FABRICATION AND CHARACTERIZATION OF HYBRID CARBON NANOTUBES / SILICA FILLED EPOXY NANOCOMPOSITES

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ABSTRACT

Carbon nanotube (CNT) was used as reinforcement to improve mechanical and thermal conductivity properties of epoxy. However, agglomeration and homogenous dispersion of CNT in the epoxy matrix remains challenge. Growth of CNT on the surface of silica gels (hybrid CNT) can reduce the agglomeration of CNT in the epoxy. Hybrid CNT was synthesized via floating and non-floating catalyst chemical vapour deposition method (CVD). Regardless reaction time and reaction temperature, growth of hybrid CNT by floating CVD method was unsuccessful. Considering the result observed from FESEM, HRTEM, TGA and Raman spectroscopy, hybrid CNT that produced from 50 ml/min hydrogen flow rates was selected to prepare the nanocomposite. Result of hybrid CNT epoxy nanocompsoite was compared with silica epoxy composite and CNT epoxy nanocomposite. Inclusion of hybrid CNT at different weight loading (0.1 wt. %, 0.5 wt. %, 1 wt. %, 1.5 wt. %, 3 wt. % and 5 wt. %) was expected to increase glass transition temperature (T_g) and thermal conductivity properties of epoxy. However, T_g values for all type of nanocomposite (hybrid CNT, CNT and silica) was lower than neat epoxy except for 1.5 wt. % silica. It also was observed that at all weight loading, silica epoxy composite has higher T_g value compared to CNT and hybrid CNT epoxy nanocomposite. Finally, addition of hybrid CNT into the epoxy also decreases the thermal conductivity value. Lowest thermal conductivity value for hybrid CNT recorded at 5 wt. % with 0.155 W/mK compared to neat epoxy (0.177 W/mK).

ABSTRAK

Nanotiub karbon (CNT) telah digunakan sebagai bahan pengukuh bagi meningkatkan sifat mekanikal dan sifat kekonduksian haba epoksi. Walaubagaimanapun, gumpalan dan penyebaran homogen CNT di dalam matriks epoksi kekal sebagai cabaran. Pertumbuhan CNT ke atas permukaan silika (CNT hibrid) boleh mengurangkan gumpalan CNT di dalam epoksi. CNT hibrid telah disintesis menggunakan kaedah pemangkin terapung dan tidak terapung penguraian wap kimia (CVD). Pertumbuhan CNT dengan menggunakan pemangkin terapung CVD tidak berjaya pada semua masa dan suhu tindakbalas. Berdasarkan keputusan daripada FESEM, HRTEM, TGA dan spektroskopi Raman, CNT hibrid yang terhasil daripada 50 ml/min kadar aliran hidrogen telah dipilih untuk menghasilkan nanokomposit. Keputusan komposit CNT hibrid dan epoksi telah dibandingkan dengan komposit silika epoksi dan komposit CNT epoksi. Kemasukan CNT hibrid pada nisbah berat yang berbeza (0.1 wt. %, 0.5 wt. %, 1 wt. %, 1.5 wt. %, 3 wt. % and 5 wt. %) dijangka dapat meningkatkan suhu peralihan kaca, Tg dan sifat kekonduksian haba epoksi. Walau bagaimanapun, nilai Tg untuk semua jenis nanokomposit (CNT hibrid, CNT dan silika) adalah lebih rendah berbanding dengan epoksi tulen kecuali pada 1.5 wt. % silika. Ia juga dapat diperhatikan bahawa pada semua kandungan berat, komposit silika epoksi mempunyai nilai Tg yang tinggi berbanding nanokomposit CNT dan cantuman CNT epoxy. Akhir sekali, penambahan CNT hibrid ke dalam epoksi juga telah mengurangkan nilai kekonduksian terma epoksi. Nilai kekonduksian terma terendah telah direkodkan pada 5 wt. % dengan 0.155 W/mK berbanding dengan epoksi mentah (0.177 W/mK).

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APPROVAL

I certify that an Examination Committee has met on 12 April 2017 to conduct the final examination of Amal Izzati binti Mohd Ismadi on her degree thesis entitled Fabrication and characterization of hybrid carbon nanotubes/silica filled epoxy nanocomposite. The committee recommends that the student be awarded the Master of Science (Mechanical Engineering).

