

Research Article

Synergetic Effects of Mechanical Properties on Graphene Nanoplatelet and Multiwalled Carbon Nanotube Hybrids Reinforced Epoxy/Carbon Fiber Composites

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Graphene nanoplatelets (GNPs) and carbon nanotubes (CNTs) are novel nanofillers possessing attractive characteristics, including robust compatibility with most polymers, high absolute strength, and cost effectiveness. In this study, an outstanding synergetic effect on the graphene nanoplatelets (GNPs) and multiwalled carbon nanotubes (CNTs) hybrids were used to reinforce epoxy composite and epoxy/carbon fiber composite laminates to enhance their mechanical properties. The mechanical properties of CNTs/GNPs hybrids on a fixed weight fraction (1 wt%) with mixing different ratio reinforced epoxy nanocomposite, such as ultimate tensile strength and flexure properties, were investigated. The mechanical properties of epoxy/carbon fiber composite laminates containing different proportions of CNTs/GNPs hybrids (0.5, 1.0, 1.5 wt%) were increased over that of neat laminates. Consequently, significant improvement in the mechanical properties was attained for these epoxy resin composites and carbon fiber-reinforced epoxy composite laminates.

1. Introduction

CNTs are one-dimensional carbon nanomaterials that possess high strength, high flexibility, low mass density, and large aspect ratio [1]. CNTs have a unique combination of mechanical, electrical, and thermal properties that render them excellent candidates to substitute or complement conventional nanofillers in fabricating multifunctional polymer nanocomposites [2–4]. However, the development of CNT reinforced polymer nanocomposites has been impeded by their cost and aggregation in polymer matrix. CNTs tend to agglomerate due to their large aspect ratio and Van der Waals forces, which leads to their dispersion difficultly in polymer matrix [5].

The GNPs planar structure provides a two-dimensional path for phonon transport, and the ultrahigh surface area allows a large surface contact area with polymer resulting in enhancement of the composite thermal conductivity [6–8].

However, the large surface area between GNPs which is GNP planar nanosheets results in large Van der Waals forces and strong π - π interactions [9–11]. Thus, the performance of GNPs based polymer composites is limited by the aggregation and stacking of GNP sheets. Since the physicochemical properties of aggregated GNPs are similar to those of graphite with its relatively low-specific surface area, the performance of GNPs will suffer significantly from reduced performance. This is an important issue if GNPs potential as a polymer composite reinforcing materials is realized [12, 13].

Epoxy is widely applied in advanced carbon fiber reinforce plastic (CFRP) due to their good mechanical performance, process-ability, compatibility with most fibers, chemical resistance, wear resistance, and low cost. However, these materials are relatively brittle, which is detrimental to the interlaminar properties between matrix and reinforcement. The addition of CNTs or GNPs to improve the interfacial

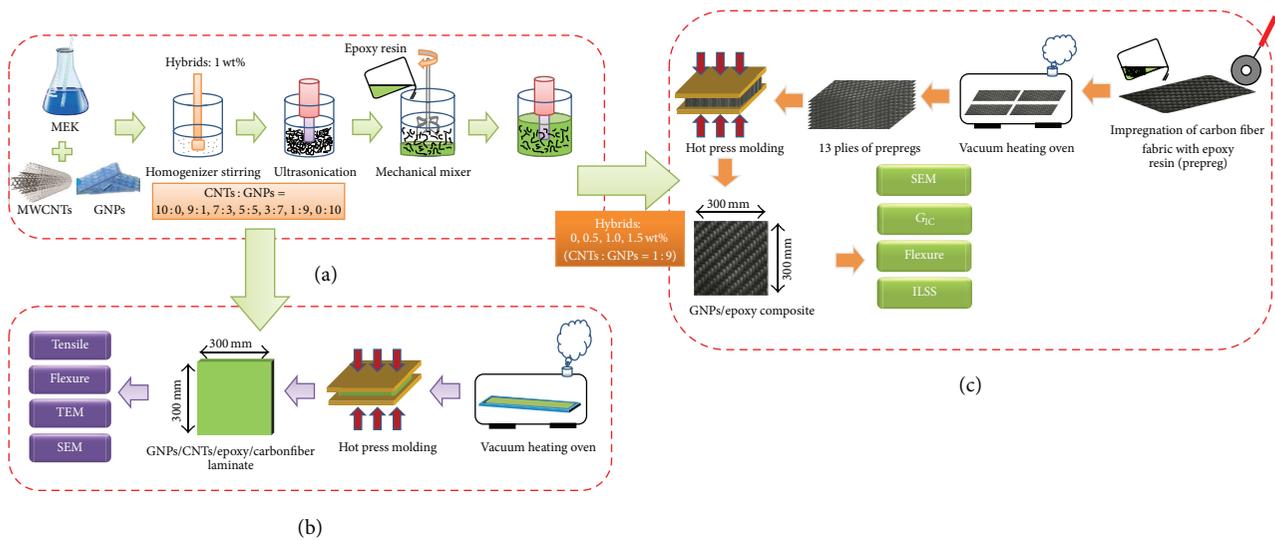


FIGURE 1: A schematic illustration of the fabrication of composites: (a) CNTs/GNPs/epoxy resin solution; (b) CNTs/GNPs/epoxy nanocomposites; (c) CNTs/GNPs/epoxy/carbon fiber composite laminates.

strength of laminates has been demonstrated. Adding carbon base nanofillers, such as CNTs and GNPs, can improve mechanical properties of polymer composites, due to their unique nanostructures, and superior properties.

