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The effects of graphene nanoplatelets and microcrystalline cellulose on the mechanical properties of carbon fiber and fiberglass composites

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Abstract

This study investigated how graphene nanoplatelets (GNP) and microcrystalline cellulose (MCC) affect the mechanical properties of epoxy (EP), carbon fiber (CF), and fiberglass (FG) composites. The tensile and flexural properties of GNP-EP and MCC-EP with varying reinforcement concentrations were examined. The results indicated significant improvements on tensile and flexural properties by GNP and MCC. At 0.1 wt% GNP, the tensile and flexural strengths increased by 10% and 21%, while 2.0 wt% GNP improved the flexural modulus by 65%. Similarly, 2.0 wt% MCC improved the tensile strength, modulus, and flexural modulus by 11%, 11%, and 46%, respectively, and a 26% increase in the flexural strength at 1.0 wt% MCC. These concentrations were used to reinforce CF and FG composites. For the CF composites, GNP effectively enhanced mechanical properties due to good dispersion and strong interface formation, resulting in a 14% increase in tensile strength and a 32% increase in modulus with 0.1 wt% GNP, while 2.0 wt% improved flexural strength by 36%. However, MCC showed a lesser impact, enhancing tensile modulus by 37% at 2.0 wt% but negatively affecting flexural performance, likely due to property mismatches. In FG composites, both additives had negative impacts, with only minimal improvements in flexural properties at specific concentrations, likely due to materials incompatibility.

1. Introduction

Polymer composites, particularly epoxy composites, are extensively used across diverse industries including automotive, aerospace, sports, and construction due to their chemical resistance, cost-effectiveness, and exceptional mechanical properties [1,2]. Hence, efforts have been focused on enhancing the mechanical properties of these composites through the integration of various reinforcements [2-4]. These reinforcements include synthetic reinforcements such as carbon nanotubes, graphene nanoplatelets, glass fibers, and carbon fibers, and natural reinforcements such as cellulose microfibrils and microcrystalline cellulose.

Glass fiber and carbon fiber composites have also been widely used as load-bearing structures in aviation and automotive applications [5,7-9]. These composites, known for their compatibility with fibers and their durability in harsh environments, have demonstrated significant improvements in flexural strength when used in hybrid scaffolds [10]. In particular, composites reinforced with carbon/graphene fillers exhibited a remarkable 95.93% increase in strength compared to pure carbon/epoxy composites [5]. The inclusion of nanoplatelets has had a notable effect on the microstructural evolution and mechanical properties of glass fiber composites. Tensile and flexural tests were conducted to characterize their mechanical behavior, revealing that the homogeneous dispersion of nanoplatelets significantly enhanced the mechanical properties of the composites. However, these reinforcements continue to face challenges related to interfacial issues with the fabric [5].

Graphene nanoplatelets (GNP) with their two-dimensional structure, offering a high surface area-to-volume ratio and low density [1-3] are particularly notable as reinforcement due to their remarkable mechanical, electrical, and thermal properties [3-6]. Incorporating GNP into epoxy could facilitate efficient load transfer between the matrix and reinforcement [5,6], significantly enhancing the tensile and flexural strengths of the composite [1,2]. With an addition of 1 wt% GNP, the epoxy composite showed a 41% increase in tensile strength and a 19% increase in tensile modulus [4]. The incorporation of graphene nanoplatelets (GNPs) into epoxy composites can offer several advantages, but it also presents some disadvantages and challenges. One significant issue is the problem of dispersion. GNPs tend to form clusters due to strong van der Waals forces, making it difficult to achieve uniform dispersion within the epoxy matrix. Poor dispersion can lead to inhomogeneous properties, which reduces the effectiveness of the GNPs [11]. Another challenge is adhesion at the interface. Ensuring strong adhesion between the GNPs and the epoxy matrix can be difficult. Poor adhesion at the interface can weaken the composite, resulting in diminished mechanical and thermal properties [12].

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Microcrystalline cellulose (MCC) has emerged as a promising natural fiber reinforcement due to its high crystallinity and compatibility with epoxy matrix [13]. An addition of MCC can significantly enhance the mechanical properties of epoxy composites, improving tensile and flexural strengths by up to 100% under optimal conditions [6]. The incorporation of cellulose into epoxy composites offers benefits such as improved sustainability and mechanical properties, but it also presents several disadvantages and challenges. One major issue is poor interfacial adhesion. Cellulose tends to absorb water (making it hydrophilic), while epoxy resins are generally hydrophobic (repelling water). This difference can result in weak interfacial adhesion between the cellulose fibers and the epoxy matrix, which reduces the mechanical strength and durability of the composite. Additionally, weak adhesion between the cellulose and the epoxy matrix can create stress concentrations, potentially leading to premature failure of the composite under mechanical loads [14].

To address some weaknesses of conventional composites, studies on hybrid composites with a combination of conventional reinforcements have been on the rise [2]. By incorporating only 0.5 wt% graphene fillers into carbon fiber composites, the tensile strength was notably enhanced by 95.93% without increasing their weight [5]. Similarly, the inclusion of Graphene nanoplatelets into glass fiber composites also resulted in a significant improvement in their mechanical properties [15], increasing tensile strength by up to 75%, flexural strength by up to 23%, tensile modulus by 116%, and flexural modulus by 38%. The enhanced bonding between glass fibers and the epoxy matrix, facilitated by the nanoplatelets, was identified as a probable cause of the improved mechanical performance of the composites [16]. This reinforcement enables efficient load transfer between the matrix and fibers [1,2], enhancing the interface between the fibers and the GNP matrix, thereby impacting mechanical properties [17]. This underscores the potential of GNP as an effective matrix in advanced epoxy composites [18]. Research has shown that adding MCC particles can significantly enhance the mechanical properties of bio composites, improving tensile and flexural strengths by up to 100% under optimal conditions [6]. The synergistic effects of MCC and CNTs further amplify these enhancements, particularly in cement composites where significant improvements in flexural and compressive strengths have been observed [13]. Additionally, the strong reinforcing effect of cellulose, leading to the formation of a networked structure above the percolation threshold through hydrogen bonding, has been noted [19].

In this work, we investigated the effects of GNP and MCC reinforcements on the mechanical properties of epoxy resin, glass fiber composites, and carbon fiber composites. The GNP and MCC fillers of various concentrations were used to reinforce the epoxy resin. The tensile and flexural properties tests were performed to determine the optimal GNP and MCC concentrations. The GNP- and MCC-reinforced epoxy of the optimal concentrations were then used as matrices in the glass fiber composites and carbon fiber composites. Lastly, the tensile and flexural properties of the composites were evaluated [20].

2. Experimental method

2.1 Materials

Graphene nanoplatelets (GNP), with a surface area of 50 $m^2 \cdot g^{-1}$ to 80 $m^2 \cdot g^{-1}$ and sizes ranging from 5 μm to 8 μm, were purchased from Merck Ltd, Thailand. Microcrystalline cellulose (MCC), was provided by SCG Packaging Pub Co., Ltd. The epoxy resin (EP) was purchased from BRP Composite Part Ltd.

2.2 Fabrication of GNP-reinforced epoxy (GNP-EP) and MCC-reinforced epoxy (MCC-EP)

To prevent aggregation of GNP in an epoxy matrix, which would result in a decrease in mechanical performance [2], the GNP was uniformly dispersed in ethanol employing the technique called "Couple Ultrasonication" [21]. The mixture underwent an ultrasonication treatment using both an ultrasonic bath (Elma Elmasonic E30H) at 37 kHz and an ultrasonic probe (Cole Parmer ultrasonic processor CP505 with a CV334 probe) at 20 kHz with a 60% amplitude for 2 h. The MCC was also subjected to a similar treatment to ensure proper dispersion before use. After the reinforcing agents were dispersed, GNP- and MCC-reinforced epoxy composites were fabricated. For GNP-EP samples, GNPs were incorporated into the epoxy at concentrations of 0.1, 0.5, 1.0, 2.0, and 3.0 wt%. The well-dispersed GNP-ethanol mixture was first combined with epoxide and mechanically stirred at 500 rpm on a hot plate at 50°C until approximately 90% of the ethanol had evaporated. An ultrasonic probe was then used to further disperse the GNPs in the epoxide for 30 min. After the mixture cooled to room temperature, the hardener was added, and the mixture was manually stirred for 5 min. The mixture was then degassed, cast into silicone molds, and left to cure at room temperature for 24 h. For MCC-EP specimens, the same procedures were followed to fabricate the samples with the concentrations of 0.5, 1.0, 1.5, 2.0, 2.5, and 3.0 wt%. The dimensions of the tensile and flexural test specimens were prepared according to ASTM D638 and ASTM D790, respectively.

