UNIVERSITI TEKNIKAL MALAYSIA MELAKA

SYNERGISTIC EFFECT OF MICRO-NANO FILLERS ON RELIABILITY PERFORMANCE OF HYBRID ELECTRICALLY CONDUCTIVE ADHESIVE

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Faculty of Mechanical Technology and Engineering

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A thesis submitted
in fulfilment of the requirements for the degree of Master of Science
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DECLARATION

I declare that this thesis entitled "Synergistic Effect of Micro-Nano Fillers on Reliability Performance of Hybrid Electrically Conductive Adhesive" is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in the candidature of any other degree.

Signature : ....

Name : Zainalfirdaus Bin Adnan

Date : 29.05.2024

APPROVAL

I hereby declare that I have read this thesis and in my opinion, this thesis is sufficient in terms of scope and quality for the award of Master of Science in Mechanical Engineering.

Signature : ....

Supervisor Name : Profesor Madya Dr. Siti Hajar Binti Sheikh Md. Fadzullah

Date : 01.07.2024

DEDICATION

To my beloved mother and father

ABSTRACT

In today's digital age, the escalating demand for efficient electronic technologies contrasts starkly with the global issue of electronic waste (e-waste) containing hazardous lead (Pb) solder. Consequently, there is a critical need for alternative materials in electronics interconnection technology, notably electrically conductive adhesives (ECA) based on polymer composites. Despite significant advancements in hybridizing various filler types in ECAs, systematic demonstration of material component optimization remains lacking. This gap jeopardizes the reliability performance of hybrid ECAs (HECA) due to polymer degradation under extreme environmental conditions of temperature and humidity. This research aims to establish an optimal HECA formulation with a low percolation threshold and enhanced reliability performance under extreme conditions, thereby investigating the failure mechanisms of HECA. The study employs a Design of Experiment (DOE) approach incorporating Analysis of Variance (ANOVA) and Response Surface Methodology (RSM) to optimize the formulation. Electrical and mechanical characterizations were conducted using a four-point probe with a Jandel RM3000+ test unit and a Hengzhun HZ-1003 universal testing machine (UTM), respectively. Samples underwent evaluation both before and after hygrothermal ageing at 85% relative humidity and 85°C for up to 504 hours. Morphological analysis utilized Scanning Electron Microscopy (SEM) and Field-Emission Scanning Electron Microscopy (FESEM). Rheological and thermal analyses were performed using an Anton Parr Rheometer and TGA-DSC1 analyser, respectively. The RSM indicated that the optimal HECA formulation achieved a 0.07 AgMF-MWCNT ratio, demonstrating enhanced electrical conductivity with a lower percolation threshold. Notably, no electrical degradation was observed over the ageing period for ratios of 0.07 and 0.17, underscoring their resilience under specified conditions. Despite epoxy cracking, micro-void formation, and delamination causing degradation, the HECA exhibited satisfactory mechanical reliability. Overall, the HECA formulation established at a 0.17 ratio in this study demonstrates superior electrical and mechanical reliability, making it well-suited for electronic packaging applications.

