HYGROTHERMAL AGING EFFECT ON RELIABILITY PERFORMANCE OF ELECTRICALLY CONDUCTIVE ADHESIVES (ECA)

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HYGROTHERMAL AGING EFFECT ON RELIABILITY PERFORMANCE OF ELECTRICALLY CONDUCTIVE ADHESIVES (ECA)

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A report submitted

in fulfillment of the requirements for the degree of

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DECLARATION

I declare that this project entitled “Hygrothermal aging effect on reliability performance of electrically conductive adhesive (ECA)” is the result of my own research except as cited in the references.

Signature : .....

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 degree in Bachelor of Mechanical Engineering (Hons)

Signature :.....

Name :.....

Date :.....

DEDICATION

To my beloved mother and father

ABSTRACT

This research investigates the hygrothermal aging effect on performance reliability of electrically conductive adhesives made from solution mixing process using epoxy matrix and MWCNT with filler loading of 5 wt.%, 6 wt.% and 7 wt.%. The test specimens which are the electrically conductive adhesives (ECA) were prepared in accordance with ASTM F390-11 using a four point probe for electrical conductivity measurement while the lap shear test was conducted with reference to ASTM D1002-10 using a universal testing machine. For the hygrothermal aging study, the ECA samples were conditioned in a humidity chamber at setting conditions of 85°C and 85 % of relative humidity (85 % RH) to assess the reliability performance of the ECA. The test specimens were subjected to 168 hours and 504 hours of hygrothermal aging and specified test specimens were characterized at normal condition as controlled specimens. Following hygrothermal aging period, the test specimens were characterized in terms of their electrical and mechanical performance. With presence of moisture attack, that is the water molecules, the electrical conductivity of the ECA increase with hygrothermal aging period. Meanwhile, lap shear results revealed contradicting trend. Regardless of the amount of MWCNT filler loading used (5-7 wt.%), due to moisture attack, voids are created in the epoxy matrix of the ECA, which results in a decrease in the shear strength of the ECA, when the samples were subjected to 168 hours and 504 hour of hygrothermal aging at 85°C and 85% RH. The trend changes at the ECA after being aged for 504 hours. The 6 wt. % and 7 wt. % of MWCNT filler loading shows an increase of shear strength while the 5 wt. % shows a decrease in shear strength. Nevertheless, all specimen shows the formation of voids in the epoxy matrix of the ECA, which contributes to the decrease in the shear strength of the ECA.

ABSTRAK

Kajian ini mengkaji tentang kesan penuaan hygrothermal terhadap prestasi kebolehpercayaan pelekat pengalir elektrik dengan kaedah pencampuran cecair menggunakan epoksi matriks dan MWCNT dengan muatan sebanyak 5 wt. %, 6 wt. % dan 7 wt. %. Spesimen ujian iaitu pelekat elektrik (ECA) telah disediakan mengikut piawai ASTM F390-11 dan menggunakan alat "4 point probe" untuk mengukur aliran elektrik manakala ujian ricih telah dijalankan dengan merujuk kepada piawai ASTM D1002-10 dengan menggunakan mesin ujian universal. Untuk kajian penuaan hygrothermal, sampel ECA telah diletakkan di dalam mesin ruang kelembapan dalam keadaan suhu 85℃ dan 85% kelembapan relatif (85% RH) untuk menilai prestasi kebolehpercayaan ECA tersebut. Spesimen ini di uji selama 168 jam dan 504 jam di dalam ruang kelembapan manakala spesimen ujian yang berada dalam keadaan normal dikenali sebagai spesimen terkawal. Selepas penuaan selesai, spesimen ujian dicirikan dari segi prestasi elektrik dan mekanikal mereka. Kajian ini mendapati bahawa keupayaan ECA untuk mengalirkan arus elektrik semakin bagus. Ini kerana molekul – molekul air yang telah meresap hasil daripada tempoh penuaan hygrothermal. Akan tetapi, hasil daripada ujian ricih telah mendapatkan trend yang bertentangan. Tanpa mengira jumlah muatan MWCNT digunakan (5-7 wt.%), hasil keresapan air terhadap ECA tersebut telah menghasilkan ruang kaviti di dalam matriks epoksi ECA, di mana telah menyebabkan penurunan dalam kekuatan ricih ECA, apabila sampel tertakluk kepada 168 jam dan 504 jam penuaan hygrothermal pada 85 ° C dan 85% RH. Perubahan trend pada ECA 504 jam penuaan, spesimen dengan muatan MWCNT 6 wt. % dan 7 wt. % mereka telah menunjukkan peningkatan kekuatan ricih manakala muatan MWCNT 5 wt. % menunjukkan penurunan dalam kekuatan ricih. Walau begitu, kesemua specimen telah menunjukkan penghasilan kaviti didalam epoksi matriks pada setiap ECA, di mana ruang kaviti ini memberi kesan terhadap penurunan dari segi kekuatan ricih pada ECA tersebut.

ACKNOWLEDGEMENTS

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LIST OF ABBREVATIONS

ACA	Anistropic Conductive Adhesive
ACF	Adhesive Conductive Film
AFM	Atomic Force Microscopy
ASTM	American Society for Testing and Materials
C	Carbon
CNT	Carbon Nano – Tube
DWCNT	Double Wall Carbon Nano – Tube
ECA	Electrically Conductive Adhesive
EMI	Electromagnetic Interface
ICA	Isotropic Conductive Adhesive
MWCNT	Multi – Wall Carbon Nano – Tube
PDA	Polydopamine
RH	Relative Humidity
SEM	Scanning Electron Microscope
SWCNT	Single – Wall Carbon Nano – Tube
TEM	Transmission Electron Microscope

LIST OF SYMBOLS

T_g	=	Glass Temperature (°C)
W_t	=	Weight after environmental test
W_i	=	Weight before environmental test
Xi	=	irritant
$W_{fraction}$	=	Weight fraction of CNT
$W_{composite}$	=	Total mass of composite
W_{filler}	=	Mass of CNT (g)
W_{matrix}	=	Mass of polymer matrix (g)
Wt%	=	Weight percentage
V	=	Potential difference (V)
I	=	Current (μA)
R	=	Sheet resistance (Ω/sq)
τ	=	Shearing stress (N/mm^2)
F	=	Force (N)
A	=	Area of contact (mm^2)

CHAPTER 1

INTRODUCTION

1.1 BACKGROUND

Electrically conductive adhesives (ECA) is one of the replacements for the use of lead-bearing solders where it is known as one of the environmentally friendly solders in electronics (Li, Lu and Wong, 2010). The characteristic of the ECA is that it has a property to electrically conduct electricity as well as heat. The electrical conductivity in the ECA is important since the adhesive must be able to create an electric connection between electric components as well as providing electromagnetic interface (EMI) or radio frequency interference functions (Metal Finishing, 2008). The ECA also has a good adhesion, high mechanical strength, impact strength and have a conductive filler.

The ECA consists of conductive metallic properties and a polymer matrix. The most typically used conductive metal in the ECA are silver and epoxy resin (Amoli, 2015). This is due to the environment-friendly properties that silver has rather than using lead which is harmful towards humans. Also, silver has the highest electrical conductivity and the lowest resistivity compared to other conductive metal such as gold, nickel and copper. However, all the metallic conductor listed are expensive. The epoxy resin that is used in the ECA is one of the conducting polymers where it is possible to obtain high dielectric constant (Lu et al., 2007). Other than silver, gold, nickel and copper as the metallic filler in the ECA, carbon nanotube (CNT) are also used as the metallic filler in the ECA. The CNT is a tube-shaped material which is made from carbon where the diameter is measured in nanometre.

The CNT varies in length, thickness and number of layers. Its characteristic can be changed depending on the layer of graphene sheet that being rolled to create the tube and changes the property whether is metallic or semiconductor (Nanoscience Instruments, 2016). There are two types of CNT, the first one is called the single-walled CNT. The single-walled CNT or SWCNT shaped as a tube with only one layer. While the second type of CNT is called the multi-walled CNT or MWCNT. The MWCNT consists of several layers of CNT which it increases in diameter. The CNT is capable for lowering the cost for developing the ECA since it helps reduces the metal content in the ECA by constructing conductive networks hence reducing the cost of ECA (Luo et al., 2016).

1.2 PROBLEM STATEMENT

It is found that ECA is greatly affected when the ambient temperature and humidity are changed. The water gain in the polymer matrixes causes an internal hydrolysis of colloids which can result in an increase of electrical resistance and reduces the bonding strength which may lead to failure. In other words, hygrothermal aging of the adhesive bonds may result in decreasing of volume (Mach, Skvor and Szaraz, 2000).

In the previous study, the mechanical and the electrical properties of the ECA were tested under a constant humidity level of 85% at 85°C for about 500 hours. The results from the studies show that the contact resistance is increased with aging time. However, it is seen that the longer the duration of hygrothermal aging results in less effect on the contact resistant of the ECA (Cui et al., 2013).

Hence, this study aims to prolong the duration of the hygrothermal aging process for about 504 hours followed by performance reliability of the ECA in terms of the electrical conductivity and mechanical properties of the ECA using varying MWCNT filler loading.

1.3 OBJECTIVES

The objectives of this project are:

i. To investigate the effect of hygrothermal aging on the reliability performance of the ECA.

ii. To examine the shear strength of the ECA following hygrothermal aging.

iii. To study the effect of temperature and humidity on the electrical conductivity of the ECA.

1.4 SCOPE OF PROJECT

The scope covered in this project is as stated below:

i. Fabrication of ECA.

ii. Electrical characterization using a 4-point probe.

iii. Mechanical characterization using Universal Test Machine (Lap Shear Test).

iv. Surface morphology.

v. Reliability study

1.5 PLANNING

The research activities that were undertaken for final year project I or PSM I are summarized in Table 1.1. The progress for about the first four weeks includes background study, literature review and fabrication of ECA. In week five, the aging process is conducted by inserting the ECA into the humidity chamber for about four weeks. Following this, the thermally aged ECA is then tested for its conductivity and the mechanical properties, via lap shear test. Then, the experimental data are analysed from week 10 and initiation of the PSM I report writing began. Week 13 is the week for the draft to be submitted to the project supervisor for further advice and amendment. Finally, in week 14, the PSM I report will be submitted.

As for PSM II, the research activity are summarized in Table 1.2. The literature review are continued from the PSM I and being updated until week 12. Next is the hygrothermal aging process which takes about three weeks depends on the availability of the humidity chamber. Following the aging process is the electrical and mechanical characterization. The electrical characterization tested on its conductivity while the mechanical characterization is tested using the lap shear test. After the lap shear test, the specimen are subjected to SEM and EDS to determine the structure of the MWCNT and the fracture after the test. The EDS is done to verify the water content in the ECA after being subjected to hygrothermal aging.

Table 1.1: Gantt chart for the research activities for PSM1

Progress	Week													
	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Background Study														
Literature Review														
Lab Visit														
ECA Fabrication														
Conduct Experiment: Hygrothermal Aging														
Conduct Experiment: Electrical Conductivity														
Conduct Experiment: Lap Shear Test														
Data Analysis														
Report Writing														
Draft Submission														
Report Submission														

Table 1.2: Gantt chart for the research activities for PSM II

Progress	Week													
	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Literature Review														
ECA Fabrication														
Conduct Experiment: Hygrothermal Aging														
Conduct Experiment: Electrical Conductivity														
Conduct Experiment: Lap Shear Test														
Conduct Experiment: SEM analysis														
Data Analysis														
Report Writing														
Draft Submission														
Report Submission														

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

In this chapter, reviews of electrically conductive adhesives (ECA) which include the type of ECA filler materials, types of Carbon Nanotube, mechanical and electrical properties, surface morphology and the reliability performance of ECA from the previous studies are presented.

