes are defined as one-dimensional, cylindrically shaped carbon allotropes with a high area and aspect ratio (length to diameter ratio). SWCNTs are considered a one-dimensional (1D) material due to their small diameter and high aspect ratio. SWCNTs are so named because of their hollow structure and number of walls. SWCNTs characteristic is low resistance, lightweight, low cost, easy to process, and high transmittance. The characteristic of Single-Walled Carbon Nano-Tubes (SWCNT) depends on the bundle, orientation of tube, and size (Yakovlev et al., 2018).

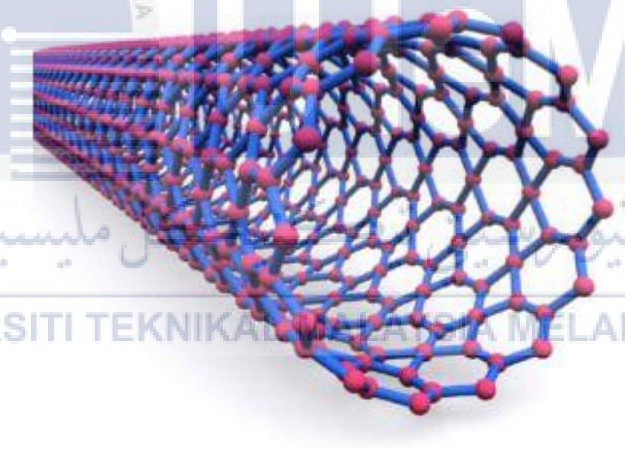

Figure 2.5: Single-Walled Carbon Nano-Tubes

2.7.8 Multi-Walled Carbon Nano-Tubes (MWCNT)

Multi-walled carbon nanotubes are hollow, cylindrically shaped carbon allotropes with a high aspect ratio (length to diameter ratio). Their name comes from their structure, and multiple one-atom-thick carbon sheets form the walls. MWNTs consist of multiple layers of dense graphene nanotubes within other nanotubes. MWCNT is suitable for uses in the electrically conductive polymer as they have very high conductivity as well as a large aspect ratio.

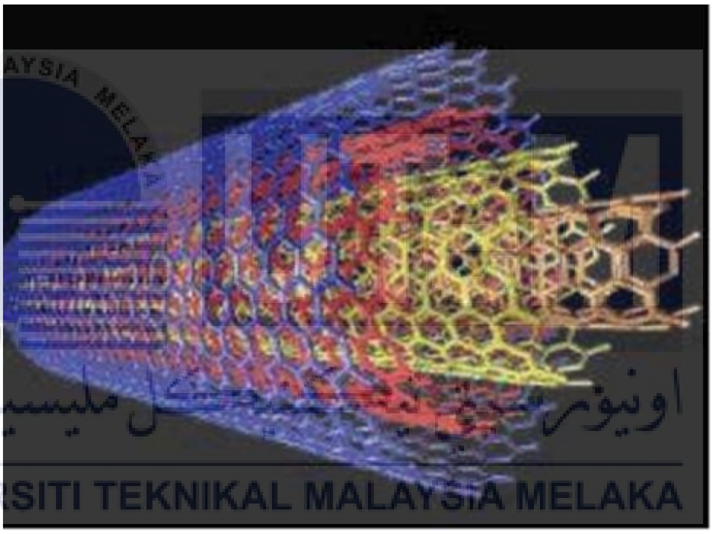

Figure 2.6: Multi-Walled Carbon Nano-Tubes.

2.8 Functional Properties of ECA Composites

Properties of the ECA can be classified into two categories, which are for electrical properties and mechanical properties.

2.8.1 Electrical properties of Electrically Conductive Adhesive (ECA)

Unlike other types of adhesives, there are two critical roles for electrically conductive adhesives. First, conductive adhesives form joints with sufficient strength to link two surfaces, and second, the two bonded surfaces form an electrical interconnection. A four-point probe was used to measure the electrical sheet resistance of electrically conductive adhesive (ECA) (Trinidad, 2016). Four-point probe four-terminal sensings or known as kelvin sensing (Chandra et al., 2017) is named after William Thomson, Lord Kelvin, a person who invented it that could measure wire resistance, show very low resistivity and eliminate inaccuracy (Amoli, 2015).

2.8.2 Mechanical Properties of Electrically Conductive Adhesive (ECA).

The mechanical properties of conducting composites are crucial for much electrical application. The conductive adhesive will form the joint with sufficient strength to link the surface to protect the component. For adhesive strength, the lap shear testing is used to determine the adhesive strength of metal to a metal joint (Trinidad, 2016). The American Society for Testing Materials (ASTM) testing standard is one of the main reference points for research and evaluating adhesive lap shear strength.

2.8.3 Hybrid Electrically Conductive Adhesive

Recently, there is increasing in developing hybrid filler for electrically conductive adhesive (ECA). The performance of a hybrid ECA is based on their type, size, and shape of fillers. Appropriate conductive fillers need to be mix with matrix resin to attain better performance of the thermal conductivity of ECA (Qiao et al., 2014). The new type of hybrid nowadays is a mixture of carbon Nano-tubes and silver flakes. Hybrid ECA combined with silver flakes and CNT has been reported to result in very high electrical and thermal conductivity. With CNT's high aspect ratio and electrical conductivity, the amount of silver needed to be compared to the single conductive filler system, such as the Ag filled adhesives, can be reduced (Marcq et al., 2011).

CHAPTER 3

METHODOLOGY

3.1 General Methodology

This chapter presents the general methodology for this research project, which includes the preparation and fabrication of the ECA composites, type of epoxy resin, Ag, MWCNT, and also the type of hardener uses. This section also will state the type of machine and apparatus need to be used in this research. Below is the general methodology involved in this research project, as shown in Figure 3.1.

i. Design of experiment.

ii. Preparation of materials, including the formulation of ECA, sample fabrication, variables, and parameters of the experiment which includes:

   a. Materials formulation of the ECA

   b. Fabrication process including, curing process using the oven

   c. Lap shear test using a Universal Test Machine

   d. Electrical conductivity measurement using a 4-point probe

   e. Scanning Electron Microscope (SEM) for surface morphology

iii. Data collection and analysis

iv. Report writing

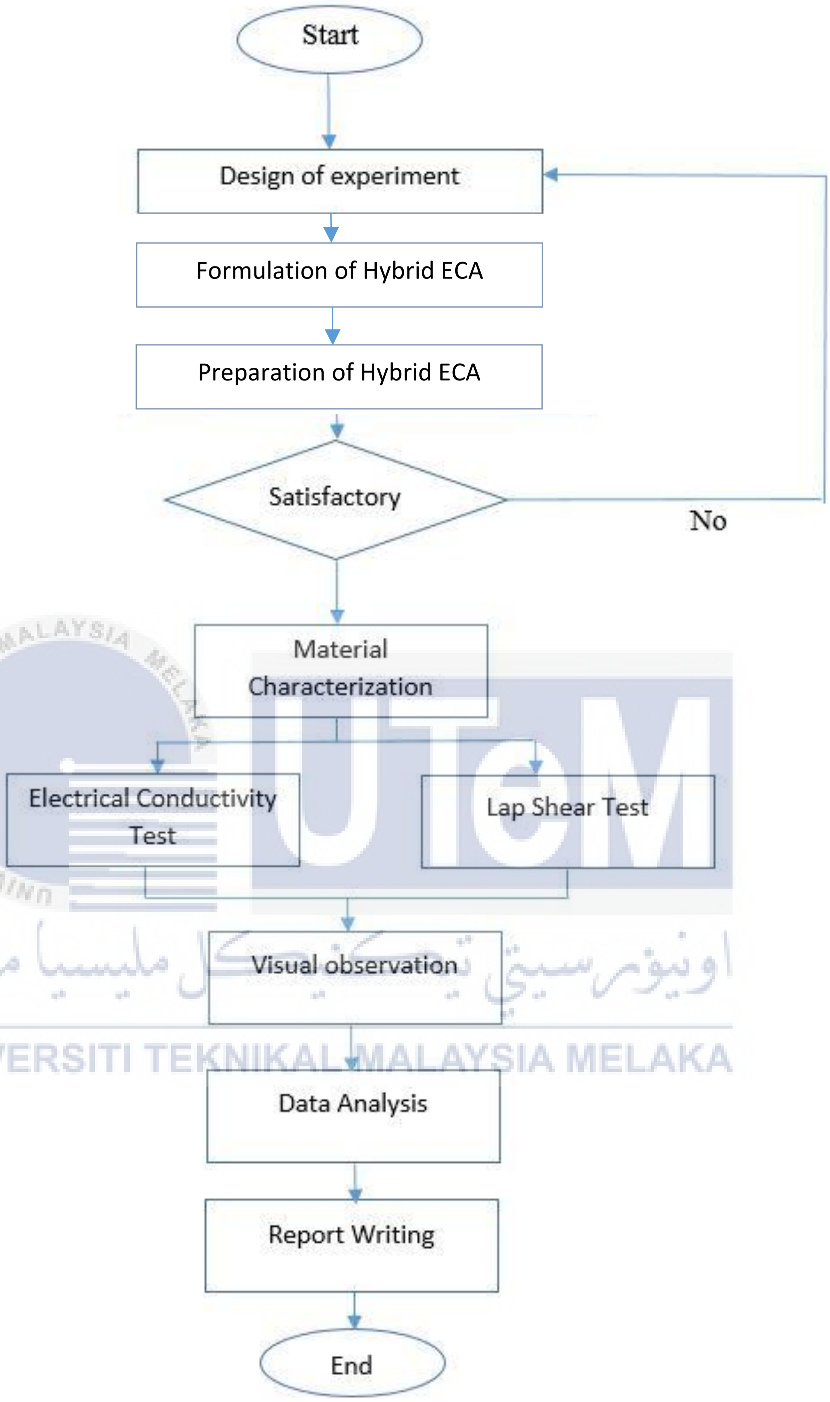

Figure 3.1: Flow Chart of Research

3.2 Raw Material

3.2.1 Polymer

The polymer used in this research is Araldite 506 epoxy resin polymer, which is supplied by Sigma-Aldrich. The selection of polymer is based on the properties. Epoxy has high tensile and modulus strength, good corrosion resistance, good chemical resistance, high adhesive, and has stability in dimension. More than that, epoxy will experience low shrinkage in cure. (Kwon, Yim, Kim, & Kim, 2011). However, the precaution must be taken when handling epoxy since the material is hazardous. The precaution that needs to be taken during handling epoxy is to wear precaution clothing and protection. Table 3.2.1a shows the GHS classification, and Table 3.2.1b shows the chemical and physical properties of Araldite 506 epoxy resin.

Table 3.1: GHS classification in accordance with 29 CFR 1910 (OSHA HSC) (Mixture et al., 2017).

Type of hazard	Category	Warning
Skin irritation	2	H315- Causes skin irritation.
Eye irritation	2A	H319- Causes severe eye irritation
Skin sensitisation	1	H317- May cause an allergic skin reaction.
Acute aquatic toxicity	2	H401- Toxic to aquatic life.
Chronic aquatic toxicity	2	H411- Toxic to aquatic life with long-lasting effects.

Table 3.2: Chemical and physical properties of Araldite 506 epoxy resin (Mixture et al., 2017).

Appearance	From: semi-solid melting to liquid Colour: colourless
Melting and freezing point	-15°C & -5 °C
Flash point	252 °C
Vapour pressure	0.04 hPa (0.03mmHg) AT 77 °C
Relative density	1.168 g/cm 3
Partition coefficient –octano/water	Long Pow: 2.8

3.2.2 Hardener

The hardener used in this research is Polyether amine D230 called JEFFAMINE D-230., which is supplied by Hunstman Singapore Pte LTD. The hardener is used as the curing agent in the process of making hybrid composites. The hardener used is colorless, has the average molecular weight around 230, and comes with low viscosity, low vapour pressure, and low Chroma. Table 3.2.2 shows the Polyether amine D230 specification, and Table 3.2.2b shows the properties of Polyether amine D230.

Table 3.3: Specification of polyether amine D230 (Materials, 2018).

Item	Standard data	Typical testing result
Appearance	Colourless to light yellow transparent liquid	Colourless to light yellow transparent liquid
Colour, alpha	60 max	10
Water (%)	0.50 max	0.26
Total amine (mmol/g)	7.40 min	7.83
Primary amine content (%)	90 min	94.90

Table 3.4: Properties of polyether amine D230 (Garibay-Vasquez, 2005).

Properties	**Polyether Amine D230**
Colour, pt-co	30
Brookfield viscosity, cps, 25°c	9
Specific gravity, 20/20°C	0.948
Density, lb/gal, 20°C	79
Refractive index,n^{20}	1.4466
Flashpoint, PMCC, °C	121
Water, wt. %	0.1
Total acetyl tables, meq/g	8.7
Total amine, meq/g	8.4
Primary amine, meq/g	8.2

Properties	Polyether Amine D230
Vapor Pressure, mm Hg/ºC	1/101
Equivalent weight with epoxies ("amine hydrogen equivalent weight," or AHEW)	60
pKa	9.46
Appearance	Colourless to slight yellow with a slight haze
Color, pt-co	60 max.
Primary amine, %	97 min.
Total amine, meq/g	8.1 min. 8.7 max.
Water, %	0.25 max

3.2.3 Multi-Walled Carbon Nano-Tubes

The Multi-Walled Carbon Nano-Tubes is supplied by nanostructure & amorphous material Inc. (NanoArmor), USA. The MWCNT has a high conductive when becoming a composites structure. MWCNT has a large aspect ratio with a length of over 100 times the diameter and comes with high chemical stability. However, MWCNT is hazardous and needs to be handle with protection. Table 3.2.3a shows the MWCNT chemical and physical properties, while Table 3.2.3b shows MWCNT specification.

Table 3.5: Chemical and physical properties of Multi-Walled Carbon Nano-Tube (Paint, 2011).

Form	Powders
Colour	Black
Odour	Odourless
Melting point	3652-3697 °C
Density	At 20 °C~ 2.1g/cm^3
Water Solubility	Insoluble

Table 3.6: MWCNT specification (Products and Nanotubes, 2018).