In our previous research [14], GNPs were used to reinforce epoxy composite and epoxy/carbon fiber composite laminates to enhance their mechanical properties. The mechanical properties of GNPs/epoxy nanocomposite, such as ultimate tensile strength and flexure properties, were investigated.

To the best of our knowledge, very few studies have so far been reported on the fabrication of CNTs/GNPs hybrids reinforced CFRP composites using manufacturing of prepregs [15]. GNPs and CNTs are difficult to process because of the issues associated with agglomeration and lack of interfacial interactions with polymers. None of the previous studies, nevertheless, have given due attention to the influence of adding CNTs/GNPs hybrids on the solvent prepreg process. Therefore, it is necessary to understand the role of adding CNTs/GNPs hybrids into the polymer matrix, which will largely affect the impregnation of fibres and prepreg processing conditions.

In this study, epoxy resin containing uniformly dispersed 1 wt% CNTs/GNPs hybrids with different mixing ratio (i.e., 10:0, 9:1, 7:3, 5:5, 3:7, 1:9, and 0:10) and prepared the CNTs/GNPs/epoxy nanocomposites. Mechanical properties of the nanocomposite, including ultimate tensile strength, flexural strength, and flexural modulus, were investigated.

The solution of epoxy resin containing the fixed mixing ratio of CNTs/GNPs hybrids (1:9) on the different contents (i.e., 0, 0.5, 1.0, and 1.5 wt%) permeate through a carbon fiber cloth was used to prepare the epoxy/carbon fiber composite laminates. The process was used to investigate if CNTs/GNPs hybrids improved the mechanical properties of carbon fiber-reinforced epoxy resin composite laminates.

Finally, the fracture surface of the specimen was investigated using scanning electron microscopy (SEM) and transmission electron microscopy (TEM) to determine the role of adding CNTs/GNPs hybrids into the epoxy and epoxy/carbon fibers composite laminates.

2. Experimental

2.1. Preparation of CNTs/GNPs/Epoxy Resin Solution. The maleic anhydride, MA, was used to modify carbon nanomaterials. Graphene nanoplatelets, GNPs with a thickness of 5–25 nm are obtained from Xiamen Knano Graphene Technology Co., Ltd. China. Multiwalled carbon nanotubes, CNTs, with 1–25 μm in length and 20 nm in diameter are obtained from Nanocyl Co., Ltd. Belgium. The GNPs and CNTs were modified by Hanyu Material Co., Ltd. Taiwan. The CNTs/GNPs hybrids were used for reinforcement in this study. The CNTs/GNPs/methyl ethyl ketone (MEK) solution was stirred for 10 minutes using a homogenizer. The solution was then vibrated by ultrasonication for 90 minutes to enable the CNTs/GNPs hybrids to disperse uniformly throughout the MEK solution. The CNTs/GNPs/MEK solution was mixed with epoxy resin and hardener (EPO-622 epoxy resin and 2-ethyl-4-methylimidazole hardener, Epotech Composite Co., Ltd, Taiwan) for 90 minutes using a mechanical stirrer and then vibrated by ultrasonication for 90 minutes to enable the CNTs/GNPs hybrids to disperse uniformly throughout the epoxy solution. A schematic illustration of the fabrication of the CNTs/GNPs/epoxy resin is shown in Figure 1(a).

2.2. Preparation of CNTs/GNPs/Epoxy Nanocomposites. The CNTs/GNPs/epoxy resin solution was placed in a heating oven for exposure at 83°C for three hours to evaporate all of the solvent and then placed in a vacuum heating oven and vacuum pumping was performed for 5 minutes

to eliminate air bubbles. The resin solution was poured into molds and then placed on a hot press machine to form the CNTs/GNPs/epoxy nanocomposites (pressed at 1500 psi and 150°C for 30 minutes). The nanocomposites were then placed in a heating oven at 140°C for 3 hours to eliminate the internal stress (postcure). A schematic illustration of the fabrication of the CNTs/GNPs/epoxy nanocomposite is shown in Figure 1(b).

2.3. Preparation of CNTs/GNPs/Epoxy/Carbon Fiber Composite Laminates

2.3.1. Impregnation of Carbon Fiber Fabric with Epoxy Resin (Prepreg). A piece of 3k carbon fiber fabric of the desired dimensions was placed on a release paper and the MEK/CNTs/GNPs/epoxy resin solution was evenly permeated on it. The carbon fiber fabric with uniformly dispersed MEK/CNTs/GNPs/epoxy resin (prepreg) was then placed in a heating oven for exposure at 83°C for three hours to evaporate all of the solvent.

2.3.2. Hot Press Molding and Postcuring. Thirteen pieces of prepregs were piled in a mold and placed on a hot press machine to prepare a CNTs/GNPs/epoxy/carbon fiber composite laminate (pressed at 1500 psi and 150°C for 30 mins). The composite laminate was then placed in a heating oven at 140°C for three hours to eliminate the internal stress of laminate (postcure) [16]. A schematic illustration of the fabrication of the composite laminate is shown in Figure 1(c).

2.4. Experimental Process

2.4.1. Experimental Process of CNTs/GNPs/Epoxy Nanocomposites. CNTs/GNPs hybrids reinforced epoxy nanocomposites containing the fixed weight fraction of CNTs/GNPs hybrids (1 wt%) with different mixing ratio (i.e., 10 : 0, 9 : 1, 7 : 3, 5 : 5, 3 : 7, 1 : 9, and 0 : 10) were fabricated. The mechanical properties of the nanocomposites, such as ultimate tensile strength, flexural strength, and flexural modulus, were investigated.