2.2 Electrically Conductive Adhesives

Electrically conductive adhesives (ECA) is a form of adhesives that provides mechanical and electrical properties which interconnects between a device, lead or chip carrier onto a circuit board. The unique ability is obtained by the composites material resides in the ECA. It is mainly composed of a polymer matrix and metallic particles (Li and Morris, 1998). The ECA is used as a replacement for soldering where the ECA is lead-free (Gilleo, n. d.). It also offers some advantages like flexiblility, simple processing and at a lower cost. There are two categories of ECA. The first one the Isotropic Conductive Adhesive (ICA) and the second one is the Anisotropic Conductive Adhesive (ACA) (Melorose, Perroy and Careas, 2015).

ICA is usually made up of silver as their metallic particle and known to be one of the substitutes for lead-based solder. The ICA possess a low-temperature processing, flexible and is compatible with non-solderable materials (Yim and Kim, 2010). However, due to its low conductivity and unstable contact resistance, it does not satisfy the similarity with the solder.

For example, the contact between silver particles is supposed to increase when the density increases due to contraction in curing of the binder and the electrical connection which results in the contraction during curing is not necessarily (Kohinata et al., 2013). The matrix shrinkage during curing does not help in development of increasing the conductivity (Lu, Tong and Wong, 1999). ACA is known as a method of connecting high density electrodes on electric components (Dou, Chan and Liu, 2004). It has the advantages of having low processing temperature, high component density and compatible with non-solderable components (Dou, Chan and Liu, 2003). Due to the adhesion between the substrate and the adhesives is one of the major concern for this type of ECA, the volume fraction is about 0.5% to 5% are below the percolation threshold. This results in unstable electrical conductivity before bonding (Mantena, 2009).

2.2.1 Matrix

The polymer matrix used in the ECA belongs to two categories. The first one is called thermoplastics. Thermoplastics consists of linear polymer chains with or without branching or side groups (Epoxy Technology, 2016). Its materials are rigid materials when at below the glass transition temperature, T_g. If the temperature is above the T_g, its starts to develop flow characteristics (E.C. Adhesives, 2010). The disadvantages of the thermoplastics is the degradation at high temperatures, where cavity is formed during the solvent evaporation.

Apart from cavity, polymer swelling also might occur when exposed to high temperature. As their redox state changes, the polymer may experience either swelling or de-swelling due to the changes in the bond length and excited by anions with oxidation and reduction (Le, Kim and Yoon, 2017). The second category of the polymer matrix is called thermoset. The thermoset consists of three-dimensional molecular network system.

The thermosets facilitate a chemical reaction between epoxy resins and hardeners when heated, which forms chemical bonds that hold each other and resists deformation (Wang, Zheng and Zheng, 2011). However, the thermosets must be mixed, in which they may be exposed to air and could not produce a good quality of mixing. The general polymer matrix that is used in ECA are listed in Table 2.1.

Table 2.1: Comparison of adhesive (E.C. Adhesives, 2010)

Materials	Advantages	Disadvantages
Epoxies	High – temperature use	Longer cure cycles with anhydride hardeners
	Good moisture and chemical resistance	
	High purity	Degassing required for two – component systems
	Low outgassing	Exotherms in large quantities for amine – curing agents
Silicones	Highest purity	Migrate to other circuit elements
	Stress absorbing	Low surface energy
	High and low temperature stability	Swelled by nonpolar solvents
Polyurethanes	Good flexibility at low temperatures	Lower thermal stability and service temperature than epoxies (150 – 163°C)
	Stress absorbing	Average bond strength unless primer is used
	Highly versatile chemistry	
Polyimides	Higher temperature stability compared to epoxies	Trapped solvent can produce voids under large ICs Multi – step curing required to volatilize solvent
		High – stress materials
	High ionic purity	May absorb moisture in cured condition
	Reduce bleed out	Cannot be B – staged
Cyanate esters	High adhesion strength	High moisture absorption
	High thermal stability, High T_E low CTE	Popcorn susceptibility

There are possibilities that the mixing between the thermoplastic material and thermoset polymers could produce a better matrix where the ECA is able to be cured in lower temperature and high temperature performance (Danish Electronics Lights & Acoustics, n.d.).

2.2.2 Filler

The commonly used metal filler in the ECA are carbon, graphite flakes and micron or nano-sized particles such as solver, nickel, copper or aluminium. The metallic filler is mixed with the polymer epoxy counterpart which forms the ECA. However, the metallic fillers have several disadvantages such as low conductivity, unstable contact resistance, low binding strength, silver migration, and difficult to be reworked. Carbon based filler usually used in low conductivity applications. Regarding this, a study by Kim et al. (2004) shows that a new type of ECA, one filled with low-melting-point alloy fillers to overcome the disadvantages, results in a good metallic interconnection and low contact resistance with 30 % of the filler volume fraction.

It is found that when there is sufficient amount of filler in the polymer matrix, the electrical properties of the ECA material changes from being an insulator to a conductor. This is caused by the linking between the metallic particles with the polymer matrix. However, if the amount of the metallic filler increases until it reaches the critical volume, V_c, it reduces the resistivity tremendously. This is called the percolation threshold (Sancaktar and Bai, 2011).

2.3 Carbon Nano-Tube

Carbon Nano-Tube, or CNT, is a nano material shaped as a tube. It has the thickness of $1/50000^{th}$ of the human hair which makes them possible to form a single atomic layer thick (Dai et al., 2003).

It is able to join with each other even with different physical structures. The rolling direction and the diameter of the CNT are the basis for their fundamental properties (Zhang, 2012). There are two main categories of CN; the first one is the Multi-Walled CNT (MWCNT) and the second one is Single-Walled CNT (SWCNT).

2.3.1 Single Walled Carbon Nano-Tube

The SWCNT is a single surface of CNT which are usually used for its electronic properties due to their varied chirality. The SWCNT is also used as a catalyst for direct ethanol or methanol fuel cells (Dong, Henderson and Field, 2012). The SWCNT are tested for its physical properties by using a simple model that comprises of SWCNT rope and an atomic force microscopy (AFM) (Sinclair, 2009). Figure 2.1 shows the method used to test the physical properties of the SWCNT while Figure 2.2 shows the relationship between elastic modulus and shear modulus as function to rope diameter.

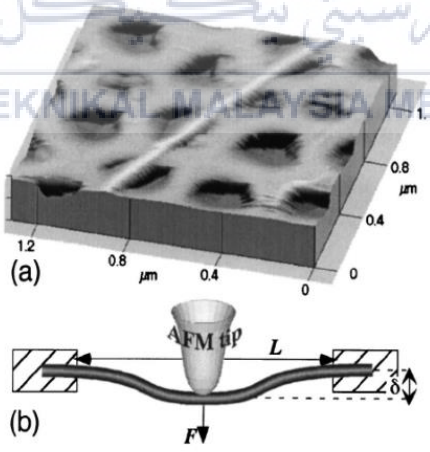

Figure 2.1: SWCNT rope on a porous membrane (a) AFM tip probe used to determine the physical properties of SWCNT (b) (Sinclair, 2009).

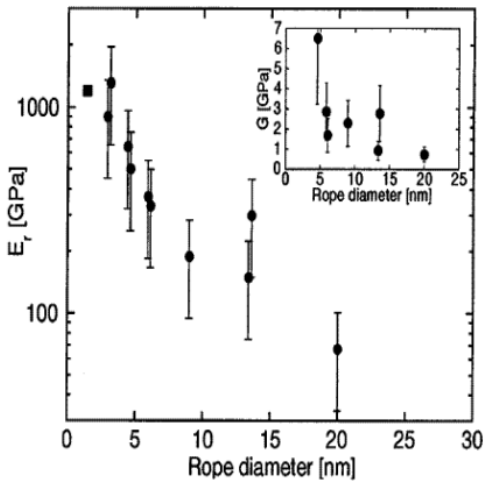

Figure 2.2 : Decreasing in elastic modulus and shear modulus as function to rope diameter (Sinclair, 2009).

The SWCNT will show its properties as a metallic material for about 1/3 of the total SWCNT while another 2/3 of the SWCNT shows semi-conducting properties if the chiral vectors are equal (Bandaru, 2007). Regarding this, a study by Sakurai et al. (2013) shows the limiting processes of the SWCNT by synthesising the SWCNT with various diameter and density. They studied the resulting growth kinetics and determined the fundamental growth of the SWCNT.

2.3.2 Multi-Walled Carbon Nano-Tube

The Multi-Walled Carbon Nano-Tube or known as MWCNT has been used in epoxy-based material due to its mechanical, thermal and electrical properties. A study was conducted where the MWCNT was used as a composite which results in addition for about 1 wt.% of MWCNT in the epoxy would increase the Young's modulus and yield strength by two times the original value and four times when the MWCNT amount is about 4 wt.% (Allaoui et al., 2002). Another study was conducted to determine the properties of the MWCNT composites. It was reported that there was an increase in the Young's modulus and

tensile strength of the ECA, regardless of whether the MWCNT filler was chemically treated or untreated (pristine). Nonetheless, due to agglomeration which occurs in the MWCNT composites, this results in a big apparent filler loading. Moreover, the agglomerates also reduces the volume fraction of the epoxy matrix (Montazeri et al., 2010). There are other methods to increase the lap shear strength of the MWCNT.

From the literature, coating Polydopamine (PDA) or acid treated to MWCNT is proven to be less effective to increase the lap shear strength. However, PDA coated pristine increases the bonding strength considerably (Subramanian et al., 2015). It was reported that with addition of small amount of the MWCNT results in an increase in the adhesive' shear strength for about 30% to 50%. Figure 2.3 shows a micrograph showing the ECA fractured surface as reported by Hsiao, Alms and Advani (2003).

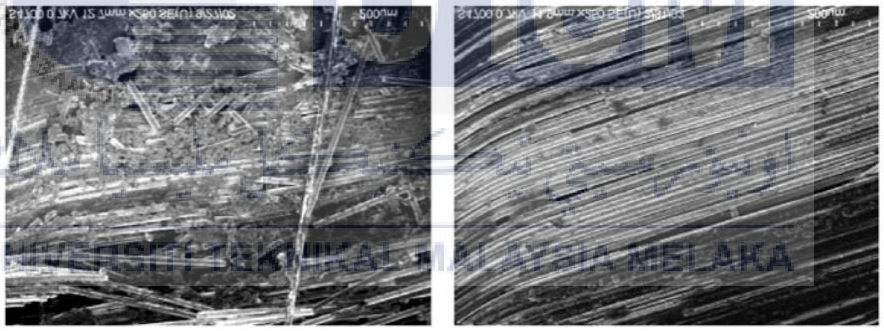

Figure 2.3: SEM image of fracture surface (Hsiao, Alms and Advani, 2003).

2.4 Mechanical Properties of Electrically Conductive Adhesives

The mechanical properties of the ECA is mostly based on the polymer matrix that had been used to fabricate the ECA. Usually the mechanical properties of the ECA were tested on the adhesion between metal-to-metal joints by using the lap shear test (Trinidad, 2016).

According to Mantena (2009), the higher the filler loading the lower mechanical strength thus increasing the electrical conductivity of the ECA. However, the results shown in Figure 2.4, obtained by Chew et al. (2016), shows that the higher the filler loading, the higher the tensile stress of the ECA.