Purity	>95 wt.% (carbon nanotubes)
Outside diameter	10-20 nm
Inside diameter	5-10 nm
Length	10-30 µm
SSA	> 200m^2/g
Ash	< 1.5wt%
Electrical conductivity	> 100 s/cm

3.2.4 Silver Flakes, Ag

The silver used in this research is in flakes form, supplied from Sigma-Aldrich. The average silver flakes size is 10 μm, with a resistance of 1.59 μΩ-cm. Table 3.2.4a shows the classification of the hazardous chemical according to CLASS regulation 2013, and Table 3.2.4b shows the chemical and physical properties of silver flakes.

Table 3.7: Classification of the hazardous chemical according to CLASS regulations 2013 (States 2020)

Type of hazard	**Category**	**Warning**
Acute hazard	1	Hazardous to the aquatic environment
Chronic hazard	1	Hazardous to the aquatic environment
H410	-	Very toxic to aquatic life with long-lasting effects.
P273	Prevention	Avoid release to the environment.
P391	Response	Collect spillage.
P501	-	Dispose of contents/ container to an approved waste disposal plant.

Table 3.8: Physical and chemical properties of silver flakes (States 2020)

Appearance	Form: flakes
Melting point/freezing point	Melting point/range: 960 °C - lit.
Initial boiling point and boiling range	2.212 °C - lit.
Relative density	10,49 g/cm3
True density	~ 2.1 g/cm^3

Figure 3.2: Epoxy, Hardener, MWCNT, Silver Flakes

3.3 Electrically Conductive Adhesive (ECA) Preparation

The hybrid electrically conductive adhesive (ECA) was prepared using the combination of the following materials:

Matrix		Filler Loading 1		Filler Loading 2
Epoxy resin	+	Multi-Walled Carbon Nano-Tubes	+	Silver Flakes

The amount of material used to prepared this hybrid composites is based on the weight percentage (wt %) and by applying the Rule of Mixture (ROM). The Equation used in RoM is expressed by Equation (3.1) and Equation (3.2), respectively.

$$Weight\ fraction = \frac{weight\ filler}{weight\ composites} \tag{3.1}$$

$$\text{Weight composites} - \text{Weight filler} = \text{Weight matrix} \tag{3.2}$$

Whereby Weight filler = mass of MWCNT and mass of silver flakes used

Weight matrix = mass of polymer used

The weight fraction used is for silver flakes and MWCNT. The weight fraction for the silver flakes used in this research is 3 wt.% 4 wt.%, and 5 wt. %. The weight fraction for MWCNT is 5wt. %. The total mass for the composites is 5 grams. For hardener, the weight used is 30 % of the weight of the matrix used. The expression used to determine the amount of hardener used in the hybrid ECA formulation is shown in Equation (3.3). The formulation for hybrid ECA is shown in Table 3.3.1.

$$\text{Weight matrix} * 30\ \% = \text{Weight of hardener in gram} \tag{3.3}$$

Table 3.9: Formulation for hybrid ECA

Silver Flakes (wt. %)	**MWCNT (wt. %)**	**Epoxy (wt. %)**	**Hardener (gram)**
3 (0.15 gram)		92 (4.60 gram)	1.380 gram
4 (0.20 gram)	5 (0.25 gram)	91 (4.55 gram)	1.365 gram
5 (0.25 gram)		90 (4.50 gram)	1.350 gram

The weight fraction and amount of material used for the hybrid ECA is shown in Table 3.3.1 above. The plastic container is used to hold the material. The material was weighed using the Mettle Toledo balance machine. The machine reading is calibrated by resetting the weight to zero every time a new material was added. After all the material preparation was readied, they were poured onto a container and place inside the Thinky Mixer adapter and finally placed inside the Thinky Mixer Model ARE 310 centrifugal mixer machine. The machine was set to run at 2000 rpm for 5 minutes in formulating the hybrid ECA studied. The flow process for the hybrid composites is shown using the flow chart in Figure 3.3.

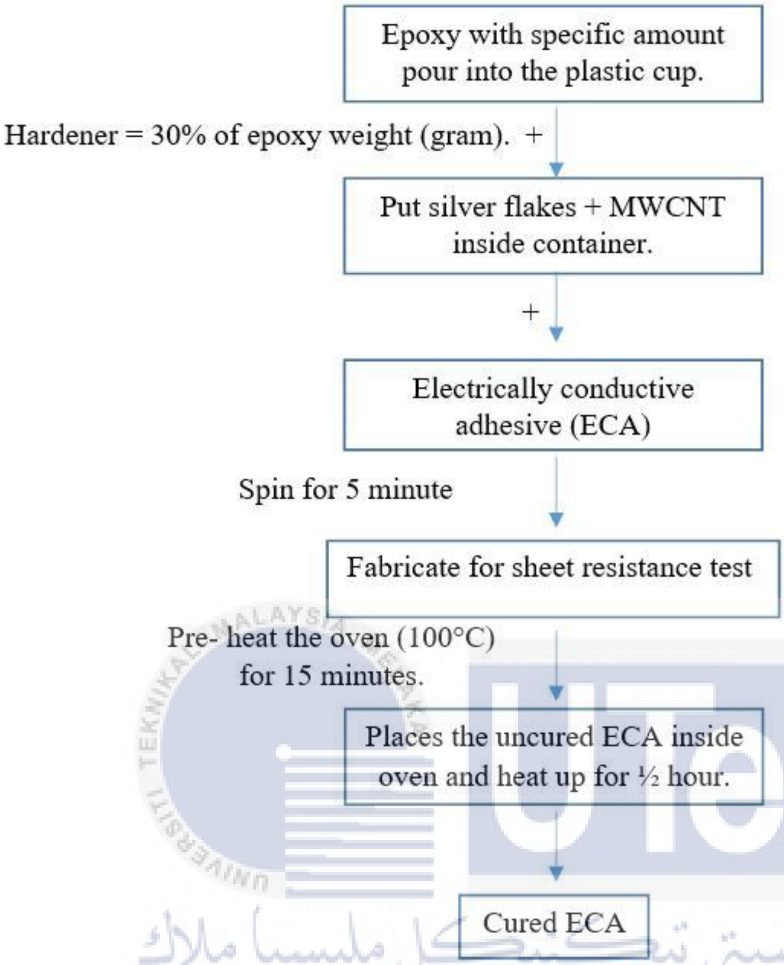

Figure 3.3: The hybrid ECA preparation flow process.

As a precaution, each specimen is fabricated in a different container from each other. The weight and amount have been determined using the RoM equation, as shown above. The Mettler Toledo is reset to "zero" each time the material is added. This procedure is repeated for each of the formulations, which are 3 wt. %, 4 wt. % and 5 wt. % of silver flakes. The Mettler Toledo balance has a high accuracy that makes this machine is very suitable to be used as the weighing mechanism. The hybrid ECA is then placed in the Thinky Mixer Model ARE 310 centrifugal planetary mixer, and the machine was set to run at 2000 rpm rotational speed for 5 minutes. The hybrid ECA is then applied onto the substrate and be put into the Memmert curing oven at 100°C for half an hour for the curing process and let it cool at air temperature.