2.3 Fabrication of GNP- and MCC-reinforced carbon fiber (CF) and fiberglass (FG) composites

The vacuum bagging technique was employed to fabricate composite samples where the reinforced matrices, i.e., GNP-EP, MCC-EP, were applied on the reinforcement fabrics, i.e., carbon fiber, fiberglass. Those layers of reinforcements were then stacked on top of each other and covered with a vacuum bag where air was completely evacuated out of the bag by a vacuum pump at a pressure of around 1 atm. The applied vacuum pressure removed excess resin and trapped air from the samples, resulting in a more uniform, compact, and high-performance composite structure. The optimal GNP and MCC concentrations, identified in Section 2.2 as providing the best mechanical performance for GNP-EP and MCC-EP composites, are selected for the fabrication in this section. The dimensions of the composite specimens followed ASTM D3039 for tensile properties testing and ASTM D7264 for flexural properties testing. The thicknesses of the tensile and flexural specimens were 2.5 mm and 4 mm, respectively. The number of carbon fibers used in the tensile and flexural specimens were 12 layer and 20 layer, respectively, and the number of fiberglass used in the tensile and flexural specimens were 16 layer and 24 layer, respectively.

2.4 Mechanical property testing

Mechanical property tests were conducted using a universal testing machine (NRI-TS500-50B). Tensile tests were performed according to procedure A of ASTM D638 for the epoxy samples reinforced with graphene nanoplatelets (GNP) and microcrystalline cellulose (MCC), and ASTM D3039 for the carbon fiber and fiberglass composites reinforced with GNP and MCC. A test speed of 5 mm·min^{-1} was used for the epoxy samples, while a test speed of 2 mm·min^{-1} was used for both the carbon fiber and fiberglass composites. Flexural tests were conducted following procedure A of ASTM D790 for the epoxy samples reinforced with GNP and MCC, and ASTM D7264 for the carbon fiber and fiberglass composites reinforced with GNP and MCC. A crosshead displacement rate of 1.28 mm·min^{-1} was used for the epoxy samples, and 1 mm·min^{-1} was used for the carbon fiber and fiberglass composites. Flexural tests were conducted until specimen failure. The test data were then plotted on stress-strain curves and analyzed to extract the mechanical properties.

2.5 Scanning electron microscopy (SEM)

The dispersion of GNP and MCC was examined using a SEM (Zeiss EVO 10, Technical Division, Directorate of Armament, Royal Thai Air Force) operating at 25 kV.

2.6 Transmission electron microscopy (TEM)

TEM was used to investigate the structure and morphology of graphene nanoplatelets (GNPs), providing detailed imaging and characterization of their atomic structure, layer thickness, and defects. Additionally, TEM offered insights into the size, shape, and quality of GNP distribution after ultrasonic treatment. The analysis was performed using a transmission electron microscope from Thermo Scientific, model TALOS F200X, located at the Synchrotron Light Research Institute (Public Organization).

2.7 X-ray tomographic microscopy (XTM)

The XTM beamline located at BL1.2W at SLRI was used to study the distribution of GNP in epoxy with the synchrotron radiation generated from the 2.2-Tesla multipole wiggler of Siam Photon Source (SPS). The XTM irradiator provides a high-intensity X-ray beam to provide detailed cross-sectional reconstructions and 3D visualizations of the samples.

3. Results and discussion

3.1 Dispersion of GNP and MCC

SEM and TEM were used to examine the dispersion quality of GNPs after the ultrasonication treatment. Figure 1 shows the SEM micrographs of the pristine GNPs and dispersed GNPs. The pristine GNPs were clustered into big chunks with an average size of 300 μm, but after the ultrasonication treatment, the GNPs were no longer clustered and more uniformly distributed, becoming smaller particles with an average size of 27.5 μm. This shows that the ultrasonication successfully addressed the aggregation issue.

Figure 2 shows TEM micrographs of the pristine GNPs and dispersed GNPs. Many layers of pristine GNP sheets appeared to stack on top of each other and aggregated into a large cluster with sharp edges, suggesting that the GNPs were tightly packed. After the dispersion, the GNP sheets were separated with only a few layers stacked. There were no longer sharp edges shown and the GNP spread out into smaller sheets with smoother surfaces, thereby reducing their overall size [16]. Owing to the smaller number of sheets stacked, they were more transparent, and different layers could easily be seen through. This confirms that the GNPs were successfully dispersed.

Similarly, the dispersion of MCC was examined with SEM. The SEM micrographs showed that there were only minimal differences observed in MCC after dispersion compared with the pristine ones, where MCC appeared as small bars and average size of 12 μm shown in as shown in Figure 3. This suggests that ultrasonication treatment may not be necessary for MCC. The size of MCC (microcrystalline cellulose) can significantly affect the properties of the composites. Smaller MCC particles generally have a higher surface area, which can enhance interfacial adhesion with the matrix, improving mechanical properties like tensile strength and modulus. However, if the particles are too large or poorly dispersed, they may act as defects, reducing composite performance [22].

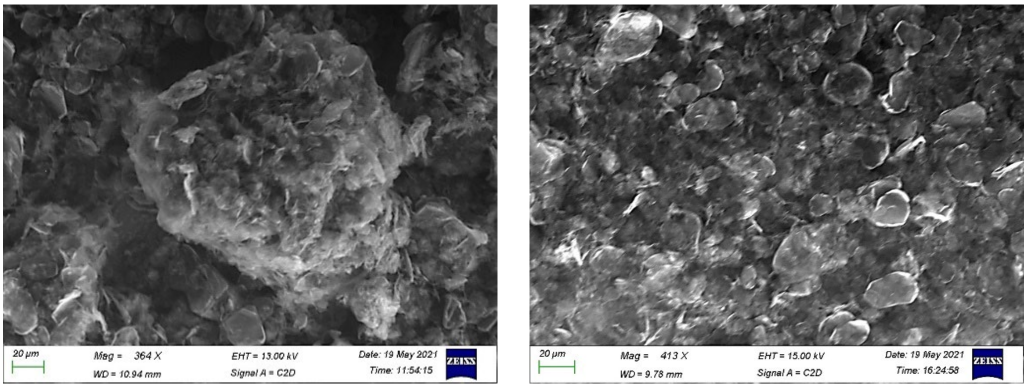

Figure 1. SEM micrographs of pristine GNPs (left) and dispersed GNPs (right).

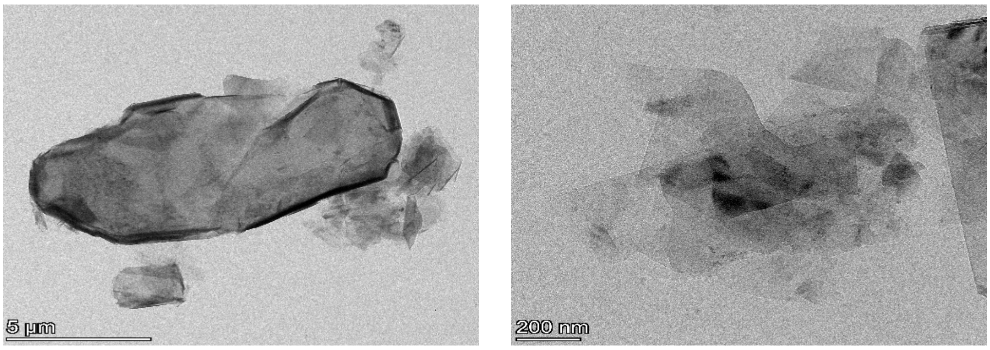

Figure 2. TEM micrographs of pristine GNPs (left) and dispersed GNPs (right).

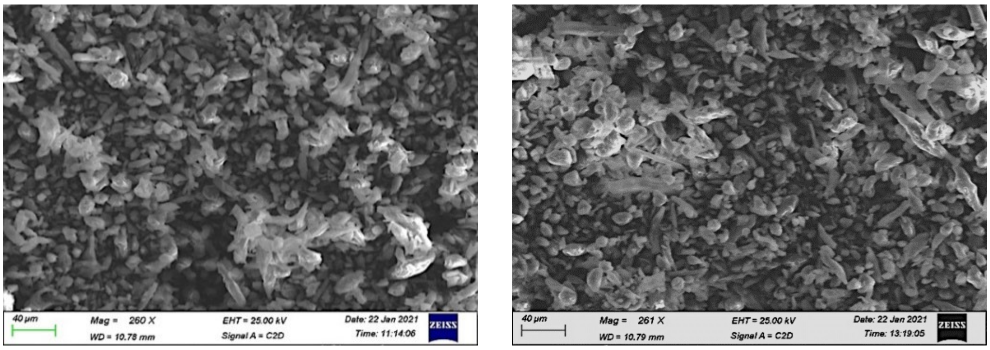

Figure 3. SEM micrographs of pristine MCC (left) and dispersed MCC (right).

