

Experimental Analysis of Epoxy and Graphene Nanoplatelet/Epoxy Composites

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Abstract: The tensile behavior of graphene nanoplatelets (GNPs) in epoxy composite under tensile loadings was characterized using uniaxial tensile test at various strain rates ranging from 0.0356 to 0.14/min at room temperature. The stresses and the accompanied strains were measured with load cell and an extensometer, respectively. This investigation covers the effect of the amount of graphene in graphene/epoxy composite material system. The uniaxial tensile tests were done to investigate the tensile behavior of the material beyond ultimate tensile strength. The results showed that both the manufacturing and experimental procedure developed are excellent methods to characterize the response of GNP/epoxy composite to tensile loadings. Additionally, the results show a clear strain rate and amount of GNP in epoxy dependencies in the material response.

Key words: Tensile, characterize, uniaxial, strain rate, graphene, manufacturing, composite, experimental.

1. Introduction

Nanoscale technology is having a more remarkable and practical application in the development and manufacturing of nanostructured based materials, particularly in the field of composite materials and structures with improved strength, lighter weight, and better electrical conduction. As the need to develop lightweight materials and products continues to increase, the need to carry out carefully designed experimental testing of newly developed nanocomposite materials and structures is increasingly more important. Graphene nanoplatelets (GNPs) are short stacks of individual layers of graphite (called graphene) [1]. In order to establish the influence of GNP on performance characteristics of composite materials, different research works have been conducted on the mechanical behavior of GNP/epoxy composites under different loading conditions as documented below.

Liang and Liechti [2] tested an epoxy resin both in tension and compression, along with conducting

multiaxial tests for validation of material models. They only used one strain rate in these tests. Behzadi and Jones [3] carried out compression test on Araldite MY720 and Araldite MY0510 resins to determine the effects of temperatures and strain rates on their yield behavior. Chen et al. [4] utilized compression test to characterize strain rate dependence on EPON 828. King et al. [5] made pure epoxy and 1-6wt % GNP in epoxy composites, and also tested these materials tensile behavior using typical macroscopic measurements. They determined their modulus and creep compliance using nanoindentation. Their experimental data showed that the tensile modulus increased from 2.72 GPa for the pure epoxy to 3.36 GPa for 6wt% (3.7 vol. %) GNP in epoxy composite. Shokrieh et al. [6] utilized a combination of a molecular dynamics (MD) and micromechanics to predict the stiffness of GNP/epoxy nanocomposites. They assumed that graphene sheets are randomly oriented in the polymer matrix. The stiffness of a multilayered GNP was achieved using MD.

Zhang and Richardson [7] employed non-contact optical measurement techniques to measure areas of damage on polymer matrix composite panels impacted by a falling weight impact test machine. They

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suggested that it be used along with conventional nondestructive testing methods to detect damage. Voyiadjis et al. [8] developed a multiscale model including damage and plasticity variables at the meso- and macro-scales.

Littell et al. [9] developed various experimental tests over a wide range of strain rate using small test specimens of epoxy resin in tension, compression, and shear. Their results have been very useful as benchmarks for numerical results. Some researchers [10-12] have shown that for polymers and polymer-based composites, modulus as determined by nanoindentation is higher than that reported by macroscopic tensile tests. Gupta et al. [13] employed FEM/micromechanics/multiscale modeling method to analyze mechanical properties of carbon nanotube reinforced polymer composites. Similar methods [14, 15] have been applied to model thermal conduction and mechanical properties in polymeric nanocomposites.

Aluko et al. [16] established the effect of temperature on the elastic and yielding behavior of epoxy using reactive force field. They [17, 18] also developed a multiscale model for predicting mechanical response of hybrid composites. Komarov et al. [19] proposed a new computational method where the polymer network is polymerized at a coarse-grained level and then mapped into a fully atomistic model. MD analyses were then carried out with the OPLS force field.

