

Fabrication and Characterization of Graphene/Epoxy Nanocomposites

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Graphene with high electric and thermal conductivities has been widely used as reinforced filler. In this study, graphene loadings in the range between 0.3 and 1.0 wt.% were added to the epoxy to fabricate the nanocomposites. The mechanical and thermal properties of nanocomposites were characterized using tensile test and differential scanning calorimeter (DSC), respectively. Experimental results show that the elastic modulus, yield strength, ultimate strength and glass transition temperature of the graphene reinforced epoxy are increasing with the increase of the graphene, while the fracture strain and toughness are decreasing with the increase of the graphene. Scanning electron microscope (SEM) was employed to investigate the dispersion and separation of graphene in the epoxy based matrix. The SEM images depict that graphene is well dispersed resulting in a significant improvement of the mechanical and thermal properties of the nanocomposites. The mechanical properties and thermal stability of epoxy nanocomposites with graphenes and multi-walled carbon nanotubes additives were then compared. Experimental results show that nanocomposite with graphene additives outperform the multi-walled carbon nanotube additives.

Keywords: graphene, nanocomposite, tensile strength, glass transition temperature.

1. INTRODUCTION

Graphene, a novel carbon material with a two-dimensional layer and single atom thickness, has received a great attention from both scientists and engineers due to its excellent mechanical and electrical properties. Theoretical prediction and experimental results show that graphene exhibits excellent properties of Young's modulus 1000 GPa, fracture toughness 125 GPa [1], thermal conductivity 5000 W/mK [2], electron charge mobility 200,000 cm²/V [3] and electrical conductivity 10⁻⁶ Ω⁻¹cm⁻¹ [4]. These properties make graphene an ideal candidate for many potential applications such as solar cells and hydrogen storage [5], sensors [6], batteries [7] and supercapacitors [8]. Graphene nanoplatelets (GNPs) composed of multiple graphene layers that are stacked together have similar properties as those of single layer graphene. However, GNPs are much easier to fabricate and hold in comparison with single-layer graphene [9]. In addition, the performance of GNP is better than that of carbon nanotube (CNT) based on the following reasons: the abundance of GNPs in nature yields in a reduce of the cost; the high specific surface area of GNPs leads to a higher degree of stress being transferred across the interface and GNPs provides a better reinforcement than that of CNTs [10].

Nanocomposites are considered as a new class of composites incorporated with nano scale reinforcements that are well dispersed in a polymer matrix. The nano scale fillers can improve the interfacial interaction, leading to the superior properties of nanocomposites in comparison with conventional composites [11]. Recently, polymer-based nanocomposites reinforced with graphenes have attracted widespread industrial interest because graphene has a great

capability of enhancing the electrical conductivity and mechanical properties of the nanocomposites [12]. Epoxy resin is one of the most commonly used polymers, due to its extremely high fracture toughness and mechanical stiffness, chemical resistance and good solvability, and better adhesion [10]. Moriche et al. [13] investigated the electrical behavior and mechanical properties of epoxy-based nanocomposites reinforced with functionalized graphene nanoplatelets. They found that the electrical conductivity of nanocomposites with GNPs of 12 wt.% was in the range of 10⁻⁴ S/m, and reported that the flexural strength increased by 11 %. Dai and Mishnaevsky [14] developed a three dimensional model of polymer-based composites reinforced with graphenes to study the influence of nano fillers, such as the volume fraction, shape, aggregation, orientation and aspect ratio of graphene nanoplatelets on the mechanical properties and failure mechanisms of nanocomposites. They found that the elastic modulus of the nanocomposites increases with the increased volume fraction, aspect ratio, interfacial properties between the polymer and graphene, and as the degree of interaction decreased. The ultimate strength exhibits similar trends, except for the aspect ratio and aggregation, where the opposite tendencies were reported. Wan et al. [15] prepared epoxy nanocomposites with homogeneously dispersed graphene using a facile surfactant-assisted process, and the tensile strength of the nanocomposite was significantly increased by 57 % at 0.1 % weight loading. Luong et al. [16] studied the incorporation of 0.38 wt.% of functionalized graphene in a polyimide nanocomposite and showed that the elastic modulus increased by an approximately 30%. Zaman et al. [17] fabricated epoxy-based nanocomposites with chemical modification of graphene nanoplatelets to study the influence of the interfacial interaction on the microstructure and properties of the nanocomposites. They found that the strain energy release rate of the

