

**ENHANCEMENT OF MECHANICAL, THERMAL AND
DIELECTRIC PROPERTIES OF HYBRID CARBON
NANOTUBES AND ALUMINA IN EPOXY NANOCOMPOSITES**

by

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TABLE OF CONTENTS

ACKNOWLEDGEMENTS	i
TABLE OF CONTENTS	ii
LIST OF TABLES	vi
LIST OF FIGURES	viii
LIST OF ABBREVIATIONS	xiii
LIST OF SYMBOLS	xiv
ABSTRAK	xv
ABSTRACT	xvii
1 INTRODUCTION	
1.1 Overview	1
1.2 Problem Statement	5
1.3 Research Objective	6
1.4 Scope of the research	7
2 LITERATURE REVIEW	
2.1 Carbon nanotubes	9
2.1.1 History and background	9
2.1.2 Structure	10
2.1.3 Properties of carbon nanotubes	15
2.1.3.1 Stability	16
2.1.3.2 Mechanical properties	16
2.1.3.3 Electrical properties	18
2.1.3.4 Thermal properties	19

2.1.3.5	Chemical characteristics	19
2.2	Epoxy resin as matrix for polymer matrix composites	20
2.3	Carbon Nanotubes filled epoxy composites	24
2.3.1	Methods of enhancing dispersion of CNT in polymer composites	26
2.3.1.1	Chemical functionalization	26
2.3.1.2	Hybridization of CNT-inorganic	27
2.4	Hybridization of CNT-inorganic via CVD	29
2.4.1	Factors that affecting the CNT growth via CVD	32
2.4.1.1	Catalyst	32
2.4.1.2	Substrates	34
2.4.1.3	Carbon sources	36
2.4.1.4	Atmosphere	37
2.4.1.5	Temperature	37
2.5	CNT-inorganic hybrid filled epoxy nanocomposites	38
2.6	Summary of the chapter	39

3 MATERIALS AND METHODOLOGY

3.1	Introduction	41
3.2	Materials	41
3.2.1	Aluminum powder	41
3.2.2	Nickel (II) nitrate hexahydrate	42
3.2.3	Sodium hydroxide	43
3.2.4	Hydrogen gas	43
3.2.5	Nitrogen gas	44

3.2.6	Methane gas	45
3.2.7	Multi-walled carbon nanotube	45
3.2.8	Alumina powder	46
3.2.9	Epoxy resin	47
3.2.10	Hardener	47
3.2.11	Silicon rubber	48
3.3	Methodology	49
3.3.1	Synthesis of CNT-alumina hybrid compound	49
3.3.2	Preparation of physically mixed CNT-alumina	53
3.3.3	Preparation of Silicon Mold	53
3.3.4	Preparation of CNT-alumina epoxy nanocomposites	54
3.4	Characterization and testings	56
3.4.1	Field emission scanning electron microscope	56
3.4.2	Energy dispersive x-ray	57
3.4.3	High resolution transmission electron microscope	57
3.4.4	X-ray diffraction analysis	57
3.4.5	Raman spectroscopy	58
3.4.6	Tensile test	58
3.4.7	Flexural test	59
3.4.8	Hardness test	59
3.4.9	Thermal conductivity	60
3.4.10	Coefficient of thermal expansion	60
3.4.11	Dielectric constant	61

4	RESULTS AND DISCUSSION	
4.1	Analysis of CNT– alumina hybrid	62
4.2	The CNT-alumina hybrid filled epoxy nanocomposite properties	76
4.2.1	Tensile Properties	76
4.2.2	Flexural Properties	82
	4.2.2.1 Failure mechanism of flexural	87
	4.2.2.2 Models of flexural modulus	89
4.2.3	Vickers micro-hardness	94
4.2.4	Morphological study of Epoxy/HYB and Epoxy/MIX	95
4.2.5	Coefficient thermal expansion	104
4.2.6	Thermal conductivity	107
	4.2.6.1 Models of thermal conductivity	110
4.2.7	Dielectric Constant	112
5	CONCLUSIONS	
5.1	Conclusions	115
5.2	Suggestion for future work	117
	REFERENCES	118
	PUBLICATIONS	133