The fracture surface of the specimen was investigated using transmission electron microscopy (TEM) and field emission scanning electron microscopy (FESEM) to determine the dispersion of the CNTs/GNPs hybrids in the nanocomposites.

2.4.2. Experimental Process of Composite Laminates. CNTs/GNPs hybrids reinforced epoxy/carbon fiber composite laminates containing the fixed mixing ratio (1 : 9) and adding three proportions of CNTs/GNPs hybrids (i.e., 0.5, 1.0, and 1.5 wt%) were fabricated. The mechanical properties of the composite laminates, such as flexure properties and interlaminar shear strength, ILSS, were investigated. A double cantilever beam (DCB) specimen was employed using corrected beam theory (CBT) method for calculating G_{IC} (Mode I) in order to determine interlaminar fracture toughness evaluation of composite laminates without and with CNTs/GNPs hybrids added.

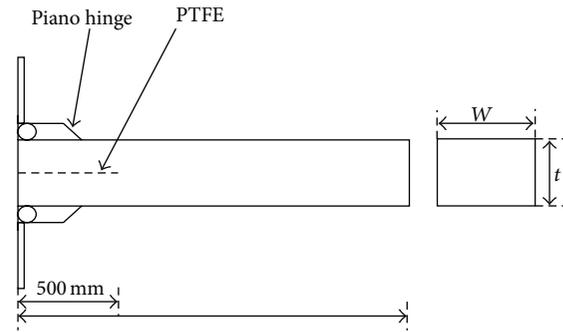


FIGURE 2: The schematic representation of the DCB specimen for Mode I (G_{IC}) test.

The fracture surface of the specimens was investigated using SEM to determine if the CNTs/GNPs hybrids would prevent the formation of pores in the laminates and makes CNTs/GNPs hybrids effective in delivering stress to improve the mechanical properties of composite laminates.

2.4.3. G_{IC} Test of Composite Laminates. Double cantilever beam (DCB) tests were used to determine the fracture resistant under Mode I opening load, in which Mode I delamination fracture energy, G_{IC} , can be followed from the British Standard, BS7991:2001. Specimen geometries were controlled to be 165 mm long beams with a 20 mm width and 3 mm thick. The specimen was prepared with a pre-inserted starter crack by a 60 μm thick nonstick film made of polytetrafluoroethylene (PTFE) to make a delamination length of 50 mm from load line, as shown in Figure 2. Two steel-alloy loading-piano hinges were bonded on either side of the specimen. DCB tests require that load, displacement, and crack length measurements are taken. To compare the G_{IC} of various amounts of CNTs/GNPs hybrids reinforced epoxy/carbon fiber composite laminates, the G_{IC} when stable crack growth reached 50 mm from the load line (i.e., the end of insert PTFE precrack) was designated as the initial G_{IC} value (G_{IC} -init). G_{IC} -init was used to analyze the influence of mixed carbon nanomaterials on the fracture toughness of epoxy resin/carbon fiber composite laminates.

G_{IC} was initially calculated by using the area method [17], which represents the area under the load-displacement curve. Area method is based on linear fracture mechanics simple beam theory, which assumes a perfectly built-in DCB specimen:

$$G_{IC} = \frac{3P\delta}{2ba}, \quad (1)$$

where P is the load (N), δ is the crack opening displacement (mm), b is the specimen width (mm), and a is the delamination length (mm). However, this underestimates the compliance ($C = \delta/P$) as the beam is not perfectly built-in. A means of correcting this effect is to apply a slightly longer delamination length of $\alpha + |\Delta|$. The $|\Delta|$ is found experimentally by plotting the cube root of compliance ($C/3$) against delamination length (α). The extrapolation of a linear fit through data yields Δ as the x -axis intercept.

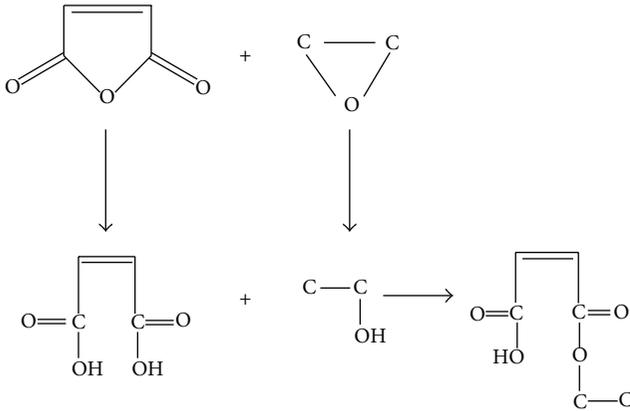


FIGURE 3: Reaction of maleic anhydride and epoxide group.

The delamination propagation values from Mode I pre-crack were used for the linear fit data. In addition, large displacement correction (F) was applied for all specimens, which contributed significantly if the δ/p ratio was larger than 0.4. This method of calculating G_{IC} is known as corrected beam theory (CBT). All initiation and propagation values were calculated according to

$$G_{IC} = \frac{3P\delta}{2b(a + |\Delta|)}F. \quad (2)$$

3. Results and Discussion

3.1. Characterization of CNTs and GNPs. Figure 3 shows the reaction of maleic anhydride (MA) and the epoxide group. This confirmed that MA can react with epoxy resin reinforcing the interfacial strength between carbon nanomaterials and the resin. The Fourier transform infrared (FT-IR) spectra images were shown in Figures 4 and 5. FT-IR was utilized to characterize the modification of CNTs and GNPs powders. The figures showed that the key absorption peak near 1,600–1,850 cm^{-1} was the $-\text{C}=\text{O}$ functional group, a standard absorption peak of acid anhydrides; these results indicated the successful MA modification of GNPs and CNTs.

