UNIVERSITI TEKNIKAL MALAYSIA MELAKA

THE NEW FORMULATION OF HYBRIDIZATION GNP/AG HIGHLY THERMAL CONDUCTIVITY

AMEERUZ KAMAL BIN AB WAHID

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**A thesis submitted
in fulfillment of the requirements for the degree of Doctor of Philosophy**

Faculty of Mechanical Engineering

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2023

DECLARATION

I declare that this thesis entitled "The New Formulation of Hybridization GNP/Ag Highly Thermal Conductivity" is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature : ....

Name : Ameeruz Kamal bin Ab Wahid

Date : 23/3/2023

APPROVAL

I hereby declare that I have read this thesis and in my opinion this thesis is sufficient in terms of scope and quality as a partial fulfilment of Doctor of Philosophy.

Signature : ....

Supervisor Name : Assoc. Prof. Ir. Ts. Dr. Mohd Azli Bin Salim

Date : 23/03/2023

DEDICATION

To my beloved mother and father and

most importantly my beloved wife.

ABSTRACT

Flexible electronics have become highly desirable for a variety of applications because of their low cost, lightweight characteristics, and flexibility. However, one of the challenges in developing a flexible electronic is depending on a suitable ink material formulation. The graphene nanoplatelets (GNP) sheets are inherently stacked together because of the strong van der Waals interactions between adjacent layers. Silver is the most popular metallic material due to its excellent electrical conductivity and chemical stability. This thesis aimed to investigate the characteristics and applications of graphene and silver conductive ink mixed with an organic solvent, which had low resistivity, high flexibility, and high thermal conductivity. Four strategies were adopted to develop such inks. The first was to formulate a new formulation of conductive ink. The second was to characterise the electrical, mechanical, and thermal behaviour of a new formulation of conductive ink. The third was to simulate the proposed formulation characteristics using the finite element method (FEM). The fourth was to validate the thermal conductivity and resistivity relationship of the new formulation. To evaluate the performance in terms of electrical, mechanical, and thermal conductivity, this research firstly developed a GNP baseline using GNP as the only conductive filler with epoxy. Finite element analysis (FEA) was used to validate the GNP baseline stretchability and thermal conductivity formulations. Following that, research was carried out on the formulation and performance of GNP hybrids using GNP, silver flakes (Ag), and silver acetate (SA) as conductive fillers mixed with organic solvents. After the twisting and bending test, the GNP hybrid formulation reliability was evaluated. The use of GNP and silver conductive ink in combination with an organic solvent result in low resistance, high flexibility, and high thermal conductivity. GNP baseline and GNP hybrid formulations were compared in terms of electrical, mechanical, and thermal conductivity. The finding that the resistivity value of 0.1 wt.% GNP of GNP hybrid is 2.35×10^{-7} Ω.m, which is significantly lower than the resistivity value of GNP baseline, 0.249 Ω.m, demonstrates that mixing GNP with silver improves the performance of electrical conductivity. The GNP hybrid had the highest shear and thermal conductivity values of 1.98 MPa and 367.28 W/m.K. The GNP hybrid formulation could minimise the amount of silver used in the production of conductive ink. The GNP hybrid also improved conductive ink flexibility and thermal conductivity, which will have an impact on circuit manufacturing in the electronics industry.