LIST OF TABLES

Table 2.1	Comparison of nanotube properties with other comparable Materials	18
Table 2.2	List of the inorganic materials used in CNT-inorganic hybrid and their potential application	32
Table 3.1	Chemical and physical information of aluminum powder	42
Table 3.2	Chemical and physical information of Nickel (II) nitrate hexahydrate	42
Table 3.3	Chemical and physical information of sodium hydroxide	43
Table 3.4	Properties of the purchased hydrogen gas	44
Table 3.5	Properties of the purchased nitrogen gas	44
Table 3.6	Information of purchased methane gas	45
Table 3.7	Properties of the purchased MWCNT	46
Table 3.8	Properties of the purchased alumina powder	46
Table 3.9	Properties of the purchased epoxy resin	47
Table 3.10	Properties of the purchased epoxy hardener	48
Table 3.11	Properties of the purchased silicon rubber	48
Table 3.12	Materials used for catalyst preparation	50
Table 3.13	Descriptions of the samples	55
Table 4.1	Raman intensity of HYB and MIX	75

Table 4.2	Fracture Strain and Toughness of a neat epoxy and the epoxy composites with 1%, 3% and 5% weight percentage of HYB and MIX	81
Table 4.3	Coefficient of thermal expansion of neat epoxy and the epoxy composites with 1 %, 3%, and 5% weight percentage of HYB and MIX	107

LIST OF FIGURES

Figure 2.1	<i>sp</i> hybridization of acetylene molecule	10
Figure 2.2	<i>sp</i> ³ hybridization of methane molecule	11
Figure 2.3	<i>sp</i> ² orbital hybridization of graphitic carbon	11
Figure 2.4	Rolling the graphene sheet results in the nanotube while folding into a sphere produces the fullerene	12
Figure 2.5	Graphene as single-walled carbon nanotube (SWCNT) and multi-walled carbon nanotube (MWCNT) structures	13
Figure 2.6	Single graphene sheet showing the chiral vector as it relates to carbon nanotubes	14
Figure 2.7	Schematic diagrams of Armchair, Zigzag, and Chiral nanotubes	15
Figure 2.8	Chemical structure of the epichlorohydrin	21
Figure 2.9	Chemical structure of the bisphenol A	21
Figure 2.10	DGEBA reaction and chemical structure	22
Figure 2.11	Diamine molecule	23
Figure 2.12	Curing reaction between molecules of DGEBA and diamine curing agent	24
Figure 2.13	Two types of nano-micro hybrid materials: (a) CNT-carbon fiber (b) CNT-alumina hybrid	28
Figure 2.14	Schematic of CVD reactor	30
Figure 2.15	Carbon nanotubes growth mode: (a) tip-growth and (b) base-growth mode	35

Figure 3.1	Chemical structure of diglycidyl ether of bisphenol-A (DGEBA)	47
Figure 3.2	Chemical structure of trimethylhexamethylene diamine (TMD)	48
Figure 3.3	Schematic equipment set up of CVD	51
Figure 3.4	The flow chart of overview process for catalyst preparation	51
Figure 3.5	The flow chart of CNT growth process to produce CNT-alumina hybrid compound	52
Figure 3.6	Schematic equipment set up of ball milling	53
Figure 3.7	The flow chart of overview process for preparation silicon mold	54
Figure 3.8	The flow chart of process for preparation of epoxy Nanocomposites	56
Figure 3.9	(a) Sample preparation for thermal conductivity measurement, (b) diagram of a TPS sensor, (c) Experimental setup of sensor and samples	60
Figure 3.10	The schematic drawing of the specimen for CTE test	60
Figure 3.11	The schematic drawing of the specimen for dielectric constant Test	61
Figure 4.1	SEM images of HYB with magnification of (a) 5000X, (b) 10000X, (c) 30000X and (d) 100000X	63
Figure 4.2	SEM images of MIX with magnification of (a) 5000X, (b) 10000X, (c) 30000X and (d) 100000X	65
Figure 4.3	HRTEM image of MWCNT of HYB (a) 9900X, (b) 38000X, (c) 1050000X, (d) 285000X and (e) 15000X	68

