INVESTIGATING THERMAL AND MECHANICAL PROPERTIES OF GRAPHENE/EPOXY NANOCOMPOSITES - EXPERIMENTS

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Abstract

The research investigated the thermal and mechanical properties of graphene/epoxy nanocomposites. Pristine graphene and functionalized graphene were used as nano-reinforcement in the nanocomposites. The functional groups grafted on the functionalized graphene were characterized through Fourier transform infrared spectra (FTIR) and X-ray photoelectron spectroscopy (XPS). Results indicated that the pristine graphene contains OH functional group. On the other hand, there are two kinds of functionalized graphene, one contains carboxyl (COOH) group and the other contains COOH and amine (NH2) groups. It was found that the graphene containing NH_2 and COOH functional groups exhibits superior reinforcement than the graphene with only COOH functional group. The improvement in mechanical and thermal properties of nanocomposites may be attributed to the enhanced interfacial thermal conductance and interfacial interaction caused by the functional groups. In addition, Young's modulus and thermal conductivity of the nanocomposites increase when the graphene loading increases. However, the fracture toughness and tensile strength of the nanocomposites attain peak values when the functionalized graphene loading is 0.1 wt%.

1. Introduction

With an exceptional Young's modulus and thermal conductivity, graphene has been extensively employed as reinforcements in polymeric nanocomposites [1-3]. Ganguli et al. [4] modified the graphene surface by using γ-APS and combined the graphene with an epoxy resin to form nanocomposites. The thermal conductivity of nanocomposites containing 20 wt% modified graphene was enhanced by 28 times compared with that of nanocomposites with pristine graphene. Other studies have also reported the enhancements of thermal conductivity achieved using modified graphene [5, 6]. In addition to thermal conductivity, the fracture toughness and moduli of nanocomposites with modified graphene have been investigated by many scholars [7-10]. In this study, the effects of pristine graphene and functionalized graphene with NH_2 and COOH functional groups on the mechanical and themal properties of nanocomposites were systematically examined. The graphene loadings employed in the nanocomposites were 0.1, 0.3, 0.5, and 1 wt%. The mechanical and thermal properties of the nanocomposites were measured using tensile tests, Mode I fracture tests, and hot disk tests, respectively.

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2. Experiments

2.1. Fabrication of Graphene/Epoxy Nanocomposites

To investigate the influences of functionalized graphene on the mechanical properties of graphene/epoxy nanocomposites, three types of graphene were introduced to the nanocomposites. The first was pristine graphene, and the remaining two were functionalized graphene. The pristine and functionalized graphene were obtained from Enerage Inc., Taiwan. Liquid phase exfoliation [11] was used to fabricate the pristine graphene. On the other hand, the functionalized graphene was fabricated through gas adsorption together with iso-phthalic acid, IPA, and melamine as a modification reagent. The epoxy resin used in this study was the diglycidyl ether of bisphenol-F (DGEBF) resin BONTEPONE 1070, which has an epoxy equivalent weight of 168, and the curing agent was the triethylenetetramine (TETA) agent BONTAMINE 7100H-100, which has an active hydrogen equivalent weight of 28. Both the epoxy resin and curing agent were supplied by the True Time Industrial Corporation in Taiwan. Four graphene loadings, 0.1, 0.3, 0.5, and 1 wt%, were used in fabricating the graphene nanocomposites. The dispersing procedure is described in [12]. Mechanical mixing by sonication is typically the main process for dispersing functionalized graphene in epoxy resin.

2.2. Characterization of Graphene

The functional groups grafted on the graphene were analyzed using Fourier transform infrared (FTIR) spectra [12]. One of the functionalized graphene samples contained the COOH functional group, and the other functionalized graphene contained both the NH_2 and COOH functional groups. Subsequently, the functionalization ratios (FRs) of the functional groups grafted onto the functionalized graphene were further quantified using an X-ray photoelectron spectroscopy (XPS) technique [13]. Figure 1 shows the XPS spectra of the functionalized graphene. For the COOH- and COOH+NH_2-modified graphene, the figure shows the peak intensities of the binding energy for carbon and oxygen atoms appearing at 284 and 534 eV, respectively.

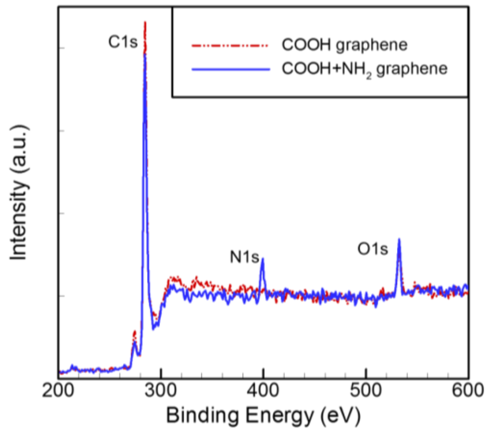

Figure 1. XPS spectra of functionalized graphene.