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nanocomposite reinforced with 4 wt.% of chemical modification GNPs was 96.1 % greater than the strain energy release rate of unmodified GNP. The glass transition temperature of the nanocomposite with 2.5 wt.% GNP was 108.6 °C, while the glass transition temperature of the neat epoxy was 94.7 °C. Pontefisso and Mishnaevsky [18] proposed a computational model to investigate the influence of microstructure, shapes and dispersion of carbon nanotubes and graphene in polymers on fracture resistance and tensile strength. Zarasvand and Golestanian [19] employed theoretical, numerical and experimental methods to study the effects of the number of layers, orientation and dispersion of GNP on the mechanical properties of nanocomposites reinforced with graphene. Spanos and Anifantis [20] developed a micromechanical hybrid finite element model to investigate the capability of stress transfer in nanocomposites reinforced with graphene. They found that stress transfer between the graphene and polymer matrix is significantly dependent on the volume fraction, polymer matrix and interfacial interaction. Liu et al. [21] designed a sandwich structure to directly measure the interfacial shear strength between polymethyl methacrylate and graphene nanoplatelets.

Jangam et al. [22, 23] investigated the effect of multi-walled carbon nanotube alignment on the mechanical, electrical and vibrational properties of nanocomposites. They found that the alignment of MWCNTs has improved the modal damping by 37 %, electrical conductivity by 200 % and fatigue life by 15 % with a small weight percentage of MWCNT loading (wt.% < 0.5 %) into the epoxy matrix. Sethi et al. [24] examined the role of aspect ratio and length of MWCNTs in determining the performance of nanocomposites. They observed that the electrical properties were proportional to length of the MWCNTs whereas the mechanical properties were dependent on the aspect ratio of the MWCNTs. Liang [25] studied the effects of graphene nanoplatelets size and content on tensile properties of polypropylene composites. Bansal et al. [26] demonstrated a promising way to improve the elastic modulus, hardness and fracture resistance simultaneously by reinforcing the epoxy (bisphenol-A) matrix with a new-age two-dimensional thin graphene oxide sheet. High improvements were observed by Zakaria et al. [27] when 1 wt.% GNPs were integrated within the epoxy system. The tensile and flexural strength of GNP nanocomposites improved up to 26 %, 29 % respectively, compared to the neat epoxy. Epoxy-based nanocomposites have been extensively used for mechanical strength applications such as aerospace, automobiles and marine industries due to their light weight, better strength and ease of processing [26].

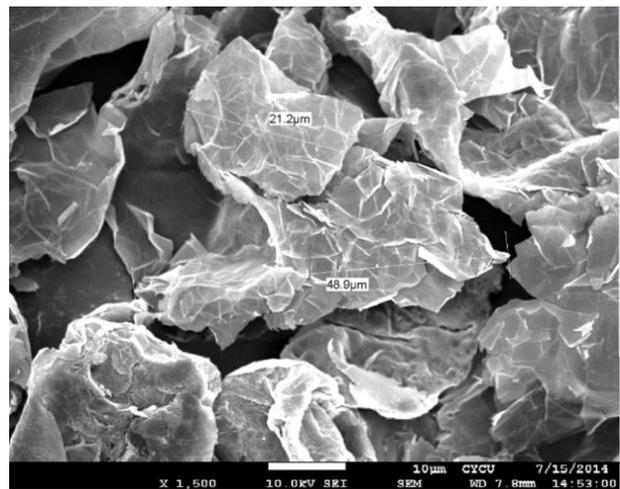
Many researchers have investigated the effects of reinforcing nanocomposite with either MWCNT or GNP. However, few works have studied and compared the performance between the MWCNT and GNP reinforced nanocomposites [28, 29]. The novelty of this study is that nanocomposites reinforced with both MWCNTs and GNPs were fabricated and characterized. In this work, epoxy nanocomposites reinforced with graphene nanoplatelets and multi-walled carbon nanotubes (MWCNTs) were prepared at various weight fractions ranging from 0.3 % to

1.0 %. The mechanical properties of nanocomposites including the elastic modulus, ultimate strength and strain at break were obtained by tensile testing. The glass transition temperature of nanocomposites was measured by differential scanning calorimeter (DSC). The mechanical and thermal properties of epoxy-based matrix with GNPs and MWCNTs additives were compared.