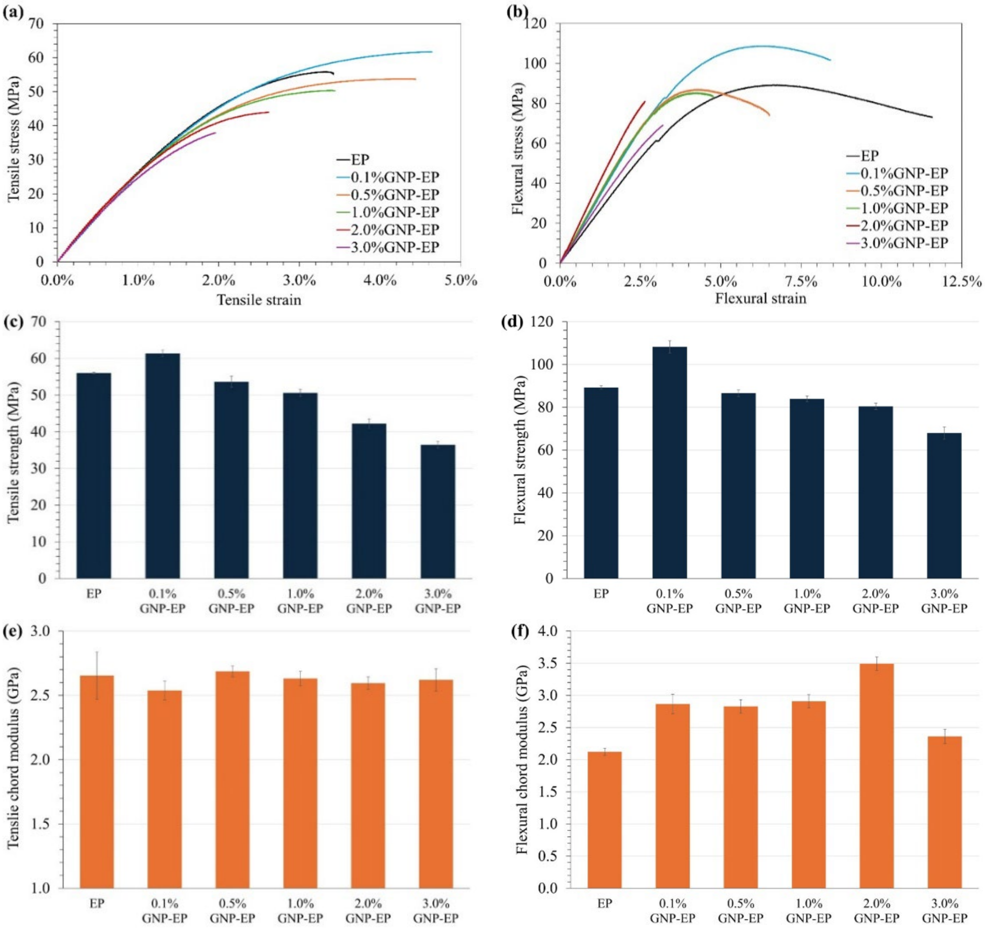

Figure 4. (a) The tensile stress-strain curves, (b) flexural stress-strain curves of the EP and GNP-EP samples, (c) their corresponding tensile strength, (d) flexural strength, and (e) tensile chord modulus and (f) flexural chord modulus.

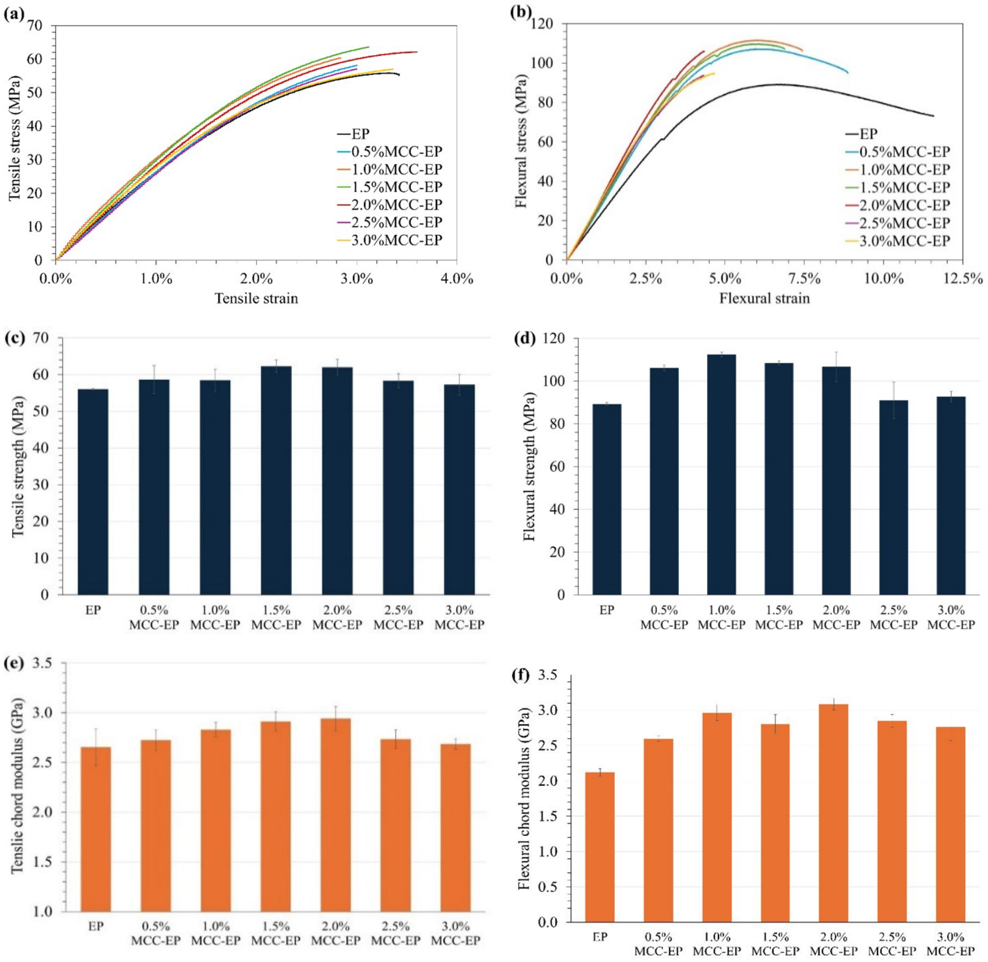

Figure 5. (a) The tensile stress-strain curves, (b) flexural stress-strain curves of the EP and MCC-EP samples, (c) their corresponding tensile strength, (d) flexural strength, and (e) tensile chord modulus, and (f) flexural chord modulus.

3.2 Mechanical properties of GNP- and MCC-reinforced epoxy

3.2.1 GNP-reinforced epoxy (GNP-EP)

The epoxy samples reinforced with 0.1, 0.5, 1.0, 2.0, and 3.0 wt% GNPs were tested for their tensile properties according to ASTM D638, compared with those of the neat epoxy (EP). The results were plotted as stress-strain curves and the tensile properties were extracted and plotted as bar charts as shown in Figure 4. The 0.1% GNP-EP exhibited the highest tensile strength of 61.3 MPa. However, as the GNP content increased, the tensile strength of the composites decreased. The ultimate tensile strain and the plastic deformation region also decreased as seen in the stress-strain curves. The tensile chord modulus, on the other hand, was roughly the same for all samples. This shows that the small amount of GNP could enhance the tensile strength of the epoxy whereas the tensile modulus was not affected. For the flexural properties, the GNP-EP samples were tested according to ASTM D790 and compared the results with those of the EP. The results were plotted as stress-strain curves and the flexural properties were extracted and plotted as bar charts as shown in Figure 4. The results showed that the addition of GNP affected both the flexural strength and modulus. The 0.1% GNP-EP exhibited the highest flexural strength of 108 MPa. However, as the GNP content increased, the flexural strength of the composites decreased. The ultimate flexural strain and the plastic deformation region also decreased as seen in the stress-strain curves, i.e., the composites became more brittle. The addition of 2.0 wt% GNP resulted in the highest flexural chord modulus of 3.49 GPa.

