Preparation of High-Performance Epoxy Hybrid Nanocomposite Composed With Graphene and Carbon Nanotube Characteristics Study

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**Abstract.** This paper describes the design, production and mechanical performance evaluation of high performance epoxy hybrid nanocomposites reinforced with graphene nanoplatelets and chopped multiwalled carbon nanotubes with nanofillers were chosen in order to take use of both the remarkable tensile strength and crack bridging capacity of CNTs and the high in-plane stiffness of GNPs. The composites were made by compression moulding with a fixed GNP content (0.5 weight %) and varying CNT loadings and ratios were 0.5:1, 0.5:1.5, and 0.5:2, respectively. Tensile strength, fracture toughness and flexural modulus were all part of the comprehensive mechanical characterization used to assess performance improvements. When compared to normal epoxy the composite with 0.5:1.5 GNP:CNT performed best increasing fracture toughness by 62% and tensile strength by up to 85%. These results demonstrate the effectiveness of hybrid filler and its applicability for demanding aerospace structures that require superior strength-to-weight ratios and impact resistance.

# Introduction

Epoxy resins are frequently employed as thermosetting matrices in sophisticated structural applications, especially in the electronics, automotive and aerospace sectors. Superior adherence to a variety of substrates, good dimensional stability and outstanding chemical resistance are among their well-known qualities. However its use is restricted in situations with significant mechanical stress or impact energy because to the inherent brittleness and low fracture toughness of cured epoxy [1-4].

In order to get beyond these restrictions, scientists have looked into using nanoparticles as reinforcements. Among these graphene nanoplatelets are made up of several layers of carbon atoms that are sp2 linked, have a tensile strength of around 130 GPa and a Young's modulus of about 1 TPa. They also have a lot of surface area for stress transfer. For improving the toughness and tensile behaviour of brittle matrices like epoxy, carbon nanotubes are multi walled varieties in chopped form that are perfect because of their high aspect ratios and remarkable tensile qualities [5-8].

Recent studies have seen a great deal of research into the incorporation of carbon nanotubes and graphene nanoplatelets into polymeric matrices, especially epoxy to improve mechanical performance, heat resistance and damage tolerance, particularly for automotive and aerospace applications. The increasing usefulness of 3Dprinted carbon fiberreinforced composites was highlighted [9-11] highlighted the structural significance of these materials in aviation applications. The investigation examined the ways in which nanoscale reinforcements, such carbon nanotubes, improve interfacial adhesion and lower failure rates in printed composites. These results were expanded [12-14] through their analysis of MWCNT polymer composites and comparison with graphene-epoxy nanocomposites. They verified that because of the synergistic interactions between the fillers, hybrid reinforcement greatly increases both tensile strength and heat conductivity.

The compatibility issues and dispersion techniques necessary to optimise property enhancement were emphasised in Farahmand's (3) thorough review of composite matrices and reinforcing fibres, such as CNTs and graphene. In their research of the fatigue and fracture behaviours of grapheme mixed nanocomposites, They [15]found crucial thresholds at which the graphene content changes from toughening to embrittlement. This information is crucial for the optimisation of aerospace-grade materials. Using both experimental and molecular dynamics methods, Bahramzadeh and Raygan (5) shown that the combination of carbon nanotubes and zirconia in a hybrid composite matrix results in damage-tolerant behaviour. They also suggested that comparable hybridisation with graphene in polymers may occur. Erol and colleagues (6) have examined carbon nanostructures for aerospace, emphasising the importance of interfacial tailoring and functionalisation between CNT, GNPs and the epoxy matrix for scalable applications. According to Monteiro and Simões (7), who examined current developments in aerospace hybrid nanocomposites, mixing 1D and 2D nanofillers results in multifunctional enhancements, including EMI attenuation thermal shielding and mechanical strength. In support of this, they [16] conducted a quantitative examination of the elastic characteristics of epoxy composites with varying weight fractions of CNT and GNP, finding that the ideal ratios for balancing stiffness and toughness were 0.5–1.5 wt%. They tackled a significant obstacle: attaining consistent dispersion of nanoparticles. In line with earlier experimental results, their work indicates that surface modification and ultrasonication have a major impact on stress transfer and overall mechanical performance [17-20].

