# DEVELOPMENT AND CHARACTERIZATION OF GRAPHENE NANOPLATELETS FILLED CARBON FIBER/BENZOXAZINE/EPOXY COMPOSITES

Poom Narongdej, Steven J. Nava, Jack T. Denk, Ehsan Barjasteh
California State University, Long Beach (CSULB)
Long Beach, California/U.S.A.

## ABSTRACT

In this study, we report an advancement in electrical, mechanical, and thermo-mechanical properties of benzoxazine-epoxy copolymer and its resultant composites by the addition of Graphene Nanoplatelets (GNPs). The GNPs were incorporated at different loadings in a copolymer consisting of bisphenol-A based benzoxazine (BZ) and bi-functional cycloaliphatic epoxy resins (CER). A three-roll mill (3RM) calendering device was utilized to disperse the GNPs in the copolymer matrix. The composites were characterized for their electrical, mechanical, and thermomechanical properties. The addition of GNPs showed an improvement of greater than 35 % in both modulus and strength with the addition of GNPs at 0.9 wt.%. A similar trend was observed in the storage moduli, where the maximum value was obtained from the sample at the same nanoparticle loading. The electrical conductivity was achieved at the electrical percolation threshold between 0.6 and 0.9 wt.% of the GNP loadings. The BZ/CER/GNPs composites were used as a matrix to impregnate carbon fibers through the vacuum-assisted resin transfer molding (VARTM) process. The resultant composite laminates were then tested for their mechanical and electrical properties.

Corresponding author: Dr. Ehsan Barjasteh

## 1. INTRODUCTION

Benzoxazine resin is a new class of thermoset resin that is suitable for high-performance applications due to its outstanding mechanical and thermal properties [1-4], good flame retardance [1,4], high char yield [1,4], low water absorption [1,3,4], excellent adhesive property [2], and electrical insulation [5,6]. It is also suitable for carbon-fiber composite manufacturing using out-of-autoclave (OOA) processes, such as vacuum-assisted resin transfer molding (VARTM). This is owed to the BZ resin's advantages, including no catalyst or coupling agent required for curing [1,4], no by-product formed in a cured sample [1,4,7], and near-zero volumetric shrinkage or expansion upon curing [1,3,4,8]. Because the OOA technique allows composite parts to be fabricated at an increasing volume with lower cost, the OOA-processable resin with developed performances is highly demanded in multiple industries.

An increase in the applications of benzoxazine and OOA manufacturing leads to multiple modifications of the resin's properties and process. Several works have reported a great compatibility and property enhancement by alloying benzoxazine with various types of epoxy resins [9,10,11]. For instance, our previous study showed that blending the bisphenol-A based benzoxazine with cycloaliphatic epoxy resin with a concentration greater than 25 wt.% exhibited

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a significant enhancement in a glass transition temperature ($T_g$) and crosslinking density. Furthermore, the resultant resin provided a suitable viscosity that eased the VARTM process [12]. However, the incorporation of epoxy led to a decrease in both tensile strength and modulus [12]. The modification of the BZ/CER system thus was required to enhance the diminished properties.

Graphene is a promising candidate as a nanofiller for enhancing the mechanical properties of the developed BZ/CER resin due to its outstanding mechanical modulus and strength of approximately 1 TPa and 130 GPa, respectively [13,14]. A single two-dimensional sheet of graphene can be obtained from overcoming the interaction force between layers of graphite. This can be achieved by multiple approaches such as ultrasonication, ball milling, and mechanical calendering [15]. In order to obtain the full advantages of utilizing graphene as a nanofiller, it is necessary for the nanoparticles to be homogenously dispersed throughout the entire matrix [16,17].