3.2. Mechanical Properties of CNTs/GNPs/Epoxy Nanocomposites. Figures 6–8 and Table 1 show the flexure properties and tensile strengths of nanocomposites with CNTs/GNPs hybrids for a fixed weight fraction (1 wt%). The mechanical properties with various mixture ratios of CNTs/GNPs hybrids were collectively superior to pure resin or single type of carbon nanomaterial reinforced resins (10 : 0 and 0 : 10). This is because the reaction of MA and epoxy resin effectively strengthened the interface between the carbon nanomaterials and the resin, noticeably increasing the mechanical properties of nanocomposites. After sufficient dispersion of CNTs/GNPs hybrids in nanocomposites, the CNTs were able to permeate between the GNPs. The GNPs were also able to infiltrate between the CNTs, forming complementary structures that are able to interact and prevent restacking as a result of Van der Waals attraction [18]. Thus, the mechanical

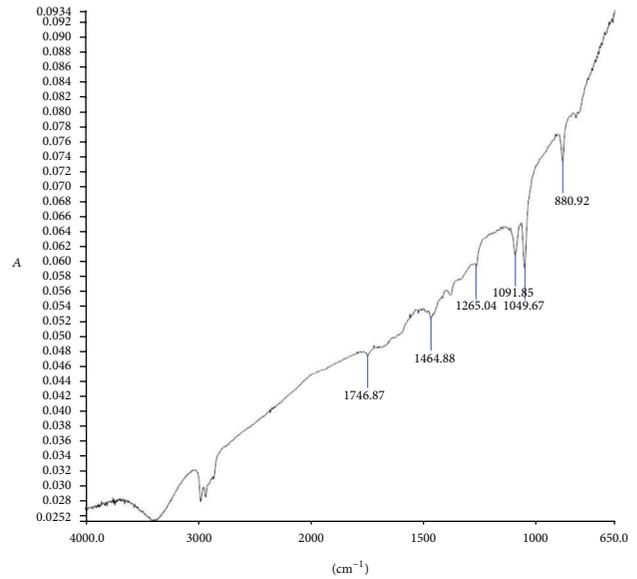


FIGURE 4: FT-IR spectra of CNTs-MA.

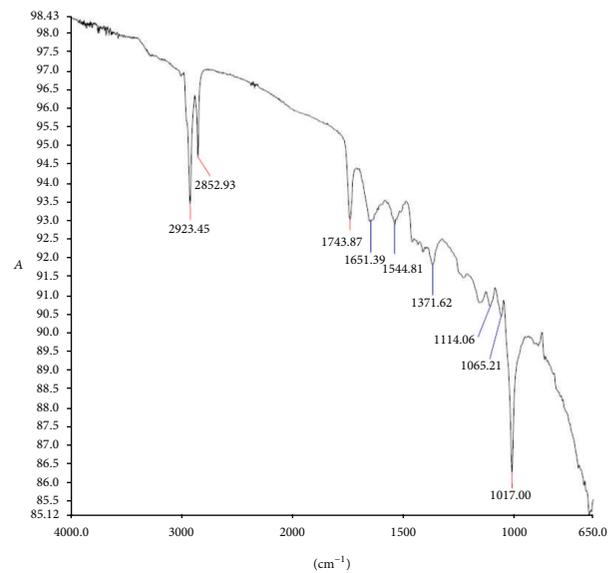


FIGURE 5: FT-IR spectra of GNPs-MA.

properties of nanocomposites containing 1 wt% CNTs/GNPs hybrids were collectively superior to nanocomposites containing 1 wt% single carbon nanomaterial and neat composite.

In Figures 7–9, when the CNTs/GNPs hybrids mixing ratios were 5 : 5 and 1 : 9 (1 wt%), the increase in mechanical properties were collectively greater than 20% of neat composite, thus inferring that these two mixing ratios can effectively inhibit the agglomeration of the two carbon nanomaterials and effectively reinforcing the epoxy resin. Two factors were proposed to explain the synergetic enhancement of CNTs/GNPs/epoxy composites: (1) flexible CNTs can construct GNPs to form 3D hybrid structure, which inhibit face to face aggregation of multigraphene platelets. This results

TABLE I: Mechanical properties of CNTs/GNPs/epoxy nanocomposites with different carbon nanomaterials mixing ratio.

Test item (unit)	CNTs/GNPs hybrids content (fixed weight fraction: 1 wt%)							
	Neat	10:0	9:1	7:3	5:5	3:7	1:9	0:10
TS (MPa)	54.9	60.2	61.2	64.1	73.4	58.1	67.0	55.6
FS (MPa)	105.9	118.1	114.5	102.7	127.8	125.7	133.8	104.5
FM (GPa)	2.2	2.59	2.62	2.40	2.71	2.64	2.76	2.53

TS: tensile strength, FS: flexural strength, and FM: flexural modulus.