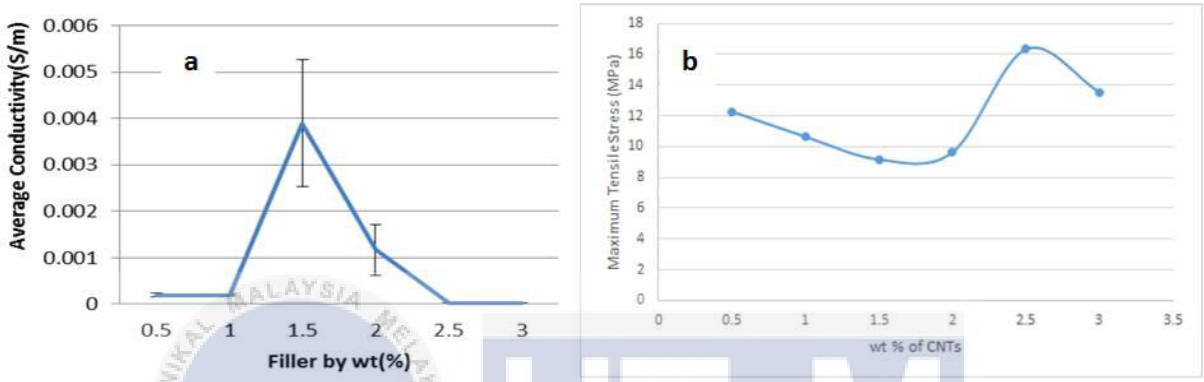

Figure 2.4 : (a) Average conductivity and (b) Maximum Tensile stress against filler loading (Chew et al., 2016).

From the findings, it can be concluded that the filler loading and the polymer matrix needed to be in a balanced amount in order to have balanced properties for mechanical and electrical properties. Proper mix or combinations may produce better performance in mechanical properties of the ECA.

2.5 Electrical Properties of Electrically Conductive Adhesives

Like the mechanical properties, the electrical properties of the ECA are present due to the metallic filler in the ECA. The higher the filler loading, the higher the electrical conductivity of the ECA (Mantena, 2009). Low filler loadings in the ECA causes a scatter

in the resin which makes it difficult to create a conductive path for electrical flow. This renders the ECA non-conductive (Li et al., 2016). Figure 2.5 shows the electrical resistivity of ECA with silver as filler. There was a study conducted on increasing the conductivity of the ICA type ECA by lowering the filler loadings or decreasing the size of the filler. Both methods proven to increase the electrical conductivity of the ICA type ECA (Wu et al., 2006) where Table 2.2 shows the results.

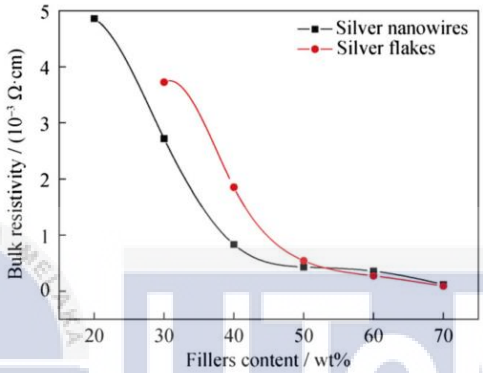

Figure 2.5: Electrical resistivity on ECA with Silver as filler (Li et al., 2016).

Table 2.2: Resistivity increasing with filler loading (Wu et al., 2006)

	Bulk resistivity (Ωcm)	Filler content (wt. %)
ICA filled with Ag nanowires	1.2×10^{-4}	56
ICA filled with 100nm Ag particles	3.65×10^{-1}	56
ICA filled with 1 μm Ag particles	3.64	56
ICA filled with 1 μm Ag particles	7.5×10^{-4}	75
ICA filled with silver dendritic	4.23×10^{-3}	70
Traditional Sn40/Pb60 solder	3×10^{-5}	>95

ACA type of the ECA has different electrical properties with different direction, which allow them to be widely used in field emission type electronics (Gao et al., 2010). As for ICA type ECA, it has a relatively high resistivity and low mechanical.

2.6 Surface Morphology of Electrically Conductive Adhesives

Surface morphology studies shows the characterization for the filler in the polymer matrix of the ECA. It shows the dispersion of the filler whether it leads from micron-sized agglomerates of filler bundles. Figure 2.6 below shows the scanning electron microscope image of hybrid composites of double wall CNT (DWCNT) and MWCNT (Marcq et al., 2011).

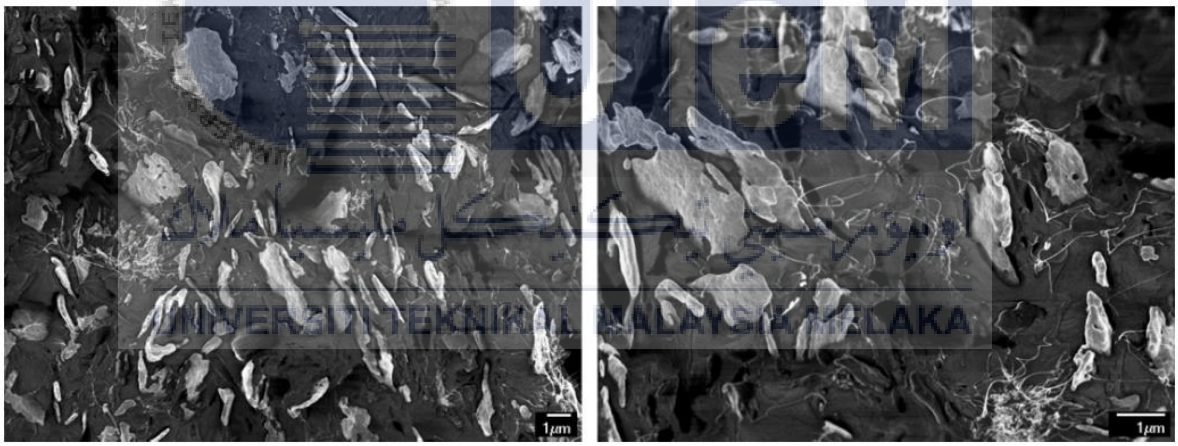

Figure 2.6: SEM image of DWCNT-Silver hybrid composites (Marcq et al., 2011).

In the study by Zhang, Chen and Xiao (2011) on the modified ECA filler, evidence of the difference in the filler after being treated by acetone centrifugation process and annealing process of silver nano-wires are apparent as shown in Figure 2.7. Most of the images were taken after the surface treatments. Since the surface treatments contributes on improving the properties of the ECA, Kornain et al. (2008) findings on treated silver nanoparticles as the

ECA filler shows that the structure of the nanoparticles is elongated but retains the spherical shape.

The modifications made by treating the surface with silane-based coupling agent causes an improvement in conductivity since the agglomerates was not formed between the particles. Figure 2.8 shows the TEM image of silver nanoparticles.

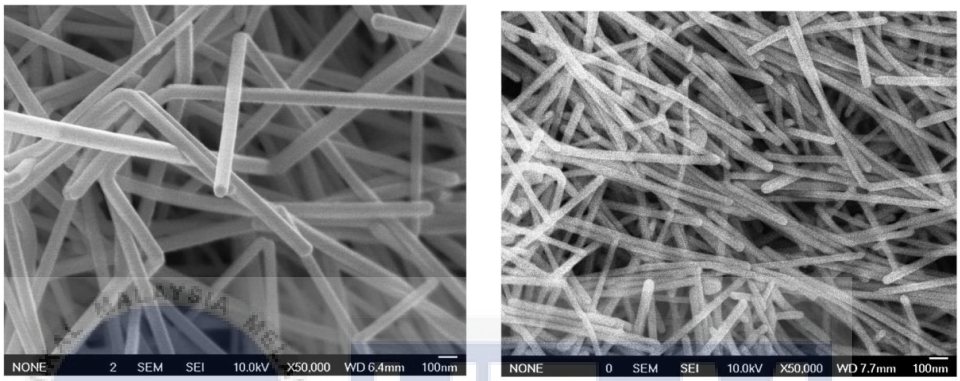

Figure 2.7: Difference between acetone centrifugation process (left) and annealing process (right) (Zhang, Chen and Xiao, 2011).

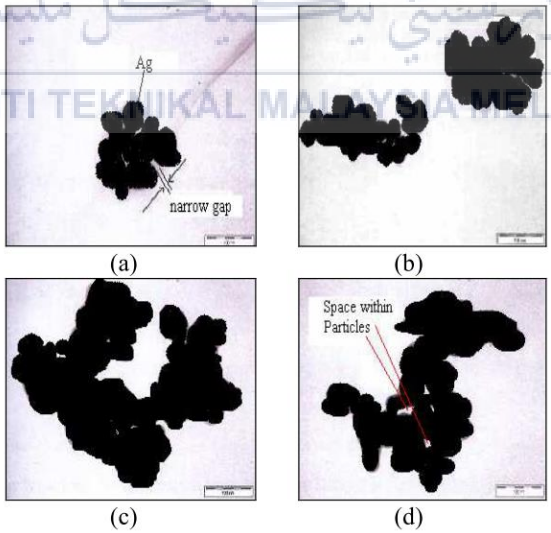

Figure 2.8: TEM image of silver nanoparticles (a) and (b) shows less elongation while (c) and (d) shows agglomerations (Koamin et al., 2008).

2.7 Reliability Performance of Electrically Conductive Adhesives

The reliability performance of the ECA is mostly known by its poor reliability of the adhesive joints. This is mainly due to the exposure to various type of environmental conditions whether a sudden increase in temperature and humidity. Other factors such as increased loadings, stress, cyclic thermal conditions, etc. effects greatly towards the ECA. For ACA type ECA, it is greatly affected by the temperature change since it could melt into a low viscosity liquid and forms a three-dimensional network. The network formed is important to characterize the ACA (Lin and Chen, 2008). As for ICA type ECA, they are known by its reliability problems between the contact resistance and non-noble metal (Li and Wong, 2006). Yim et al. (2008) stated that between ICA and ACA, ICA is more suitable for flip chip bonding than ACA.

2.7.1 Effect of Hygrothermal Aging on Electrically Conductive Adhesives

Hygrothermal aging the ECA under operating conditions and accelerated conditions may increase the joint resistance of the ECA. The Hygrothermal aging initially improves the physical property of the composite in the ECA. After some time, the volume of the polymer matrix decreases and increasing the contact between the matrix and filler. However, it degrades the physical properties of the ECA (Barto, Cinert and Mach, 2011). Higher filler loadings contributes to the lowering of the joint resistance of the ECA (Nicolicis and Mündlein, 2007). Another study conducted to improve the joints by hygrothermal aging the ECA at temperature ranges at 160 °C to 200°C which results in better contact and decreasing the non-linearity (Mach, Skvor and Szaraz, 2000).

Cui et al. (2013) found that the ECA absorbs the moisture that is on the ECA surface and continues along the aging time. The aging of the ECA also proved to increase the resistance of the adhesive joints which are caused by partial damage contact with the conductive particles (Mach, Radev and Pietrikova, 2008).

Copper based ECA has increased in volume resistivity with aging time when under elevated temperature and humidity (E.C. Adhesives, 2010). Wong and Lu (2000) found that the increased resistance due to the silver flakes in the ICA started to experience galvanic corrosion during the hygrothermal aging.

2.7.1.1 Effect of Hygrothermal Aging on Mechanical Properties

The mechanical properties of the ECA after being thermal aged shows a significant drop of mechanical performance in increasing of aging time. The S_n37Pb and the adhesive has decreased in shear strength after the hygrothermal aging (Liu et al., 1997). The hygrothermal aging is mostly affects the micro-structure and the mechanical properties of the epoxy resin (Gao, Chen and Gao, 2012). A study on the conductive adhesive film (ACF) finds that the Young's modulus and the tensile strength declines rapidly at the earlier duration of the hygrothermal aging. As time increases, the decreasing rate declines slightly (Gao, Chen and Gao, 2012). Figure 2.11 shows the results of the relationship mention earlier. Xu (2002) reported that the hygrothermal aging results slightly reduces the strength and failure stain which the samples experienced thermal degradation and embrittlement on the matrix.