3.2.2 MCC-reinforced epoxy (MCC-EP)

The epoxy samples reinforced with 0.5, 1.0, 1.5, 2.0, 2.5 and 3.0 wt% MCC were tested for their tensile properties according to ASTM D638, compared with those of the EP. The results were plotted as stress-strain curves and the tensile properties were extracted and plotted as bar charts as shown in Figure 5. There was not much change in shape of the stress-strain curves as the MCC content increased. The tensile strength of the samples increased as the MCC content increased. The highest strength of 62.0 MPa was reached by the 2.0% MCC-EP sample and then declined as the content increased. Similarly, the tensile modulus increased as the MCC content increased and reached the highest values of 2.94 GPa by the 2.0% MCC-EP. Then, the modulus decreased with more MCC content. For the flexural properties, the MCC-EP samples were tested according to ASTM D790 and compared the results with those of the EP. The results were plotted as stress-strain curves and the flexural properties were extracted and plotted as bar charts as shown in Figure 5. The flexural strength and modulus increased as the MCC content increased and reached their peaks at 112 MPa and 3.09 GPa by the 1.0% MCC-EP and 2.0% MCC-EP samples, respectively. Increasing the MCC content further reduced the flexural strength and modulus. The ultimate flexural strain and the plastic deformation region also decreased as seen in the stress-strain curves, i.e., the composites became more brittle.

From the results, it was clearly demonstrated that the addition of GNP and MCC into epoxy resin improved both tensile and flexural properties significantly. Specifically, the GNP reinforcement notably enhanced the tensile strength, flexural strength and flexural modulus of the composite. Particularly at 0.1 wt% concentrations, the results showed an increase in tensile strength and flexural strength over those of the EP by about 10% and 21%, respectively, as shown in Figure 6 and Table 1. At 2.0 wt%, a substantial increase in the flexural modulus by 65% compared to EP was shown. This suggested that the addition of carbon structures enhanced epoxy's flexibility, contrasting its inherent brittleness [1,2,7]. Effective dispersion of the reinforcing agents within the epoxy is crucial for enhancing composite properties, as confirmed by XTM analysis as shown in Figure 7, which revealed well-dispersed GNP throughout the epoxy matrix [10]. The 2D carbon structure of GNP, with its large surface area, increased contact with the polymer matrix, facilitating optimal stress transfer from the polymer to the nanoflakes.

Figure 7 showed the XTM images of the pristine GNPs mixed in epoxy compared with the dispersed GNPs that underwent ultrasonication treatment before mixing in the epoxy. Figure 7(a-b) showed epoxy background in brown while in Figure 7(c-d), the epoxy background was removed to allow a clearer observation of GNPs. As seen from the images, the pristine GNPs were visibly clustered together, forming large aggregates within the epoxy, while the dispersed case, the XTM could only showed a few tiny spots of the smaller GNP particles. It was indicated that most of them were too small and more uniformly spread throughout the epoxy. The results from the XTM images were consistent with the SEM and TEM results, confirming the effectiveness of the dispersion methods in uniformly dispersing GNPs.

The incorporation of MCC significantly enhanced the tensile and flexural properties. Particularly evident at the 2.0 wt% MCC concentration where the tensile strength, tensile modulus and flexural modulus were enhanced by about 11%, 11% and 46%, respectively, compared to those of EP. Also, at 1.0 wt%, the flexural strength was improved by about 26% compared to that of EP. This may be due to the high crystallinity of MCC and the formation of hydrogen bonds with the polymer matrix enhance interfacial interactions, thereby improving composite mechanical properties [8,14]. However, increasing the ratio of reinforcing agents can lead to inhomogeneous distribution and filler agglomeration, causing stress concentrations and restricting polymer chain mobility. This results in reduced elongation at break and partial decreases in tensile strength at higher filler loadings [16].

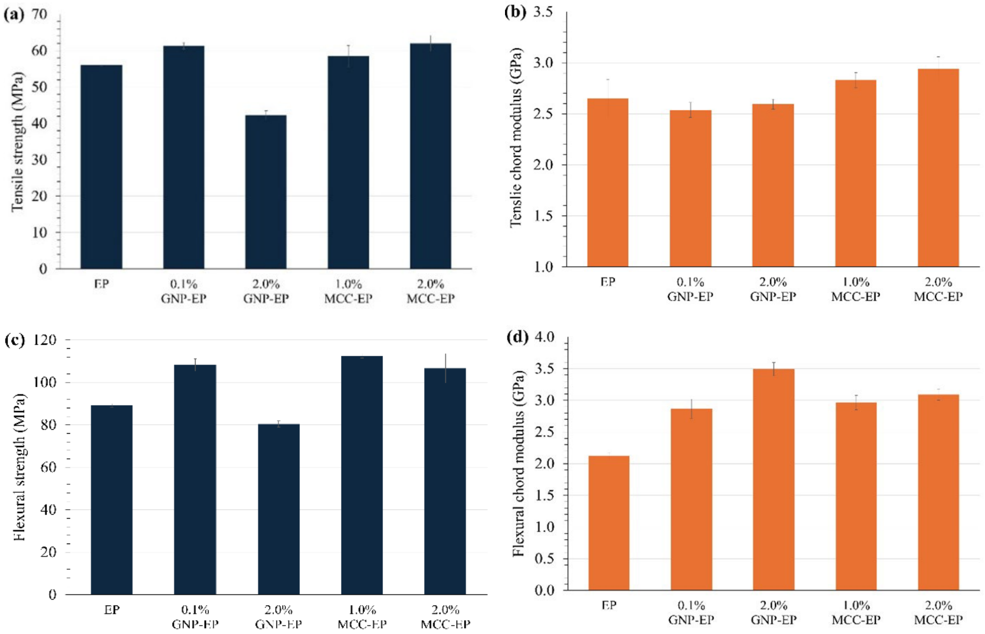

Figure 6. Comparison of (a) tensile strength, (b) tensile chord modulus, (c) flexural strength, and (d) flexural chord modulus of the selected GNP-EP and MCC-EP samples.

Table 1. The mechanical properties of the selected GNP-EP and MCC-EP samples.

Sample	**Tensile properties**				**Flexural properties**			
	Strength [MPa]	**SD**	**Modulus [GPa]**	**SD**	**Strength [MPa]**	**SD**	**Modulus [GPa]**	**SD**
EP	56.0	0.20	2.65	0.18	89.20.78	2.12	0.05	EP
0.1%GNP-EP	61.3	0.88	2.54	0.07	108 2.87	2.87	0.15	0.1%GNP-EP
2.0%GNP-EP	42.2	1.50	2.59	0.04	80.41.46	3.49	0.10	2.0%GNP-EP
1.0%MCC-EP	58.5	2.96	2.83	0.07	113 1.07	2.96	0.84	1.0%MCC-EP
2.0%MCC-EP	62.0	2.19	2.94	0.12	107 6.96	3.09	0.97	2.0%MCC-EP

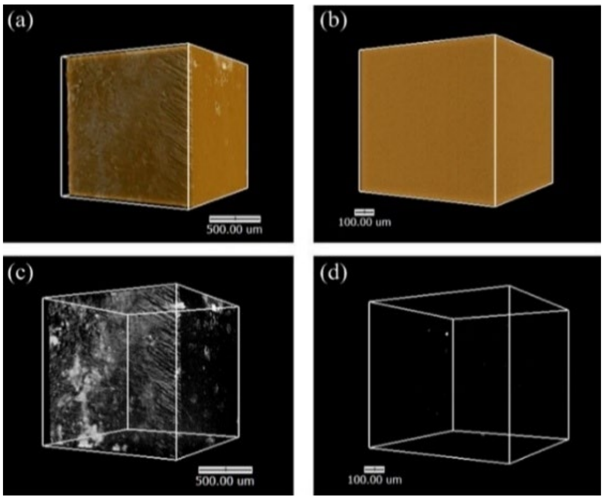

Figure 7. XTM images of (a) the pristine GNPs, (b) dispersed GNPs in the epoxy background in brown and the XTM wireframe images of, (c) the pristine GNPs, and (d) dispersed GNPs with the epoxy background removed.

In comparison, with optimal concentrations, both GNP and MCC improved tensile strength of the epoxy composites. However, in the GNP-EP case, the tensile strength increased slightly, and the tensile modulus decreased slightly. Graphene nanoplatelets (GNPs) enhance the epoxy by effectively bridging microcracks [23] and facilitating efficient load transfer when adequately dispersed. Nonetheless, due to their platelet-like structure, they primarily reinforce the composite in the plane of stress application rather than substantially in the tensile direction. Furthermore, any occurrence of agglomeration may create stress concentration sites, thereby limiting the potential for strength enhancement. The reasons for its decrease in tensile modulus may result from the nature of GNPs. Although GNPs exhibit rigidity, their distribution and orientation within the matrix are essential factors influencing the modulus. If GNPs align in a more random manner or result in polymer chain slippage, they can diminish stiffness, causing a slight reduction in modulus. Additionally, certain GNPs increase the free volume between polymer chains, which reduces the overall stiffness of the composite material. In the case of MCC-EP, both tensile strength and modulus were slightly higher than those of GNP-EP. MCC is made of crystalline cellulose microfibers that strongly bond with epoxy through hydrogen bonding. This bonding facilitates efficient load transfer, resulting in a moderate increase in tensile strength. Additionally, MCC are rigid, elongated reinforcements that align along the tensile axis, unlike GNPs. Their ability to limit epoxy chain mobility and enhance load-bearing capacity leads to a greater improvement in modulus. While MCC is less stiff than GNP, it interacts better with epoxy, resulting in a greater tensile modulus improvement.