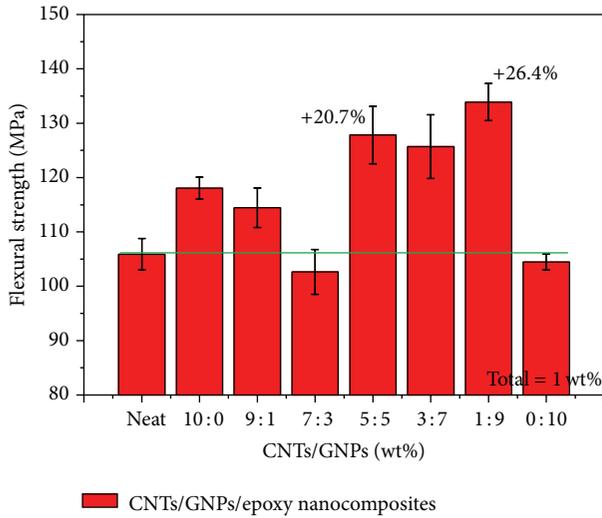


FIGURE 6: Flexural strength of CNTs/GNPs/epoxy nanocomposites.

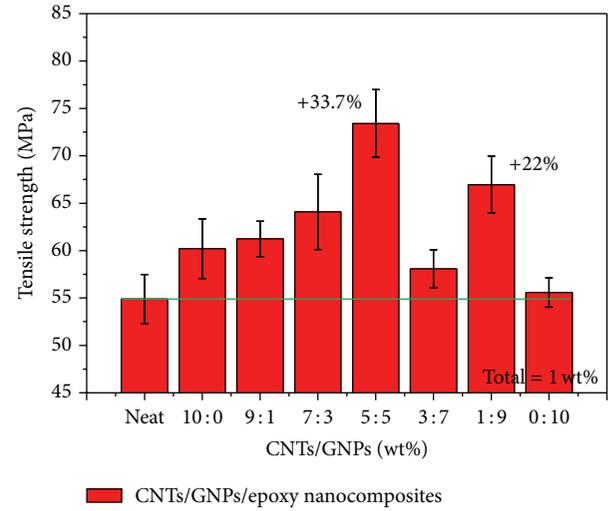


FIGURE 8: Tensile strength of CNTs/GNPs/epoxy nanocomposites.

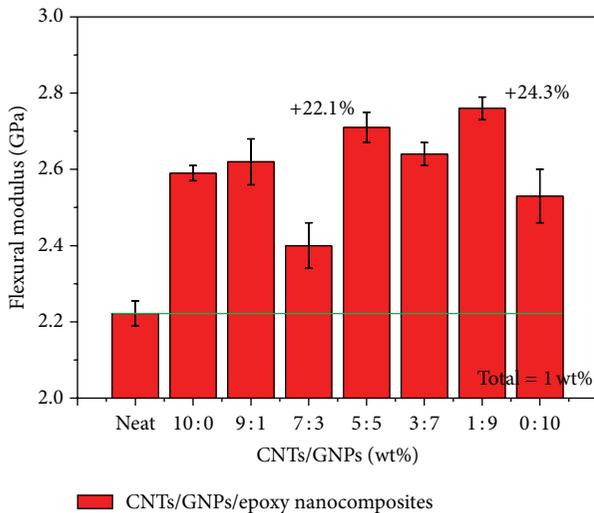


FIGURE 7: Flexural modulus of CNTs/GNPs/epoxy nanocomposites.

in a large surface area, thus increasing the contact surface area between CNTs/GNPs hybrids and epoxy matrix; (2) the CNTs can act as extended tentacles for the 3D hybrid architectures, which can become entangled with the polymer chain resulting in better interaction between CNTs/GNPs and the epoxy matrix [18].

However, the lower mechanical properties of nanocomposite with CNTs/GNPs hybrids mixing ratios were 7:3 and 0:10 (1 wt% of GNPs) which can be attributed to following effects. (1) The properties of GNPs rapidly devolve as sheets aggregate, because aggregated sheets behave like micrometer-size fillers with relatively low surface area. (2) The GNPs agglomerates would form steric obstacles, restricting polymer to flow into the agglomerates and resulting in the formation of holes and voids between GNPs and epoxy [18].

3.3. Fracture Surface of CNTs/GNPs/Epoxy Nanocomposites. Figure 9 shows the SEM images of nanocomposites containing fixed weight fraction (1 wt%) with a CNTs/GNPs hybrids mixing ratio of 5:5. Figure 10 is SEM images of nanocomposites with a CNTs/GNPs hybrids mixing ratio of 1:9. Figures 9(a) and 10(a) show that CNTs and GNPs cross-link in crevices among the corrugation area to restrain the creviced growth. Corrugation and CNTs/GNPs hybrids can increase the interfacial friction between carbon fiber and matrix to enhance the mechanical properties. However, when 0.5 wt% (5:5) and 0.1 wt% (1:9) of CNTs are mixing with GNPs and added to epoxy resins, CNTs bridged the GNPs layers as well as resin cracks, as seen in Figures 9(b) and 10(b). Moreover, Figures 9(b) and 10(b) also showed that major cracks in the laminates were restrained by GNPs; CNTs can effectively suppress minor cracks regardless of the CNTs contents which were 0.5 wt% or 0.1 wt%. Figure 11 shows the TEM images of the fracture surfaces of CNTs/GNPs/epoxy