Hadden et al. [20] studied mechanical properties of GNP/carbon fiber/epoxy hybrid composites using multiscale modeling simulations. The multiscale modeling that was developed in their studies consisted of MD and micromechanical modeling that was validated with experimental testing. Their results agreed with experimental data and also provided more useful information into the composite mechanical behavior. The effect of GNP volume fraction and dispersion on the axial properties of the hybrid composite was shown to be insignificant, while the

results showed a substantial impact on transverse mechanical properties. Joel et al. [21] developed a molecular model for the structure of the carbon fiber/polymer interphase for multiscale analysis of composites.

While different computational modeling simulations have been applied to study the impact of GNP on the mechanical behavior of GNP/epoxy composite, there are no experimental data available in literature to document the combined effects of volume fraction of graphene and strain rate on the mechanical behavior of GNP/epoxy composite. For this reason, the goal of this study is to experimentally characterize the behavior of GNP/epoxy systems over a range of volume fractions of GNP in epoxy, and their response to strain rates. In addition, this investigation covers a new methodology for manufacturing the GNP/epoxy composite tensile bars, the process for testing, and the techniques used for measuring the output data.

The paper is arranged as follows: Section 2 explains the methodology of the research work. Section 3 documents the results and discussion. Section 4 presents the conclusions.

2. Methodology

The materials and experimental methods are documented below.

2.1 Materials and Specimen Fabrication

The specimens that are made by machining often consist of defects and flaws due to the surface asperities caused by the cutting tools. These defects may often lead to experimental errors in characterizing the mechanical performance of the nanocomposite. To circumvent this issue and improve the experimental method, a rapid prototyping technique was utilized to make the tensile bars that are free from any type of post-preparation processes. At first, several 3-D print tensile bars pattern using ABS (Acrylonitrile Butadiene Styrene) plastic were fabricated. The bars were then placed into a boxed

frame, covered with high heat resistant silicone mold rubber (Mold Max 60) and allowed drying for a day. The resulted mold is shown in Fig. 1. This rectangular flat-shape mold was made according to ASTM Type I tensile bars (165 mm long by 19 mm wide by 3.3 mm thick) for high volume test sample production.

To make pure epoxy bars, a mixture of 72 g of resin and 28 g of hardener was prepared by hand using a wooden stick for about 3 minutes. Then the mixture was degassed under vacuum in an oven at a temperature of 75 °C for 10 minutes. The mixture was poured into the mold and allowed to gradually cure at room temperature for five days.

To fabricate the GNP/epoxy composite, a predetermined weight fraction of GNP (XG Science) [1] was added to the epoxy resin in a beaker. The material was mixed using 2-in diameter disperser blade in a Ross high shear mixer HSM-100 LSK-I at a rotating speed of 2,000 rpm for 30 min. The hardener was then added to the GNP/resin materials and mixed by hand for 3 minutes. The mixture was degassed under vacuum in an oven at a temperature of 75 °C for 5 minutes before it was poured into the mold. The GNP/epoxy was allowed to gradually cure at room temperature for five days. It should be noted that the GNP utilized in this study is xGnP-C-300, available from XG Sciences. It has grade C particles with a 2 µm average platelet diameter and a thickness of 2 nm. It has a density of 2.0 g/mL and average surface areas of 300 m²/g [1].

2.2 Tensile Test Method

As stated before, the rapid prototyping techniques employed in fabricating the bars allowed a large volume of the bars to be made into precision of ASTM Type I tensile bar without any secondary machining operations. The tensile properties of both the pure epoxy and GNP/epoxy composite were determined using ASTM D638 at strain rate of 0.1400, 0.0712, and 0.0356/min (corresponding to displacement rates of 3.556, 1.80848, and 1.0 mm/min, respectively).

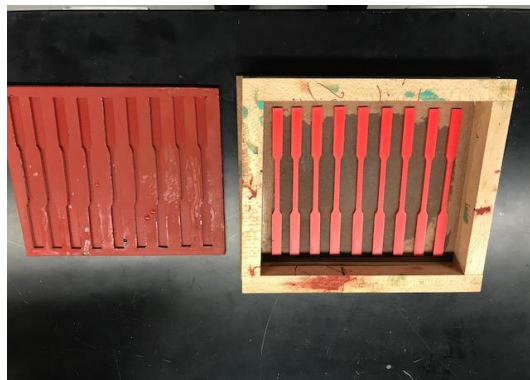


Fig. 1 The rapid prototyping tool for making the tensile bars.