FORMULASI BAHARU PENGHIBRIDAN GNP/AG KEBERALIRAN HABA TINGGI

ABSTRAK

Elektronik fleksibel telah menjadi sangat diperlukan untuk pelbagai aplikasi kerana kosnya yang rendah, ciri yang ringan dan fleksibiliti. Walau bagaimanapun, salah satu cabaran dalam membangunkan elektronik yang fleksibel adalah bergantung kepada formulasi bahan dakwat yang sesuai. Kepingan nanoplatelet graphene (GNP) sememangnya disusun bersama kerana interaksi Van Der Waals yang kuat antara lapisan-lapisannya. Perak adalah bahan logam yang paling popular kerana kekonduksian elektrik yang sangat baik dan mempunyai kestabilan kimia. Tesis ini bertujuan untuk mengkaji ciri-ciri dan aplikasi dakwat konduktif graphene dan perak yang dicampur dengan pelarut organik, mempunyai kerintangan rendah, kelenturan tinggi, dan kekonduksian haba yang tinggi. Empat strategi telah digunakan untuk membangunkan dakwat tersebut. Yang pertama adalah untuk merumuskan formulasi baru dakwat konduktif. Yang kedua ialah untuk mencirikan perlakuan elektrik, mekanikal dan haba bagi rumusan baharu dakwat konduktif. Yang ketiga adalah untuk mensimulasikan ciri-ciri formulasi yang dicadangkan dengan menggunakan kaedah unsur terhingga (FEM). Keempat adalah untuk mengesahkan hubungan kekonduksian haba dan kerintangan formulasi baru. Untuk menilai prestasi dari segi kekonduksian elektrik, mekanikal dan haba, penyelidikan ini mula-mula membangunkan garis dasar GNP yang menggunakan GNP sebagai satu-satunya pengisi konduktif dengan epoksi. Analisis unsur terhingga (FEA) telah digunakan untuk mengesahkan kebolehregangan garis dasar GNP dan formulasi kekonduksian haba. Berikutan itu, penyelidikan telah dijalankan terhadap perumusan dan prestasi hibrid GNP dengan menggunakan GNP, plat perak (Ag), dan asetat perak (SA) sebagai pengisi konduktif yang dicampur dengan pelarut organik. Selepas ujian memusing dan membengkok, kebolehpercayaan formulasi hibrid GNP dinilai. Penggunaan GNP dan dakwat konduktif perak dalam kombinasi dengan pelarut organik menghasilkan rintangan yang rendah, fleksibiliti tinggi dan kekonduksian haba yang tinggi. Garis dasar GNP dan formulasi hibrid GNP dibandingkan dari segi kekonduksian elektrik, mekanikal dan haba. Dapatan bahawa nilai kerintangan 0.1 wt.% GNP daripada hibrid GNP ialah 2.35 x 10^{-7} *Ω.m, yang jauh lebih rendah daripada nilai kerintangan garis dasar GNP, 0.249 Ω.m, menunjukkan bahawa campuran antara GNP dengan perak dapat meningkatkan prestasi kekonduksian elektrik. Hibrid GNP mempunyai nilai ricih dan kekonduksian haba yang tinggi iaitu 1.98 MPa dan 367.28 W/m.K. Formulasi hibrid GNP boleh meminimumkan jumlah perak yang digunakan dalam pengeluaran dakwat konduktif. Hibrid GNP juga dapat meningkatkan kelenturan dakwat konduktif dan kekonduksian haba, seterusnya akan memberi kesan kepada pembuatan litar dalam industri elektronik.*

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LIST OF ABBREVIATIONS

PCB	Printed circuit boards
GNP	Graphene nanoplatelets
Ag	Silver flake
SA	Silver acetate
FEA	Finite element analysis
FEM	Finite element method
CNT	Carbon nanotube
ECG	Diagnostic electrocardiogram
PTF	Polymer thick films
POC	Point-of-care
EC	Ethyl cellulose
PI	Polyimide
NPM	N-methyl-2-pyrrolidone
DMF	Dimethylformamide
ECA	Electrical conductive adhesives
SEM	Scanning Electron Microscopy
EDX	Energy Dispersive X-ray
SCI	Stretchable conductive ink
Au	Aurum (Gold)
Pt	Platinum

AgNP	Silver nanoparticles
AgNW	Silver nanowires
PEDOT:PSS	Poly (3,4-ethylenedioxythiophene)-poly (styrenesulfonate)
PAN	Polyaniline
PET	Polyethylene terephthalate
PVP	Polyvinylpyrrolidone
EC	Ethyl cellulose
TCPM	Thermally conductive polymer materials
GO	Graphene oxides
rGO	Reduced graphene oxides
IoT	Internet of things
0D	Zero dimensional
1D	One-dimensional
2D	Two-dimensional
3D	Three-dimensional
FLG	Few-layer graphene
EMI	Electromagnetic interference
NaCl	Sodium chloride
NH_2	Nitrenium ion (amine)
OH	Hydroxyl
D_k	Dielectric constants
T_g	Glass-transition temperatures
T_d	Thermal decomposition temperatures
SSWNT	Single-walled carbon nanotubes
CPI	Chemical process industries

Te-PEDOT:PSS	Thermoelectric poly (3,4-ethylenedioxythiophene) polystyrene sulfonate
CFD	Computational fluid dynamics
PDMS	Polydimethylsiloxane
Au-Sn	Gold-tin
Au-Ge	Gold-germanium
AgIn	Silver-indium
MT	Mori-Tanaka
Gr-Ag	Graphene - silver
RMC	Recharging microchannel
FVM	Finite volume method
GNP-Ag	Graphene nanoplatelets and silver
xGnP H-5	GNP powder
SMD	Surface-mount device
SiC	Silicon carbide
TPC	Tetrahedrons patch confirming
SAC	Tin/silver/copper
wt.%	Weight percentage
h_{max}	Maximum depth of the indentation
F_{max}	Indentation maximum force
R_{Th}	Thermal resistance
R_m	Material resistance
UTM	Universal Testing Machine
SPP	Silk-sericin photoresist