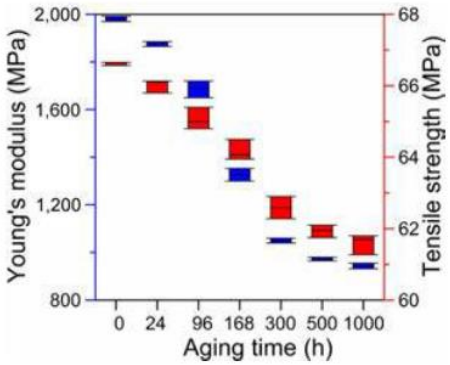

Figure 2.9 : Tensile strength and young's modulus decreases with increasing aging time (Gao, Gao and Chen, 2015).

2.7.1.2 Effect of Hygrothermal Aging on Electrical Properties

The hygrothermal aging could cause a significant effect on the electrical properties of the ECA. The resistivity of an ECA is greatly affected by the hygrothermal aging. During the initial aging time, the resistivity is reduced greatly. Over the hours, the decreased resistivity started to reach a plateau or either increases. This is caused by the degradation process that occurred in the ECA. It disrupts the conductive paths between the matrix and the conductive filler which results in an increased resistivity (Barto, Cinert and Mach, 2011). Figure below shows the resistivity of the ECA after being hygrothermal aged for about 840 hours.

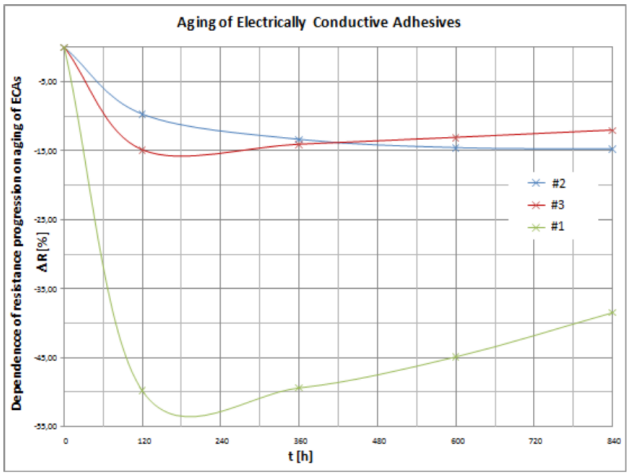

Figure 2.10 : Resistivity dependence over time (Barto, Cinert and Mach, 2011).

Klosterman, Li and Morris (1998) used an ICA type ECA on the effect of the hygrothermal aging at 85°C and 85% RH. The results that had been obtained shows the interfacial resistance increases. He also stated that the interfacial resistance is different with the volume resistivity behaviour. Figure 2.11 shows the comparison between the interfacial resistivity and volume resistivity.

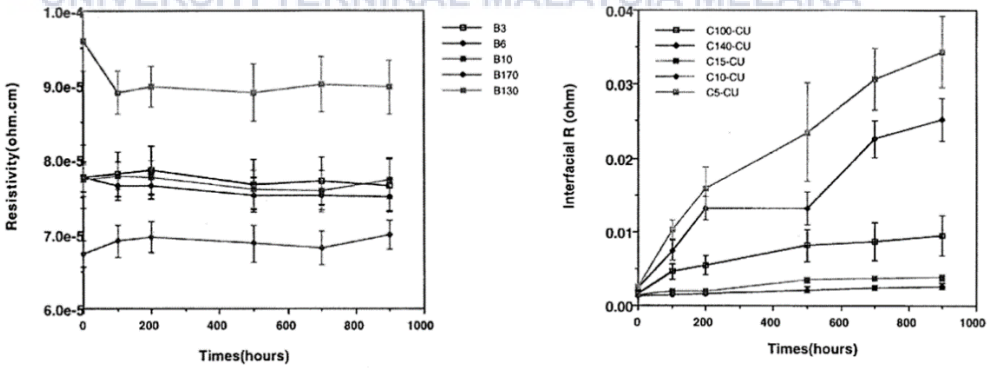

Figure 2.11 : Difference between resistivity behaviour(left) and interfacial resistance(right) (Klosterman, Li and Morris, 1998).

2.7.2 Moisture Effect on Electrically Conductive Adhesives

The ECA may be affected by the moisture for which it is the key for the ECA to determine for its long-term usage since the ECA will be used in various condition which may deteriorate the adhesive. The adhesive strength will be reduced when the adhesive matrix absorbs the moisture (Coughlan and Lewis, 2006). Also, due to incomplete curing, the epoxy could absorb more water by a significant amount. With the absorption of the moisture, the performance of the ECA would be changed permanently. The moisture intake can be measured by the weight gain relation with the total weight gain (Tan et al., 2006) as given in the expression in Equation (2.1) below:-

$$\frac{W_t - W_i}{W_i} \times 100\% \quad (2.1)$$

Where the W_t is the weight after environmental test, and W_i is the weight before the environmental test.

The internal stress of the ECA cause some changes in the polymer chains in the matrix where the amount or the size of the polymer increases in size. This is called the swelling process. Figure 2.12 shows the moisture absorption increases with the temperature and time. The pressure however, affects the balance between the differences in pressure which results in more moisture intake (Cao, Lai and Liu, 2005).

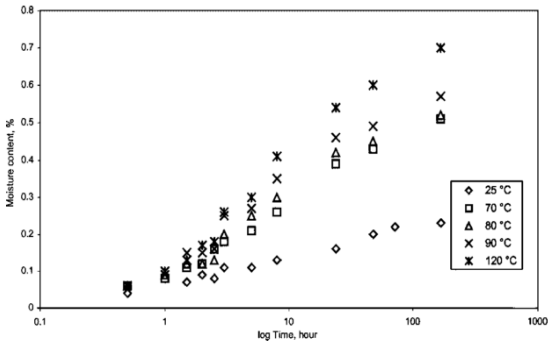

Figure 2.12 : Moisture absorption increases proportionally with temperature and time (Cao, Lai and Liu, 2005)

CHAPTER 3

METHODOLOGY

3.1 Overview of Research

The methods for ECA fabrication, machine and apparatus used, type of test being done to the specimen are shown. Figure 3.1 shows the flow of this research that being conducted. It starts with the literature review until final report writing. The research activities that is carried out during the final year project or known as PSM is as summarized below:

i. Fabrication of ECA, substrate for electrical test and aluminium plate for lap shear test.

ii. Procedures of equipment usage that will be used for the research which includes:

   a. Oven for curing process.

   b. Humidity chamber for hygrothermal aging process.

   c. Universal test machine for mechanical test.

   d. 4-point probe for electrical test.

   e. Scanning electron microscope (SEM) for surface morphology

iii. Sample preparation before inserting into the humidity chamber.

iv. Conduct electrical and mechanical test before and after hygrothermal aging.

v. Data collection and data analysis.

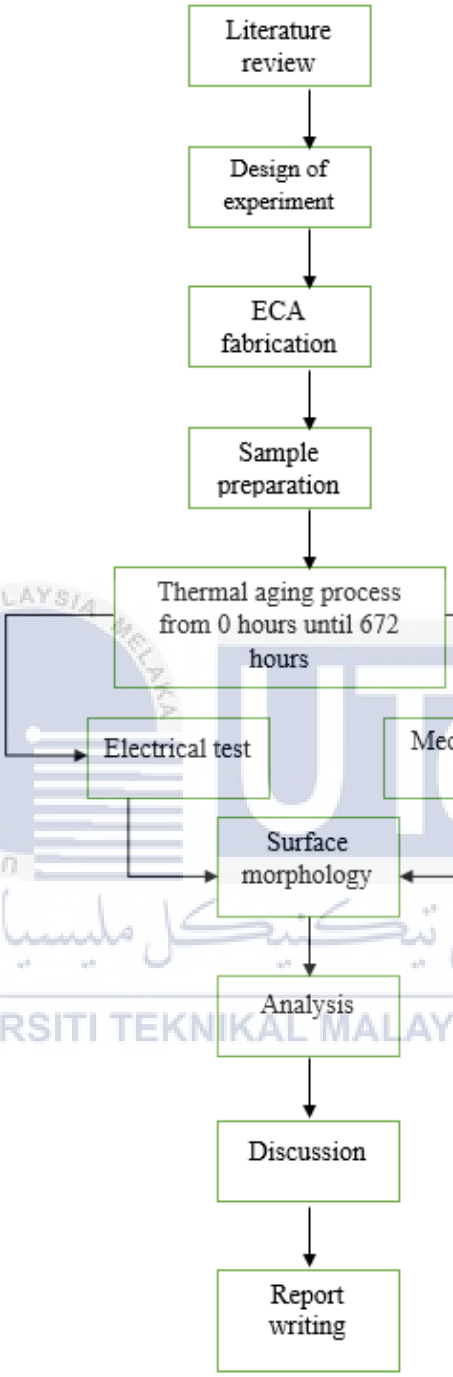

Figure 3.1: Research activity flow.

3.2 Raw Materials

3.2.1 Polymer

Araldite 506 epoxy resin is used as the polymer matrix for the ECA. This epoxy is one of the three mixing components for fabricating the ECA. It is responsible for the ECA to develop a connection between the ECA and the substrate. The epoxy are supplied by Sigma – Aldrich. This epoxy resin is suitable for research and development usage. It is colourless and it has several hazard category which is:

a. Skin corrosion and irritation (Category 2)
b. Serious eye damage and eye irritation (Category 2)
c. Skin sensitisation (Category 1)
d. Hazardous to the aquatic environment - chronic hazard (Category 2)

Handling the epoxy resin requires safety masks and impervious gloves. Table 3.1 shows the properties of the epoxy resin.

Table 3.1 Sigma – Aldrich epoxy resin properties[59].

Material Properties	
Physical Properties	Araldite 506 epoxy resin
Form	Semi solid melting to a liquid
Colour	Colourless
Melting point/ Freezing point	-15 – 5 °C
Flash point	252°C
Vapour pressure	0.04 hPa
Relative Density	1168 g/cm^3
Incompatible materials	Strong oxidizing agents, acids, Amines, Bases
Toxicity	LD50 Oral - Rat - 13.600 mg/kg
Carcinogenicity	none

3.2.2 Hardener

The hardener that is used for fabricating the ECA is the Polyether amine D230. The hardener is the second component that is used to fabricate the ECA. It is used as the curing agent for the ECA. It has an average molecular weight of 230. The molecular structure of the Polyether amine is shown in the Figure 3.2.

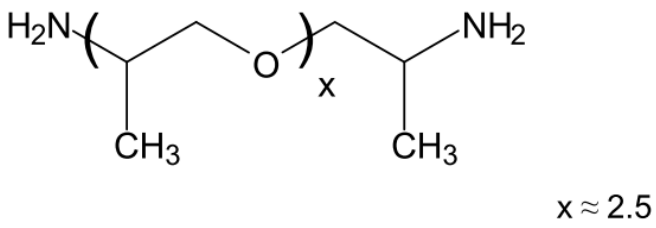

Figure 3.2: Molecular structure of Polyether amine

The advantages of this hardener is that it has a low viscosity, colour and vapour pressure, provides tough, clear, impact resistant coatings and adhesives (Technical Bulletin). Table 3.2 shows the physical properties of the hardener.

Table 3.2: Polyetheramine D – 230 physical properties.

Material Properties	
Physical Properties	Polyetheramine D – 230
Form	Semi solid melting to a liquid
Colour	Colourless to pale yellowish
Viscosity	9.5cSt
Flash point	121°C
Vapour pressure	1/100 – 10/133 mm Hg/°C
Relative Density	1168 g/cm^3
pH	11.7
Refractive Index	1.4466

3.2.3 Carbon Nano – Tube

In this research, the MWCNT is used as the metallic filler for the ECA. The powdered MWCNT supplied by Nanostructured & Amorphous Material Inc. (NanoAmor), USA. The MWCNT is categorized as irritant (Xi) hazard since it will cause irritation to the eyes and the respiratory system. Hence, proper safety measures were taken such as wearing impervious gloves and mask when handling the MWCNT. Table 3.3 shows the MWCNT specifications while Table 3.4 shows the physical properties of the MWCNT:

Table 3.3: NanoAmor MWCNT specification (Air Liquid Canada Inc., 2007).