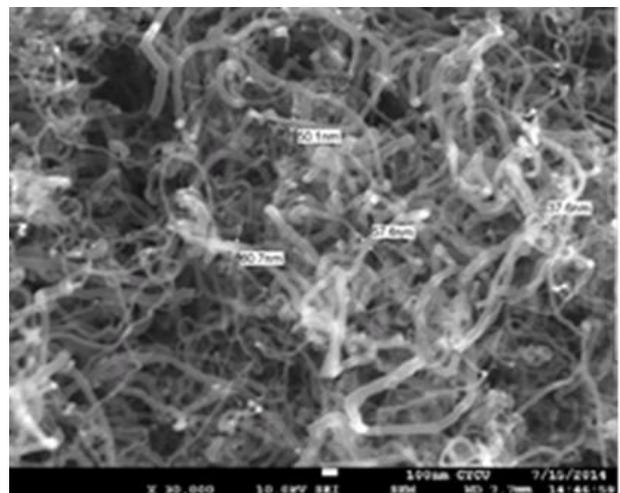
2. PREPARATION OF EPOXY NANOCOMPOSITES

2.1. Materials

Both the graphene nanoplatelets and multi-walled carbon nanotubes were purchased from Uchess Co. Taiwan. The GNPs had thickness ranging from 1 to 10 nm, length ranging 0.5–20 µm, specific surface area 40–60 m²/g and a purity of > 99.5 wt.%. The diameter of MWCNTs is in the range between 40 ~ 60 nm, while the length is 5–10 µm and a purity of > 95 wt.%. The matrix used in this work consisting of part A: epoxy Mungo 4200A and part B: hardener Mungo 4200B, was provided by Golden Root Co., Ltd Taiwan. Deionized water was employed in all processes if necessary. The morphologies of pristine GNPs and MWCTNs were examined using scanning electron microscopy (SEM) as shown in Fig. 1.



a



b

Fig. 1. SEM images: a–pristine GNPs; b–MWCNTs

2.2. Fabrication of GNP/epoxy nanocomposites

Epoxy-based nanocomposites reinforced with different weight loadings of GNPs and MWCNTs were fabricated using the following procedures. A desired concentration of GNPs or MWCNTs was first dispersed in ethanol and sonicated in an ultrasonic bath for 30 min at temperature of 40 °C. Then the suspension was poured into a liquid epoxy that was preheated at a temperature of 40 °C for 30 minutes. The solution was further processed with mechanical stirring and sonicating for 3 hours to separate the aggregation of nanofillers (GNPs or MWCNTs) and achieve good dispersion. The dispersions were degassed in a vacuum chamber maintained at pressure of 20 mmHg for 30 min to remove the ethanol, followed by addition of the hardener with a resin-to-hardener weight ratio of 2:1 and softly stirred it for about 10 minutes. Afterwards the mixture was placed in a vacuum chamber for about 30 min to remove the bubbles induced from the stirring. After degassing, the blend was poured into a dog-bone-like aluminum mold to fabricate the tensile test specimen. The specimen was cured in a vacuum oven at a temperature of 40 °C for 24 hours. Samples of neat epoxy, GNPs/Epoxy and MWCNTs/Epoxy were prepared in the same procedures. Three samples were prepared for each concentration. The experimental results reported in this work are the averaged values. The standard deviation is also provided to demonstrate the consistency of the experimental tests.

2.3. Characterization and instruments

The dispersion of GNPs and MWCNTs in epoxy was characterized by a field emission-scanning electron microscope (FE-SEM, Joel JSM-7600F, Japan) with an accelerating potential of 10.0 kV. The mechanical properties of the nanocomposites were obtained by uniaxial tensile test using a Hounsfield Model 10KS universal test machine according to the ASTM standard D638. The thermal stability of epoxy nanocomposites was determined by differential scanning calorimeter (DSC) measurements performed on a Mettler Toledo DSC 1 under a nitrogen atmosphere. Each sample (about 4 mg) was heated from -10 to 200 °C at a heating rate of 10°C/min.