Figure 3.4: The equipment used in formulating and preparing the hybrid ECA paste showing (a) a Mettle Toledo Analytical Balance (b) The Thinky Mixer Model ARE 310 centrifugal planetary mixer and (c) the Memmert Curing Oven

3.4 Fabrication of Printed Hybrid ECA on Substrate.

The substrate dimension 45mm x 12.7mm is used as the print surface for the composites. The substrate is being cut using the (Trotec® Speedy 〖300TM〗 laser cutter machine. The laser cutter machine was used since the machine offers an excellent cutting edge and uniform size of the substrate need. The 305 mm x 305 mm substrate is placed into the laser cutter machine, and the desired size of the substrate input will be inserted into the machine software, and the cutting process will begin.

Figure 3.5: A laser cutter machine to cut the acrylic substrate

Once the desired substrate size is prepared, the scotch tape is used to create the template for the printing purpose for the hybrid ECA. One piece of the substrate was used for each specimen. The uncured hybrid ECA is applied onto the prepared substrate sheet and will be a squeeze and flatten to the desired thickness by a razor blade. The printed hybrid ECA is then placed in the 100°C oven for half an hour. The cured hybrid ECA product is shown in Figure 3.7 below

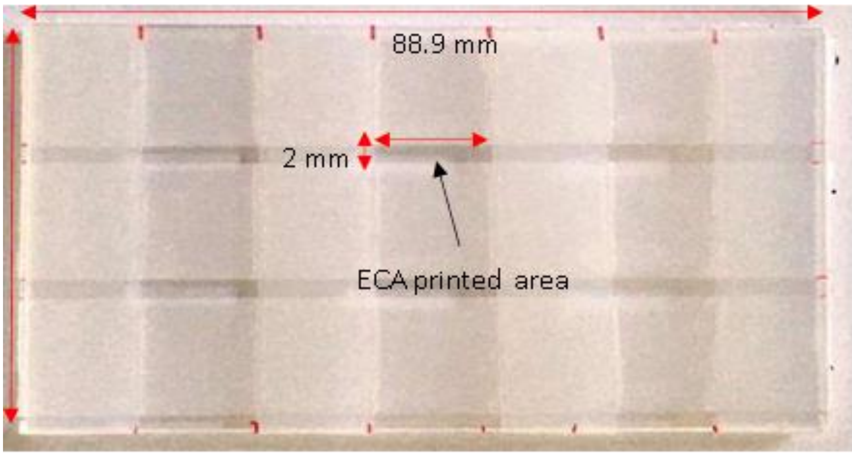

Figure 3.6: Substrate Sheet

Figure 3.7: Printed hybrid ECA

3.5 Electrical Conductivity Test

For the electrical conductivity test, a JANDEL rm3000+ 4-point probe was used to measure the resistivity of the printed hybrid ECA. The standard used for this electrical conductivity test is by referring to ASTM F390 as the standard guideline. There are six strips printed for each type of specimen. A minimum of three readings was taken for each of the strips, with a total of 36 readings taken for each specimen. The average reading for each specimen was then calculated to determine the conductivity of the specimen. The data from the JANDEL RM3000+ 4-point probe was then transferred to the MS Excel software to plot the resulting graph for each specimen. The precaution that needs to be taken when handling this machine is to reset the machine before taking reading for each specimen to avoid the data being mixed up with other specimens.

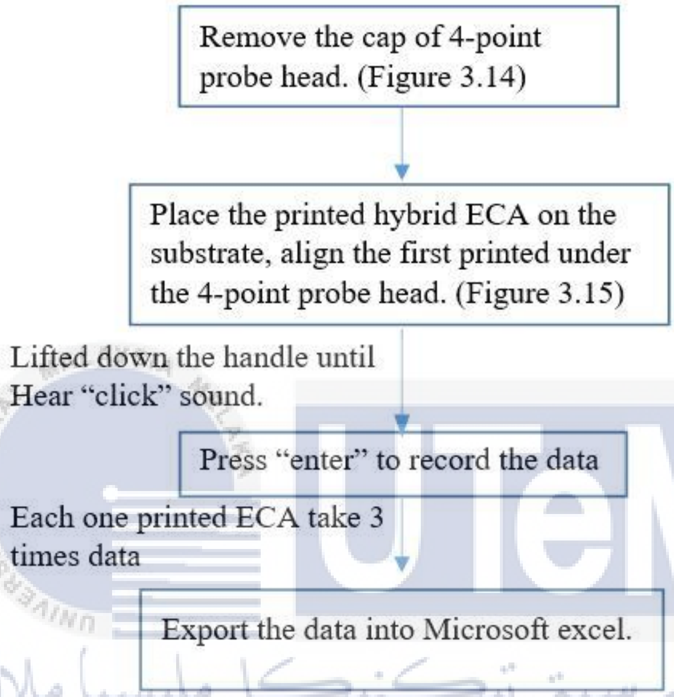

Figure 3.8: The flow process on how the resistivity data is measured and collected for analysis using the Microsoft software.

Figure 3.9: JANDEL model rm3000+ 4-point probe

The data recorded in the experiment was imported to Microsoft Excel, and the average reading will be calculated. The sheet resistance was calculated using equation (3.4), while the volume resistivity was calculated using equation (3.5), respectively.

$$R = G\ \frac{v}{I} \tag{3.4}$$

Where; R is sheet resistance $\Omega/\ sq$

G is correction factor; 1.9475

V is voltage

I is current.

$$\mathrm{P} = \frac{v}{I}\mathrm{Gt} \tag{3.5}$$

Whereby P is volume resistivity, Ω.cm;

t is the thickness of printed ECA

3.6 Lap Shear Test

An Instron Instron 8872 Universal Testing Machine (UTM) was used to conduct the lap shear test as per ASTM D1002 (Performance et al. 2019). This method is the most popular and commonly used to determine the bonding and performance of the materials (Trinidad et al., 2017). A minimum of five specimens was used to run the test for hybrid ECA containing Ag flakes of 3 wt. %, 4 wt.%, and 5 wt.% filler loadings blended with the 5 wt.% MWCNT in the epoxy binder. Figure 3.12 shows the flow process is done using Microsoft software.

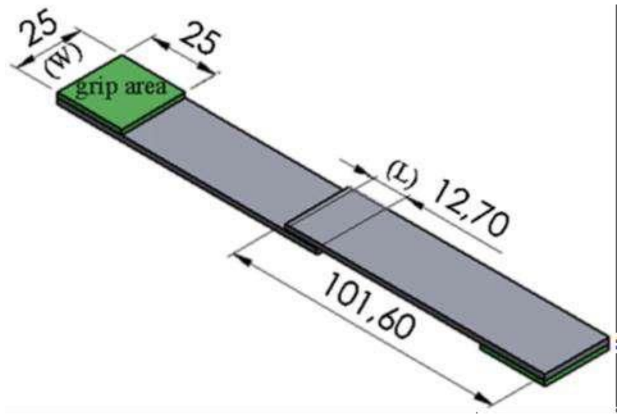

Figure 3.10: Schematic view of the tensile lap shear Nano adhesive joint sample (Ekrem et al. 2017)

The hybrid ECA is printed on the aluminum substrate, referring to the schematic view in Figure 3.10. The printed hybrid ECA then been placed into the designed jig and be cured in the oven at 100°C for 30 minutes. For the grips area, two supporting parts of the same sample thickness are used to prevent the bending impact on the joint zone. The flow process of the fabrication of the lap shear test specimen shown by the figure below.