MWCNT	Outer diameter, OD (nm)		Length, L (μm)		Aspect Ratio, A.R (L/OD)		
	Min.	Max.	Min.	Max.	Min.	Max.	Avg.
	10	20	10	30	1000	1500	1250

Table 3.4: NanoAmor MWCNT physical properties (Air Liquid Canada Inc., 2007).

Material Properties	
Physical Properties	MWCNT (10-20nm,OD)
Form	Powder
Colour	Black
Odour	Odourless
Density	~2.1 g/cm^3
Bulk density	0.04 – 0.05 g/cm^3
SSA	>200 m^2/g
Purity	>95%
Melting Point	3652 – 3697 °C

3.3 Electrically Conductive Adhesives preparation

The ECA is made up from three raw materials. Polymer matrix, curing agent and conductive filler. All these materials needed to be mixed properly to produce an ECA. Firstly the amount of the material in terms of weight percentage are calculated by using the Rule of Mixture, as given in Equations (3.1) and (3.2) below:-

$$W_{fraction} = \frac{W_{filler}}{W_{composite}} \tag{3.1}$$

$$W_{composite} - W_{filler} = W_{matrix} \tag{3.2}$$

Where $W_{fraction}$ is the weight fraction of the CNT for example 5wt. %, 6wt. % and 7wt. %. $W_{composite}$ is the total mass of the composite which is 5g, is used for all specimens. W_{filler} is the mass of the CNT required and W_{matrix} is the mass of the polymer matrix. For hardener, the mass required is determined by multiplying 30% from the W_{matrix}, as shown in Equation (3.3) below:-

$$W_{matrix} \times 0.3 = weight\ or\ mass\ of\ hardener \tag{3.3}$$

After the mass of the materials are determined, they are then mixed according to specific sequence starting with the polymer matrix, hardener and lastly conductive filler. Before starting the fabrication, the mass of the container is measured by using the Mettler Toledo balance and after the measurements are calibrated, the polymer matrix is poured into the container as shown in Figure 3.3.

After pouring the epoxy into the container, the hardener is poured afterwards. When the amount of the required hardener has been added, the mixture is then stirred for one minute as shown in Figure 3.4. During stirring, the mixture starts to become murky. This indicates the chemical reaction has started between the polymer matrix and hardener. The stirring is continued until the murkiness dissipates.

Figure 3.3: Mettler Toledo Balance (left) and pouring the epoxy into container (right)

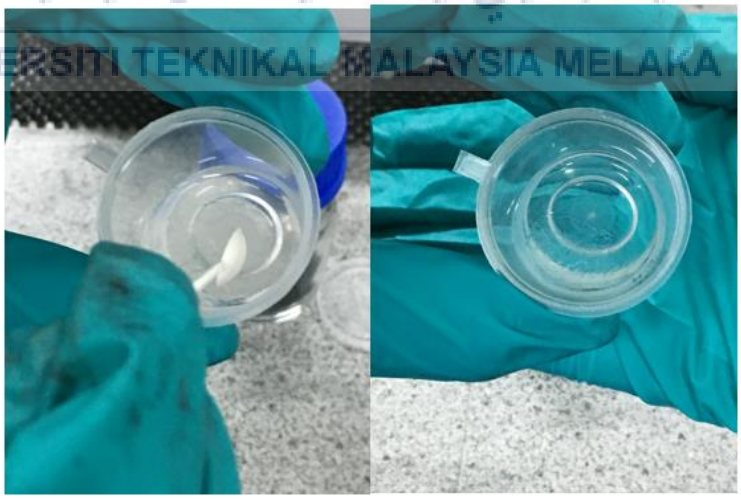

Figure 3.4: Stirring the mixture of the epoxy and hardener (left), murkiness as a sign of chemical reaction (right).

Lastly, MWCNT is added into the mixture until the required amount. Then, it is stirred for five minutes as shown in Figure 3.5.

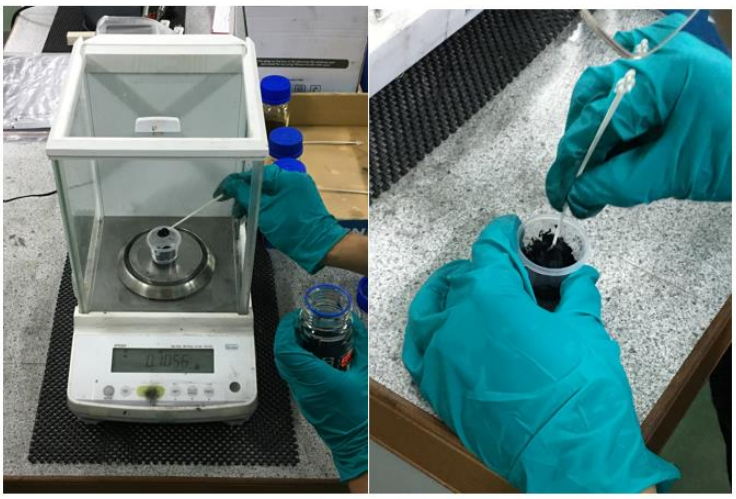

Figure 3.5: Inserting MWCNT into the mixture (left), mixing of ECA (right)

3.3.1 Sample Preparation

There are two types of sample needed to be prepared. The first one is for the electrical test and the second one is for the lap shear test. For the electrical test, the substrate is prepared using a Scotch™ tape and Perspex™ board. The Perspex™ board is cut using a Perspex™ laser cutting machine. The dimensions of the substrate are shown in Figure 3.6.

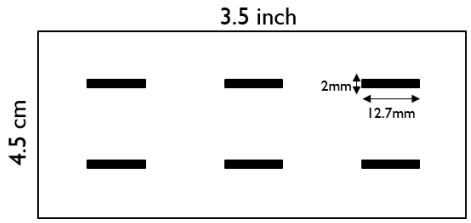

Figure 3.6: Electrical test substrate dimension

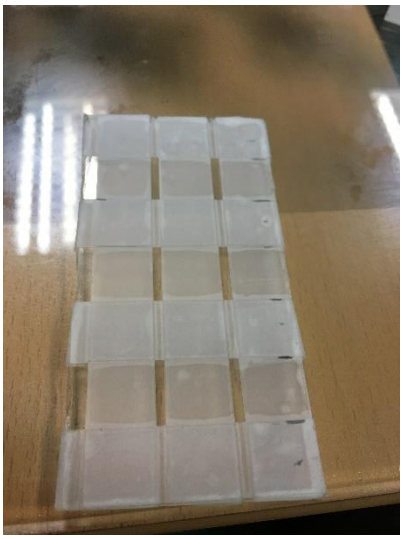

Figure 3.7: Substrate for electrical test.

The ECA is spread on the substrate evenly so it fills the cavity in Figure 3.7, with the thickness of 0.01 mm – 0.02 mm. The test specimen is prepared for each filler loadings as shown in Figure 3.8.

Figure 3.8: Placing the ECA on the substrate (left), Spreading the ECA evenly (Right).

The specimen that is used for the lap shear test is the aluminium strip. The dimensions and method required to place the ECA on the specimen is done by following the ASTM D – 1002 – 05 (2005) guideline. The specimen is prepared by cutting the aluminium sheet using the shearing machine shown in Figure 3.9.

Figure 3.9: Shearing machine.

After cutting the aluminium sheet, it is then taped with the Scotch™ tape to indicate the area of the ECA will be spread on. Figure 3.10 shows the dimension of the specimen preparation for the lap shear test. The ECA is then spread on both specimen at one side. Afterwards, it is mounted on a holder so the specimen would be held in place with the help of the clipper.

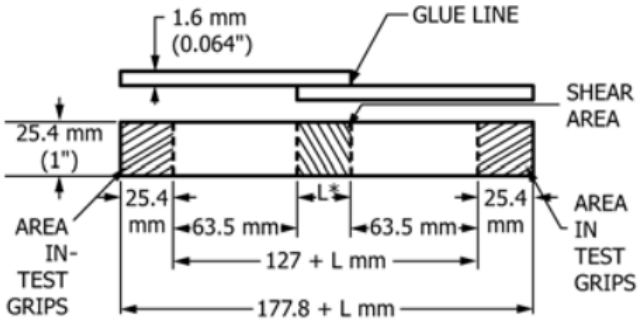

Figure 3.10: Specimen dimension for lap shear test (ASTM D 1002-05, 2005).

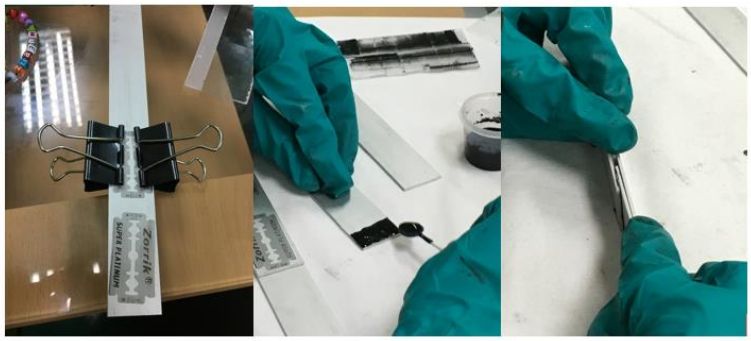

Figure 3.11: Specimen holder with clipper (left), Spreading the ECA on the specimen (middle), Mounting the specimen on the specimen holder (right)

Before inserting the specimen into the oven, the specimen is clipped to ensure the force exerted on the ECA is distributed evenly as can be seen in Figure 3.11 and 3.12. Both electrical and lap shear test specimens are inserted into the oven for about 30 minutes for curing process at 100°C. After the curing process, the tests specimen is left to cool down so that the ECA are cured properly. There are about 4 test specimens per filler loading which consists of 1 test specimen for electrical test and 3 for lap shear test. The test specimens were prepared for 0 hours, 168 hours and 504 hours of hygrothermal aging.

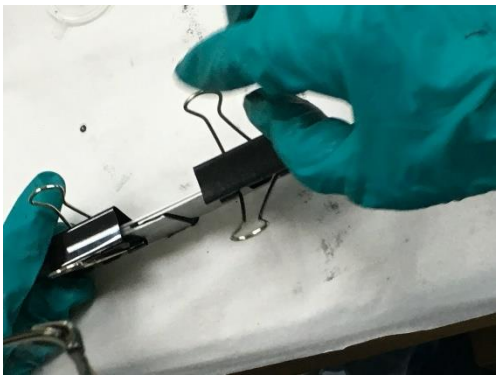

Figure 3.12: Clipper clipped on the test specimen and the specimen holder.

3.4 Hygrothermal Aging Process

The 0 hours (as-received condition) test specimen was tested under normal condition for the controlled test. The data obtained following the hygrothermal aging process is compared with the controlled test specimen. All the test specimen conditioned in the humidity chamber were recorded in terms of their mass before and after hygrothermal aging process.

The hygrothermal aging process are conducted before the electrical and the lap shear test are conducted except for 0 hours test specimens, since the test specimens are used as the controlled data for this research. Test specimens for 168 hours and 504 hours are inserted into the humidity chamber. The humidity chamber is set at 85°C for its temperature and 85% relative humidity.

3.5 Electrical Conductivity Test

The test specimen for electrical test are conducted after the hygrothermal aging process. The electrical conductivity test is conducted by using the 4-point probe shown in Figure 3.13. The measurement of the conductivity are done by directly flow the current through the test specimens which the potential difference can be determined (ASTM, 2015). As for the sheet resistance, it is determined by calculating the measured current and voltage values using correction factors.

Figure 3.13: Jandel model RM3000+ 4 point probe.