KESAN SINERGISTIK PENGISI MIKRO-NANO TERHADAP PRESTASI KEBOLEHPERCAYAAN PELEKAT KONDUKTIF ELEKTRIK HIBRID

ABSTRAK

Di era digitalisasi ini, permintaan untuk teknologi elektronik yang cekap dan canggih meningkat, sementara sisa elektronik (e-waste) yang mengandungi timah (Pb) penuh beracun menjadi isu global. Oleh itu, terdapat keperluan mendesak untuk bahan alternatif dalam teknologi penyambungan elektronik, terutamanya pelekat konduktif elektrik berdasarkan komposit polimer yang lebih dikenali sebagai pelekat konduktif elektrik (ECA). Walaupun banyak kajian membuktikan peningkatan luar biasa dalam hibridisasi pelbagai jenis pengisi dalam ECA, optimisasi komponen bahan dalam komposit tidak dipamerkan secara sistematik. Ini mengancam prestasi kebolehpercayaan hibrid ECA (HECA) kerana degradasi polimer di bawah keadaan suhu dan kelembapan ekstrem. Kajian ini bertujuan untuk menetapkan formulasi HECA optimum dengan ambang perkolasi yang rendah dan prestasi kebolehpercayaan yang diperkukuhkan di bawah keadaan ekstrem, serta menyiasat mekanisme kegagalan HECA. Kajian ini menggunakan pendekatan Reka Bentuk Eksperimen (DOE) yang menggabungkan Analisis Varians (ANOVA) dan Metodologi Permukaan Tindak Balas (RSM) untuk mengoptimumkan formulasi. Pencirian elektrik dan mekanikal dilakukan menggunakan probe empat titik dengan unit ujian Jandel RM3000+ dan mesin ujian serba guna Hengzhun HZ-1003, masing-masing. Sampel dinilai sebelum dan selepas penuaan hidrotermal pada kelembapan relatif 85% dan 85°C selama hingga 504 jam. Analisis morfologi menggunakan Mikroskop Elektron Pemindahan Medan (SEM) dan Mikroskop Elektron Pemindahan Medan (FESEM). Analisis reologi dan termal dilakukan menggunakan Rheometer Anton Parr dan analis TGA-DSC1, masing-masing. RSM menunjukkan formulasi HECA optimum diperoleh pada nisbah 0.07 AgMF-MWCNT, menunjukkan pengaliran elektrik yang dipertingkatkan dengan ambang perkolasi yang lebih rendah. Tidak kurang pentingnya, tidak ada degradasi elektrik yang diperhatikan sepanjang tempoh penuaan untuk nisbah 0.07 dan 0.17, menunjukkan keupayaan tahan degradasi di bawah keadaan yang dinyatakan. Walaupun retakan epoksi, pembentukan mikro-void, dan delaminasi menyebabkan degradasi, HECA menunjukkan kebolehpercayaan mekanikal yang memuaskan. Secara keseluruhannya, formulasi HECA yang ditetapkan pada nisbah 0.17 dalam kajian ini menunjukkan prestasi kebolehpercayaan elektrik dan mekanikal yang unggul, menjadikannya sesuai untuk aplikasi pembungkusan elektronik.

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LIST OF ABBREVIATIONS

1D	-	One-dimensional
2D	-	Two-dimensional
3D	-	Three-dimensional
4T	-	Four-point probe
ACA	-	Anisotropic conductive adhesive
Ag	-	Silver
AgDn	-	Silver dendrites
AgMF	-	Silver micro-flakes
AgNP	-	Silver nanoparticle
ANOVA	-	Analysis of variance
ASTM	-	American Society for Testing and Material
BN	-	Boron nitride
BNNP	-	Boron nitride nanoparticle
BBD	-	Box-Behnken design
CCD	-	Central composite design
CLASS	-	Classification, Labelling and Safety Data Sheet of Hazardous Chemical
CMF	-	Common failure

CNF	-	Corner failure
CNH	-	Carbon nanohorns
CNT	-	Carbon nanotube
CSV	-	Comma-separated variable
Cu	-	Copper
CVD	-	Chemical vapour deposition
DC	-	Direct Current
DGEBA	-	Bisphenol-A diglycidyl ether
DOE	-	Design of Experiment
DSC	-	Differential Scanning Calorimetry
DWCNT	-	Double-walled carbon nanotube
ECA	-	Electrically conductive adhesive
EG	-	Expanded graphite
EMI	-	Electromagnetic interference
FESEM	-	Field-Emission Scanning Electron Microscopy
f-GNP	-	Functionalized graphene nanoplatelets
GNP	-	Graphene nanoplatelets
Gr	-	Graphite
HECA	-	Hybrid electrically conductive adhesive
ICA	-	Isotropic conductive adhesive
ICP	-	Intrinsically conducting polymers
MWCNT	-	Multi-walled carbon nanotube