Figure 4.4	Schematic diagram of CNT growth on alumina particle via tip growth mechanism	71
Figure 4.5	EDX spectrum of HYB	72
Figure 4.6	XRD patterns of (a) HYB and (b) MIX	73
Figure 4.7	Raman spectrum of HYB and MIX	75
Figure 4.8	Typical tensile stress–strain curves of a neat epoxy and the epoxy composites with 1%, 3%, and 5% weight percentage of HYB and MIX	76
Figure 4.9	Typical Stress–strain curves of epoxy composites with different weight percentages of the HYB and MIX (a) 1%, (b) 3% and (c) 5%	77
Figure 4.10	Ultimate tensile strength of a neat epoxy and the epoxy composites with 1%, 3% and 5% weight percentage of the HYB and MIX	79
Figure 4.11	Tensile modulus of a neat epoxy and the epoxy composites with 1%, 3% and 5% weight percentage of the HYB and MIX	81
Figure 4.12	Typical flexural stress–strain curves of neat epoxy and the epoxy composites with 1%, 3%, and 5% weight percentage of HYB and MIX	83
Figure 4.13	Typical flexural stress–strain curves of epoxy composites with different weight percentages of HYB and MIX (a) 1%, (b) 3% and (c) 5%	83
Figure 4.14	Flexural strength of neat epoxy and the epoxy composites with 1%, 3% and 5% weight percentage of HYB and MIX	86

Figure 4.15	Flexural modulus of neat epoxy and the epoxy composites with 1%, 3% and 5% weight percentage of HYB and MIX	87
Figure 4.16	Schematic representation of the crack propagation in the Epoxy/HYB	88
Figure 4.17	Schematic representation of the crack propagation in the Epoxy/MIX	88
Figure 4.18	Comparison of predictions of elastic modulus by Hashin-Shtrikman model, Paul model, Einstein model and Guth model with experiments for Epoxy/HYB and Epoxy/MIX composites	93
Figure 4.19	Vickers micro-hardness of Epoxy/HYB and Epoxy/MIX as a function of filler loading	94
Figure 4.20	SEM images of fracture surfaces of Epoxy/HYB 3 with magnification of (a) 1,000X, (b) 5,000X, (c) 10,000X and (d) 30,000X	96
Figure 4.21	SEM images of fracture surfaces of Epoxy/MIX 3 with magnification of (a) 1,000X, (b) 3,000X, (c) 10,000X and (d) 30,000X	98
Figure 4.22	HRTEM micrographs of Epoxy/HYB 3 with magnification of (a) 9900X, (b) 15,000X, (c) 29,000X and (d) 9,900X	100
Figure 4.23	HRTEM micrographs of Epoxy/MIX 3 with magnification of (a) 9900X, (b) 15,000X, (c) 29,000X and (d) 9,900X	102
Figure 4.24	Dimensional changes of the neat epoxy and the epoxy composites with 1%, 3% and 5% weight percentage of HYB and MIX as a function of temperature	106

Figure 4.25	Thermal conductivity of the baseline epoxy and the epoxy composites with 1%, 3% and 5% weight percentage of HYB and MIX	108
Figure 4.26	Heat flow through (a) Epoxy/HYB and (b) Epoxy/MIX	109
Figure 4.27	A proposed model of hybrid SWCNT and GNP	109
Figure 4.28	Frequency dependent dielectric constant of neat epoxy and epoxy filled with HYB and MIX at various different filler loadings	113

LIST OF ABBREVIATIONS

ASTM	American Society for Testing and Materials
CNT	Carbon Nanotube
CTE	Coefficient of Thermal Expansion
CVD	Chemical Vapor Deposition
DGEBA	Diglycidyl Ether of Bisphenol-A
EDX	Energy Dispersive X-ray
FESEM	Field Emission Scanning Electron Microscope
HRTEM	High Resolution Transmission Electron Microscope
HYB	CNT-Alumina hybrid compound
MWCNT	Multi-walled Carbon Nanotube
MIX	CNT- Alumina physically mix
SWCNT	Single-walled Carbon Nanotube
UTS	Ultimate Tensile Strength
TMD	Trimethylhexamethylene Diamine
XRD	X-ray Diffraction