The formula that is used to calculate the sheet resistance is given in Equations (3.4) and (3.5) shown below:-

$$V = IR \tag{3.4}$$

Rearranging,

$$R = \frac{V}{I} \tag{3.5}$$

Where,

i. V = Voltage, volt, V

ii. I = Current, ampere, A

iii. R = Sheet resistance, Ω/sq

When the sheet resistance is obtained, it is multiplied with the correction factor which is 1.9475. The test is done by indenting the ECA strip on the test specimen for about 3 to 6 times per strip. The movement from one point to another takes about 0.05 mm to 0.1 mm. For each specimen, 6 ECA strip must be taken its conductivity. The 4 point probe is calibrated before start conducting the electrical conductivity test as shown in Figure 3.14. In this research, the current is set for the electrical test is 1μA. After the data is obtain the average for sheet resistance times correction factor is determined.

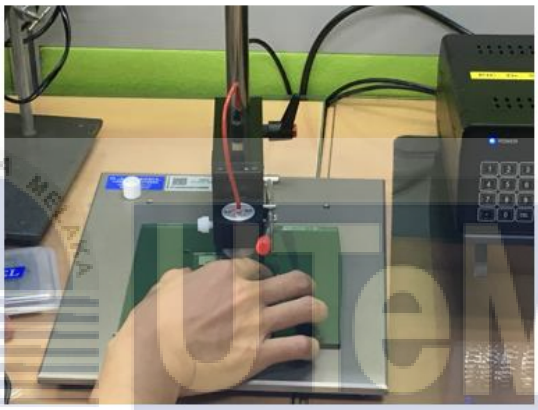

Figure 3.14: Measuring the electrical conductivity.

3.6 Lap Shear Test

The lap shear test is conducted using the universal test machine shown in Figure 3.15. The test specimen is placed at the slot of the machine and then being tighten by turning the wrenches. The distance between the test specimens can be adjusted by the lowering or rising the upper jaw of the universal test machine.

Figure 3.15: Placing the test specimen on at the UTM machine (left), Lap shear test in progress (right)

Each of the test specimen are tested accordingly after the hygrothermal aging process is completed. During the lap shear test, the machine may break the test specimen since high force is exerted on the test specimen. Hence, the safety distance between the UTM must be concerned. The test is completed when the test specimen is separated. Equation (3.6) shows the shearing stress that is calculated following the lap shear testing.

$$\tau = \frac{F}{A} \tag{3.6}$$

Where,

i. τ = Shearing stress, MPa or N/mm^2

ii. F= Force, N

iii. A= Area of contact, mm^2

3.7 Surface Morphology

The surface morphology study is conducted by using the Scanning Electron Microscope machine (SEM). It uses a beam with high amount of electron to obtain various signals at the surface of the test specimen. The SEM reveals information about the texture, chemical composition, structure and the constituents between the polymer matrix and conductive filler.

Before using the SEM, the specimen must be prepared by cutting into smaller size or suitable sizes for about 1cm x 1cm size for the SEM to analyse the specimen. The specimens were chosen for its lowest and highest shear strength. The specimens were placed on a stub where it is coated with a platinum layer. This is to prevent charging of electron when the beam ray is sprayed on the specimen.

In this study, SEM analysis is conducted to examine the type of failure occurred in the ECA after the lap shear test. In addition, it also can examine the bridges or linkages between the polymer matrix and the metallic filler that contributes to the electrical conductivity of the ECA.

CHAPTER 4

RESULTS AND DISCUSSION

4.1. Introduction

In this chapter, the results of the experiment are explained by comparing the differences between the controlled condition of ECA and the hygrothermal aged ECA. These were explained for its electrical resistivity of the ECA and the shear strength of the ECA.

4.2. Effect of Hygrothermal Aging on the Electrical Resistivity of the ECA

The electrical conductivity test is conducted to access the ECA using three different filler loadings, which is 5 wt. %, 6 wt. % and 7 wt. %. There are 3 groups of test specimens which is the controlled condition or 0 week of hygrothermal aging , the 168 hours or 1 week of hygrothermal aging and the 504 hours or 3 weeks of hygrothermal aging with 85°C temperature and 85% RH of humidity. The tests were conducted accordingly and the data is tabulated in Table 4.1.

The experimental result following the electrical conductivity test for the controlled specimens suggest that the sheet resistance of the ECA decreases with increasing filler loadings. From the result obtained during the electrical conductivity test, it is found that the resistance for each filler loading decreases from 5 wt. %, 6 wt. % and 7 wt. % sequentially due to the amount between the conductive filler and the non-conductive polymer differs in each filler loadings (Barto, Cinert and Mach, 2011).

For the 5 wt. % filler loading, the ECA does not reach the percolation threshold. The transition between insulator–to–conductor is still not enough since the amount of the polymer matrix is still higher than the conductive filler (Ma et al., 2010). This explains why the resistance of the 5 wt. % filler loading being the highest. With increasing filler loadings, the percolation threshold is reached at 6 wt. % and 7 wt. % results in both filler loading have developed a conductive path which reduces the resistivity of the ECA and increasing the conductivity of the ECA.

Table 4.1: Electrical Conductivity Data.

Electrical Conductivity in kilo-ohms/square			
Filler Loadings	0 hours (0 Week) of Hygrothermal aging	168 hours (1 Week) of Hygrothermal aging	504 hours (3 Week) of Hygrothermal aging
5 wt.%	64.96 ± 29.04	39.17 ± 9.16	48.74 ± 11.22
6 wt.%	5.61 ± 1.19	5.55 ± 2.08	9.17 ± 1.32
7 wt.%	1.75 ± 0.30	1.34 ± 0.14	2.15 ± 0.33

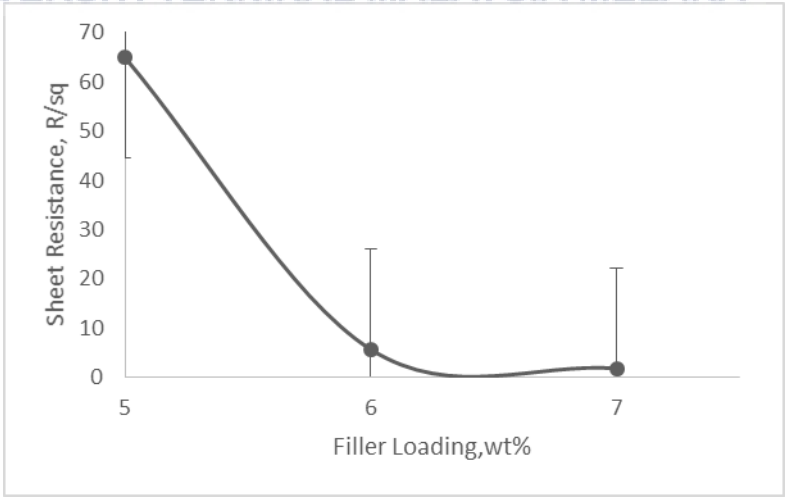

Figure 4.1: Results for electrical conductivity for 0 hours of hygrothermal aging.

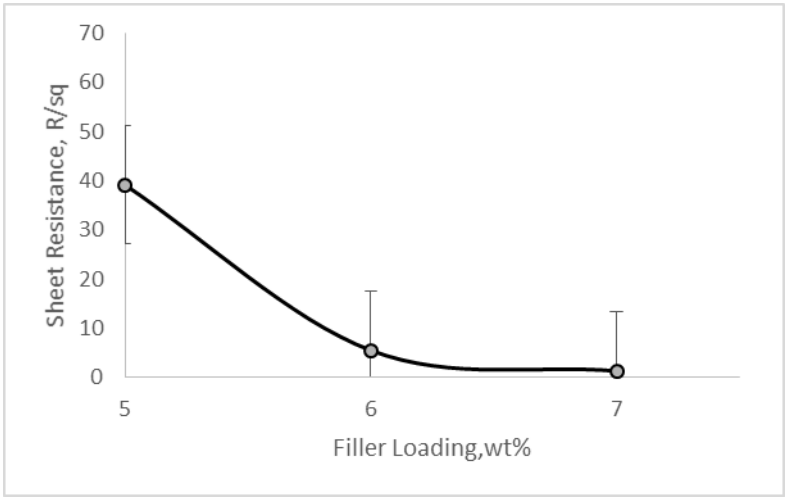

Figure 4.2: Results for electrical conductivity for 168 hours of hygrothermal aging

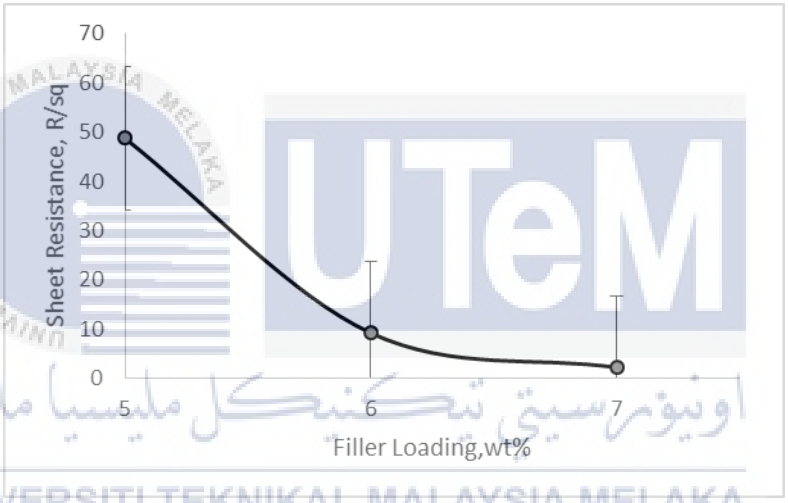

Figure 4.3: Results for electrical conductivity for 504 hours of hygrothermal aging

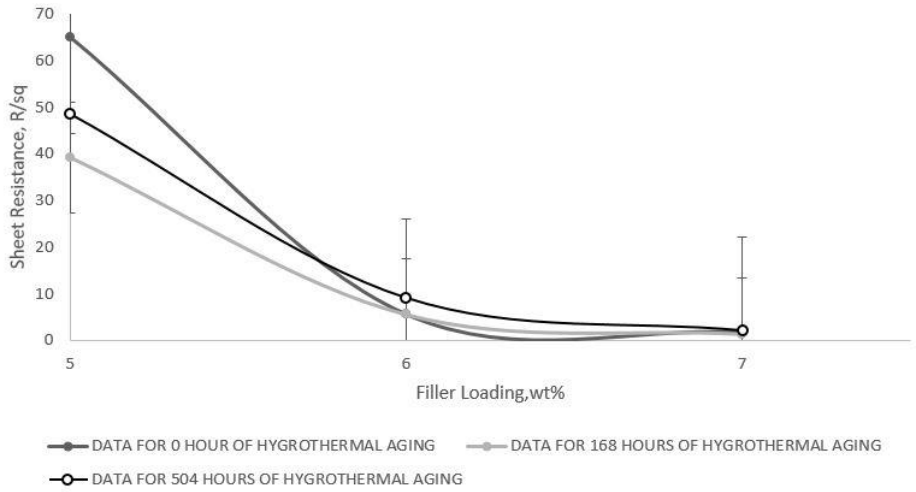

Figure 4.4: Comparison between electrical conductivity for controlled specimen, 168 hours and 504 hours of hygrothermal aging.

As for the results in Figure 4.1, there is a slight decrease in the sheet resistance for 6 wt. % and 7 wt. % filler loadings. The cause of the decreasing is the moisture absorption during the hygrothermal aging which either breaks or construct the conductive path of the conductive filler, while the 5 wt. % filler loading decreases significantly. This shows that the hygrothermal aging process on the 5 wt. % ECA absorbs more moisture compared with the 6 wt. % and 7 wt. % ECA. The mass of the ECA also increases after the hygrothermal aging process for each of the filler loadings. However, in Figure 4.4, the 504 hours specimen increases in resistance of compared to the 168 hours specimen. Due to longer aging time, the moisture absorption also increases as well as disrupting the individual conductive paths in the adhesive and the conductive filler. Thus, it increases the resistivity (Barto, Cinert and Mach, 2011).