Fig. 2 The experimental set up for measuring the tensile modulus.

These tensile tests were performed on a Zwick tensile tester incorporated with a load cell that measured the varied loadings, while the extensometer attached to the specimens measured the strain as shown in Fig. 2. On completion of each test, the tensile modulus was calculated from the linear portion of the stress-strain curve for three different samples. Also, the extensometer was only attached beyond ultimate tensile strength of the material. This is to prevent the damage to the extensometer that might arise due to the snapping of the specimens. However,

the specimens were tested to the point of failure to study the fracture surfaces.

2.3 Scanning Electron Microscope (SEM) Examination

To further compare and understand the toughening mechanism in these composites, the fracture surfaces from several of these bars, including one for pure epoxy were observed using a JOEL 6330F Scanning Electron Microscope (SEM). The fracture surfaces were sputter coated with gold to allow a better observation of the surface morphology.

3. Results and Discussion

3.1 Tensile Test Analysis

As stated before, the engineering stress was plotted against engineering strain for the tensile test within the linear range to obtain the Young's modulus. Table 1 shows the effect of GNP reinforcement and strain rate on elastic modulus of epoxy. Generally, the Young's modulus increases with increased percent weight of GNP in epoxy, an indication of better performance for fiber/epoxy composite when GNP is embedded into the epoxy matrix. Fig. 3 shows the effect of GNP reinforcement on epoxy at a strain rate of 0.14/min. It can be seen in this figure that the pure epoxy has the least value of stiffness while this value increases with GNP weight percent reinforcement. This result confirms the claim that the mechanical integrity of composite structure is remarkably improved when the size of reinforcement is reduced to nanoscale. This increase in stiffness can be attributed to the preparation of homogenous dispersions of GNP in the nanocomposite which were obtained using high shear mixer processing technique, thereby increasing the GNP/epoxy performance significantly due to load transfer of the nanomaterials (GNP). The high modulus, strength of nanofillers, and robust interfacial adhesion between the nanofillers and matrix also contributed to the reinforcement.

Also, Fig. 4 documents the effect of strain rate on the elastic modulus of epoxy containing 1% by weight

of GNP. This figure clearly shows that the tensile modulus increases with increased value of strain rate. This suggests that the uniform dispersion GNP particles restricted the mobility of polymer chains. The restriction of this immobilized epoxy material systems around nanoparticles increases with strain rate because an increased rate gives less time for epoxy chains to move past GNP particles.

For comparison purposes, the present result for the rate of deformation equal 1 mm/min is shown in Table 2 with King et al.'s [5] experimental findings. The results from the present study is slightly higher and this might be due to the difference in the curing process. Littell et al. [9] also obtained 2.52 and 2.77 GPa when the rate of deformation is 0.0019 and 0.19 mm/min for pure epoxy. These values are lower than the two results presented in Table 2 because the Young's modulus for epoxy is strain rate dependent.

Additionally, Tables 3 and 4 document the ultimate tensile strengths and their corresponding strains, respectively. The addition of GNP causes the ultimate tensile strength to increase with the increased amount

Table 1 Young's modulus of pure and reinforced epoxy at different strain rate and weight fraction of GNP.