One of the commonly used methods to disperse the nanoparticles is ultrasonication. This approach requires a low-viscosity medium to assist the dispersion of the nanoparticles; therefore, solvents such as chloroform [18], acetone [19], ethanol [20], and tetrahydrofuran (THF) [21,22] are commonly used in several studies. However, these solvents are required to be removed from the system prior to the curing process to prevent negative effects on thermal and mechanical properties processed by any residual solvent [23,24]. This adds a complexity and extra cost to the fabrication process. For this reason, a solvent-free nanocomposite manufacturing approach becomes a key research interest. In our previous study [12], both ultrasonication and mechanical calendering using the 3RM were implemented to disperse GNPs in the BZ/CER matrix. Both mechanical and electrical performances of the 3RM samples were found to outperform the ultrasonication samples. Furthermore, this solvent-free mechanical calendering approach was suitable for high-volume nanocomposite manufacturing due to its simplicity, shorter manufacturing time, and uniform dispersion of nanoparticles in a host matrix.

In this study, GNPs at various ratios were incorporated in the BZ/CER resin using the 3RM calendering technique. The cured samples were characterized for their mechanical, thermomechanical, and electrical properties. The resultant nanocomposite with the most favorable ratio of GNPs was chosen to reinforce carbon fibers using the VARTM method. The cured composite was tested for its mechanical and electrical performances.

# 2. EXPERIMENTATION

## 2.1 Materials

Bisphenol A-based benzoxazine resin (BZ) was supplied from Huntsman Corporation with a density of 1.22 g/cm$^3$. Cycloaliphatic epoxy resin (CER), 3,4-epoxycyclohexylmethy 3,4-epoxycyclohexane carboxylate, was obtained from Lindau Chemicals Inc. with a density of 1.165 g/cm$^3$ under the chemical name of Lindoxy 190 (131-143 g/equiv.). Unidirectional carbon fiber fabrics were received from Textile Products Inc. with an areal weight of 335 gsm. Graphite nanoparticles were purchased from Ashbury Graphite Mills with a surface area of 350 m$^2$/g, under its commercial name of Nano 24.

## 2.2 Nanocomposite Fabrication

The ratio between BZ and CER (62.5:37.5) was chosen from our previous research [12], which provided a suitable viscosity for the VARTM process as well as improved thermal properties. Therefore, the same mixing ratio was investigated further with the incorporation of GNPs in this study.

The reference BZ/CER sample was mixed in a temperature-controlled oil bath at 110 ˚C with a magnetic stirrer rotating at 375 rpm until a homogenous mixture was achieved. The BZ/CER/GNP nanocomposite was fabricated by first adding the GNPs in CER at different ratios (0.6, 0.9, 1.8, and 2.4 wt.%). The GNP/CER mixture was then milled nine cycles in total until a uniform dispersion was obtained. The mixture was then transferred to a 20 ml vial and was immersed in the temperature-controlled oil bath at 110 ˚C. The BZ powder was gradually added to the GNP/CER mixture under a control shear force generated by a magnetic stirrer rotating at 375 rpm. Both reference and BZ/CER/GNP mixtures were placed between aluminum plates covered with the non-porous Teflon-coated fiberglass sheet to manufacture a thin film. The rests were casted in the aluminum mold for a surface resistivity test. The samples were cured at 180 ˚C for 2 hours, 200 ˚C for 2 hours, and 220 ˚C for 2 hours.

### 2.3 Composite Fabrication

The reference carbon-fiber composite laminate was manufactured without any GNP incorporation. The resin was prepared by mixing CER and BZ using a high-shear mechanical mixer in a temperature-controlled oil bath. The temperature was controlled at 110 ˚C, and the stirring speed was maintained at 800 rpm.

A specific ratio of GNP was incorporated in the CER. The mixture was then poured into a 3RM calendaring device (Torrey Hills Technologies) for nine cycles to ensure a uniform dispersion of GNPs. The BZ was then added gradually to the GNP/CER mixture to prevent a rapid increase in viscosity using the same mixing condition as the reference panel.