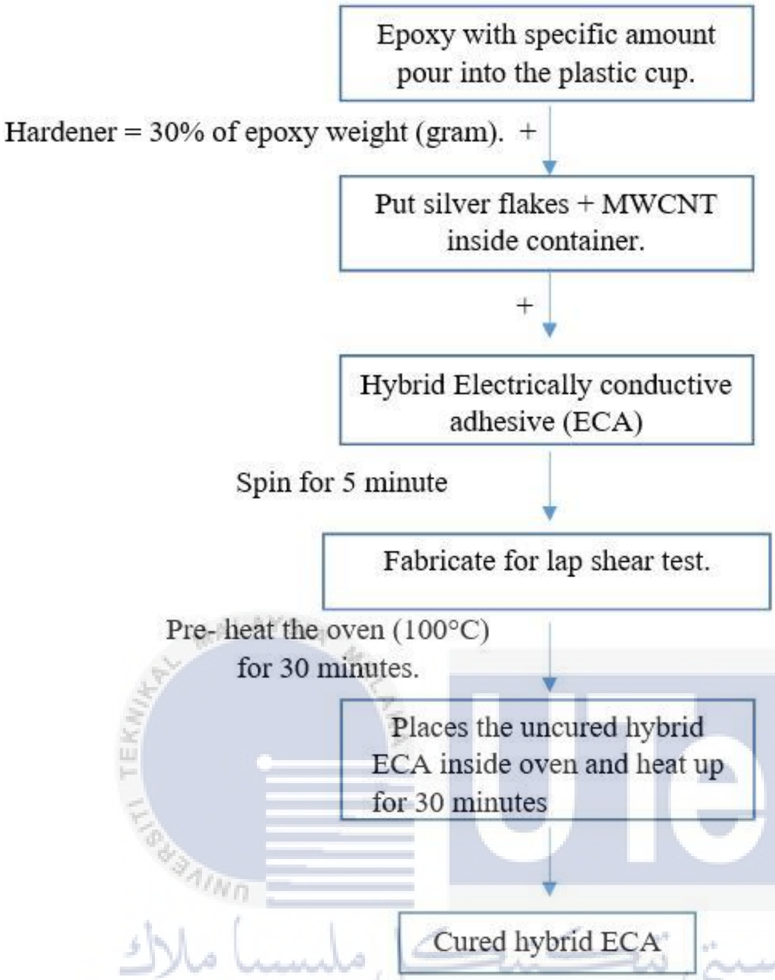

Figure 3.11: Fabrication of Lap Shear Test

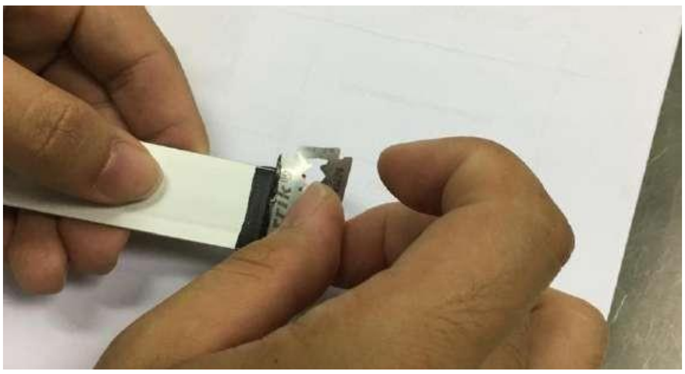

Figure 3.12: Printed hybrid ECA on an aluminum substrate.

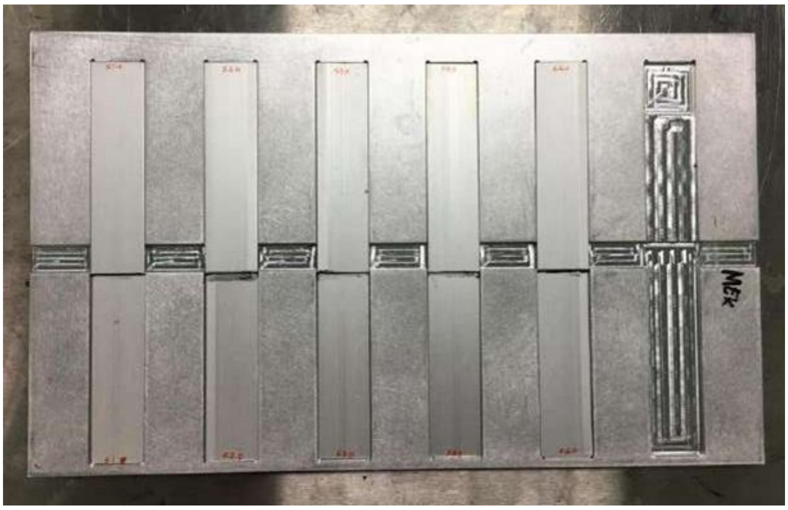

Figure 3.13: Designed jig used to cure the hybrid ECA

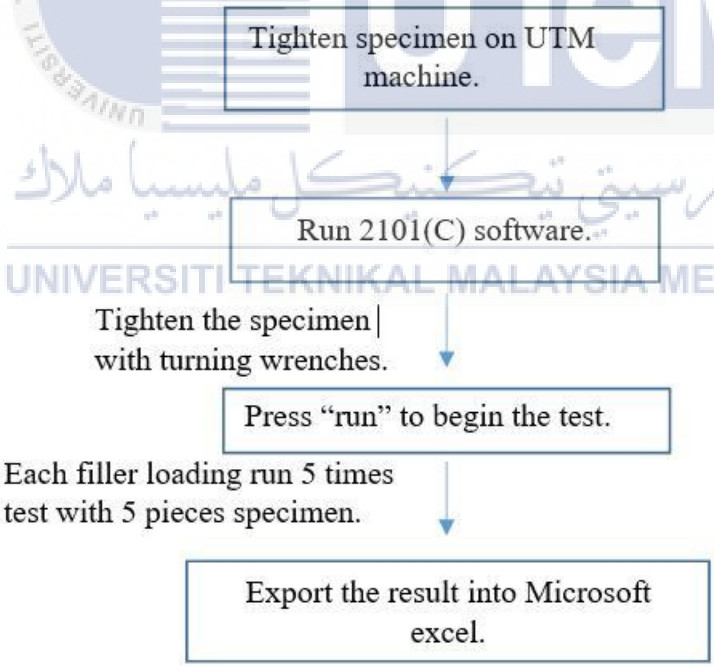

Figure 3.14: The flow process on how the lap shear test result is conducted and collected for analysis using the Microsoft software (Sow et al. 2019)