Comparing the flexural properties, GNP significantly increases flexural strength and flexural modulus. GNPs are known for their ability to resist bending forces due to their high aspect ratio. In flexural tests, the composite undergoes both tensile (bottom) and compressive (top) stresses, and GNPs effectively resist both. Their capacity to bridge cracks [23] and enhance stress transfer [24] is more evident in flexural loading than in tensile loading. Moreover, under flexural loading, graphene nanoplatelets (GNPs) exhibit better alignment within the composite structure [25], thereby enhancing stiffness. The layered configuration of GNPs offers superior resistance to deformation during bending, resulting in a significant increase in modulus. Contrary to tensile tests—where polymer chains may slip around GNPs—in flexural tests, GNPs contribute more directly to stiffness. In the case of MCC-EP, MCC significantly increases flexural strength (slightly higher than GNP's) and flexural modulus (but less than GNP's). MCC bonds better with epoxy than GNPs, enhancing load transfer in bending. Its fibrous nature distributes stress evenly, improving flexural strength more than GNPs. MCC also increases toughness by absorbing energy through micro-fibrillation and deflecting cracks. While MCC stiffens the composite, it doesn't match GNP's intrinsic stiffness. Due to their planar structure and high stiffness, GNPs enhance the modulus in bending more effectively. MCC also introduces energy absorption mechanisms, like micro-fibril sliding [26,27], which reduces the stiffness gain compared to GNPs.

Choosing the appropriate concentrations of GNP and MCC is crucial for optimizing the mechanical properties of epoxy resin composites, as it involves balancing improvements in strength and modulus while considering the effects of dispersion and agglomeration. Based on their performance on the strength and modulus improvements, we selected 0.1% and 2.0% GNP, and 1.0% and 2.0% MCC for further investigation on the effects of GNP and MCC on the mechanical properties of fiberglass and carbon fiber reinforced composites in the next sections.

3.3 Mechanical properties of the GNP-CF and MCC-CF composites

3.3.1 Tensile properties

The tensile properties of the CF, 0.1% GNP-CF, 2.0% GNP-CF, 1.0% MCC-CF, and 2.0% MCC-CF were tested in accordance with ASTM D3039. The results were plotted as stress-strain curves and the tensile properties were extracted and plotted as bar charts as shown in Figure 8. The 0.1% GNP-CF exhibited the highest tensile strength of 506 MPa which is a 14% improvement over the CF. This was to be expected as the tensile strength of the epoxy was also enhanced with the addition of 0.1 wt% GNP by about 10%. This may be due to the interfacial interaction between the GNP and CF surfaces. The graphite-like structure, which is characterized by hexagonal covalent bonds and interplanar Van der Waals forces, results in highly anisotropic mechanical properties. The GNP-CF composites benefit from the carbon structure of the GNPs, which enhances the interface between the graphene sheets and carbon fiber edges, leading to increased tensile strength [9,10]. Additionally, the enhanced adhesion between the reinforcement and the matrix, achieved by uniformly distributing GNPs within the epoxy and integrating them with the carbon fibers, improves load transfer and consequently increases tensile strength. Interestingly, while the tensile modulus was not improved in the 0.1% GNP-EP, it was improved by about 32% (14.4 GPa) in the case of the 0.1% GNP-CF. GNPs enhance the flexural modulus by increasing the stiffness of the matrix, thereby improving the overall stiffness of the composite, particularly at low to medium concentrations. Nevertheless, adding GNPs at high concentration can decrease the modulus. Like in the case of 2.0% GNP-CF, the excess GNPs could not be uniformly distributed within the matrix, leading to agglomeration which limits the exposure of the GNP to the CF surfaces and causes uneven stress distribution [11], resulting in a slight increase in tensile modulus and a dramatic decrease in tensile strength [8]. Hence, incorporation of GNP at appropriate levels can increase the composite's flexural modulus, resulting in greater stiffness and improved resistance to flexural deformation.

For the 1.0% MCC-CF and 2.0% MCC-CF, we could see a significant decrease in tensile strength as the MCC content increased. This contrasted the results from the MCC-EP samples where their tensile strengths were improved. The reduction in strength was due to the lower strength of cellulose fibers compared to carbon fibers. In composites with a balanced fiber content, using the right proportions can reduce the weight of the composite without significantly affecting its tensile strength [14]. Also, this could be because of the lack of interaction between the MCC and CF. On the contrary, the tensile modulus increases as the MCC content increased. The 2.0% MCC-CF exhibited the highest tensile modulus of 15.0 GPa which was about 37% greater than that of the CF. The tensile modulus of the composite falls between that of carbon fiber and cellulose, with a higher carbon fiber content resulting in a stiffer composite [28].

3.3.2 Flexural properties

The flexural properties of the CF, 0.1% GNP-CF, 2.0% GNP-CF, 1.0% MCC-CF, and 2.0% MCC-CF were tested in accordance with ASTM D7264. The results were plotted as stress-strain curves and the flexural properties were extracted and plotted as bar charts as shown in Figure 9. The flexural strength of the composites increased as the GNP content increased. Particularly, the 2.0% GNP-CF showed the highest flexural strength of 348 MPa which was about 36% higher than that of the CF. Nevertheless, the flexural moduli of the 1.0% GNP-CF and 2.0% GNP-CF were not improved but slightly reduced instead. These results contradicted those of the GNP-EP samples where their moduli were dramatically enhanced and only the strength of the 0.1% GNP-EP was enhanced. This may suggest that the interfacial interaction between the GNP and CF surfaces was more beneficial to the flexural strength than modulus. Its high length-to-width ratio, 2D structure, good dispersion, and compatibility with fibers all contributed to the enhancement of stress transfer and slows down crack propagation in the material. The enhanced adhesion between the reinforcement and the matrix, achieved through the uniform distribution of GNP within the epoxy and its integration with the carbon fibers, contributed to better load transfer and resulted in the observed increase in flexural strength [10].

In the case of 1.0% MCC-CF and 2.0% MCC-CF samples, both the flexural strength and modulus decreased as the MCC content increased. This contrasts with the results from the MCC-EP counterparts where both flexural strength and modulus were dramatically enhanced. This reduction in strength and modulus may be attributed to the incompatibility between the MCC and CF [29], e.g., differences in hydrophilicity, causing adhesion issues at the interface, i.e., MCC and CF do not adhere well to the matrix [30], reducing load transfer ability and hence the composite's overall strength. Also, MCC and CF possess significantly different mechanical properties—CF are much stronger and stiffer than MCC. As a result, mixing them could lead to uneven stress distribution within the composite, ultimately diminishing mechanical performance [31-33].