Twelve plies of dry carbon fiber with a dimension of 30.5 cm × 30.5 cm were stacked in a unidirectional sequence and were fabricated using the VARTM process. Both reference and GNP-filled composite laminates were infused at 110 ˚C and were cured at 180 ˚C for 2 hours, 200 ˚C for 2 hours, and 220 ˚C for 2 hours.

### 2.4 Characterization

A tensile test for thin-film samples was conducted at room temperature, in accordance with ASTM D1708, using the ADMET eXpert 4200 micro-tensile tester with a 445 N load cell. The average thickness and width of the specimens were 0.1 and 5 mm, respectively. The gauge length was initially set at 22 mm with the movement speed of 1 mm/min. Five specimens were tested for each GNP loading, and stress-strain curves were plotted to calculate the modulus of elasticity, tensile strength, and percent elongation.

A dynamic mechanical analysis of the nanocomposite was conducted using a DMA instrument (Q800, TA Instruments) equipped with a tensile clamp, in accordance with ASTM D4065. The temperature was changed from 30 ˚C to 350 ˚C with a heating rate of 5 ˚C/min.

An electrical property of the nanocomposite was determined based on surface resistivity of the casted samples using the four-point probe (Signatone Pro4) connected with the Keithley 2461 sourcemeter. The distance between measuring pins was fixed at 1 mm. The resistivity of the carbon fiber composites was measured by a two-point measurement with electrodes attached on the specimens' surfaces by conductive carbon tapes. The resistivity of the samples was measured on a rectangular bar with average dimensions of 8.9 cm × 3.2 cm × 0.39 cm.

A short beam shear test was determined using a universal mechanical test frame (Shimadzu) equipped with a 10 kN load cell, in accordance with ASTM D2344. The interlaminar shear strength (ILSS) of the carbon fiber composite was determined from samples that were tested

at the room temperature with a span-to-thickness ratio of 4:1 and a crosshead speed of 1.27 mm/min. At least five samples were tested, and the average values were reported.

A flexural test was performed on the carbon fiber composite in order to measure the flexural modulus and strength. The test was conducted at the room temperature using a universal mechanical test frame (Shimadzu) mounted with a 10 kN load cell. A support span-to-thickness ratio of 32:1 and crosshead speed of 1 mm/min were applied in accordance with ASTM D7264.

# 3. RESULTS AND DISCUSSION

## 3.1 Dispersion of Graphene Nanoplatelets in Epoxy Matrix

A use of solvent is essential in easing the dispersion and preventing sedimentation of GNPs in a host matrix. In order to prevent any possible negative effects caused by remaining solvent prior to the curing process and to minimize the manufacturing steps, CER was chosen as a dispersing medium in this study to alleviate the distribution of GNPs due to its low viscosity.

It is important to ensure a uniform dispersion of GNPs in the matrix and maintain homogeneity without any sign of re-agglomeration. A small ratio of GNPs (i.e., 0.01 wt.%) was initially added and dispersed in the CER under nine cycles with the 3RM until no obvious cluster of the nanoparticles was observed in the mixture. A reference sample was prepared by physically mixing 0.01 wt.% GNPs with CER for a comparison as shown in **Figure 1**. The reference sample was observed to have several large agglomerations in the mixture and sediments at the bottom of the container. On the other hand, the mixture dispersed by the 3RM showed a homogenous mixture with a uniform dispersion of GNPs in the CER matrix.

After a three-day storage, the sample dispersed by the 3RM showed no signs of re-agglomeration among GNP particles and the mixture remained completely homogeneous. In contrast, the sample mixed by hand showed discontinuity in particle distribution and sediment at the bottom of the container. Therefore, it could be concluded that CER is a suitable candidate to replace typical solvents used for dispersion and the 3RM method is sufficient for dispersion of GNPs.