LIST OF SYMBOLS

%	Percentage
°C	Degree Celsius
>	More Than
<	Less Than
GPa	Giga Pascal
MPa	Mega Pascal
GHz	Giga Hertz
KHz	Kilo Hertz
ppm/°C	part per million/Degree Celsius
W/mK	Watt/meterKelvin
µm	Micrometer
nm	Nanometer
L	Length
Wt %	Weight Percent
mol	mole
Pa.s	Pascal-second
g/cm ³	Gram per cubic centimeter
g/l	Gram per litre

**PENAMBAHBAIKAN SIFAT-SIFAT MEKANIK, TERMA DAN
DIELEKTRIK BAGI HIBRID NANOTIUB KARBON DAN ALUMINA
DALAM NANOKOMPOSIT EPOKSI**

ABSTRAK

Penambahan nanotub karbon (CNT) dalam nanokomposit polimer telah memberikan cabaran kepada penyelidik disebabkan oleh taburannya dalam matriks polimer. Kajian ini memberikan fokus terhadap sebatian hibrid CNT-alumina yang dihasilkan melalui pemendapan wap kimia (CVD) yang digunakan untuk mempertingkatkan taburan dan menambah baik sifat-sifat mekanik, terma dan dielektrik bagi nanokomposit epoksi. Sebatian hibrid CNT-alumina telah berjaya disintesis melalui kaedah CVD dengan menggunakan pemangkin nikel di bawah atmosfera metana pada suhu 800 °C. Bagi tujuan perbandingan, campuran CNT-alumina secara fizikal juga disediakan dengan menggunakan kaedah pengisaran bebola bagi tujuan perbandingan. Nanokomposit epoksi terisi sebatian hibrid CNT-alumina dan CNT-alumina yang dicampurkan secara fizikal telah dicirikan berdasarkan muatan pengisi (iaitu 1% - 5%). Sebatian hibrid CNT-alumina masing-masing mempunyai saiz antara 10 – 30 nm dan 12 % berat karbon berdasarkan analisis medan pancaran mikroskop imbasan electron pancaran medan (FESEM), mikroskop pemancaran elektron resolusi tinggi (HRTEM) dan serakan tenaga sinar-x (EDX), manakala analisis pembelauan sinar-x (XRD) mendedahkan kewujudan fasa karbon antara beberapa fasa alumina. Penilaian bagi sebatian hibrid CNT-alumina terisi nanokomposit epoksi menunjukkan sifat-sifat mekanik, terma dan dielektrik yang lebih tinggi berbanding CNT-alumina yang dicampurkan secara fizikal serta terisi nanokomposit epoksi. Peningkatan ini berkaitan dengan taburan seragam

sebatian hibrid CNT-alumina seperti yang diperhatikan daripada FESEM dan HRTEM. Penggunaan sebatian hibrid CNT-alumina terisi nanokomposit epoksi telah dibuktikan mampu untuk meningkatkan kekuatan tegangan sehingga 30%, modulus tegangan sebanyak 39%, kekuatan lenturan sebanyak 30%, modulus lenturan sebanyak 35%, kekerasan sebanyak 17%, konduktiviti terma sebanyak 20%, nilai suhu peralihan kaca sebanyak 25% dan pemalar dielektrik sebanyak 20% apabila dibandingkan dengan epoksi yang tulen.

ENHANCEMENT OF MECHANICAL, THERMAL AND DIELECTRIC PROPERTIES OF HYBRID CARBON NANOTUBES AND ALUMINA IN EPOXY NANOCOMPOSITES