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LIST OF ABBREVIATIONS AND SYMBOLS

CNT	Carbon Nanotube
CNF	Carbon Nanofibre
CVD	Chemical Vapour Deposition
DMA	Dynamic Mechanical Analysis
DMTA	Dynamic Mechanical Thermal Analysis
DGEBA	Diglycidyl Ether of Bisphenol A
FESEM	Field Emission Scanning Electron Microscope
HRTEM	High Resolution Transmission Electron Microscope
ID	Inner Diameter
MWNT	Multiwall Carbon Nanotube
OD	Outer Diameter
SWNT	Single Wall Carbon Nanotube
TGA	Thermal Gravimetric Analysis
Al_2O_3	Aluminium Oxide
Co	Cobalt
d	Diameter
E'	Storage Modulus
Fe	Iron
g	Gram
I_D	Raman Intensity for D Peak
I_G	Raman Intensity for G Peak
l	Length
min	Minute

mK	Meter Kelvin
ml	Mililiter
mg	Milligram
Ni	Nickel
nm	Nanometer
rpm	Revolution per Minute
SiO_2	Silicon Dioxide
T_g	Glass Transition Temperature
W	Watts
wt. %	Weight loading percent
tan δ	Loss factor
°C	Degree Celcius
µm	Micrometer
K_c	Effective Thermal Conductivity of Composite
K_f	Thermal Conductivity of Filler
K_m	Thermal Conductivity of Epoxy
Φ_f	Volume Fraction of Particle
V_f	Volume Fraction of Filler
W_f	Weight of Filler
W_m	Weight of Matrix

CHAPTER 1

INTRODUCTION

1.0 Introduction

In 1991, carbon nanotube (CNT) was introduced by Iijima in a paper entitled "Helical microtubules of graphitic carbon" [1]. CNT was described as a graphitic carbon needle that has a diameter ranging from 4 to 30 nm. Since then, extensive researches on CNT properties were done to discover its potential. Several methods to synthesize CNT have been designed and developed such as arch discharge [2], laser ablation [3] and chemical vapour deposition (CVD) [4]. Among these three methods, CVD was widely used because of its advantages which are high yield CNT production, low cost and ability to control CNT morphology by varying the growth parameters.

Development in nanomaterials and nanotechnology provides opportunities to introduce CNT in almost all material as filler and reinforcement including in epoxy. Epoxy which was discovered in 1909 is a type of thermosetting polymer resin and has a wide variety of application. Potential application of epoxy ranges from electronic devices, aeronautic components [5], coating materials [6] and automotive parts [7]. However, epoxy has low thermal conductivity properties and poor tribological materials. In fact, this disadvantage has reduced the performance of epoxy in some applications. Many researcher and industrial expert have discovered a way to improve the epoxy properties which is by addition of CNT. CNT was recognized to improve the mechanical, thermal conductivity and electrical properties of polymer [8]. This is because CNT possess remarkable mechanical and thermal conductivity properties that are comparable to diamond. There are studies that were successful in improving the mechanical and thermal conductivity of epoxy.

Present study attempts to grow CNT onto the surface of silica by using chemical vapour deposition (CVD) system. Hybrid CNT produced was then introduced into the epoxy matrix. Hybrid CNT are expected to increase the mechanical and thermal conductivity properties of epoxy. Hybrid CNT epoxy nanocomposite was characterized by dynamic mechanical thermal analysis (DMTA) and thermal conductivity analysis.

1.1 Background of problem

Epoxy has good mechanical properties but low in thermal conductivity properties (~0.2 W/mK) [9]. Thermally conductive epoxies are needed in some applications such as in electronic devices. Therefore, many high thermal conductivity particles including CNT was incorporated into the epoxy. The developments in nanocomposite and nanotechnology have provided opportunities to incorporate CNT in the epoxy matrix. However, processing the CNT and epoxy nanocomposite remains to be a challenge. During nanocomposite processing, CNTs tend to agglomerate due to their strong Van der Waals interactions, high surface area and high aspect ratio. Agglomeration will hinder the performance of CNT in the epoxy. In addition, agglomerations of CNT also create defect sites that will initiate failure of nanocomposite. Therefore, CNT need to be dispersed homogenously in the epoxy matrix in order to take advantage on its outstanding properties. Many studies were performed in improving the dispersion of CNT including applying high shear force during processing. As a result, addition of 0.25 wt. % SWNT has increased epoxy Young's modulus by 30% compared to neat epoxy [10]. In addition, CNT chemical functionalization is also one of the methods to improve dispersion within epoxy matrix. For example, 1 wt. % of poly-4-aminostyrene CNT increases epoxy tensile strength by34% compared to neat epoxy. On the other hand, tensile strength for pristine CNT nanocomposite decreases by 8.8% [11]. This indicates that chemical functionalization has enhanced CNT dispersion.