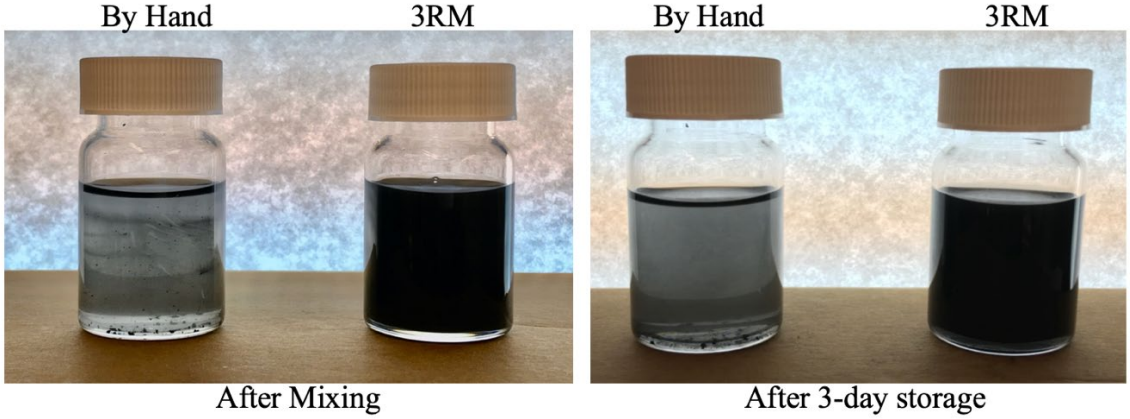



**Figure 1.** Three-day storage of the CER/GNPs mixture at room temperature for the agglomeration observation.

### 3.2 Graphene Nanoplatelets Filled Benzoxazine-Epoxy Nanocomposite

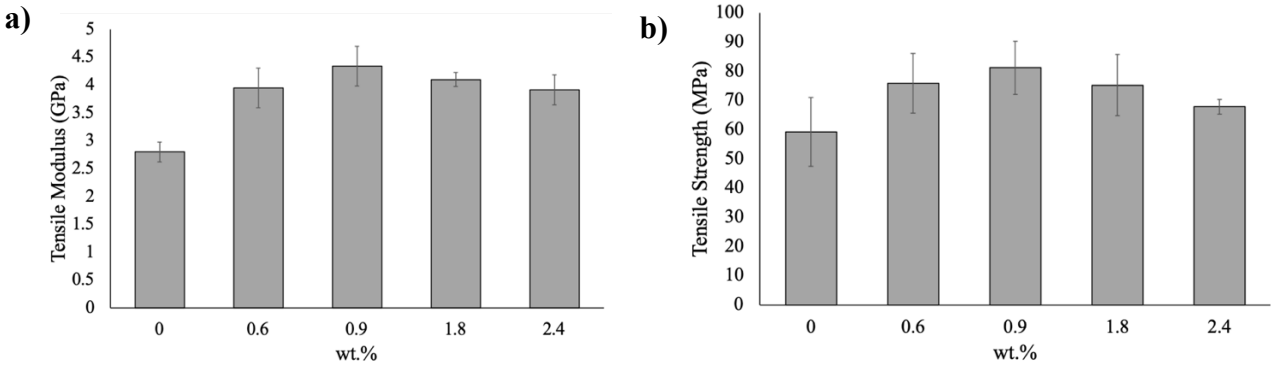
#### *3.2.1 Tensile Properties*

A reference specimen was manufactured without an addition of GNPs represented as 0 wt.% in **Figure 2**, followed by samples with increasing GNP contents. The tensile test was performed to determine the effect of GNP addition on the specimen's stiffness, strength, and elongation.

