CHARACTERISATION OF HYBRID GRAPHENE NANOPLATELETS WITH FUNCTIONALISED BORON NITRIDE AS THERMAL INTERFACE MATERIAL

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DECLARATION

I declare that this thesis entitled "Characterisation of Hybrid Graphene Nanoplatelets with Functionalised Boron Nitride as Thermal Interface Material" 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 : **SOLEHAH BINTI JASMEE**

Date : **19/10/2023**

APPROVAL

I hereby declare that I have read this thesis and in my opinion, this thesis is sufficient interms of scope and quality for the award of Doctor of Philosophy.

Signature : .....

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Date : **19/10/2023**

DEDICATION

To my beloved mother, father, siblings and my best friend.

ABSTRACT

Conductive fillers with a polymer matrix or Thermal Conductive Adhesive (TCA) have met most of the ideal criteria due to being ecologically friendly, reliable, and affordable. However, this effort may not yet lead to a significant technology breakthrough due to the low thermal conductivity of the polymer and the high filler loading used in a single-filled composite. Hence, it has heightened the need for hybrid fillers with extraordinary properties, such as boron nitride (BN) and graphene nanoplatelets (GNPs), at varying sizes with the incorporation of silane-modified BN (fBN) to improve composite's properties. Yet, the affirmative review shows very few studies have reported on the hybrid composite GNPs with fBN at different filler sizes and lower filler loading (<20 wt.%), as well as its suitable processing parameter and important characterization such as rheological, water absorption, and adhesion strength with and without moisture. Thus, this study seeks to determine the curing time, temperature, and mixing method of the composite, percolation threshold of thermal conductivity of single composite GNPs, BN, and fBN, and thermal conductivity of hybridized GNPs with BN or fBN below filler loading of single composite and also to characterize the rheological properties, adhesion strength (single lap joint (SLJ) approach) and failure mechanism of hybridized GNPs with BN or fBN composite at ambient and 100% humidity conditions. The hybrid composites were prepared by hybridizing different ratios of GNPs (5 µm and 15 µm) with BN or fBN (10 µm) that were modified using silane coupling agents (KH550 and KH560). KD2 Pro and Anton Paar Rheometer MCR 72 were used to measure thermal conductivity and rheological properties, while the SLJ test was prepared and accordance with ASTM D1002, ASTM D 1151, ASTM D 5229/D 5229M, and EN ISO 10365 standards. The result indicates that hybrid GNPs15/fBN_KH560 composites exhibit the highest thermal conductivity (1.16 ± 0.02 W/mK) at a GNPs15 ratio 0.75. However, hybrid GNP5 composites have a higher shear strength than hybrid GNP15 composites at all GNPs ratios, with the highest values reported being 1.20 ±0.10 MPa and 1.14 ± 0.15 MPa at GNPs ratio of 0.75 with fBN_KH560. The bigger filler size of GNPs may help to transfer heat, but smaller fillers are preferable to transfer stress. The failure mechanisms like plastic void growth, crack deflection, particle debonding, and pull-out mechanism also help promote cohesive failure in the hybrid composite. Hybrid GNP15 composites have high shear strength and absorb more water than hybrid GNP5 but can sustain for 30 days before reaching equilibrium. The larger filler particle fills the gaps between the epoxy, reducing the formation of cracks or voids. Nevertheless, moisture still caused the shear strength of composites to reduce (0.39 ± 0.05 MPa). In summary, the properties of hybrid composites containing fBN_KH560, especially at GNPs ratio of 0.75, show outstanding properties due to high compatibility between fBN_KH560 and the polymer matrix, proven by the lowest measured contact angle, and Hansen solubility parameter difference, well-dispersed filler, and better wetting properties (lowest viscosities varies shear rate) in rheological analysis.