The mass difference were done to determine the mass of moisture absorption after the hygrothermal aging process. The diffusion of water molecule into the ECA is more active due to longer aging time. The mass difference of the 168 hours of hygrothermal aging can be seen in Tables 4.2 and 4.3 and 504 hours of hygrothermal aging in Tables 4.4 and 4.5 respectively. The difference in percentage can be seen in Table 4.6.

Table 4.2: Mass of test specimen before and after hygrothermal aging.

Filler Loadings	Mass before 168 hours of Hygrothermal aging, (g)	Mass after 168 hours of Hygrothermal aging, (g)
5 wt.%	12.51	12.61
6 wt.%	12.90	13.00
7 wt.%	13.13	13.23

Table 4.3: Mass difference and percentage difference for 168 hours of hygrothermal aging.

Filler Loadings	Difference in mass (g)	Percentage difference in mass (%)
5 wt.%	0.09	0.76
6 wt.%	0.10	0.79
7 wt.%	0.10	0.76

For the 5 wt. % filler loading, such observation could possibly be due to the act of the water molecules, which aid in developing a conductive path which allows current to flow through which it reduces the sheet resistance of the ECA. Due to the amount of the polymer matrix in the ECA is larger than the conductive filler, the rate of moisture intake is also increases, as reported elsewhere in the literature (Navabizadehrafsanjan, Hoa and Rosca, 2016). The polymer matrix in the ECA also swells due to the moisture intake which increases

the resistivity of the ECA. When the polymer swells, the epoxy expands and the bond length of epoxy were changed where it increases the total volume of the epoxy (Le, Kim and Yoon, 2017). When the volume increases the mass also increases.

Table 4.4: Mass of test specimen before and after hygrothermal aging.

Filler Loadings	Mass before 504 hours of Hygrothermal aging, (g)	Mass after 504 hours of Hygrothermal aging, (g)
5 wt.%	12.94	13.05
6 wt.%	12.92	13.01
7 wt.%	12.95	13.04

Table 4.5: Mass difference and percentage difference for 504 hours of hygrothermal aging.

Filler Loadings	Difference in mass (g)	Percentage difference in mass (%)
5 wt.%	0.11	0.85
6 wt.%	0.09	0.70
7 wt.%	0.09	0.69

Table 4.6: Percentage difference in mass of test specimen between 168 hours and 504 hours of hygrothermal aging.

Filler Loadings	Percentage difference in mass 168 hours of Hygrothermal aging (%)	Percentage difference in mass for 504 of Hygrothermal aging (%)
5 wt.%	0.76	0.85
6 wt.%	0.79	0.70
7 wt.%	0.76	0.69

Table 4.6 shows the difference in mass between 168 hours and 504 hours of hygrothermal aging. As shown in the table above, the longer the aging duration, the higher the moisture intake. At MWCNT filler loading of 5 wt. %, the ECA specimens subjected to 504 hours of hygrothermal aging show a 0.85 % of mass difference before and after aging. It increases for about 0.09 % when compared to the 168 hours of hygrothermal aging specimen. The increase in mass were caused by the polymer content in the ECA, in which the epoxy were in much higher volume than the conductive filler. This results in more water absorption by the polymer matrix. The decrease in mass of the ECA specimens following 504 hours of hygrothermal aging could also be seen at MWCNT filler loading of 7 wt. % where it had a decrease for about 0.07 %. It could be seen that the percentage difference were different with the 5 wt. % of conductive filler where the conductive filler content were much greater than the polymer matrix.

4.3. Effect of Hygrothermal Aging on the Shear Strength of the ECA

For the lap – shear test, three test specimens for each filler loadings named specimen 5 – 0W – A, 5 – 0W – B, and 5 – 0W – C for 5 wt. %, 6 – 0W – A, 6 – 0W – B, and 6 – 0W – C for 6 wt.%, and 7 – 0W – A, 7 – 0W – B, and 7 – 0W – C for 7 wt.%. The test specimen labelled 0W indicates controlled condition specimen. While test specimens labelled 1W and 3W represents specimens for the 168 hours and 504 hours of hygrothermal aging respectively. Tables 4.7 and 4.8 shows the experimental data obtained from the lap shear test, while Figure 4.5 shows the comparison bar chart of the result.

Figure 4.5 shows the controlled and the 168 hours of hygrothermal aging specimen, it shows a decrease in shear stress with each filler loadings. The decrease is due to the amount of polymer resin which acts as the adhesive decreases in each filler loading. Both of the 5 wt.% filler loading has the highest average shear stress because it has the most polymer matrix inside the ECA. This makes the bonds between the conductive filler and the polymer matrix able to transfer load efficiently (Jojibabu et al., 2016). The ECA with filler loading of 6 wt.% and 7 wt.% is poor in load transfer, possibly because of the agglomerates formed at the conductive filler. This makes the movements in the ECA easier to be initiated (Chew et al., 2015). The result from the 168 hours of hygrothermal aging specimen, all the ECA with varying filler loadings exhibit a significant decrease in the shear stress. This could possibly be mainly due to the moisture absorption which makes the conductive filler to loosen towards the polymer matrix and causes them to be detached by developing voids and fracture (Mir and Kumar, 2012).

However, the results following 504 hours of hygrothermal aging of the ECA shows a different trend. The data shows an increases the shear strength in the ECA with MWCNT filler loadings of 6 wt.% and 7 wt.% filler loadings while at much lower filler loading of 5 wt.%, there is a decrease in shear strength due to moisture absorption when compared to the controlled condition and 168 hours specimen. This is possibly due to the agglomeration of the ECA which results in improper distribution between the epoxy and CNT during the mixing process. Other possible reason for such observation is due to polymer swelling, caused by moisture absorption and thermal expansion. The outcome of the polymer swelling results in an increase in the resistance of interconnection and decreasing contact between the surface and ECA.

Table 4.7(a): Lap shear test data.

Properties / Specimen		Maximum Shear Strength (N/mm^2)
Hours	Specimen	
0	5 – 0W – A	10.23
	5 – 0W – B	9.13
	5 – 0W – C	9.30
	Average	9.55
	6 – 0W – A	6.74
	6 – 0W – B	7.75
	6 – 0W – C	7.17
	Average	7.22
	7 – 0W – A	6.50
	7 – 0W – B	6.36
	7 – 0W – C	6.15
	Average	6.34
168	5 – 1W – A	3.31
	5 – 1W – B	3.19
	5 – 0W – C	4.53
	Average	3.68
	6 – 1W – A	3.59
	6 – 1W – B	3.49
	6 – 1W – C	3.34
	Average	3.47

Table 4.7(b): Lap shear test data.

Properties / Specimen		Maximum Shear Strength (N/mm^2)
Hours	Specimen	
168	7 – 1W – A	2.68
	7 – 1W – B	3.23
	7 – 1W – C	2.99
	Average	2.97
504	5 – 3W – A	2.79
	5 – 3W – B	4.51
	5 – 3W – C	3.19
	Average	3.49
	6 – 3W – A	4.08
	6 – 3W – B	5.34
	6 – 3W – C	4.72
	Average	4.72
	7 – 3W – A	3.90
	7 – 3W – B	3.80
	7 – 3W – C	3.66
	Average	3.79

Table 4.8: Average values of shear stress.

Average Shear Strength, N/mm^2			
Week / Filler loadings	0 hours of hygrothermal aging (Controlled specimen)	168 hours of hygrothermal aging	504 hours of hygrothermal aging
5 wt.%	9.55 ± 0.59	3.67 ± 0.74	3.49 ± 0.90
6 wt.%	7.22 ± 0.50	3.47 ± 0.13	4.72 ± 0.63
7 wt.%	6.34 ± 0.18	2.97 ± 0.27	3.79 ± 0.12

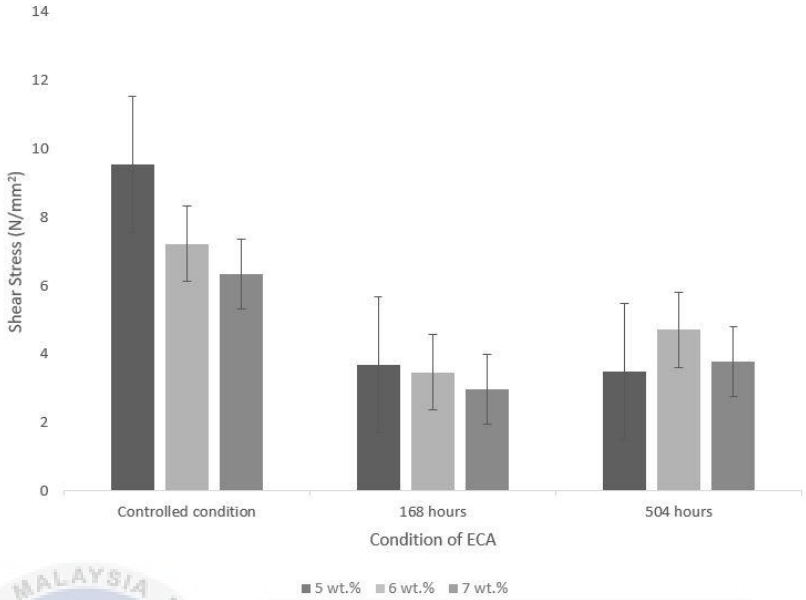

Figure 4.5: Shear strength for the ECA subjected to 0 hours, 168 hours and 504 hours of hygrothermal aging.

Figure 4.6 to 4.9 reveal the presence of fracture following the lap shear test which results in cavitation or voids in the ECA. The SEM images were taken on the ECA top side of the specimen. The voids formed in the ECA occurred during the curing process where the ECA and the aluminium strip that being bonded together may form air bubbles before the curing process. After the curing process were completed, the specified area became a void. The voids result in a decrease in shear strength of the ECA.

Moreover, in Figure 4.10, some traces of swelling or agglomeration in the ECA are also apparent, which decreases the shear strength of the ECA. The agglomerations weakens the interlayer strength between the ECA and the aluminium substrate. In addition, Figure 4.10 also shows a strand of MWCNT isolated from the polymer matrix due to poor technique during mixing process.

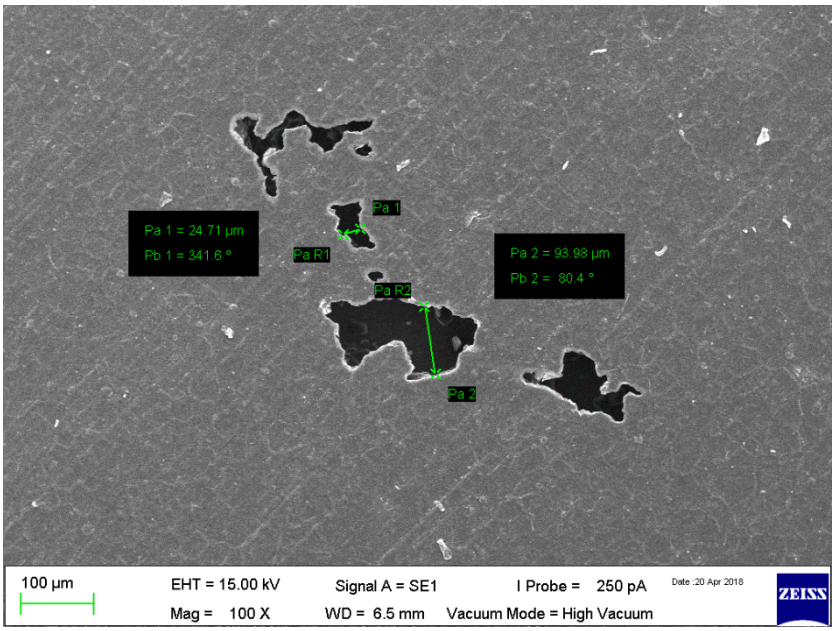

Figure 4.6: SEM image at 100x magnification of ECA with 5 wt. % of MWCNT subjected to 168 hours of hygrothermal aging (Top view).