**Figure 2a** showed an improvement in the modulus of elasticity with an increasing GNP content up to 0.9 wt.%. At this ratio, the modulus reached 4.3 GPa, which was equivalent to a 53.6 % increase from the reference. This significant enhancement of the tensile properties was due to the outstanding stiffness of graphene (~1 TPa [13,14]) that constricted the mobility of polymer chains and shared a portion of load from the host matrix [17,24,25,26,27]. However, a further addition of GNPs beyond this ratio resulted in a decline of sample's stiffness. This was a result of GNP agglomerations at high nanofiller contents that minimized the effectiveness of the GNP's aspect ratio, which reduced the stress transfer from the matrix [24].

The trend in tensile strength was similar to the modulus of elasticity with a maximum increase of 36.9 % at 0.9 wt.%. This enhancement in the tensile strength was the result of a uniform dispersion of GNPs in the BZ/CER matrix as well as the graphene's exceptional tensile strength of nearly 130 GPa [13,14]. A further increase in the GNP content resulted in a reduction of the tensile strength as it had a high tendency to re-agglomerate. The agglomerate acted as a defect center and stress-concentration point when a load was applied [22,23,25].

The elongation of the GNP-filled nanocomposite was found to decrease with an increased GNP concentration, as shown in **Figure 2c**. At higher GNP contents, the particles began to contact each other and restricted the degree of chain formation under loading [21,22,28]. The resultant nanocomposite therefore became more brittle.



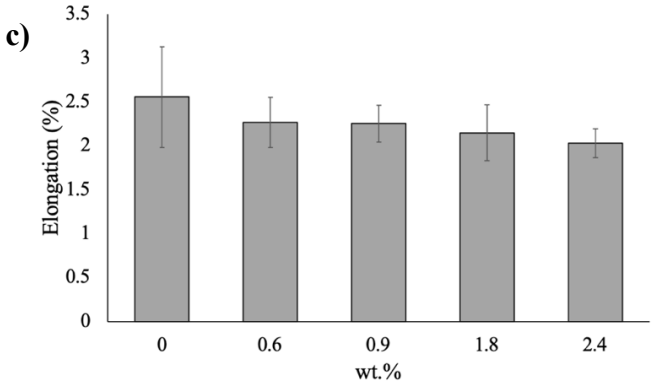



**Figure 2.** Tensile moduli (a), tensile strength (b), and percent elongation (c) of nanocomposites with different GNP contents dispersed by 3RM approach.

### *3.2.2 Thermomechanical Properties*

The effect of GNP addition on the thermo-mechanical properties was investigated. **Figure 3** shows the storage modulus and loss factor (Tan $\delta$) values with respect to temperature. It was observed that the initial storage modulus values at 35 ˚C increased with the increasing GNP concentration up to the maximum at 0.9 wt.%. With a further inclusion of GNPs beyond this content, a decrease in the storage modulus value was observed. The increase in storage modulus at 35 ˚C corresponded with the results obtained in the static tensile test, indicating a stronger stiffness due to a greater dispersion of GNPs in the specimens. The $T_g$ values of all samples were determined and compared at the peak of the Tan $\delta$ curves. No significant change in the $T_g$ was observed with the incorporation of GNPs.