3. RESULTS AND DISCUSSION

3.1. Mechanical properties

Uniaxial tensile tests were performed to evaluate the mechanical properties of neat epoxy, GNPs/epoxy and MWCNTs/epoxy nanocomposites with various weight fractions ranging from 0.3 % to 1.0 % under the loading

rate of 1 mm/min. To ensure the reproductivity of the results, three samples of each group were prepared and tested. The results presented in this study are the average values of these samples. The stress and strain relationships for neat epoxy and epoxy reinforced with GNPs at various contents ranging from 0.3 wt.% to 1 wt.% are plotted in Fig. 2. The figure shows that both the elastic modulus and tensile strength of GNP/epoxy are higher than that of neat epoxy. However, the fracture strain of GNP/epoxy exhibits in the opposite trend which is decreasing with the increase of the GNP loading. The numerical values of elastic modulus, yielding stress, ultimate tensile strength, fracture strain and fracture modulus are presented in Table 1. The elastic modulus (2.80 GPa) of GNPs/Epoxy nanocomposite with 1 % weight fraction is about 40.7 % larger than the neat epoxy (1.99 GPa). Incorporation of 1 % weight fraction of GNPs increases the tensile strength of the neat epoxy by 25.8 % from 40.29 MPa to 50.67 MPa. At the same time, the fracture strain of the neat epoxy decreased by 33.7 % from 0.0418 to 0.0277 as the concentration of GNPs increases to 1 wt.%. The elastic modulus and ultimate strength are significantly improved with a comprised fracture strain. The results also show that the stiffness and strength of the epoxy are increased as the concentration of GNPs increased while the ductility is reduced as the GNPs increased. The increase in the elastic modulus and ultimate strength of the GNPs/Epoxy nanocomposites could be related to the uniform dispersion of GNPs and its anisotropic orientation in epoxy matrix, resulting in an efficient stress transfer from epoxy matrix to the nanofillers.

Fig. 3 plots the stress and strain relationships of MWCNTs/epoxy nanocomposites with different MWCNT loadings varying between 0.3 and 1 wt.%. The elastic modulus, yielding stress, ultimate tensile strength, fracture strain and fracture modulus of nanocomposites can be determined from the stress and strain relationships as listed in Table 2. In particular, the table shows that both the elastic modulus and ultimate tensile strength increase as the MWCNT loading increased, while the fracture strain and fracture modulus decrease as the MWCNT loading increased. The elastic modulus of the MWCNT reinforced epoxy increased from 1.99 GPa to 2.40 GPa as the MWCNT loading increased from 0 % to 1.0 wt.%. In addition, the ultimate tensile strength of the MWCNT reinforced epoxy increased from 40.29 MPa to 48.87 MPa as the MWCNT loading increased from 0 % to 1.0 wt.%. Meanwhile, the elongation at break decreased from 4.18 % to 3.49 % as the MWCNT loading increased from 0 % to 1.0 wt.%.

Table 1. The averaged values of the mechanical properties of GNPs/Epoxy nanocomposites with different weight fractions of GNPs

Weight fraction	0 %	0.3 %	0.5 %	0.8 %	1 %
Mechanical properties					
Young's modulus, GPa	1.99 ± 0.02	2.23 ± 0.01	2.42 ± 0	2.64 ± 0.03	2.8 ± 0.05
Yield stress, MPa	31.18 ± 0.27	34.89 ± 0.18	35.46 ± 0.91	36.29 ± 0.05	37.86 ± 0.46
Tensile strength, MPa	40.29 ± 0.13	44.23 ± 0.29	45.13 ± 0.17	49.49 ± 0.38	50.67 ± 0.47
Fracture strain	0.0418 ± 0.0007	0.0333 ± 0.0026	0.031 ± 0.0025	0.0298 ± 0.0015	0.0277 ± 0.0009
Fracture modulus, MPa	1.18 ± 0.01	1.05 ± 0.02	1.02 ± 0.01	0.96 ± 0.06	0.86 ± 0.06

Table 2. The averaged values of the mechanical properties of MWCNTs/Epoxy nanocomposites with different weight fractions of MWCNTs

Weight fraction	0 %	0.3 %	0.5 %	0.8 %	1 %
Young's modulus, GPa	1.99 ± 0.02	2.17 ± 0.01	2.27 ± 0.02	2.34 ± 0.02	2.40 ± 0.01
Yield stress, MPa	31.18 ± 0.27	33.55 ± 0.22	34.20 ± 0.03	35.78 ± 0.27	36.69 ± 0.22
Tensile strength, MPa	40.29 ± 0.13	44.66 ± 0.14	45.26 ± 0.2	47.42 ± 0.27	48.87 ± 0.15
Fracture strain	0.0418 ± 0.0007	0.0401 ± 0.0001	0.0384 ± 0.0003	0.0355 ± 0.0002	0.0349 ± 0.0001
Fracture modulus, MPa	1.18 ± 0.01	1.27 ± 0.01	1.22 ± 0.02	1.15 ± 0.01	1.16 ± 0.01