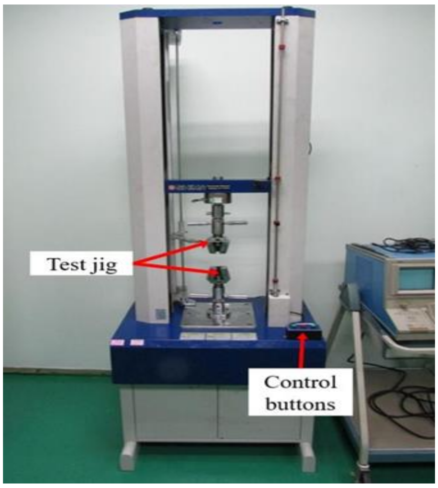

Figure 3.15: Universal Material Testing Machine

Figure 3.16: Set up for lap shear test

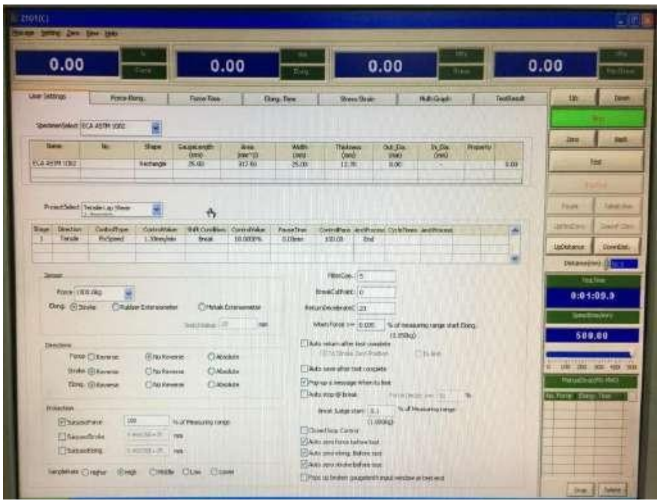

Figure 3.17: 2101 software for lap shear test

During the lap shear test, each sample is inspected by using the visual observation to determine the type of failure for different weight percent of filler loading. The result is discussed in Chapter 4.

CHAPTER 4

RESULT AND DISCUSSION

4.1 Introduction

In this chapter, the experimental results from the electrical and mechanical characterization of the hybrid ECA containing a constant 5 wt. % MWCNT and varying micron-sized silver flakes (3 wt. %, 4 wt. %, and 5 wt. %) conductive fillers are presented and discussed by correlating with the literature.

4.2 Electrical Performance of the Hybrid ECA with Varying Ag Filler Loading.

The conductivity performance of the hybrid ECA is presented in terms of the sheet resistance attained by the JANDEL 4-point probe. Table 4.1 shows the result for the sheet resistance for the hybrid ECA. Based on Ohm's law, the volume resistivity also can be defined as electrical resistance. Table 4.2 shows the result for the volume resistivity of the hybrid ECA composites. In general, it can be suggested that the electrical resistivity decreases with increasing filler loading for the hybrid ECA studied.

4.2.1 The Effect of Filler Loading on the Hybrid ECA Sheet Resistance

From the results, it is apparent that the average sheet resistance decreases from 63.97 ± 16.12 kΩ/sq for the hybrid ECA with 3 wt. % of silver flakes, to 41.84 ± 7.53 kΩ/sq for those with 4 wt. % silver flakes and 27.07 ± 10.16 kΩ/sq for the hybrid ECA containing 5 wt. % of the silver flakes. This trend is in good agreement with the literature (Marcq et al. 2011; Sow et al. 2019). It was argued that at the minimum resistivity, a percolation threshold is reached, which, in this study, is found at 10 wt.% of the filler loading, whereby there is 5 wt.% of Ag in the hybrid ECA. At the percolation threshold, there is a formation of a conductive path, thus resulted in a decrease in the resistivity of the hybrid ECA (Sow et al. 2019).

Based on Figure 4.1, increasing the filler loading from 3 wt. % to 5 wt. % results in a decrease in the average sheet resistance of the hybrid ECA. The highest sheet resistance is observed at 3 wt. % Ag filler loading, with a value of 63.97 kΩ/ sq while the lowest sheet resistance is noted at 5 wt. % Ag filler loading with a value of 27.07 kΩ/ sq.

Table 4.1: Data for average sheet resistance for hybrid ECA with varying filler loading.

Ag Filler Loading, wt. %	**Total Filler Loading (wt.%)**	**Average Sheet Resistance, kΩ/ sq**
3	8	63.97 ± 16.12
4	9	41.84 ± 7.53
5	10	27.07 ± 10.16

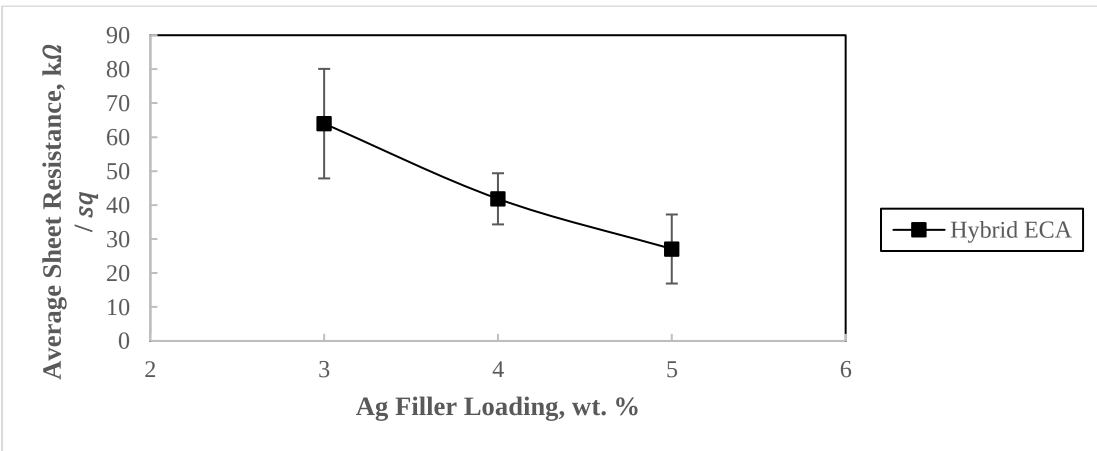

Figure 4.1: Graph of Average sheet resistance against Silver flakes (Ag) filler loading

Therefore, overall, from the results from the electrical properties for hybrid ECA, a total of 10 wt. % filler loading (5 wt. % silver flakes, 5 wt. % MWCNT) gives the lowest sheet resistance compared to 9 wt. % filler loading (4 wt. % silver flakes, 5wt. % MWCNT) and 8 wt.% filler loading (3 wt. % silver flakes, 5 wt. % MWCNT). This result shows that with increasing the filler loading, the electrical properties of the hybrid ECA resulted in decreasing resistivity and become a better electrical conductor (Marcq et al., 2011). From the previous research by Sow (2019), it was argued that increasing the fillers loading of a hybrid ECA will decrease the electrical resistivity of the hybrid ECA (Sow et al. 2019). Such observation is due to the percolation threshold reached, creating a conductive path in decreasing hybrid ECA resistivity and increasing conductivity (Patole and Lubineau, 2014). The trend suggests that increasing filler loading will cause a decrease in sheet resistance and volume resistivity, as stated by Mantena (2009).