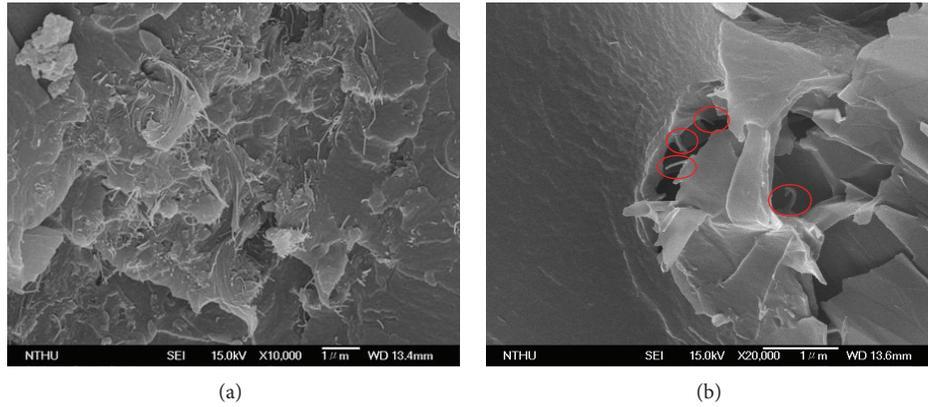


FIGURE 9: SEM images of CNTs/GNPs/epoxy nanocomposite (CNTs/GNPs = 5 : 5). (a) $\times 10,000$. (b) $\times 20,000$.

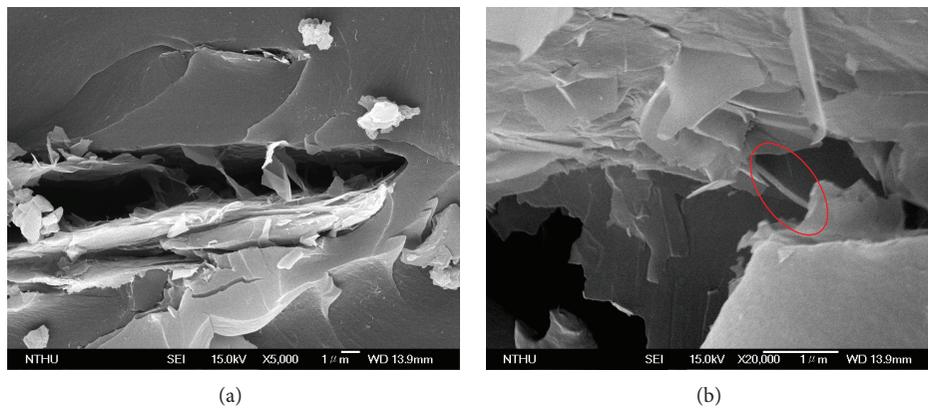


FIGURE 10: SEM images of CNTs/GNPs/epoxy nanocomposite (CNTs/GNPs = 1 : 9). (a) $\times 5,000$. (b) $\times 20,000$.

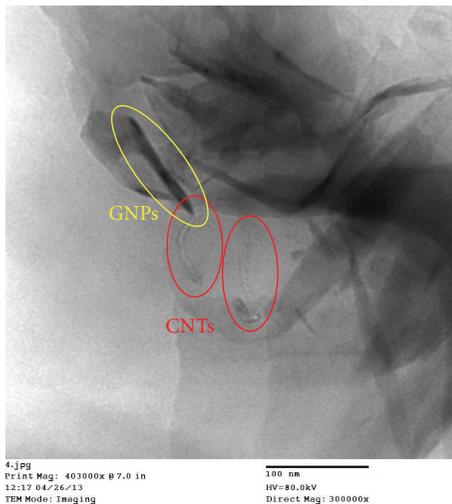


FIGURE 11: TEM image of CNTs/GNPs/epoxy nanocomposite. (CNTs/GNPs = 1 : 9).

resin nanocomposite (CNTs/GNPs = 1 : 9). For nanocomposite containing CNTs/GNPs hybrids, the TEM shows the CNTs entangled around the GNPs.

3.4. Mechanical Properties of CNTs/GNPs/Epoxy/Carbon Fiber Composite Laminates. In the previous section, CNTs/GNPs hybrids reinforced epoxy nanocomposite containing the fixed content of CNTs/GNPs hybrids (1 wt%) with different mixing ratio were fabricated and investigate their mechanical properties. The results showed that CNTs/GNPs hybrids mixing ratios of 5 : 5 and 1 : 9 were superior to other ratios. Based on the average of each mechanical property and the reference data obtained from the literature [18], CNTs/GNPs hybrids ratio of 1 : 9 with different added proportions (i.e., 0, 0.5, 1.0, and 1.5 wt%) was used for fabricating CNTs/GNPs/epoxy/carbon fiber composite laminates. Subsequently, static mechanical properties such as flexure, interlaminar shear strength (ILSS), and fracture toughness (G_{IC}) were investigated. This provided further understanding regarding the mixtures of 1D and 2D carbon nanomaterials and how their synergetic effects reinforce carbon fiber composite laminates.

Figure 12 and Table 2 show the influence of adding CNTs/GNPs hybrids on the flexural strength of epoxy/carbon fiber composite laminates. The results showed that the flexural strength of carbon fiber composite laminates increased when the amount of CNTs/GNPs hybrids added was increased. When 1 wt% of CNTs/GNPs hybrids was added,

TABLE 2: Mechanical properties of CNTs/GNPs/CFRP composite laminates.

Test item (unit)	CNTs/GNPs hybrids content (wt%) CNTs : GNPs = 1 : 9			
	0	0.5	1.0	1.5
FS (MPa)	580.6	624.8 (+7.6%)	682.1 (+17.5%)	633.8 (+9.2%)
FM (GPa)	31.3	33.4 (+6.8%)	33.7 (+7.7%)	34.4 (+10.0%)
ILSS (MPa)	54.6	76.1 (+39.3%)	76.5 (+40.0%)	72.5 (+32.7%)
G_{IC} (J/m ²)	468.9	648.7 (+38.3%)	866.8 (+84.9%)	938.0 (+100%)

FS: flexural strength, FM: flexural modulus, and ILSS: interlaminar shearing strength. G_{IC} : initial fracture toughness G_{IC} .