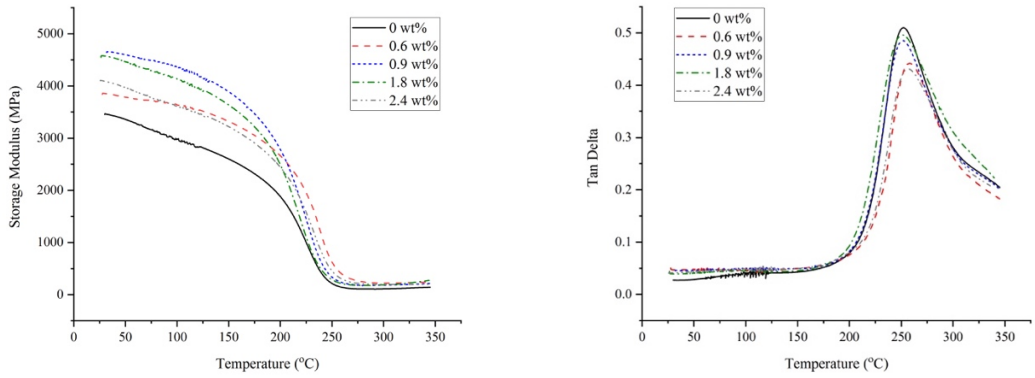



**Figure 3.** Storage moduli (left) and Tan $\delta$ (right) curves of different GNP contents by 3RM approach.

### *3.2.3 Electrical Properties*

In general, neat epoxy and benzoxazine resins exhibit an insulative behavior [5,6,17,22,29]. However, electrically conductive composites are highly demanded in the electronic and aerospace industries. One approach to overcome this limitation was to incorporate conductive nanofillers such as GNPs into the BZ/CER resin. In this study, the resistivity of the GNP-filled

nanocomposites was found to reduce by several orders of magnitude due to a continuous formation of conductive networks of the graphene sheets.

Various GNP contents were incorporated to the BZ/CER resin up to the point where its resistivity decreased dramatically. This point, known as a percolation threshold, indicated a pathway formation for electron transfer in the nanocomposite [22]. The governing factors that determined the percolation threshold included a specific surface area, aspect ratio, dispersion uniformity, and interparticle distance of GNPs [17,22,30].

The electrical resistivity for each concentration of GNP-filled BZ/CER nanocomposite can be seen in **Figure 4**. A significant drop in electrical resistance was observed between 0.6 and 0.9 wt.% GNPs. This sudden drop in resistivity suggested a concentration where the percolation threshold could be found. The insulative behavior at low GNP contents was due to a large distance between GNPs that resulted in a discontinuous electrical path for electron mobility. With further addition of GNPs, the interparticle distance became small enough to promote the electron transfer [17,21].

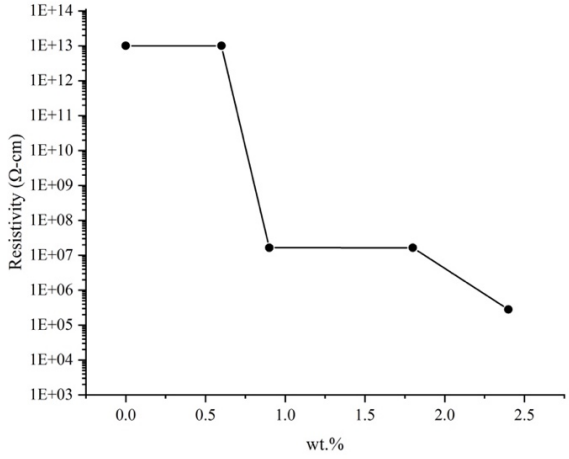



**Figure 4.** Electrical resistivity at different GNP contents dispersed by 3RM approach.

## 3.3 Composites

### *3.3.1 Mechanical Properties*

Based on the results from different GNP contents in the nanocomposite, 0.9wt.% showed the optimum performances in all mechanical, thermo-mechanical, and electrical properties. A further study aimed to incorporate the GNP-filled nanocomposite in a carbon-fiber laminate. Short beam shear and flexural tests were then conducted to determine the mechanical properties of the resultant laminate.

The ILSS values were determined from the short beam shear test, which directly linked with the interfacial adhesion between fiber and matrix [31]. The reference laminate showed an ILSS value at 76.6 MPa. The incorporation of GNPs increased the value up to 80 MPa, which was a 4.5 % increase from the reference. The addition of GNPs could have influenced the mechanical interlocking between the fiber and matrix leading to an increase in shear modulus and reduced the interlaminar stress concentrations at the interphase [31].