4.2.2 Correlation of the hybrid ECA electrical property with the literature

Based on the results presented in Figure 4.3 and Table 4.3, in general, it is apparent that the average sheet resistance of hybrid ECA decreases with increasing filler loading for both types of hybrid ECA. Such observation is in good agreement with the work reported by Mantena et al. (2009) (Mantena et al. 2009).

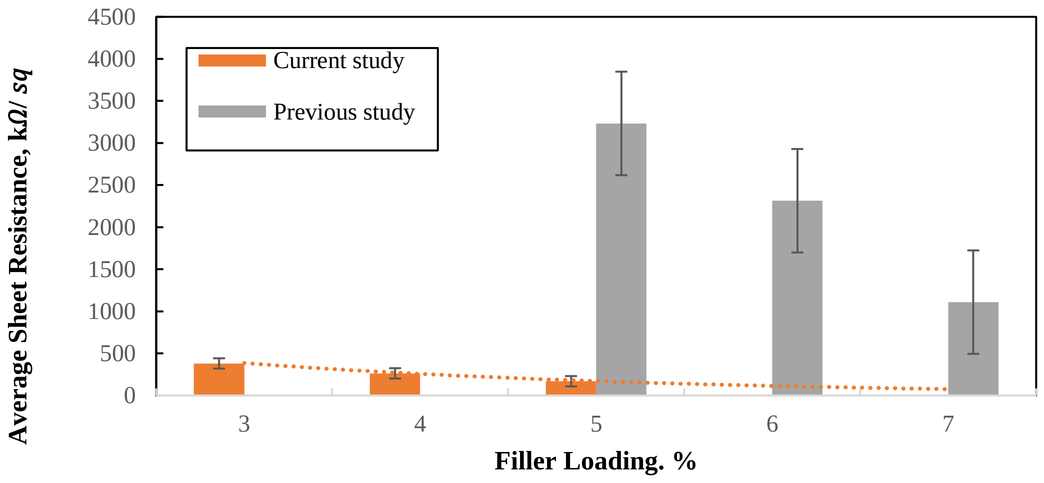

Figure 4.2 Correlation of the hybrid ECA's average sheet resistance with the literature.

Furthermore, the result shows that better electrical conductivity can be achieved with respect to an increase in the filler loading (Marcq et al. 2011; Nasaruddin et al. 2019; Sow et al. 2019). It was argued that by controlling of the filler loading wt. % can improve mechanical performance while enhancing the composite's electrical properties (Board 2019; dal Lago et al. 2020). The electrical conductivity exhibited in the hybrid ECA of the current study is superior to those reported in the past research, an indication that a synergistic effect is achieved in the hybrid ECA formulated in this present study. As an example, at total filler loading of 10 wt.%, there is more than fifty percent improvement in the electrical sheet resistance obtained in the current study.

Table 4.2 Comparison of the hybrid ECA's average sheet resistance

Reference	**Ag Filler Loading (wt.%)**	**MWCNT Filler Loading (wt.%)**	**Total filler loading (wt.%)**	**Average Sheet Resistance (kΩ/ sq)**
This study	3	5	8	381.14 ± 100.78
	4	5	9	263.02 ± 47.04
	5	5	10	169.20 ± 63.48
(Sow et al. 2019)	5	5	10	3233.17 ± 243.06
	5	6	11	2314.06± 962.34
	5	7	12	1109.45 ± 1333.95

4.3 Mechanical Performance of Hybrid Electrically Conductive Adhesive (ECA) With Varying Filler Loading.

4.3.1 The Effect of Filler Loading on the Lap Shear Strength of Hybrid ECA.

Based on Figure 4.3 and Table 4.5, in general, there is an increase in the average lap shear strength of the hybrid ECA. More specifically, from 3 to 4 wt.% of Ag, there is a gradual increase in the lap shear strength, with an optimum value of 5.12 ± 0.87 MPa exhibited by the ECA containing 4 wt.% Ag. Following this, with a further increase of 1 wt.% of Ag in the hybrid ECA system (total filler loading of 10 wt. %), there is a marginal reduction in the hybrid ECA average lap shear strength, with a value of 5.05±0.43 MPa. Such observation is possible because, after the specific composition of resin and filler loading in the ECA system, the filler is unable to carry the external load and results in decreasing shear strength of the composite (Ekrem et al. 2017), in which the strength of the composite is lower than the strength of the neat polymer matrix (Gabr et al. 2014). Moreover, it was argued that beyond a specific limit of filler loading content, the adhesion bond between the epoxy resin and filler loading would decrease, resulting in the strength reduction of the composite (Shakuntala, Raghavendra, and Samir Kumar 2014). In other words, it suggests that the composition of filler loading will increase the mechanical properties of the composite until it reaches a specific level or percolation and result in a decrease in the mechanical properties of the composite when exceeding the limit or threshold. A similar observation was reported by Mergen, Arda, and Evingür (2020); Shakuntala, Raghavendra, and Samir Kumar (2014).

Table 4.3: Data of average lap shear strength with varying filler loading

Ag Filler Loading (wt. %)	MWCNT Filler Loading (wt.%)	Maximum Lap Shear Stress (MPa)
3	5	2.85 ± 0.80
4	5	5.12 ± 0.87
5	5	5.052 ± 0.43

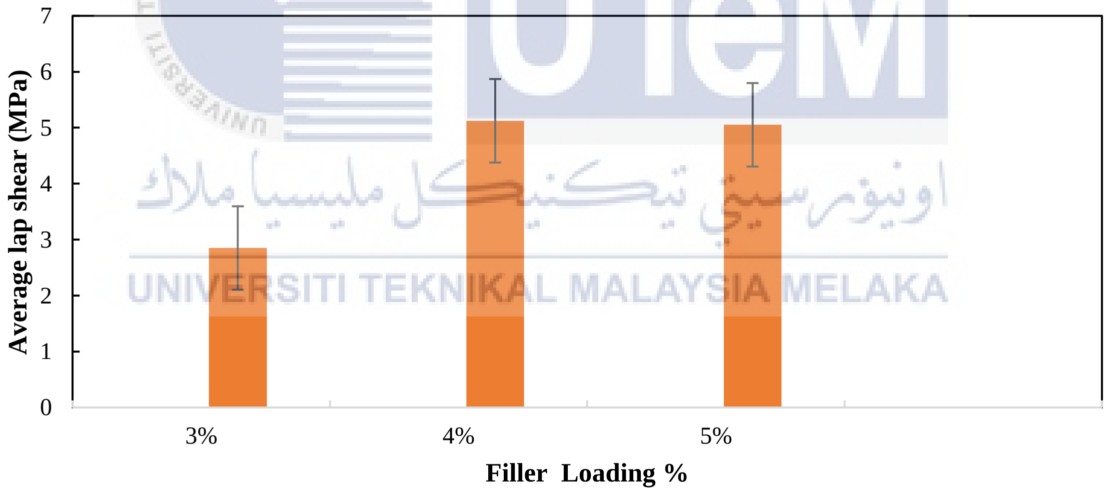

Figure 4.3 Average lap shear strength of the Hybrid ECA with varying Ag filler loadings

4.3.2 Correlation of the hybrid ECA lap shear strength with the literature

Based on the plot in Figure 4.4 and data in Table 4.4, there is a decreasing trend in the average lap shear strength with increasing filler loading reported by the previous study by (Sow et al. 2019)because the density of the composite is decrease compared to the polymer itself (Mergen, Arda, and Evingür 2020). Higher filler loading will lead to poor wetting and a reduction in the stress transfer efficiency across the filler resin interface (Gabr et al. 2014). However, at a total filler loading of 10 wt.%, the correlation between the hybrid ECA lap shear strength shows that the lap shear strength of the current study is inferior compared to those by Sow et al. (2019). Moreover, with an increasing amount of MWCNT in the hybrid ECA, greater lap shear strength is achieved in their work.