They broadened their focus by working on metal matrix composites, but their statistical optimization method for mechanical tuning provides useful information that can be used to polymer systems, particularly in frameworks for multi-objective material design. In order to model the elastic behavior and viscoelastic behavior of GNP/CNT-reinforced nanocomposites [21-24], Research [25-28]used sophisticated finite element micromechanical modelling. The two fillers' observed synergies were confirmed by their models. According to [29-30] the development of functionally graded epoxy nanocomposites requires the use of additive manufacturing techniques, especially 3D printing. This strategy supports the aerospace industry's need for structural components that are lightweight and customizable in design. Epoxy composites with both CNT and GNP additives were directly studied [31-33] who confirmed improved impact resistance and increased tensile strength (~30%). As seen by SEM analysis, they also reported better interfacial load transfer. In their study of micro cracking problems in epoxy matrices showed that the combination of nano-structured block copolymers and GNPs significantly improved resistance to crack initiation and propagation. The formulation for aerospace-grade toughness was optimized using their Design of Experiments (DoE)-based approach. Last but not least, research [34-39] provided a thorough analysis of nanosilica for strengthening epoxy composites. They recognised that, despite their silica focus, GNP/CNT hybrids are susceptible to similar mechanisms, such as particle bridging, plastic deformation zones and crack tip blunting, highlighting the potential of multiscale filler integration [40-43].

Prior research has documented the distinct impacts of either GNPs or CNTs on epoxy behaviour. On the other hand, these two nanofillers may hybridise to produce synergistic benefits. GNPs' 2D structure and CNTs' 1D tubular shape combine to create a multidimensional reinforcement network that simultaneously prevents cracks from starting, bridges cracks that are spreading and improves load transfer effectiveness [44-49]. The combined reinforcement impact of chopped CNTs and GNP in different ratios is examined in this work. The materials are made via compression moulding, a scalable and applicable technology. Creating and describing nanocomposites that satisfy the exacting mechanical and dependability requirements for aerospace panel applications is the aim.

# Materials and Methods

## Materials

The material used in current investigation was a commercially available diglycidyl ether of bisphenol A built epoxy resin which was chosen for its outstanding mechanical capabilities, chemical resilience, and compatibility with nanofillers. The resin was cured with an aliphatic amine hardener in a stoichiometric ratio resulting in optimum crosslinking and consistent network development throughout the composite. Graphene nanoplatelets were used for reinforcement with a high explicit surface area of 300 m²/g, a nominal thickness of 5 nm and average lateral dimensions ranging from 5 to 10 µm. These features enable GNPs to provide a wide interfacial contact area with the epoxy matrix resulting in efficient load transmission and better mechanical reinforcement [50-51].

In addition to GNPs, chopped multiwalled carbon nanotubes were used as a secondary nanofiller. The MWCNTs had average length (approx) of 20µm and a diameter of 10-15 nm. Their chopped form was chosen precisely to improve dispersion uniformity and eliminate entanglement concerns associated with longer CNT resulting in better interfacial interaction with the epoxy matrix. The accumulation of GNP and MWCNT contributed to in plane strength and barrier characteristics, while MWCNTs supplied out-of-plane reinforcement and crack bridging. These nanofillers were designed to improve the overall mechanical and functional performance of epoxy-based nanocomposites [52].

## Composite Preparation

The composite production technique was developed to ensure that nanofillers were evenly dispersed and that they formed strong interfacial bonds with the epoxy matrix with the process initiates with the dispersion of graphene nanoplatelets and chopped multi walled CNT in acetone using a probe sonicator at 20 kHz and 200 W for 30 minutes. This process was essential for breaking down agglomerates and generating a stable colloidal suspension of nanofillers and after dispersion the filler acetone mixture was gradually added to the diglycidyl ether of bisphenol A epoxy resin and manually agitated for 15 minutes at 2000 rpm confirming that the nanofillers remained dispersed consistently across the resin matrix.

To remove any remaining solvent and air the resultant mixture was vacuum dried at 80°C. This process aided in the complete evaporation of acetone while also degassing the mixture which is critical for preventing porosity in the final cured composite. The degassed and homogenised slurry was placed into moulds and compressed for 60 minutes at 100°C and 20 MPa pressure and to increase crosslinking density and improve mechanical and thermal stability, the composites were post cured for two hours at 120°C.