However, the peak intensity of nitrogen atoms (approximately 400 eV) was observed only in the COOH+NH_2 graphene. The peak area of carbon atoms was further differentiated by deconvolutions associated with different C groups. The C1s peaks for the COOH-modified graphene could be divided into sp^2 C–C (284.5eV) and COOH (288.9eV), as shown in Figure 2(a). Therefore, the FRs of the different groups could be determined using the ratio of the corresponding area to the full area of carbon atoms [13]. Table 1 shows the corresponding results. Similarly, the total C1s peaks for the COOH+NH_2 functionalized graphene were divided into C–C (284.5 eV), C–N (286.9 eV), and COOH (288.9 eV), as shown in Figure 2(b) and the FRs are also listed in Table 1. The COOH modified graphene contains a 4.1% FR for the COOH group, and COOH+NH_2-modified graphene shows 4.7% and 9.7% FRs for the COOH and NH_2 groups, respectively.

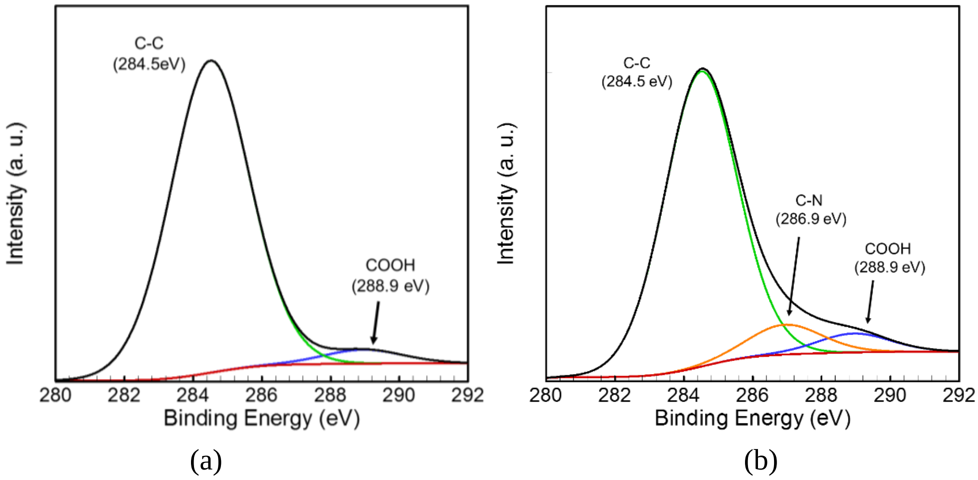

Figure 21. XPS spectra of C1s for: (a) COOH modified graphene; (b) COOH+NH_2 modified graphene.

Table 1. Functionalization ratio of functionalized graphene.

Group	FR (%)	
	COOH-graphene	COOH+NH_2 - graphene
COOH	4.1	4.7
C-N	0.0	9.7

2.3. Tensile Test

The mechanical properties of the graphene/epoxy nanocomposites were measured by means of tensile tests based on the ASTM D638-10 standard [14]. Back-to-back strain gages were adhered on the center of the specimens to eliminate the bending effect and measure the strain history during the tests. The tensile tests were performed on a hydraulic MTS machine at a displacement rate of 2 mm/min, and the corresponding stress histories were obtained from a load cell mounted on the loading fixture. Figures 3 illustrates the influences of the pristine graphene and functionalized graphene on the tensile modulus of nanocomposites. At the same graphene loading, the nanocomposites with functionalized graphene exhibit apparently higher moduli than those of nanocomposites with pristine graphene. Furthermore, the functionalized graphene with COOH and NH_2 functional groups can provide superior reinforcement than that only containing the COOH group. The enhancement of moduli achieved by adding the functional groups could be attributed to the modification of the interfacial properties between the graphene and surrounding matrix.