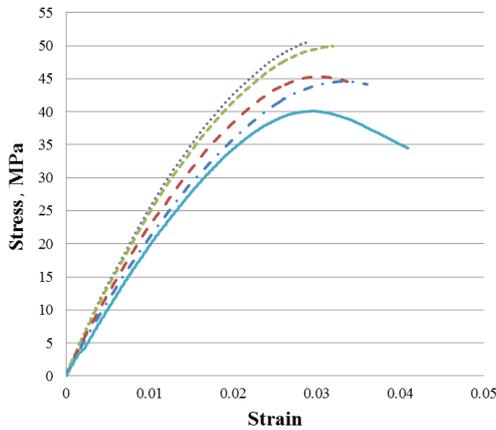


Fig. 2. Stress and strain relationships for GNPs/epoxy nanocomposites with different weight fractions of GNPs

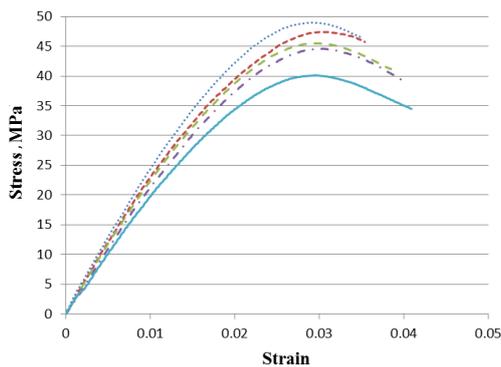


Fig. 3. Stress and strain relationships for MWCNTs/epoxy nanocomposites with different weight fractions of MWCNTs

Graphene can be used to generate a variety of carbon-based nanostructures. For example, a single layer of graphene can be rolled up to construct a single-walled carbon nanotube (SWCNT). Multiple graphene layers are rolled up to form multiple concentric SWCNTs with the same axis comprising multi-walled carbon nanotube (MWCNT). All the above carbon-based nanostructures exhibit excellent mechanical properties (such as elastic modulus and ultimate tensile strength) due to the sp^2 carbon bonding network. The aim of this work is to investigate which of these nanofillers has the best performance of transferring their mechanical properties to the polymer-based nanocomposites. To examine this, we compared the elastic modulus, yielding stress, ultimate tensile strength, and fracture strain of GNPs/Epoxy and MWCNTs/Epoxy nanocomposites for various weight fractions ranging from 0.3 % to 1.0 %, as shown in Fig. 4, Fig. 5, Fig. 6, and Fig. 7, respectively.

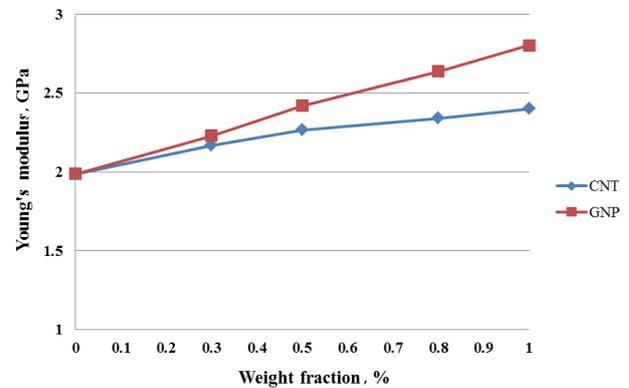


Fig. 4. Comparison of Young's modulus between GNPs/Epoxy and MWCNTs/epoxy nanocomposites with various weight fractions

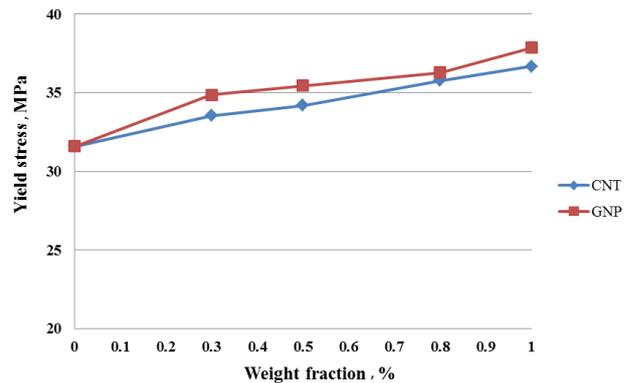


Fig. 5. Comparison of yield stress between GNPs/Epoxy and MWCNTs/epoxy nanocomposites with various weight fractions