PENCIRIAN HIBRID GRAFEN NANOPLATLETS BERFUNGSIKAN BORON NITRIDA SEBAGAI BAHAN ANTARA MUKA TERMA

ABSTRAK

Pengisi konduktif dengan matriks polimer atau Pelekat Konduktif Terma (TCA) telah memenuhi kebanyakan kriteria ideal seperti mesra alam, boleh dipercayai dan berpatutan. Namun, usaha ini belum mampu membawa kepada kejayaan teknologi yang ketara disebabkan oleh kekonduksian haba polimer yang masih rendah dan pemuatan pengisi yang tinggi dalam sesuatu komposit. Oleh itu, ia telah meningkatkan keperluan untuk menghibrid pengisi yang mempunyai sifat yang luar biasa seperti boron nitrida (BN) dan grafen nanoplatelets (GNPs), pada saiz yang berbeza dan dengan penggabungan BN yand diubahsuai (fBN) dengan silane untuk meningkatkan ciri-ciri komposit. Namun, semakan afirmatif menunjukkan sangat sedikit kajian melaporkan tentang GNP komposit hibrid dengan fBN pada saiz pengisi yang berbeza dan pemuatan pengisi yang rendah (<20 berat.%), serta parameter pemprosesan yang sesuai dan pencirian penting seperti reologi, penyerapan air, dan kekuatan lekatan dengan dan tanpa kelembapan. Justeru, kajian ini dijalnkan untuk menentukan tempoh pengesetan, suhu pengesetan, dan kaedah campuran komposit yang sesuai, ambang perkolasi keberkondukton terma bagi komposit tunggal GNPs, BN, dan fBN juga apabila dihibridkan dibawah pengisi pemuatan komposit tunggal dan juga untuk mencirikan sifat reologi, kekuatan lekatan (pendekatan ujian sendi tunggal (SLJ)) dan mekanisme kegagalan hibrid GNPs dengan BN atau fBN pada suhu sekeliling dan 100% kelembapan. Komposit hibrid disediakan dengan mengacukan berbeza nisbah antara GNPs (5µm dan 15µm) dengan BN atau fBN (10 µm) yang diubah suai dengan ejen gandingan silane (KH550 dan KH560). KD2 PRo dan Anton Paar Rheometer MCR 72 digunakan untuk mengukur kekonduksian terma dan sifat reologi manakala SLJ disediakan berdasarkan ASTM D1002 ASTM D 1151, ASTM D 5229/D 5229M dan EN ISO 10365. Hasilnya menunjukkan bahawa hibrid komposit GNPs15/fBN_KH560 mempamerkan kekonduksian terma tertinggi (1.16 ± 0.02 W/mK) pada nisbah GNPs15 0.75. Namun, alisis kekuatan ricih menunjukkan hibrid komposit GNPs5 lebih tinggi berbanding hibrid komposit GNPs15 pada semua nisbah GNPs dengan nilai tertinggi adalah 1.20 ±0.10 MPa dan 1.14 ± 0.15 MPa pada GNPs ratio of 0.75 dengan fBN_KH560. Saiz pengisi GNP yang lebih besar membantu memindahkan haba, tetapi pengisi yang lebih kecil adalah lebih baik untuk memindahkan tekanan. Mekanisme kegagalan seperti pertumbuhan lompang plastik, pesongan retak, penyahikatan zarah, dan mekanisme tarik keluar juga membantu menggalakkan kegagalan kohesi dalam komposit hibrid. Walau bagaimanapun, komposit GNP15 hibrid dilihat menyerap lebih banyak air daripada GNP5 hibrid tetapi mampu bertahan sehingga 30 hari sebelum mencapai keseimbangan. Pengisi yang lebih besar mengisi jurang antara epoksi, mengurangkan pembentukan retakan atau lompang. Namun, lembapan masih menyebabkan kekuatan ricih komposit berkurangan (0.39 ± 0.05 MPa). Ringkasnya, komposit hibrid yang mengandungi fBN terutamnaya pada nisbah GNPs 0.75 menunjukkan ciri yang menonjol kerana keserasian yang tinggi antara fBN_KH560 dan matriks polimer, dibuktikan oleh sudut sentuhan terukur dan perbezaan parameter keterlarutan Hansen yang rendah, pengisi tersebar dengan baik, dan sifat pembasahan yang lebih baik (kelikatan terendah pada kadar ricih berbeza) dalam analisis reologi.

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LIST OF ABBREVIATIONS

0D	-	Zero-dimensional
1D	-	One-dimensional
2D	-	Two-dimensional
3D	-	Three-dimensional
AF	-	Adhesion failure
AEW	-	amine equivalent weight
APCF	-	Adhesion and cohesion failure with peel
ATR-FTIR	-	Attenuated total reflection –Fourier transform infrared spectroscopy
BN	-	Boron nitride
BS	-	Bath sonication
BeO	-	Berium oxide
BLT	-	Bond line thickness
C_2H_5OH	-	Ethanol
CB	-	Carbon black
CF	-	Carbon fibre
CF	-	Cohesive failure
Cu	-	Copper
CFS	-	Cohesive substrate failure
CNT	-	Carbon nanotube
C-O-C	-	Epoxide group
CPU	-	Central processing unit
CTE	-	Coefficient of thermal expansion
CVD	-	Chemical vapour deposition
DF	-	Delamination failure
DBC	-	Direct bound copper
DSC	-	Differential scanning calorimetry
EC	-	Electrical conductivity
ECA	-	Electrically conductive adhesives
EE	-	Energy-efficient
EEW	-	Epoxy equivalent weight
EV	-	Electric vehicles
fBN	-	Functionalised BN
FLG	-	Few layer graphene
GO	-	Graphene Oxide
GNPs	-	Graphene nanoplatelets
HDPE	-	High density polyethene
HEVs	-	Hybrid electrical vehicles
HSP	-	Hansen solubility parameter
ICs	-	Integrated circuits
IGBTs	-	insulated gate bipolar transistors
IHS	-	Integrated heat spreader
ITR	-	Interface thermal resistance
KH550	-	3-Aminopropyltriethoxysilane

KH560	-	3-Glycidoxypropyl-trimethoxysilane
LED	-	Light-emitting diode
LTJT	-	low temperature joining
MLG	-	Multi-layer graphene
MOSFETs	-	Metal–oxide–semiconductor field-effect transistors
MWCNT	-	Multi-wall carbon nanotube
NaOH	-	Sodium hydroxide
Ni	-	Nickel
-OH	-	Hydroxide functional groups
OLED	-	Omitted- light emitting diode
PDMS	-	Polydimethylsiloxane
Pb	-	Lead
PCM	-	Phase change materials
PCT	-	phase change temperature
PCTM	-	Planetary Centrifugal Thinky Mixer
RoHS	-	Restriction of hazardous substances directive
Sn	-	Tin
SF	-	substrate failure
SEM	-	Scanning electron microscopy
SET	-	Single-electron transistor
SiC	-	Silica carbide
SLJ	-	Single lap joint
SLG	-	Single layer graphene
SSA	-	Specific surface area
TBR	-	Thermal boundary resistance
TC	-	Thermal conductivity
TCA	-	Thermally conductive adhesives
TCE	-	Thermal conductivity enhancement
TCR		Thermal contact resistance
TIMs	-	Thermal interface materials
UH	-	Ultrasonic homogenizer
UTM	-	Universal tensile machine