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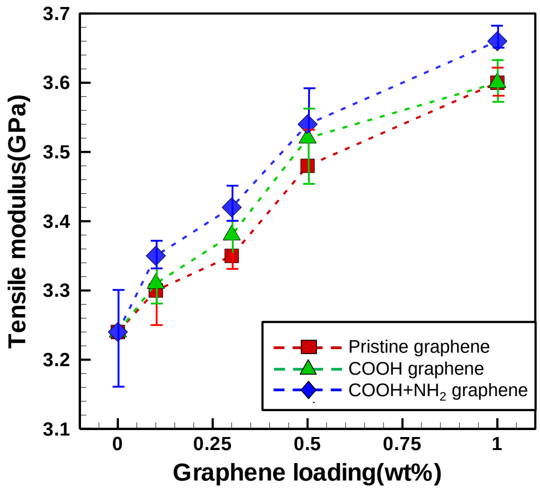

Figure 3. Tensile moduli of graphene nanocomposites with various graphene loadings.

2.4. Fracture Test

To investigate the effect of graphene on the Mode I fracture toughness (K_{IC}) of nanocomposites, single-edge-notch bending (SENB) specimens were fabricated and then employed in three-point bending tests. The three-point bending experiments were conducted according to ASTM D5045-99 standards [15]. The procedure for preparing the SENB specimens is described elsewhere [16]. Figure 4 illustrates the fracture toughness of the nanocomposites with different loadings of graphene and functionalized graphene.

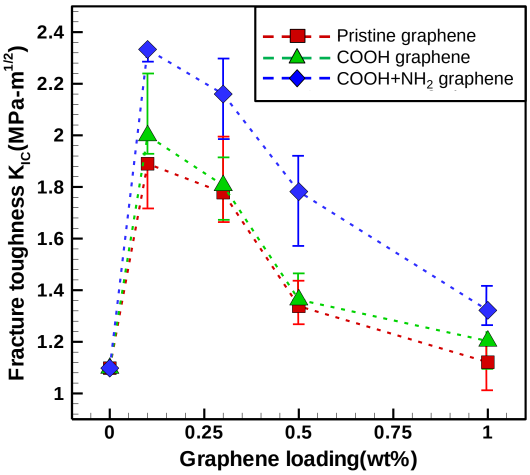

Figure 4. Mode I fracture toughness of graphene nanocomposites with various graphene loadings.

The fracture toughness of the nanocomposites reaches peak values at the graphene loading of 0.1 wt% and then decreases as the graphene loading increases until the graphene loading is 1 wt%. Moreover, at the same graphene loading, the nanocomposites with modified graphene exhibit higher fracture toughness than that of the nanocomposites with pristine graphene. In addition, the graphene nanocomposites with the COOH and NH_2 functional groups also provide greater fracture toughness than those with the COOH functional group.

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2.5. Thermal Conductivity

The thermal conductivity of the graphene nanocomposites was measured using a hot disk machine [17]. During the tests, a heating coil with a radius of 3.2 mm was sandwiched between the specimens. The electrical power was set as 0.008 W, and the test duration was 40 seconds. Specimens with various loadings of pristine and functionalized graphene were tested, and the thermal conductivity is shown in Figure 5. The thermal conductivity of the nanocomposites increases as the graphene loading increases. Furthermore, at the same graphene loading, the nanocomposites with COOH and NH_2 modified graphene exhibit higher thermal conductivity than that of the nanocomposites with COOH modified or pristine graphene.

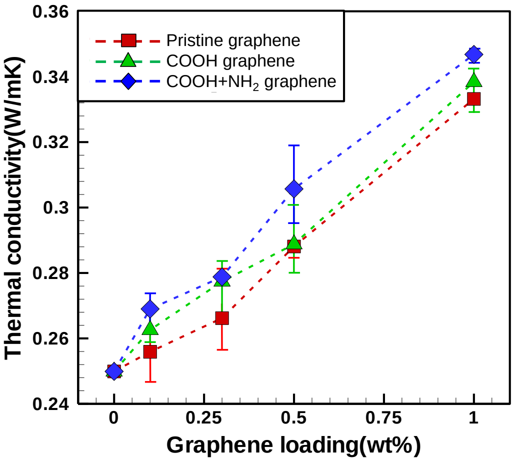

Figure 5. Thermal conductivity of graphene nanocomposites with various graphene loadings.

3. Conclusions

The mechanical and thermal properties of graphene/epoxy nanocomposites with different loadings of functionalized graphene were investigated. The experimental results indicated that the moduli of the nanocomposites increase as the graphene loading increases. However, the fracture toughness of the nanocomposites attain peak values at the graphene loading of 0.1 wt% and then decrease as the graphene loading increases. The COOH and NH_2 modified graphene exhibits superior reinforcement as compared to the pristine and COOH modified graphene in terms of the moduli, thermal conductivity, and fracture toughness of the nanocomposites.

Acknowledgments

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