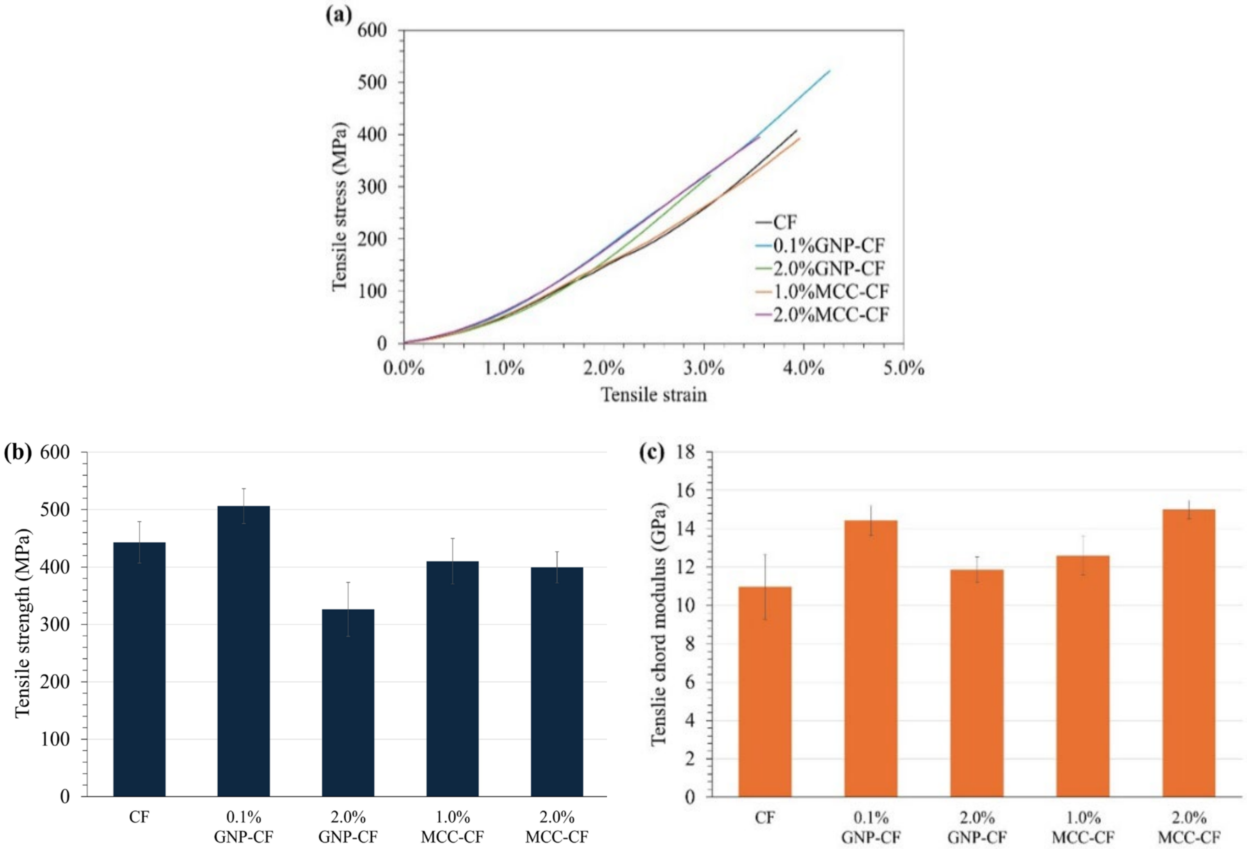

Figure 8. (a) The tensile stress–strain curves of the CF, GNP-CF and MCC-CF samples, (b) their corresponding tensile strength, and (c) tensile chord modulus.

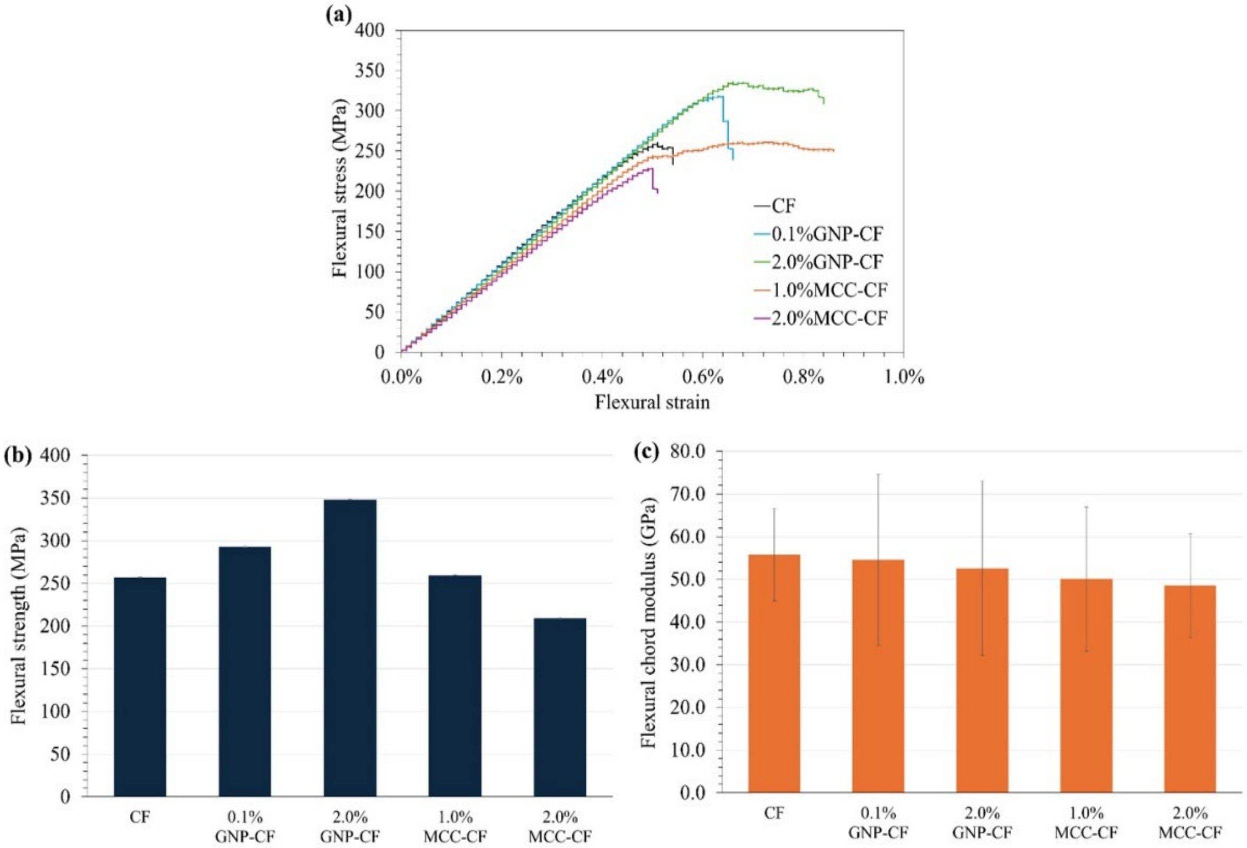

Figure 9. (a) The flexural stress–strain curves of the CF, GNP-CF and MCC-CF samples, (b) their corresponding flexural strength, and (c) flexural chord modulus.

Table 2. The mechanical properties of the CF, GNP-CF and MCC-CF composites.

Sample	Tensile properties				Flexural properties			
	Strength [MPa]	SD	Modulus [GPa]	SD	Strength [MPa]	SD	Modulus [GPa]	SD
CF	443	36.15	10.96	1.70	257	0.51	55.8	10.76
0.1%GNP-CF	506	30.35	14.42	0.77	293	0.57	54.6	19.96
2.0%GNP-CF	326	47.09	11.86	0.67	348	0.69	52.6	20.43
1.0%MCC-CF	410	39.63	12.60	1.02	259	0.68	50.1	16.87
2.0%MCC-CF	399	27.18	14.99	0.47	209	0.89	48.6	12.02

Table 2 summarizes the tensile properties and flexural properties of the CF, GNP-CF, and MCC-CF composites.

Figure 10 shows the SEM micrographs of the fracture surfaces of the tested CF, GNP-CF, and MCC-CF specimens at 1000x and 10000x magnifications. The CF specimen showed a sharp fracture as shown in Figure 10(a) which was to be expected due to the brittle nature of carbon fibers. The surface of the matrix groves left by the fers were rather clean and smooth as shown in Figure 10(b). For the GNP-CF case, the fractured fibers were not as sharp and clean as the CF case, i.e., the fractured fibers were not completely separated and there was more residual resin attached to the fibers as shown in Figure 10(c). Moreover, the matrix surface was rougher with tiny particles attached as shown in Figure 10(d). This suggested that the addition of GNPs helped improve mechanical properties by enhancing the interfacing between the epoxy and carbon fibers and also acted as crack deflectors and stress distributors. In the case of MCC-CF, the fracture was not as sharp as that of the CF, but the fractured fibers were separated as shown in Figure 10(e). The surface of the matrix was much rougher than that of the GNP-CF and contained larger particles on the surface as shown in Figure 10(f). This suggested weak interfacial adhesion between the EP and CF due to its incompatibility with the CF, leading to decreases in mechanical properties in general. However, with the strong hydrogen bonding between MCC and epoxy—hence the larger residual resin particles—and its ability to restrict polymer chain movement, it was able to improve the tensile modulus of the composite.

3.4.2 Flexural properties

3.4 Mechanical properties of the GNP-FG and MCC-FG composites

3.4.1 Tensile properties

The tensile properties of the FG, 0.1% GNP-FG, 2.0% GNP-FG, 1.0% MCC-FG and 2.0% MCC-FG were tested in accordance with

ASTM D3039. The results were plotted as stress-strain curves and the tensile properties were extracted and plotted as bar charts as shown in Figure 11. The tensile strength of the GNP-FG samples slightly decreased as the GNP content increased while the tensile modulus significantly decreased. The tensile strengths of the 0.1% GNP-FG and 2.0% GNP-FG were reduced by about 2% and 6%, respectively, compared to that of the FG. The poor tensile strength of the 0.1% GNP-FG may result from poor dispersion and adhesion issues at the interface. Consequently, the GNP in the matrix cannot sufficiently penetrate or enhance the efficiency of the interface between the EP and FG, thereby reducing the tensile strength [34,35]. Their tensile moduli, however, were considerably reduced by about 15% and 20%, respectively, which is not beyond expectation as the 0.1% GNP-EP and 2.0% GNP-EP did not possessed such an improvement either.