Both types of approaches give disadvantages to the industry because multiple steps were involved in the process of composite preparation. In addition, functionalization of CNT and high shear force can distort the CNTs structure [12,13]. In order to achieve high dispersion of CNT with fewer processing steps and CNT distortion, alternative methods have been approached by researchers. In these few years, direct growth of CNT onto a substrate (hybrid CNT) has caught researches attention. Alumina (Al_2O_3) [13], silicon [14] and carbon fibre [15] are common materials that are used as substrate. Substrate will work as a carrier to disperse CNTs into polymer matrices as well as reduce the entanglement between the CNT.

This study tackles the dispersion of CNT by grafting the CNT onto porous silica gels. The silica gels will function as a transportation medium of CNT inside the epoxy. This hybrid CNT is expected to increase the degree of dispersion and thus homogenous dispersion of CNT could be achieved. This CNT arrangement is also expected to reduce the viscosity of epoxy even at high filler loading. As a result, there will be an increase in the epoxy's high thermal mechanical properties and thermal conductivity properties.

1.2 Objective of study

The purpose of this study is to achieve several objectives as follows:

a) To synthesize CNT on silica gel and characterize the particles with FESEM, HRTEM, Raman spectroscopy and TGA.

b) To evaluate and investigate the mechanical thermal properties of hybrid CNT epoxy nanocomposite and compare with commercial CNT and silica nanocomposite.

c) To evaluate and investigate the thermal conductivity properties of hybrid epoxy nanocomposite and compare with commercial CNT and silica nanocomposite.

1.3 Scope of the study

The scope of this study is as follows:

1) The methods used to synthesize hybrid CNT are floating chemical vapour deposition and non floating chemical vapour deposition.

2) Reaction temperature and reaction time with varied parameters for floating chemical vapour deposition. The temperature variation was 800 °C, 850 °C and 900 °C. Reaction time was varied at 1 hour, 2 hours and 3 hours.

3) Hydrogen flow rate with varied parameters for non floating chemical vapour deposition. Hydrogen flow rate was varied at 0 ml/min, 50 ml/min and 100 ml/min.

4) Weight percentage of filler (hybrid CNT, commercial CNT and silica) used to prepare nanocomposite were 0.1 wt. %, 0.5 wt. %, 1 wt. %, 1.5 wt. %, 3 wt. % and 5 wt. %.

5) The mechanical properties of epoxy nanocomposite were characterized by using dynamic mechanical analysis (DMA 8000) instrument. Glass transition temperature (T_g), glassy region and rubbery region of epoxy nanocomposite were determined by DMA. Then, T_g value of hybrid CNT epoxy nanocomposite was compared with two other fillers (silica and commercial CNT).

6) Thermal conductivity value of epoxy nanocomposite was analyzed by KD2 pro analyzer. Value of hybrid CNT epoxy nanocomposite was compared with silica and commercial CNT.

1.4 Contribution of study

There are many studies on hybrid CNT; however, based on author's best knowledge the study on incorporation of these hybrid CNT with epoxy polymer is still lacking. Therefore, the properties of hybrid CNT with epoxy polymer must be studied and observed. This study focuses on how hybrid CNT can improve thermal mechanical properties and thermal conductivity properties of epoxy. In addition, this study not only focuses on nanocomposite processing and characterization, it's also focuses on synthesizing hybrid CNT using two methods (floating CVD and non floating CVD). In the present study, the growth parameters that affects CNT morphology such as effects of reaction temperature, reaction time and effects of hydrogen were included. Based on knowledge gained, high quality and high yield hybrid CNT can be produced. Furthermore, knowledge on hybrid CNT synthesis can be added to the existing studies.

1.5 Outline of the thesis

- **Chapter 1** begins with the introduction, background of problem, objectives of the study, scope of the study and outline of the thesis.

- **Chapter 2** is a literature review that focuses on properties of CNT, CNT catalyst and substrate, effects of hydrogen, effects of temperature, CNT growth mechanism and study on mechanical and thermal conductivity of nanocomposites.

- **Chapter 3** provides description on experimental techniques used in this study. The descriptions include methods used to synthesize hybrid CNT and epoxy nanocomposites, and characterization technique for both hybrid CNT and nanocomposite.

- **Chapter 4** presents the results obtained on hybrid CNT characterization, thermal mechanical analysis, and thermal conductivity of nanocomposite.

- **Chapter 5** concludes the overall findings and provides suggestion for the future work.