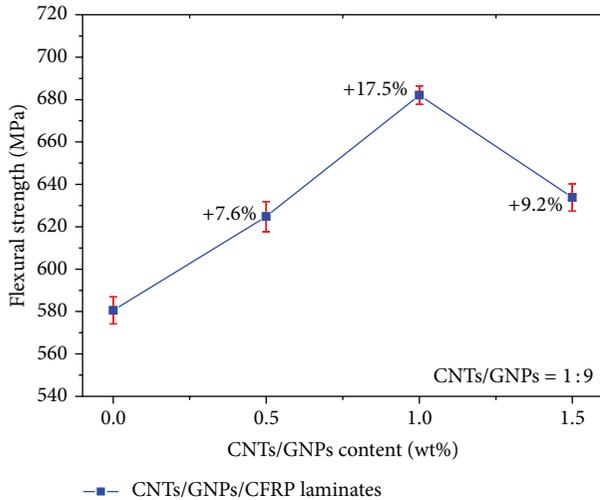


FIGURE 12: Flexural strength of CNTs/GNPs/carbon fiber composite laminates.

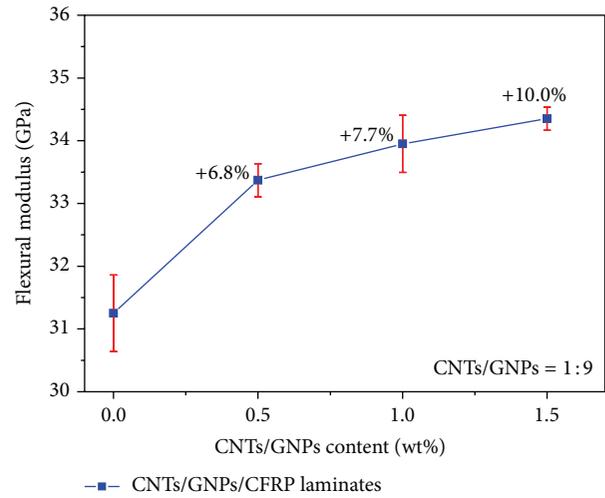


FIGURE 13: Flexural modulus of CNTs/GNPs/carbon fiber composite laminates.

the flexural strength of composite laminates increased up to 17.5%.

Figure 13 and Table 2 show the flexural modulus measured from the flexural test of CNTs/GNPs/epoxy/carbon fiber composite laminates, which is plotted as a function of CNTs/GNPs hybrids content. The modulus grew rapidly with increasing CNTs/GNPs hybrids content. When the hybrids content increased to 1.5 wt%, the highest flexural modulus was reached, and the enhancement was increased by 10%.

Figure 14 and Table 2 present the experimental results of the interlaminar shear strength (ILSS) of the epoxy/carbon fiber composite laminates without and with the three proportions of CNTs/GNPs hybrids contents. The ILSS test have seen implemented in accordance with ASTM D2344. Adding CNTs/GNPs hybrids effectively increased the ILSS of composite laminates. The ILSS of composite laminates were similar when 0.5 and 1.0 wt% of CNTs/GNPs hybrids were added, both increased up to 40%. When 1.5 wt% of CNTs/GNPs hybrids was added to composite laminates, the ILSS were slightly inferior compared to those of the former compositions, but still increased by 32.7%.

Based on the flexure properties of epoxy/carbon fiber composite laminates and results from ILSS, several contributing factors were identified.

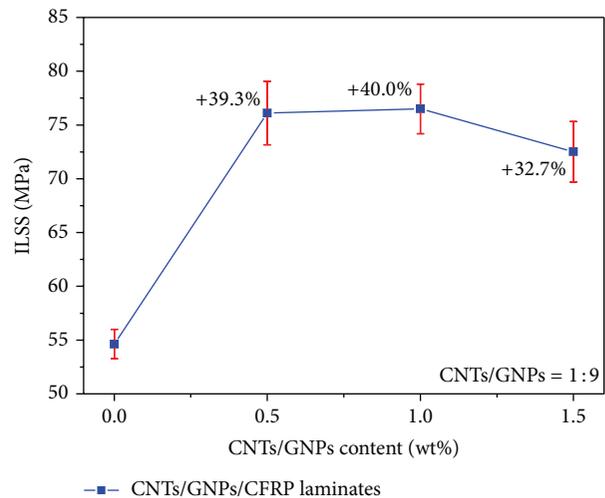


FIGURE 14: Interlaminar shear strength (ILSS) of CNTs/GNPs/carbon fiber composite laminates.

