

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

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UNIVERSITI TEKNIKAL MALAYSIA MELAKA

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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 solders. 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 rintangan ECA 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 1×10^{-4} .cm, while 6×10^{-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 solders. 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 ($< 0.001 \text{ ohm-cm}$) 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														