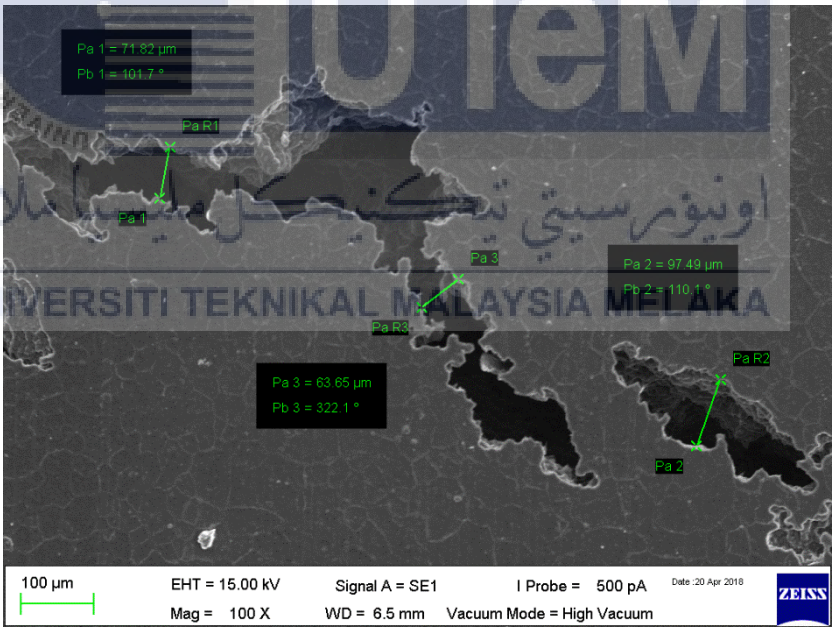

Figure 4.7: SEM image at 100x magnification of ECA with 7 wt. % subjected to 168 hours of hygrothermal aging (Top view).

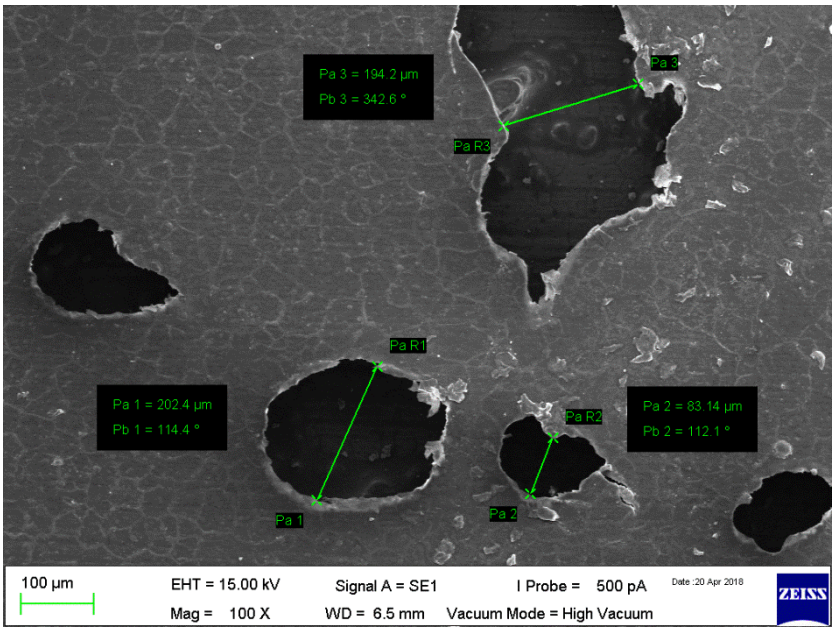

Figure 4.8: SEM image at 100x magnification of ECA with 5 wt. % subjected to 504 hours of hygrothermal aging (Top view).

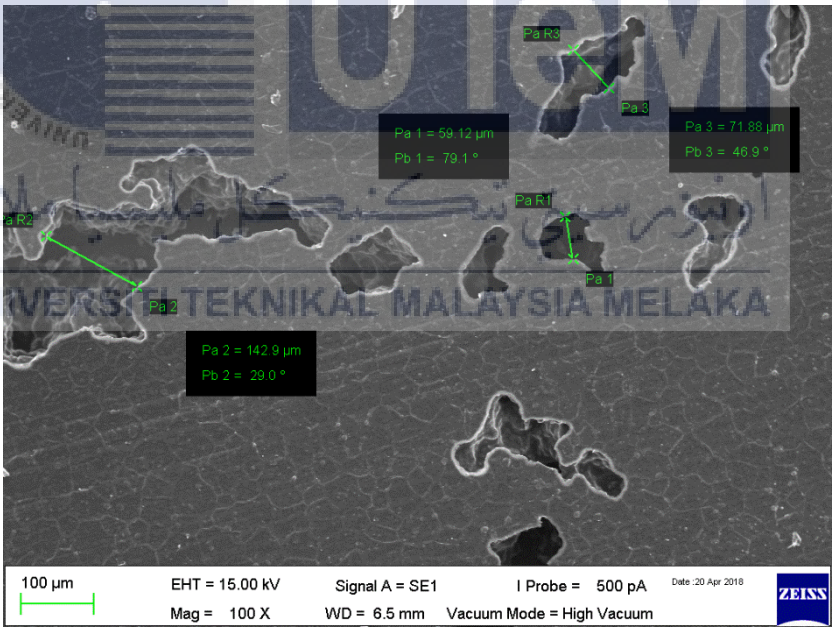

Figure 4.9: SEM image at 100x magnification of ECA with 6 wt. % subjected to 504 hours of hygrothermal aging (Top view).

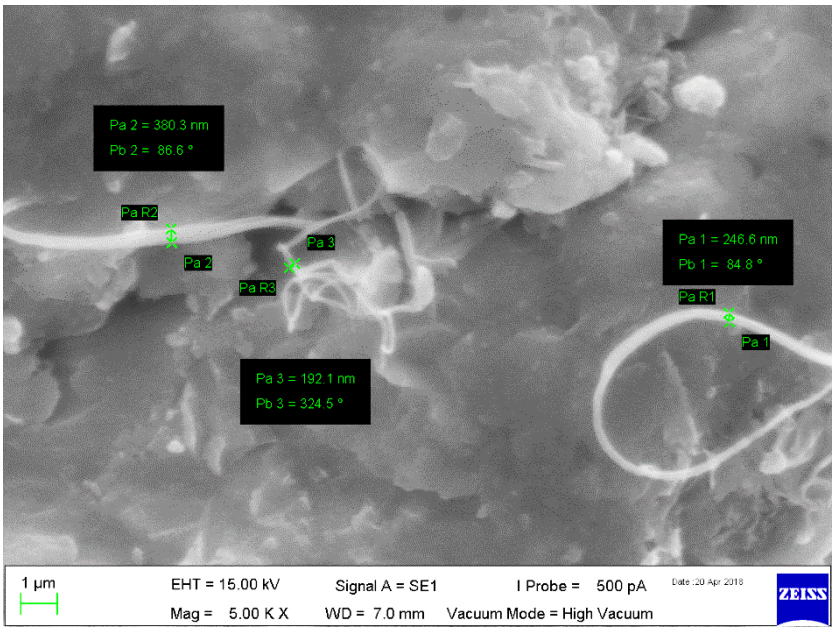

Figure 4.10: SEM image at 5000x magnification of ECA with 7 wt. % subjected to 168 hours of hygrothermal aging (Top view).

4.4. Chapter Summary

The reliability performance of the ECA is reported in terms of the electrical conductivity and shear strength following the hygrothermal aging process. With the presence of moisture, lower contact resistance between the ECA and the substrates are observed, possibly due to the act of the moisture absorption, that aids in developing a conductive path, allowing current to flow through which reduces the sheet resistance of the ECA. Regardless of the amount of MWCNT filler loading (5-7 wt.%), due to moisture attack, voids are created in the epoxy matrix of the ECA, which results in a decrease in the shear strength of the ECA, when the samples were subjected to hygrothermal aging up to 504 hours at 85°C and 85% RH.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1. Conclusion

The reliability performance of the ECA is based on the electrical conductivity and shear strength after being subjected to hygrothermal aging process. The difference between the controlled condition and the following 168 hours and 504 hours of hygrothermal aging process were apparent in the results from electrical conductivity and shear strength.

Table 5.1: Summary of the results obtained from the relevant test conducted for the reliability performance of ECA before and after hygrothermal aging.

Filler loadings (wt. %)	Mass difference after hygrothermal aging (g)			Electrical conductivity (kilo-ohms/square)			Shear strength (N/mm^2)		
	0 hours	168 hours	504 hours	0 hours	168 hours	504 hours	0 hours	168 hours	504 hours
5	0	0.09	0.11	64.96 ± 29.04	39.17 ± 9.16	48.74 ± 11.22	9.55 ± 0.59	3.67 ± 0.74	3.49 ± 0.90
6	0	0.10	0.09	5.61 ± 1.19	5.55 ± 2.08	9.17 ± 1.32	7.22 ± 0.50	3.47 ± 0.13	4.72 ± 0.63
7	0	0.10	0.09	1.75 ± 0.30	1.34 ± 0.14	2.15 ± 0.33	6.34 ± 0.18	2.97 ± 0.27	3.79 ± 0.12

Table 5.1 shows the summarized findings of this study which consists of mass difference after hygrothermal aging, electrical conductivity and shear strength. There were three durations of hygrothermal aging time which is 0 hours, 168 hours and 504 hours of hygrothermal aging. The increase of water intake decreases the electrical conductivity and shear strength. With the presence of moisture, lower contact resistance between the ECA and the substrates are observed, possibly due to the act of the moisture absorption, that aids in developing a conductive path, allowing current to flow through which reduces the sheet resistance of the ECA. However, due to moisture intake, it also disrupts the individual conductive path of the ECA which results in the increase of sheet resistance as reported elsewhere in the literature.

However, dissimilar trend is found for the shear strength of the ECA. Regardless of the amount of MWCNT filler loading (5-7 wt.%), due to moisture attack, voids are created in the epoxy matrix of the ECA, which results in a decrease in the shear strength of the ECA, when the samples were subjected to 168 hours and 504 hours of hygrothermal aging at 85°C and 85% RH. The trend changes at the ECA after being aged for 504 hours. The 6 wt.% and 7 wt.% of MWCNT filler loading shows an increase of shear strength, due to improper mixing process of the ECA while the 5 wt. % shows a decrease in shear strength. Nevertheless, all specimen shows the formation of voids in the epoxy matrix of the ECA, which contributes in the decrease in the shear strength of the ECA.

5.2. Recommendation for future works

The aim of using the ECA is for it to be considered an environment-friendly interconnection technique by replacing the more harmful lead solder that nowadays being used, as one of the two potential alternatives. However, the ECA that being developed were not fully utilised since the mixture of polymer matrix and the conductive filler inside the ECA were affected by the changes in temperature and humidity. As the humidity increases, the moisture content in the ECA increases due to moisture absorption. Where it results in a decrease in their mechanical properties while increasing their electrical properties.

The ECA fabrication through the solution mixing process could be further improved since the formation of ECA differs where it is not consistent for every ECA produced. Proper measurement of the amount of the polymer matrix and conductive filler should be more precise to avoid defects in the ECA produced. Also, during the printing process, it is much favourable to use a stencil so the ECA could be spread more consistent for both mechanical and electrical testing.

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