Four distinct compositions were created for comparison testing which included a clean epoxy resin as a reference and three nanocomposites reinforced with fixed GNP content and increasing MWCNT ratios: 0.5:1, 0.5:1.5, and 0.5:2 and this formulation technique was designed to assess the influence of MWCNT loading on the inclusive performance of the epoxy matrix while maintaining GNP concentration constant.

# Results and Discussion

## Tensile Strength Analysis

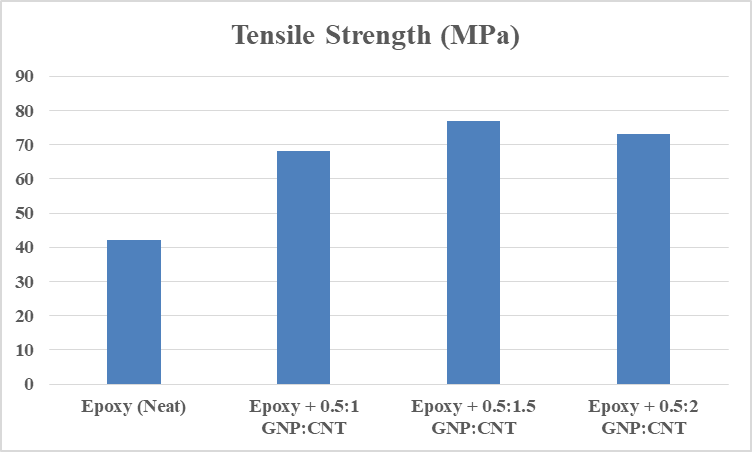
Tensile strength testing was carried out in line with ASTM D638 standards to assess the mechanical belongings of the epoxy and its nanocomposites and the unreinforced epoxy had an average tensile strength of 42 MPa which is characteristic of brittle thermosetting polymers with minimal plastic deformation before fracture. The tensile strength increased significantly to 68 MPa after using hybrid nanofillers with a GNP:CNT ratio of 0.5:1 representing a 62% improvement. This improvement can be attributable to the combination effect of fracture deflection by graphene nanoplatelets and efficient load transfer via multiwalled carbon nanotubes. The planar structure of GNP served as physical barriers to crack formation but CNT high aspect ratio and mechanical strength allowed them to bridge microcracks and carry some of the load transferred from the matrix.

The composite with a GNP and CNT ratio of 0.5:1.5 demonstrated the greatest improvement, reaching a peak tensile strength of 77 MPa. This represents an 85% increase over the neat epoxy implying that this specific mix promoted excellent filler contact. At this load a well developed and continuous hybrid reinforcing network was likely generated inside the matrix limiting fracture propagation while also increasing stiffness and interfacial bonding.

However increasing the CNT content to a GNP:CNT ratio of 0.5:2 resulted in a modest decrease in performance with tensile strength plateauing at roughly 73 MPa. This pattern shows the onset of filler saturation and CNT clumping which might introduce stress concentration sites and disturb the composite's uniform stress distribution. As a result reinforcing efficiency decreased marginally highlighting the significance of balancing filler content to avoid negative impacts on mechanical performance.

**TABLE 1.** Mechanical Properties Summary Table

|  |  |  |  |
| --- | --- | --- | --- |
| **Composite** | **Tensile Strength**  **MPa** | **Flexural Modulus GPa** | **Fracture Toughness (MPa·m^1/2)** |
| Epoxy (Neat) | 42 | 2.1 | 1.12 |
| Epoxy + 0.5:1 GNP-CNT | 68 | 3 | 1.65 |
| Epoxy + 0.5:1.5 GNP-CNT | 77 | 3.3 | 1.82 |
| Epoxy + 0.5:2 GNP-CNT | 73 | 3.5 | 1.75 |
| Epoxy (Neat) | 42 | 2.1 | 1.12 |