ABSTRACT

The incorporation of carbon nanotube (CNT) in polymer nanocomposites has become challenges for researchers due to its dispersion in polymer matrix. This work focuses on CNT-alumina hybrid compound prepared via chemical vapor deposition (CVD) which is used to improve dispersion and enhance the mechanical, thermal and dielectric properties of epoxy nanocomposites. The CNT-alumina hybrid compound was successfully synthesized via CVD by using nickel catalyst under methane atmosphere at 800 °C. The physically mixed CNT-alumina was also prepared by ball milling method for comparison. The CNT-alumina hybrid compound and physically mixed CNT-alumina filled epoxy nanocomposites were characterized according to their filler loadings (i.e. 1% - 5%). The CNT-alumina hybrid compound had the size between 10 – 30 nm and 12 Wt % of carbon according to field emission scanning electron microscope (FESEM), high resolution transmission electron microscope (HRTEM) and energy dispersive x-ray (EDX) analysis respectively, while x-ray diffraction (XRD) revealed the existence of carbon phase among several phases of alumina. The CNT-alumina hybrid compound filled epoxy nanocomposites assessments showed higher mechanical, thermal and dielectric properties than the physically mixed CNT-alumina filled epoxy nanocomposites. This increase is associated with the homogeneous dispersion of CNT-alumina hybrid compound as observed from FESEM and HRTEM. It was demonstrated that the CNT-alumina hybrid compound filled epoxy nanocomposites are capable of increasing tensile

strength by up to 30%, giving tensile modulus of 39%, flexural strength of 30%, flexural modulus of 35%, hardness of 17%, thermal conductivity of 20%, glass transition temperature value of 25% and dielectric constant of 20% when compared to a neat epoxy.

CHAPTER 1

INTRODUCTION

1.1 Overview

The extensively growing area of nanoengineering materials will create many perspectives for polymer and composites determined by the final product of the polymer nanocomposites. In the late 1980s, polymer nanocomposites were commercially produced in both academic laboratories and research organisations (Komarneni, 1992). Roy (1992) and Komarneni et al. (1997) started to use the term 'nanocomposites' in 1984 to describe the fact that polymeric product contains two or more distinct materials in the nanoscale. Since that time, the term 'nanocomposites' has been globally recognized and described as a very large group of materials involving structures in the nanometer range between 1-100 nm (Gleiter, 1995). The properties of the nanocomposites have become an attraction to researchers due to size of the structures which is totally different from those of the bulk matrix. In 1988, the Toyota Group became the first company to utilize nanocomposites parts for their novel car models by using polymer/layered nanocomposites (Fukushima et al., 1988). Subsequently, numerous other companies also started to look into nanocomposites and this led to a dramatic development of research in this novel class of materials.

The reinforcement of polymer by using fillers such as inorganic or organic materials is typical in the production of modern polymer composites. Generally, conventional composites use micrometer particles, platelets or fibres as fillers in polymer composites. These conventional composites have been explored for many

years to be fully utilized in various industrial applications (Schadler, 2003). For examples, composites based on thermosetting resins are extensively used for structural materials product such as polymer, concrete, glass fibre reinforced epoxy, inorganic particle filled epoxy, etc. Usually, micro-filler particles are inactive and their main purpose is to reduce the cost of the final product. The most crucial factors that control the properties of polymer composites which contain inactive fillers are size, shape and distribution of the filler, while surface morphology and chemistry play a minor role. On the other hand, active fillers in polymer composites demonstrate a reinforcing effect of filler on mechanical properties depending on the morphology and interaction between filler and polymer matrix (Ajayan et al., 2006). Typically, polymer reinforced with micrometer active fillers exhibits improvement in hardness, however its tensile properties worsen due to stress concentration resulting from the presence of filler particle.

Polymer nanocomposites are developed by incorporating a small amount of nano-filler dispersed at a molecular level in the polymer matrix as a replacement for the conventional polymer composites. The homogenous dispersion of nano-filler particles provides very high surface area to volume ratio between filler and polymer matrix. A special characteristic of polymer nanocomposites is that a significant enhancement in properties can be achieved at low filler loading which consequently minimizes the weight of the final product (Mai and Yu, 2006).

Epoxy resins are a type of versatile polymer materials characterized by the existence of more than one epoxy group or oxirane ring located on their molecular structure. Similar to other thermosets, epoxy resin also forms a network on curing

with various types of curing agents. Nowadays, epoxy resin is recognized as one of the interesting class of polymers and used as a matrix for polymer composites due to its strong adhesion and excellent overall mechanical properties, including high strength and stiffness, high chemical, thermal, and dimensional stability, as well as good creep and solvent resistance (Hussain et al., 2006). The performance of epoxy resin can usually be improved by incorporating various type of including particulate filler and fibrous reinforcement. Recently, carbon nanotubes, as particulate filler in epoxy resin, have been widely used due to their remarkable improvements in mechanical, electrical, optical and thermal properties (Coleman et al., 2006a).