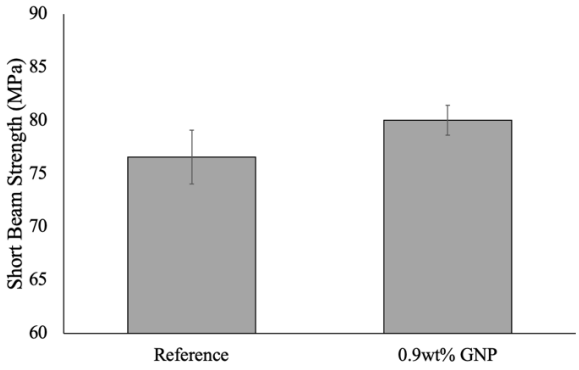



**Figure 5.** Interlaminar shear strength of composite laminates with and without GNPs.

Flexural properties of both the reference and GNP-filled composites were determined by conducting a flexural test. **Figure 6** compares the flexural modulus between the two specimens, with an increase from 101.03 GPa up to 111.82 GPa. This was equivalent to 10.68 % increase from the reference. Similarly, the flexural strength showed an increase of 4.8 %, from 1311.74 MPa to 1374.97 MPa. A small increase in both flexural modulus and strength values was also reported by Qin et al. [31] determining that the flexural properties in the 0˚ direction were primarily dominant by the properties of fibers.

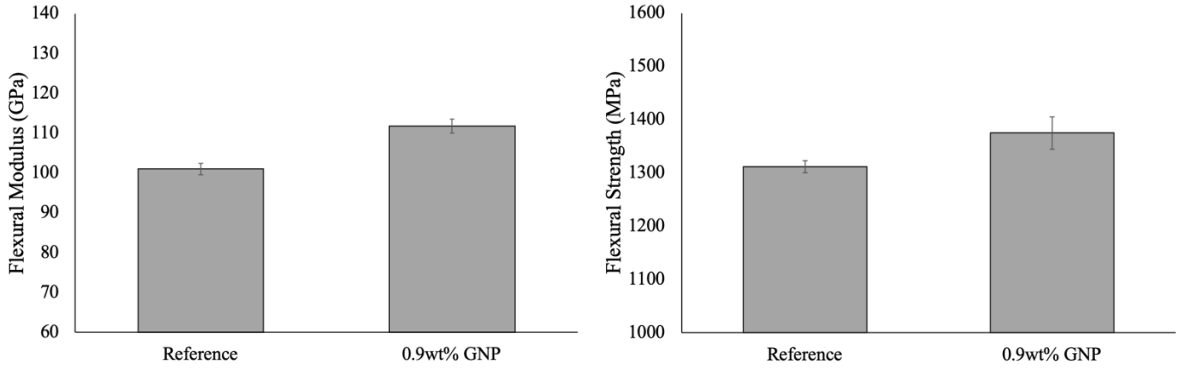



**Figure 6.** Flexural properties of composite laminates with and without GNPs: flexural modulus (left), flexural strength (right).

### *3.3.2 Electrical Properties*

In general, dry carbon fibers are electrically conductive, but both BZ and CER resins possess an insulative behavior [5,6,17,22,29]. When the BZ/CER copolymer was infused into the laminate, it blocked the conductive path between the ply layers as well as surfaces. As mentioned in the previous section, the electrical percolation threshold was found at 0.9 wt.% GNPs. The same GNP content was then added to the BZ/CER resin to investigate the effect of conductive nanofillers on electrical resistivity.

The electrical resistance of both reference and GNP-filed composites were measured by a two-point probe method. Both of the reference and GNP samples were cut into a rectangular shape.

The length of each specimen bar was parallel to the direction of fibers, and the width was perpendicular to the fiber direction.

**Table 1** shows the resistance results of each laminate. The incorporation of GNPs led to a reduction in electrical resistance in all directions. The reference laminate was observed to have an average volume resistivity of 2.44 Ω-cm, while the addition of 0.9 wt.% GNPs resulted in a 41.5 % reduction of the composite's volume resistivity to 1.42 Ω-cm. The electrical resistance was influenced by the GNPs within the composite panel, which may have assisted in creating more electrical networks for the electron transfer [31]. A conductive path between the probe points was created on both sides of the composites' surfaces to determine the surface resistivity in each coordinate direction and volume resistivity of the entire panel.