- (1) The first factor was the entanglement and the complementary between CNTs and GNPs. When CNTs were added to the GNPs, they entangled the GNPs and filled gaps between GNPs' interlayer. In addition

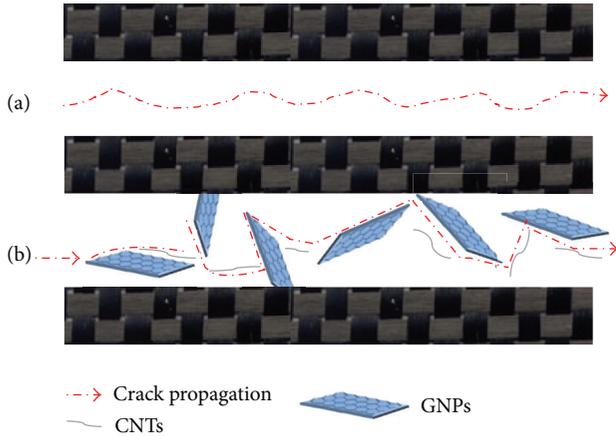


FIGURE 15: Crack propagation on carbon nanomaterials reinforced epoxy/CFRP laminate interface. (a) Neat CFRP laminate; (b) CNTs/GNPs/CFRP composite laminate.

to that CNTs/GNPs hybrids restricting cracks growth in resin, these hybrids could effectively suppress or deter crack propagation. Specifically, when GNPs restrained the development of major cracks, CNTs suppress minor cracks, as shown in Figure 15.

- (2) In general, the addition of inorganic nanoparticles (e.g., GNPs or CNTs), which results in the decrement of mobility of polymer chains, gradually became brittle and increased the modulus of the composite. Thus, the modulus of the polymer composites followed the increase with increasing proportions of carbon nanomaterials added [19, 20].

3.5. Fracture Toughness of CNTs/GNPs/Epoxy/Carbon Fiber Composite Laminates. A double cantilever beam was used in this study to investigate Mode I interlaminar fracture toughness of various amounts of CNTs/GNPs hybrids reinforced epoxy/carbon fiber composite laminates.

Figure 16 and Table 2 indicate that the G_{IC} of composites laminate without added CNTs/GNPs hybrids is approximately 468.9 J/m^2 . The G_{IC} of the CNTs/GNPs hybrids added reinforced composite laminate was 100% higher at 938 J/m^2 than without added CNTs/GNPs hybrids when the carbon nanomaterials content was 1.5 wt%.

A comprehensive knowledge about the influence of nanoparticles on the micromechanics is required in order to explain the observed toughening effect of nanoparticles. The mechanisms of increasing the fracture toughness of polymers via the incorporation of particles have been extensively studied within the last three decades [21–24]. The application of microparticles (spherical or fibrous) exhibits the highest effect in brittle (e.g., thermosetting) matrix systems. Several theories have been developed to explain and understand the effects of particle-toughening and they are often in good agreement with experimental results. The most important micromechanical mechanisms leading to an increase in fracture toughness are (i) localised inelastic matrix deformation and void nucleation, (ii) particle/fibre debonding, (iii) crack

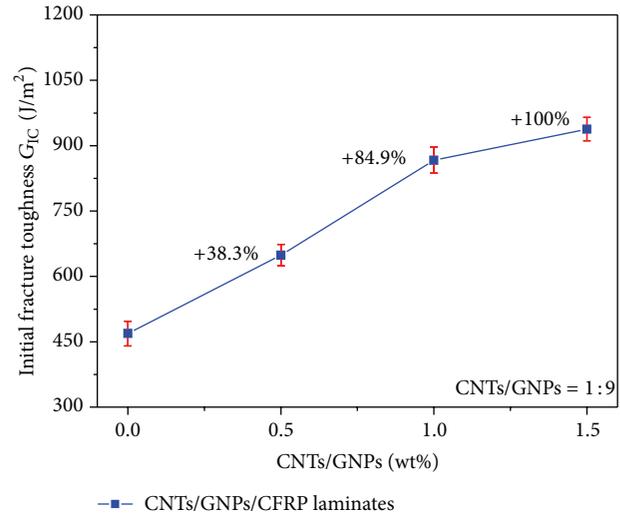


FIGURE 16: Initial fracture toughness (G_{IC}) of CNTs/GNPs/CFRP composite laminates.

deflection, (iv) crack pinning, (v) fibre pull-out, (vi) crack tip blunting (or crack tip deformation), and (vii) particle/fibre deformation or breaking at the crack tip. In this study, CNTs/GNPs hybrids mixed to reinforce epoxy/carbon fiber composite laminates and investigate their interlaminar crack suppression properties. Based on the aforementioned literature and experimental results, the crack growth suppression behavior in composite laminate was primarily caused by the crack deflection of GNPs. When cracks begin to grow, the cracks in the interlaminations of composite laminate deflect due to the interactions of GNPs, consequently suppressing crack growth effectively. In addition, regarding size effect, GNPs primarily suppress the growth of major cracks and CNTs resist the propagation of smaller cracks. Thus, under the synergetic effect of CNTs and GNPs, the interlaminar energy of the fiber composite laminate can be suitably reinforced. This is the reason that the reinforcement effect of Mode I fracture toughness became increasingly noticeable as the carbon nanomaterial content increased.

4. Conclusion

The experimental results showed that the mechanical properties of CNTs/GNPs/epoxy nanocomposites and CNTs/GNPs/epoxy/carbon fiber composite laminates have optimal characteristics with reinforcement through CNTs/GNPs hybrids addition; furthermore, the ultimate tensile strength, flexure properties, and interlaminar shear strength (ILSS) were all improved. Based on the experimental results, adding the present CNTs/GNPs hybrids to the epoxy resin and epoxy/carbon fiber composite laminates provides a considerable synergetic effect. Therefore, when CNTs were added to the GNPs, they entangled the GNPs and filled gaps between GNPs' interlayer. In addition to that CNTs/GNPs hybrids restricting cracks growth in resin, these hybrids could effectively suppress or deter crack propagation in the composite laminates.

Conflict of Interests

The authors declare that they have no conflict of interests.

Acknowledgment

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