Over the last decades, carbon nanotubes have remained attractive to both engineers and scientists who are interested in their applications due to the unique combination of their properties (Meyyappan, 2004). Carbon nanotubes have progressed into one of the most intensively studied materials and they play a major role in co-triggering the polymer nanocomposites (Andrews and Weisenberger, 2004). Carbon nanotubes are regarded as seamless cylinders formed by wrapping graphene sheets with carbon atoms that are covalently bonded with each other through sp² hybridization. Based on the number of layer of graphene sheets, they can be categorized as single-walled or multi-walled carbon nanotubes. These carbon nanotubes molecular systems are nanometer in diameter and up to centimeter in length which demonstrate length/diameter aspect ratio of more than 10⁷ (Ebbesen, 1996). The contiguous carbon nanotubes possess a high degree of atomic-scale perfection and this makes them chemically inert as their close relationship with graphene. As for graphene under tension, the strength of the carbon nanotubes are two times higher than steel (Gojny et al., 2004). The melting point of the carbon

nanotubes in vacuum is approximately 3700 °C which is almost similar to graphite and greater than any metal (Jorio et al., 2007). Carbon nanotubes can act as ballistic conductors of electrons or behave as a semiconductor depending on the atomic structure and the diameter (Javey et al., 2003). In addition, carbon nanotubes are an excellent conductor of heat and reported to surpass the thermal conductivity of pure diamond (Jorio et al., 2007). Furthermore, carbon nanotubes also seem to be biocompatible in many environments (Smart et al., 2006).

In the meantime, a wide range of carbon nanotubes based hybrid materials have also been developed by the combination of carbon nanotubes with other materials. These hybrid materials with attractive features exhibit promising applications in the fields of nanobiotechnology, energy conversion, fuel storage, catalysis and electronic nanodevices (He et al., 2009). In particular, the micro supports could be either micrometric inorganic particles or various fibers. Instead of conventionally mixing two types of materials in a random way, carbon nanotubes are directly grown on the surface of the microscopic supports to form the nano-micro hybrid structures (Xie and Gao, 2007; Wang et al., 2008; Jiang et al., 2007). The nano-micro hybrid structures have a reinforced interface between each carbon nanotubes and the micrometer support. At the same time, rather than in entangled state, carbon nanotubes could be distributed on the surface of micrometer particles. This is desired to avoid the agglomeration of carbon nanotubes and to obtain good dispersion in composites. Furthermore, hybrid materials in the composites favor to transfer efficiently the loads between the matrix and the fillers. Thus, enhanced properties of composites are expected to be achieved by using the hybrid materials as fillers in composites.

1.2 Problem Statement

The extraordinary properties of the CNT make it an ideal candidate to be used as multifunctional filler system in epoxy composites. Nevertheless, there are huge challenges for researchers to impart the intrinsic properties of the CNT due to the difficulty to disperse CNT in the epoxy matrix. CNT is usually present in the form of bundles or ropes due to the intrinsic van der Waals force attraction, its high surface area and its high aspect ratio (HowardáEbron, 2004). These characteristic of CNT can easily result in poor dispersion and significance level of agglomeration. Poor dispersion of the CNT will significantly reduce their reinforcement efficiency due to the CNT slipping each other when load is applied which will result in the formation of micro-voids in the epoxy composites.

Many researches have been conducted to overcome the above mentioned challenges i.e; to improve the dispersion of CNT in epoxy matrix (Du et al., 2007). Most of the techniques are dealing with processing issues rather than looking at the filler design aspect or architecture. The research includes investigation of different methods such as chemical functionalization (Yang et al., 2009), and physical blending (ultrasonication or high shear mixing) (Park et al., 2002). Chemical functionalization focuses on the surface treatment to the CNT structure to improve their chemical interaction with polymer matrix, and it has enhanced the dispersion of CNT. However, chemical functionalization method often causes structural defect that diminishes the intrinsic properties of CNT (Park et al., 2002). Hybridization of CNT with an inorganic filler is implemented to improve dispersion of CNT without damaging its structure. Nevertheless, conventional hybridization method by milling requires longer time which may damage the CNT structure. Furthermore, the