Strain rate/min	E (GPa) pure epoxy	E (GPa) 1% wt GNP/epoxy	E (GPa) 2% wt GNP/epoxy	E (GPa) 3% wt GNP/epoxy
0.1400	4.1	4.344	5.297	7.602
0.0712	3.244	3.844	4.927	5.041
0.0356	2.84	3.722	4.356	4.736

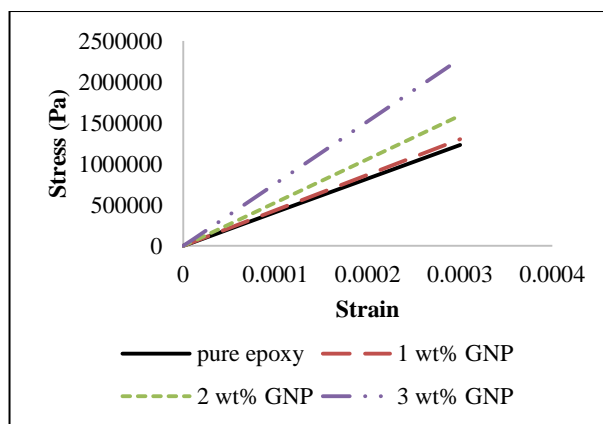


Fig. 3 The effect of GNP in epoxy at a strain rate of 0.14/min.

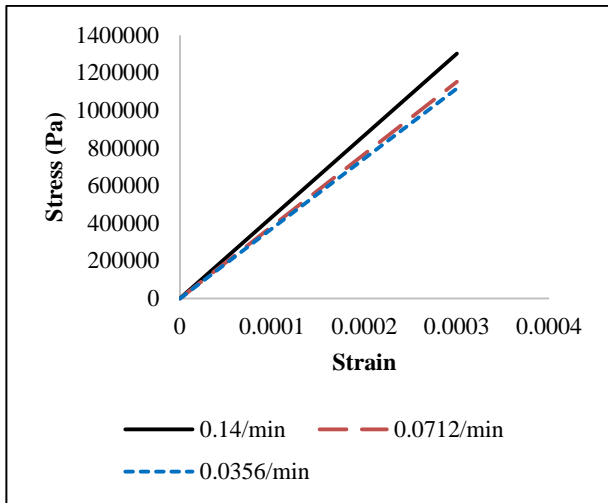


Fig. 4 The effect of strain rates on tensile modulus for 1 wt% of GNP in epoxy.

Table 2 The effect of GNP on epoxy when the rate of displacement is 1 mm/min.

Wt.% of GNP	E (GPa) Present	E (GPa) King et al. [5]
0 (pure epoxy)	2.84	2.72
1	3.722	2.82
2	4.356	2.92
3	4.736	3.04

Table 3 Ultimate tensile strength (UTS) of pure and reinforced epoxy at different strain rates and weight fractions of GNP.

Strain rate/ min	UTS (GPa) Pure epoxy	UTS	UTS	UTS
		(MPa) 1% wt GNP/ epoxy	(MPa) 2% wt GNP/ epoxy	(MPa) 3% wt GNP/ epoxy
0.1400	21.762	28.456	31.274	37.481
0.0712	19.746	24.922	27.651	30.696
0.0356	16.227	22.813	32.744	28.134

Table 4 Strain at ultimate tensile strength (ϵ_{UTS}) of pure and reinforced epoxy at different strain rates and weight fractions of GNP.

Strain rate/ min	ϵ_{UTS} (%) Pure epoxy	ϵ_{UTS} (%)	ϵ_{UTS} (%)	ϵ_{UTS} (%)
		1% wt GNP/ epoxy	2% wt GNP/ epoxy	3% wt GNP/ epoxy
0.1400	4.432	1.234	1.123	1.069
0.0712	4.334	1.238	1.108	1.074
0.0356	4.974	1.169	1.145	1.131

of GNP. However, their corresponding strains decreases slightly. The effect of GNP addition on epoxy is more predominant on ultimate tensile

strengths than their corresponding strains. The results confirmed that the homogeneously distributed GNPs improve the toughness of the GNP/epoxy composite. These improvements suggest that the interparticle distance was smaller than the nanoparticle diameter. The small interparticle distance enhances the formation of an immobilized epoxy material around nanoparticles thereby, contributing to the strength of nanocomposites.