The results of tensile property tests of 1.0% MCC-FG and 2.0% MCC-FC showed that both the tensile strength and tensile modulus of MCC-FG samples were significantly reduced. These decreases may be attributed to the inability of the MCC in the epoxy to effectively penetrate and enhance the interfacing between the EP and the FG. This inefficiency can lead to a reduction in tensile strength [35,36].

The flexural properties of the FG, 0.1% GNP-FG, 2.0% GNP-FG, 1.0% MCC-FG, and 2.0% MCC-FG were tested according to ASTM D7264. The results were plotted as stress-strain curves and the flexural properties were extracted and plotted as bar charts as shown in Figure 12. The results showed that the 0.1% GNP-FG showed the highest flexural strength, which was about 8.0% greater than that of the FG, but it was significantly reduced by about 15% for the 2.0% GNP-GF. Their flexural chord moduli were also significantly reduced by about 14% and 18%, respectively. This may result from an excessive GNP content leading to poor dispersion, poor interfacial bonding, and poor processing can degrade performance [18].

The flexural strengths for the MCC-FG samples decreased as the MCC concentration increased. The flexural strength of the 1.0% MCC-FG and 2.0% MCC-FG were reduced by about 15% and 31%, respectively, compared to that of the FG. Though, the flexural modulus of the 1.0% MCC- FG improved slightly by about 4%. However, the flexural modulus was lowered by about 4% for the 2.0% MCC-FG. This again was possibly caused by the lack of interfacial interaction between the MCC and fiberglass.

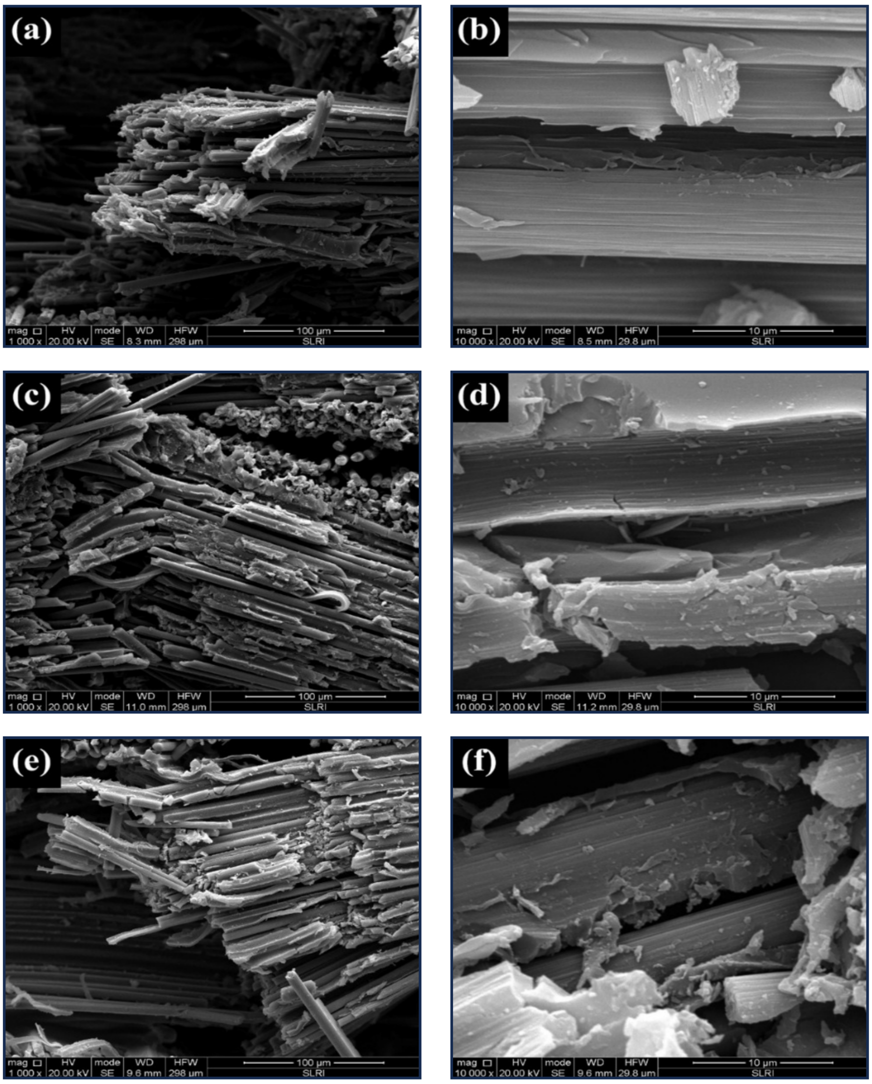

Figure 10. SEM micrographs of the fracture surfaces of CF at (a) 1000x, (b) 10000x, GNP-CF at (c)1000x, (d) 10000x, and MCC-CF at (e) 1000x (f) 10000x after mechanical properties testing.

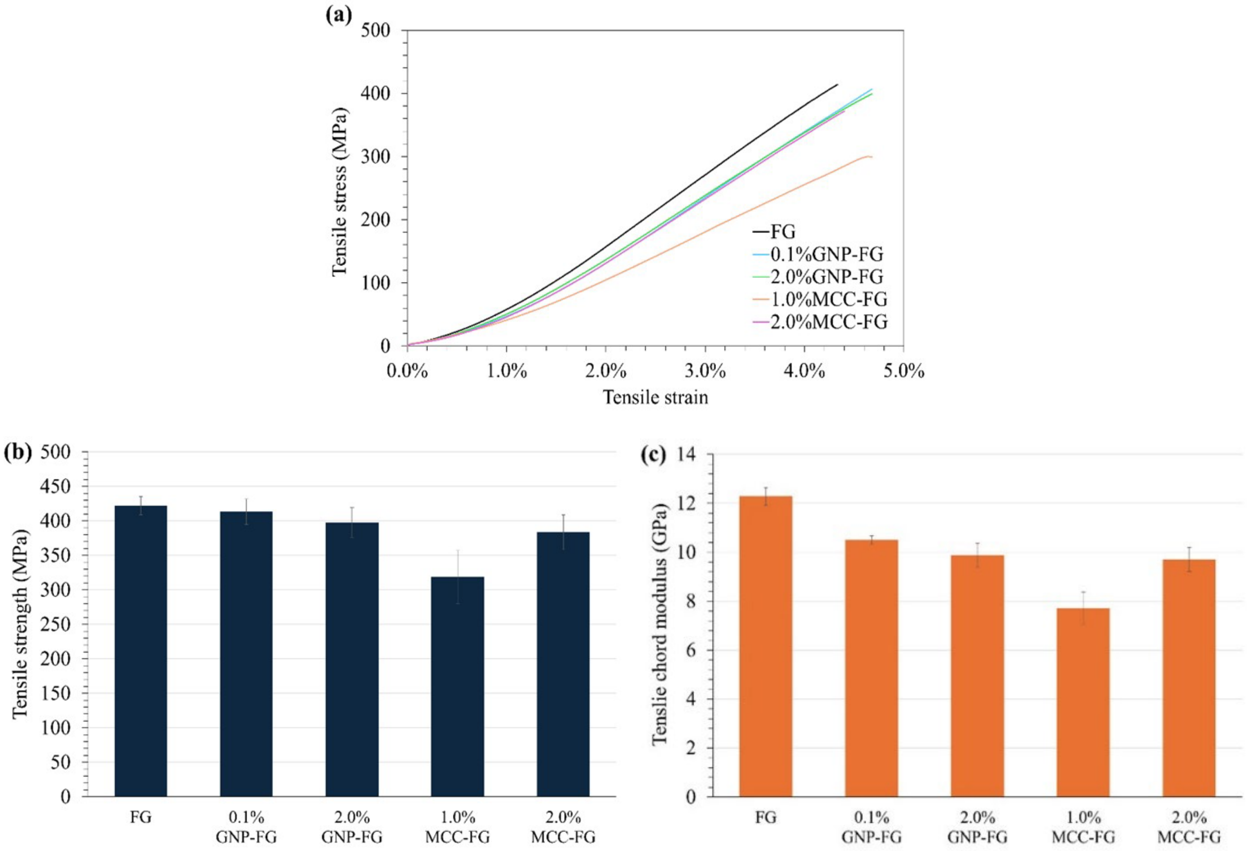

Figure 11. (a) The tensile stress–strain curves of the FG, GNP-FG and MCC-FG samples, (b) their corresponding tensile strength, and (c) tensile chord modulus.