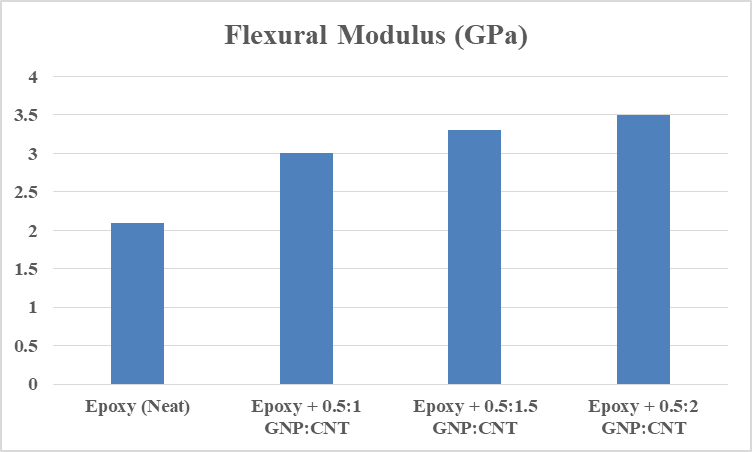
**Figure 1.** Tensile strength

## Flexural Strength

According to ASTM D790 criteria the flexural behaviour trended in line with their tensile performance and flexural modulus of 2.1 GPa demonstrated its distinctive brittleness and comparatively low resistance to bending deformation. Flexural stiffness gradually increased with the addition of hybrid nanofillers mostly as aoutcome of the intrinsic planar rigidity of graphene nanoplatelets and the reinforcing action of multi walled carbon nanotubes. Under bending stress the CNT functioned as load-bearing nanostructures, while the GNP helped by preventing matrix deformation due to their large surface area and aspect ratio.

Flexural modulus significantly improved when the CNT content rose from the 0.5:1 to 0.5:1.5 GNP:CNT ratio suggesting improved stiffness and structural integrity. The composite with a GNP:CNT ratio of 0.5:2 had the highest flexural modulus measuring 3.5 GPa. With a 67% increase in comparison to clean epoxy shows a significant reinforcing impact and the dual filler technique helped create a strong internal network that could withstand flexural loads.

The difference in flexural modulus between the 0.5:1.5 and 0.5:2 compositions indicates that the benefits of having more CNT begin to diminish at the ideal dispersion point but at higher concentrations CNT agglomeration is more likely to happen which can obstruct efficient load transfer and result in localised stiffness changes and this behaviour highlights how important it is to maintain the proper filler balance to maximise reinforcing and avoid dispersion-induced constraints.

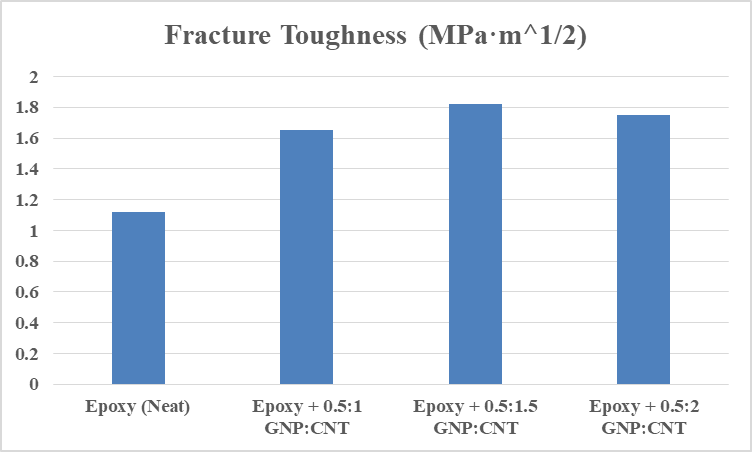


**Figure 2.** Flexural strength

However, at 7.5 wt% n-SiC, the flexural strength slightly declined to 45 MPa. This reduction was likely due to the onset of particle agglomeration at higher filler concentrations. Clustering of n-SiC disrupted the homogeneity of the matrix, leading to local stress concentrations and diminished reinforcement efficiency. Nonetheless, all reinforced composites demonstrated superior flexural performance compared to pure HDPE.

## Fracture Toughness

Testing fracture toughness using ASTM D5045 showed notable increases in the epoxy composites enhanced with hybrid nanofillers' crack resistance. Consistent with its brittle fracture behavior and limited capacity to absorb energy during crack propagation, the plain epoxy shown a somewhat low critical stress intensity factor (K\_IC) of 1.12 MPa•m^½. The fracture toughness rose significantly to 1.82 MPa•m^½ when GNP and MWCNTs were included at a ratio of 0.5:1.5, therefore reflecting a 62% increase over the empty matrix. This development is suggestive of the efficient toughening systems the nanofillers bring about.