3.2 Fracture Surface Examination through SEM

Fig. 5 shows the micrograph of the fracture surfaces of samples made with pure epoxy, sample contains 1wt% GNP, and sample contain 3wt% of GNP. It is clear that the fracture surface of the sample made with pure epoxy is smooth and very similar to the typical fracture surfaces observed by other investigator [22]. In contrast, the fracture surfaces of the samples containing 1wt% or 3wt% GNP are much rougher than those of pure epoxy. It is clear that for samples containing GNP, a large number of fracture surfaces are present. These large numbers of fracture surfaces are the major contributing factor for improving the mechanical integrity of this composite structure. It is also interesting to point out that some of these fracture surfaces have different heights and show some separation of GNP platelets at the crack boundary [1].

4. Conclusion

The results showed that the rapid prototyping methodology developed for specimen preparation to circumvent the machining of the tensile bars, proves to be very fast and efficient. It also reduces the amount of surface asperities due to machining effect on the specimens. The method can be used for other types of resin or materials that are completely different. The combined effect of GNP and strain rates on tensile modulus of GNP/epoxy composite was experimentally investigated using Zwick tensile test at room temperature. The results showed that Young's modulus remarkably increases with increased strain

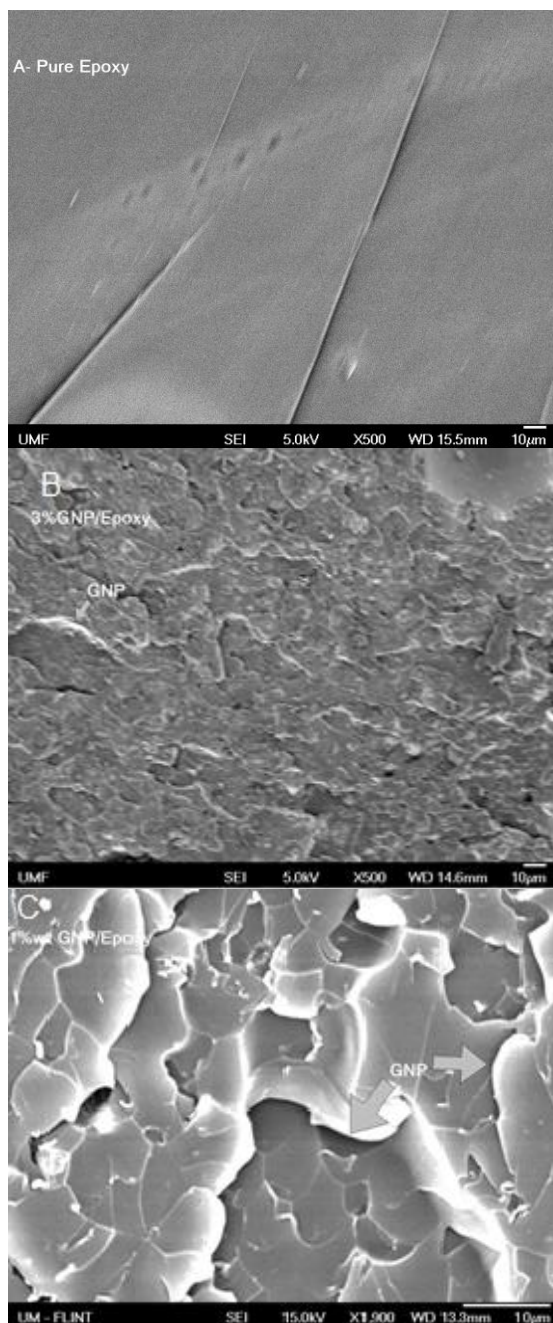


Fig. 5 SEM micrograph of the fracture surfaces of three different samples: (A) Pure Epoxy and (B) Samples contain 3%wt GNP at same magnification. Sample (C) shows the fracture surfaces at higher magnification for 1% GNP.

rate and weight fraction of GNP in epoxy. The results confirmed that the stiffness of epoxy matrix composite can be increased when it is reinforced with nanoparticles. The data generated can be used as a benchmark for the development of numerical models for the analysis of GNP/epoxy composite.

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