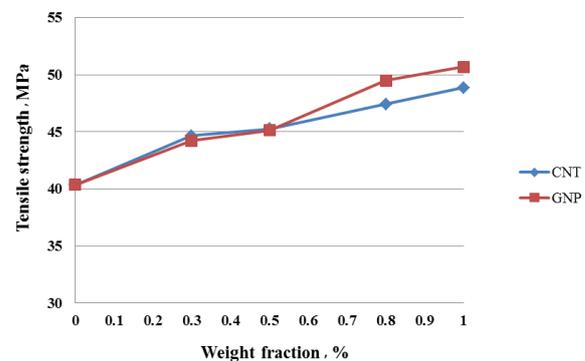


Fig. 6. Comparison of tensile strength between GNPs/Epoxy and MWCNTs/epoxy nanocomposites with various weight fractions

Experimental results indicate that the elastic modulus, yielding stress and ultimate tensile strength of the GNPs/Epoxy nanocomposite are higher than that of

MWCNTs/Epoxy nanocomposite, while the fracture strain is lower than that of the MWCNTs/Epoxy nanocomposite. It can be observed that the capability of GNPs in strengthening the nanocomposite is clearly superior to MWCNTs. The better performance of GNPs over MWCNTs in terms of mechanical properties could be attributed to the high specific surface area resulting in an improvement of nanofiller-matrix adhesion and interlocking. The reinforcement effect of the GNP fillers is the good interfacial interactions between GNPs and epoxy macromolecules.

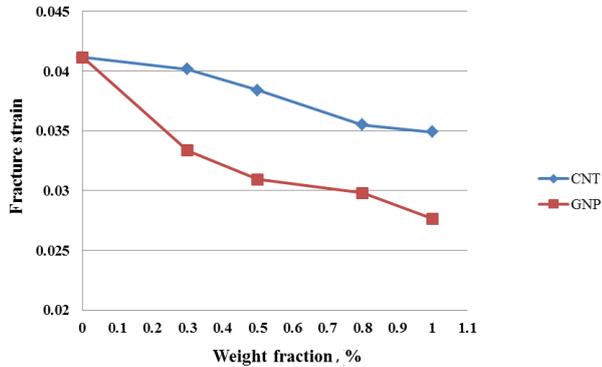


Fig. 7. Comparison of fracture strain between GNPs/Epoxy and MWCNTs/epoxy nanocomposites with various weight fractions

3.2. Fracture surface morphology

To better understand the enhancement of mechanical properties of GNPs/Epoxy nanocomposites, the morphologies of the fractured surfaces of neat epoxy and GNPs/Epoxy nanocomposites were examined using SEM. Figure 10 shows the SEM images of the fractured surfaces. It can be seen that neat epoxy possesses a smooth fractured surface as illustrated in Fig. 8 a. In comparison, the GNPs/Epoxy nanocomposites with 0.3 wt.%, 0.5 wt.% and 1.0 wt.% of graphene contents exhibit rough fractured surfaces as shown in Fig. 8 b–d, and this can be attributed to the strong interfacial adhesion and good compatibility between the epoxy matrix and GNPs. Such strong interfacial adhesion is in favor of the stress transfer from the epoxy matrix to the graphene, resulting in an improvement of the Young's modulus and tensile strength of the nanocomposites while compares with those of the neat epoxy. Another important factor concerning the reinforcement effect of the GNPs is the better dispersion of the GNPs in the epoxy matrix as shown in Fig. 8 b–d, than the dispersion of the MWCNTs as shown in Fig. 9.

3.3. Thermal property

Differential scanning calorimeter (DSC) can be used to study the thermal stability of carbon-based materials and the relative information of the molecules that are grafted to the graphene. In this work, the influences of the GNPs and MWCNTs on the glass transition temperature (T_g) of epoxy-based nanocomposites were investigated by DSC. The DSC thermograms of the GNPs/Epoxy and MWCNTs/Epoxy nanocomposites are shown in Fig. 10 and Fig. 11, respectively.