**Table 1.** Electrical properties of composite laminates with and without GNPs.

| Resistance | Reference laminate | GNP-filled laminate |
|---|---|---|
| **Average 0˚-direction resistance (Ω)** | 15.13 | 11.76 |
| **Average 90˚-direction resistance (Ω)** | 2.47 | 1.36 |
| **Average through thickness resistance (Ω)** | 2.58 | 1.28 |
| **Volume resistivity ($\rho$) (Ω-cm)** | 2.44 | 1.42 |

### *3.3.3 Analysis and Optimization of Manufacturing Process*

It was observed from the resultant panel that it was difficult to obtain a uniform dispersion of the GNP-filled laminate. Although a uniform dispersion of GNPs in the matrix could be obtained by the use of 3RM at 0.9 wt.%, during the resin transfer infusion, the GNPs did not flow uniformly. This resulted in a high concentration of GNPs at the inlet and both edges of the panel, as shown by the arrows in Figure 7. Therefore, the manufacturing process of GNP-filled composites should be further investigated and optimized.

In order to optimize the manufacturing process, it is beneficial to initially visualize the flow of GNPs during the infusion process. This can be conducted by infusing the GNPs at a low weight fraction (e.g., 0.01 wt.%) into a lighter color material, such as fiberglass. Once the flow of GNP is observed, there are several methods can be further implemented to improve the dispersion quality. One method is to reduce the infusion pressure in order to ensure that the GNPs can more easily flow in between dry fiber plies. Another approach is to replace the infusion step with a B-staging technique by placing resin films in between carbon fiber plies. The layup can then be cured with a suitable pressure and temperature using a heat press [32].

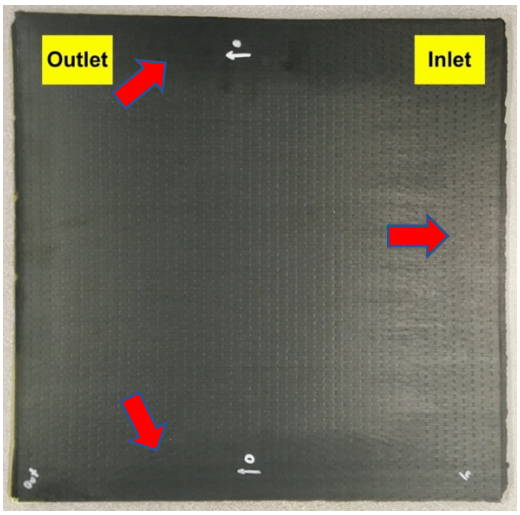



**Figure 7.** Bottom side of the resultant GNP-filled composite laminate.

## 4. CONCLUSIONS

The incorporation of GNPs in the BZ/CER copolymer was investigated in this study. It was found that increasing GNP contents up to 0.9 wt.% enhanced both mechanical and thermo-mechanical properties. A further nanoparticle addition beyond this content resulted in a decrease in both tensile performances and storage moduli. No significant change in the glass transition temperature was observed with the incorporation of GNPs. The electrical properties were determined based on the resistivity of the nanocomposite, and the percolation threshold was found between 0.6 and 0.9 wt.%.

The GNP content of 0.9 wt.% was determined to provide the optimum properties; therefore, the same ratio was used during composite fabrication. The GNP-filled composite showed improvements in both flexural properties and interlaminar shear strength. The electrical resistance decreased in the composite containing GNPs. Although improvements in mechanical and electrical properties could be obtained with the addition of GNPs, the GNP content was found to be ununiform throughout the entire laminate via the VARTM process. Therefore, an optimization of manufacturing processes should be further investigated.

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