NiBN	-	Nickel boron nitride
OMD	-	Optimal mixture design
PANI	-	Polyaniline
Pb	-	Lead
PCB	-	Printed circuit board
Ppy	-	Polypyrrole
RH	-	Relative Humidity
RSM	-	Response surface methodology
RT	-	Room temperature
SEM	-	Scanning Electron Microscopy
SiC	-	Silicon carbide
SIF	-	Side failure
SMT	-	Surface-mount technology
Sn	-	Tin
SWCNT	-	Single-walled carbon nanotube
TGA	-	Thermogravitational analysis
UTM	-	Universal testing machine

LIST OF SYMBOLS

A	-	Area of overlap
$E_{wt.\%}$	-	Weight percentage of epoxy
F_{max}	-	Maximum tensile force
$F_{wt.\%}$	-	Weight percentage of filler
G	-	Correction Factor
$H_{wt.\%}$	-	Weight percentage of hardener
I	-	Current
l	-	Sample's length
$M_{wt.\%}$	-	Weight percentage of matrix
R	-	Electrical resistance
R_0	-	Initial electrical resistance
R^2	-	Multiple correlation coefficient
T	-	Current temperature
T_d	-	Degradation temperature
T_g	-	Glass transition temperature
t_s	-	Thickness of the sample
V	-	Voltage
V	-	Voltage

V_f	-	Volume percentage of filler
V_m	-	Volume percentage of matrix
w	-	Sample's width
W_c	-	Weights of composite
W_e	-	Weight of epoxy
W_f	-	Weights of filler
W_{f1}	-	Weight of first hybrid filler
W_{f2}	-	Weight of second hybrid filler
W_h	-	Weight of hardener
W_m	-	Weights of matrix
x	-	Model coefficient of estimates
X_c	-	Density of composite
X_f	-	Density of filler
X_m	-	Density of matrix
Y1	-	Volume resistivity response (in Scheffé equation)
ρ	-	Volume resistivity
ρ_0	-	Initial volume resistivity
τ	-	Lap shear strength
τ_0	-	Initial lap shear strength

LIST OF PUBLICATIONS

Adnan, Z., Fadzullah, S. H. S. M., Omar, G., Mustafa, Z., Ramli, M. B., Razali, N., and Kamarolzaman, A. A., 2023. Hygrothermal Durability of Hybrid Electrically Conductive Adhesive at Optimal Micro-nano Fillers Ratio. *Proceedings of the 9th International Conference and Exhibition on Sustainable Energy and Advanced Materials*, Lecture Notes in Mechanical Engineering. Springer, Singapore. 179-183.

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CHAPTER 1

INTRODUCTION

1.1 Research background

Conductive silver paste is the most sought-after material in the electronic industry for making circuits and terminal assembling. Despite the highly conductive benefit carried by silver as a conductive filler, however, the applications of an electrically conductive adhesive (ECA) are at a crucial state due to the high amount of silver needed, as demonstrated by many researchers in attempts to achieve excellent electrical properties of the ECA (Marcq et al., 2011; Cui and Du, 2013; Ji et al., 2015; Luo et al., 2016; Zhou et al., 2016), which conceives a need to overlook for metal and non-metal filler combination in ECA. The idea of hybridizing fillers in ECA has disclosed a more comprehensive range of studies regarding interface dissociation and synergistic effect, including various incorporation of metal and non-metal filler materials. However, numerous hybrid electrically conductive adhesive (HECA) projects achieving high electrical performance does not actually provides an optimum formulation (Amoli et al., 2015). Moreover, since HECA comprises multiple filler elements, the distribution of the material components is rather crucial due to the incorporation of different particle size, geometry and aspect ratio that are responsible for the formation of electrical network inside the HECA.

There are many works attempted in monitoring and enhancing different filler elements' dispersion behaviour. For instance, using dispersant agent for carbon nanotube (CNT) (Marcq et al., 2011), direct mixing and sonication