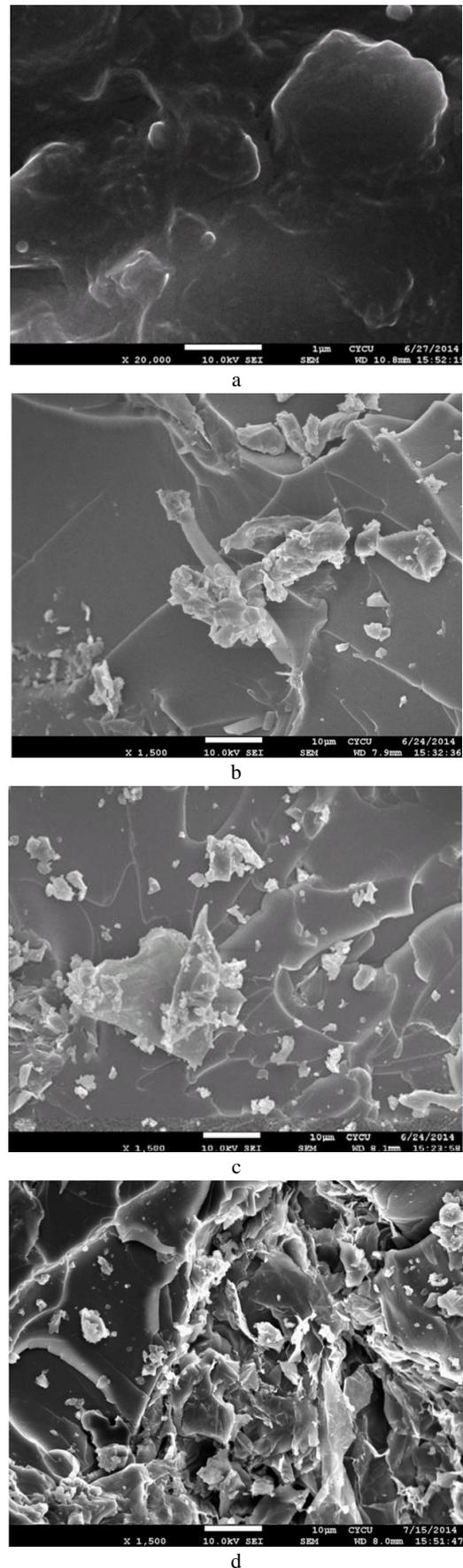
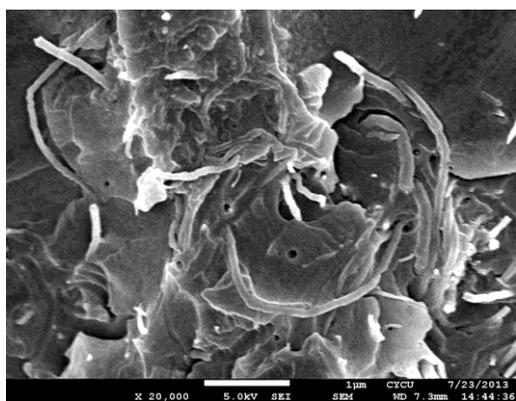
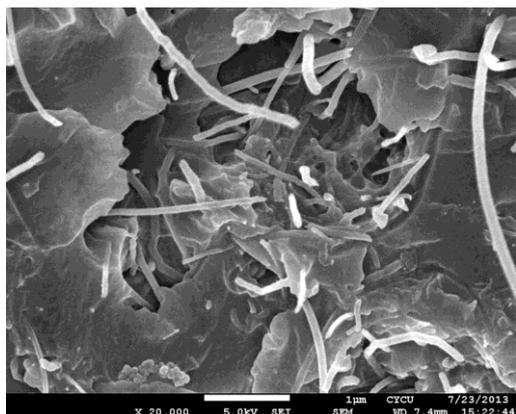


Fig. 8. SEM images of fractured surfaces of: a–neat epoxy; b–0.3 wt.% GNP; c–0.5 wt.% GNP; d–1.0 wt.% GNP