Besides, as shown in Figure 4.7 on the SEM images reported by Performance et., al (2019) the neat polymer shows the smooth fracture compare to the composite (Performance et al. 2019). The smooth fracture indicates the ductility of the material. Still, the composition of polymer and MWCNT as the filler loading delivers a rough surface, which shows the composite has become a brittle type of material. The matrix material will inherit some of the filler loading material properties, which explain why the strength of the neat polymer is better than composite (Performance et al. 2019).

Table 4.4: Correlation of the results with the previous study on average maximum lap shear strength

Reference	**Ag Filler Loading (wt.%)**	**MWCNT Filler Loading (wt.%)**	**Total filler loading (wt.%)**	**Average Max. Lap Shear Strength (MPa)**
This study	3	5	8	2.85 ± 0.80
	4	5	9	5.124 ± 0.87
	5	5	10	5.052 ± 0.43
(Sow et al. 2019)	5	5	10	8.61± 1.21
	5	6	11	7.79 ± 1.05
	5	7	12	7.03 ± 1.96

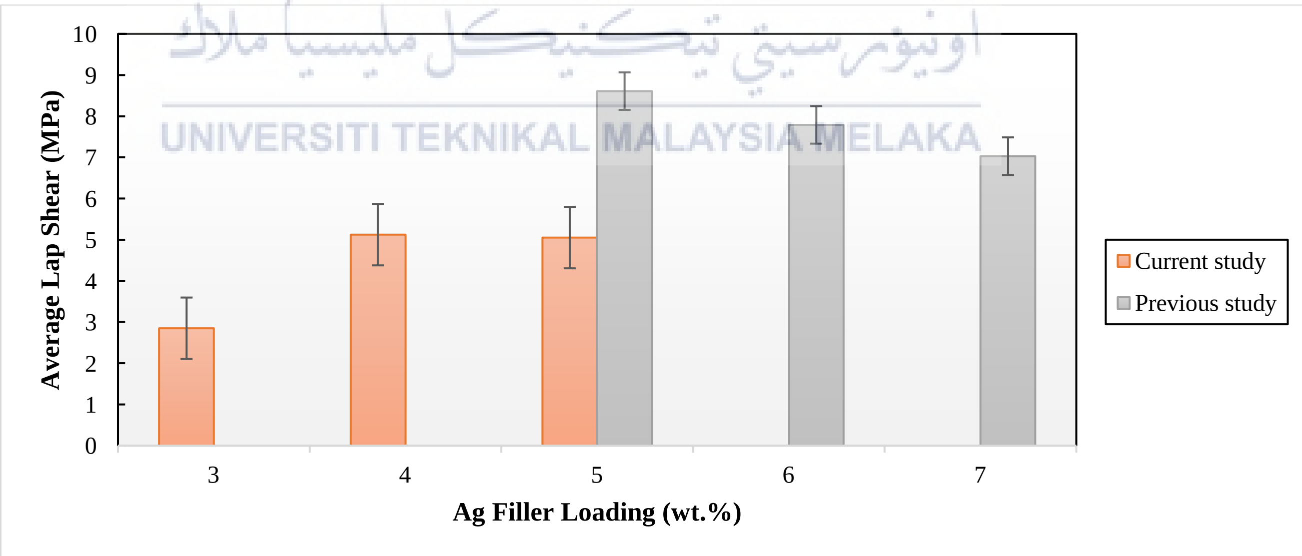

Figure 4.4 Comparison of the hybrid ECA lap shear strength with the literature

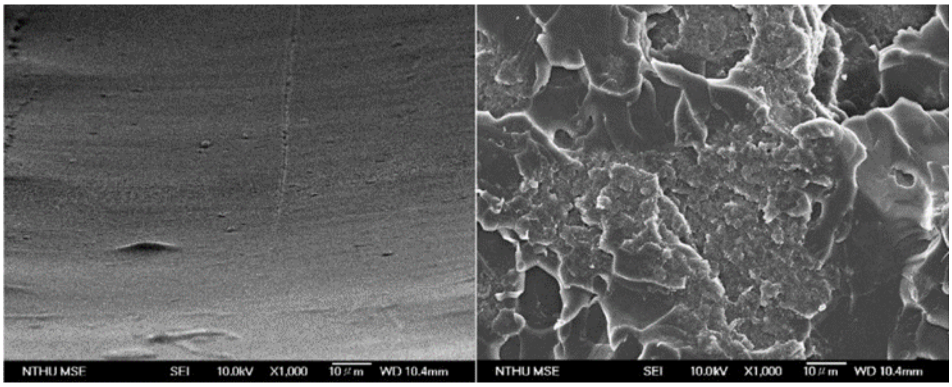

Figure 4.5 Tensile fracture surface of (a) 0 wt. % and (b) 5 wt.% of MWCNT filled epoxy polymer (Performance et al. 2019)

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

Based on the experimental results attained in this final year project, several conclusions can be drawn:

- In terms of the electrical property of the hybrid ECA, for the range of filler loading considered, that is with 3-5 wt.% micron-sized Ag flakes in combination with a 5 wt.% MWCNT, there is a gradual reduction in the average sheet resistance of the hybrid ECA, an indication that the adhesive conductivity is improved with increasing Ag filler.
- As for the mechanical property, the results from lap shear test revealed that increasing the Ag filler from 3 to 4 wt.% in the hybrid ECA system, (which correspond to 8-9 wt.% total filler in the hybrid ECA), shows a gradual increase in the average maximum lap shear strength. However, beyond this filler loading, there is a gradual decrease, suggesting a saturation state for the hybrid ECA system, therefore adding more filler does not further enhance the mechanical strength of the hybrid ECA.

5.2 Recommendation for future work

An extension of the current research work shall be carried out with suggestions as below:-

- The use of different filler types or other filler geometry of the conductive filler, such as silver nanowire or functionalized MWCNT, to evaluate the synergistic effect of the electrical properties of the hybrid ECA with varying filler loading.
- The use of a customized stencil printing template to produce better printing quality of the hybrid ECA
- Interfacial bonding of the hybrid ECA investigation by analyzing the ECA sample cross-section following the lap shear testing using appropriate imaging techniques such as Scanning Electron Microscope or Transmission Electron Microscope.

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