**Figure 3.**Fracture toughness

Several reinforcing events occurring at the microstructural level explain the noted increase in K\_IC. Though not displayed here scanning electron microscopy (SEM) photographs of the fracture surfaces usually show important characteristics including matrix-filler debonding, CNT pullout and GNP sheet bridging. These methods increase the fracture route and hence demand more energy to overcome interfacial bonds, so contributing to energy dissipation during crack propagation. Because of their planar design, GNPs deflect and twist the fracture path; CNTs act as nanoscale bridges resisting crack opening. Further energy absorption and blanking of fracture tips by localized debonding at the filler–matrix interface helps to contribute to the general toughness of the composite.

At higher CNT loading levels, especially at the GNP:CNT ratio of 0.5:2 the fracture toughness indicates a decrease in energy absorption capability. CNT clumping is mostly responsible for this reduction since it causes poor filler distribution and the development of stress concentration sites. Such agglomerates can function as early failure sites therefore restricting the strength of the toughening processes. These results support the vital need of preserving homogeneous nanofiller dispersion to completely realize the mechanical benefits of hybrid reinforcement systems.

## Application in Aerospace Composite Panels

Fuselage panels, fairings, and internal stiffeners in aerospace structural design call for materials with a high strength-to-weight ratio, excellent damage tolerance, and long-term constancy under mechanical and thermal stresses. Well-suited to satisfy these performance criteria, the epoxy-based composites created in this work are reinforced with chopped multi-walled carbon nanotubes and graphene nanoplatelets. With a GNP:CNT ratio of 0.5:1.5, the composite showing the most favourable balance of mechanical properties including amplified tensile strength, better fracture toughness and sufficient flexural stiffness among the formulations examined. For aerospace uses where both load bearing capacity and resistance to crack propagation are vital and mix makes the perfect contender.

By means of mechanisms including fracture bridging and pullout the hybrid reinforcing technique efficiently uses the individual advantages of both fillers: planar GNPs offering stiffness and barrier characteristics and CNTs help to dissipate energy. Consequently the 0.5:1.5 composition provides a structurally strong matrix that can resist mechanical loads and suppress failure under dynamic or cyclic conditions generally found in flight situations.

Significantly, the technique used compression molding adds useful value to the material system. This method is compatible with the current infrastructure used in aircraft part production, scalable, reasonably priced. Essential for structural uses, it enables consistent processing of composite panels with complicated geometries and controlled fiber orientation. Particularly under heat cycling and vibrational loads typical in operation the ability to create lightweight yet structurally strong components using this technique offers the potential to increase aircraft efficiency by lowering total weight and improving structural durability.

# Conclusion

This work effectively shows the synthesis and characterising of epoxy based hybrid nanocomposites mixed with chopped multi-walled CNTs and graphene nanoplatelets (GNPs). When compared to pure epoxy, the composite with a GNP:CNT ratio of 0.5:1.5 showed to be the most successful among the several formulations examined producing appreciable fracture toughness, tensile strength and flexural modulus. The combined action of GNPs and CNTs which together create a multidimensional reinforcing network attributes these improvements. This network improves stress transfer, controls fracture and deformation resistance of the composite and inhibits crack propagation.

Up to the 1.5 ratio, mechanical characteristics increased with increasing CNT content; additional addition (0.5:2) produced performance plateauing or even somewhat deteriorating results. This behavior is ascribed to filler agglomeration, which can induce local stress concentrations and impede matrix uniform stress dispersion. Thus, optimizing the advantages of nanoscale reinforcement at the macroscale depends critically on reaching the correct balance in filler ratio and guaranteeing high-quality dispersion.

Future research should concentrate on assessing the thermomechanical stability of the composites under different temperature and humidity conditions, evaluating fatigue resistance under cyclic mechanical loads, and investigating surface functionalization techniques to improve filler matrix compatibility and dispersion, so advancing the application of these materials in aerospace and other high performance sectors. Furthermore, attempts should be made to validate the performance of these nanocomposites in useful service conditions by including them into actual aircraft laminate constructions.

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