a



b

Fig. 9. SEM images of fractured surfaces of: a–0.3 wt.% MWCNT; b–0.5 wt.% MWCNT

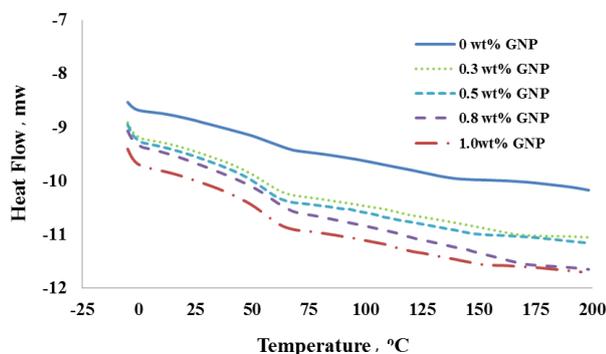


Fig. 10. DSC thermograms of GNP/Epoxy nanocomposite with various GNP contents

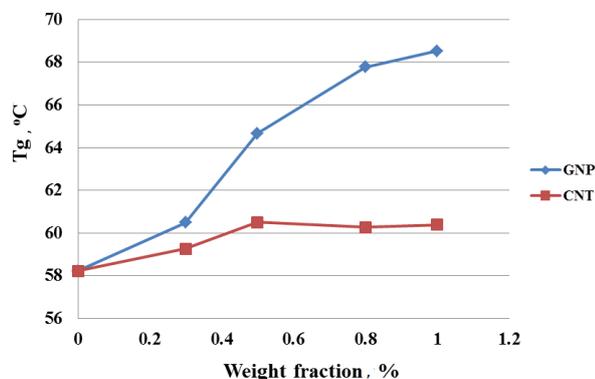


Fig. 11. DSC thermograms of MWCNTs/Epoxy nanocomposite with various MWCNT contents

The results of the glass transition temperature of MWCNTs/Epoxy and GNPs/Epoxy nanocomposites with various weight fractions are listed in Table 3.

Table 3. The averaged values of the glass transition temperature of MWCNTs/Epoxy and GNPs/Epoxy nanocomposites with various weight fractions

Weight fraction	Glass transition temperature T_g , °C	
	MWCNTs/Epoxy	GNPs/Epoxy
0 %	58.22 ± 0.53	58.22 ± 0.53
0.3 %	59.26 ± 0.62	60.49 ± 0.25
0.5 %	60.50 ± 0.48	64.67 ± 0.36
0.8 %	60.27 ± 0.24	67.77 ± 0.091
1 %	60.38 ± 0.37	69.18 ± 0.53

It shows that the glass transition temperature of GNPs/Epoxy nanocomposite increases from 58.22 °C to 69.18 °C as the weight fraction of GNP increases from 0 % to 1 % which is about 18.8 % higher than that of the neat epoxy. This could be attributed to a large aspect ratio and strong interaction between graphene and the epoxy matrix, restricting the movement of the polymer chains. In contrast, the addition of MWCNTs on the epoxy exhibits a little effect on the glass transition temperature of the nanocomposite. As the weight fraction of the MWCNT increases from 0 % to 1 %, the glass transition temperature increases from 58.22 °C to 69.18 °C which is only 3.7 % higher than that of the neat epoxy. The comparison of the glass transition temperature between MWCNTs/Epoxy and GNPs/Epoxy nanocomposites for a variety weight fractions is shown in Fig. 12. It can be seen that GNPs perform significantly better than that of MWCNTs.

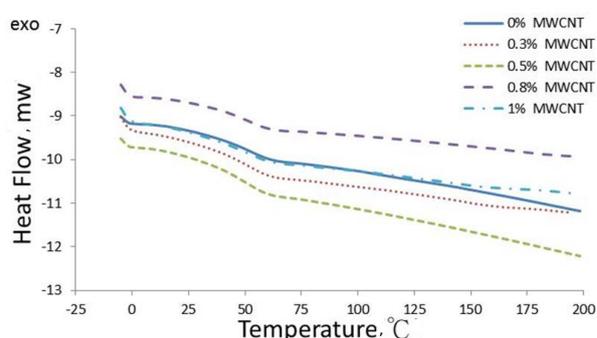


Fig. 12. Comparison of glass transition temperature between GNPs/Epoxy and MWCNTs/epoxy nanocomposites with various weight fractions

4. CONCLUSIONS

Two different types of nanocomposites were successfully prepared by incorporating GNPs and MWCNTs into epoxy matrix. The effects of GNPs and MWCNTs on the mechanical and thermal properties of the nanocomposites were studied. The elastic modulus of the GNPs/Epoxy nanocomposite was 40.7 % greater than the neat epoxy as compared to 20.6 % increase for MWCNTs/Epoxy nanocomposite at the same weight fraction of 1 %. The ultimate tensile strength of the neat epoxy was increased by 25.8 % with 1 wt.% of GNPs compared to an increase of 21.3 % for 1 wt.% of MWCNTs. The glass transition temperature of neat epoxy

was increased by 18.8 % with GNPs compared to an increase of 3.7 % for MWCNTs. Experimental results demonstrate that the GNPs is superior to the MWCNT. The GNPs/Epoxy nanocomposites exhibit a significant improvement on the mechanical properties such as elastic modulus, yielding stress and ultimate tensile strength. The outperformance of GNPs over MWCNTs in terms of mechanical properties could be attributed to the high specific surface area resulting in an improvement of nanofiller-matrix adhesion and interlocking.

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