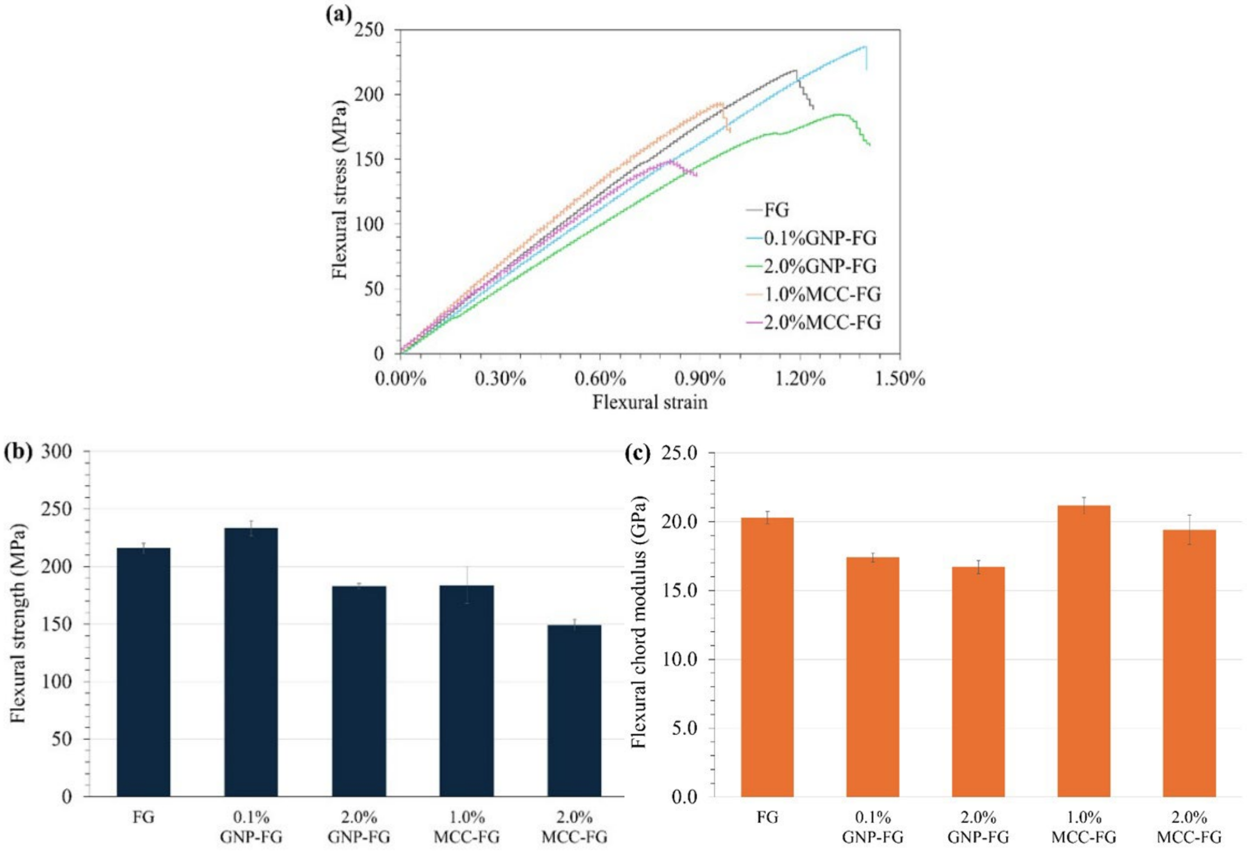

Figure 12. (a) The flexural stress–strain curves of the FG, GNP-FG and MCC-FG samples, (b) their corresponding flexural strength, and (c) flexural chord modulus.

Table 3 summarizes the tensile properties and flexural properties of the FG, GNP-CF, and MCC-CF composites.

Figure 13 shows the SEM micrographs of the fracture surfaces of the tested FG, GNP-FG, and MCC-FG specimens at 1000x and 10000x magnifications. As seen in Figure 13(a), the fractured fiberglass fibers were more separated from each other than those of CF and there was less matrix remaining attached. The surfaces of the matrix and fiber were also very smooth as shown in Figure 13(b). This indicated a weaker interfacing than that of CF, resulting in poorer mechanical properties in general, in addition to the lower fiber strength. As shown in Figure 13(c-d), there were more residual resin that stayed intact, and they attached to the fibers slightly more than in the case of FG. This suggested that the GNPs' crack-bridging effect helped strengthen the matrix slightly, resulting in a small increase in flexural strength. Nonetheless, due to poor adhesion with the fiberglass, the improvement is minor. In the case of MCC-FG, there was less residual resin and there was less interfacing between the matrix and fibers, i.e., the matrix was clearly separated from the fibers. This suggested a mismatch between MCC and fiberglass. However, despite the reduction in mechanical properties of the fiberglass composites in general, MCC's rigid crystalline structure still provides minor stiffness enhancement

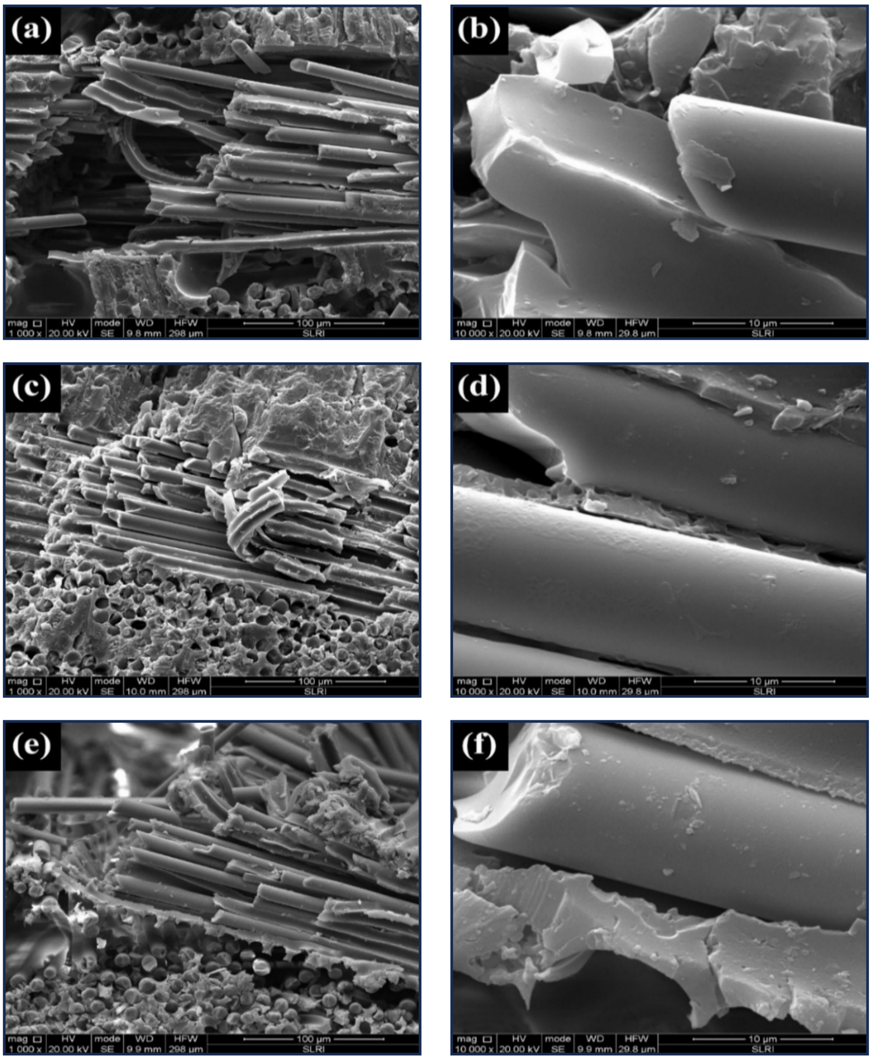

Figure 13. SEM micrographs of the fracture surfaces of FG at (a) 1000x and (b) 10000x, GNP-FG at 1000x and 10000x, and MCC-FG at 1000x and at 10000x after mechanical properties testing

Table 3. The mechanical properties of the FG, GNP-FG and MCC-FG composites.

Sample	Tensile properties				Flexural properties			
	Strength [MPa]	SD	Modulus [GPa]	SD	Strength [MPa]	SD	Modulus [GPa]	SD
FG	422	13.23	12.3	0.36	216	4.31	20.3	0.45
0.1%GNP-FG	413	18.30	10.5	0.17	233	6.44	17.4	0.32
2.0%GNP-FG	398	21.54	9.88	0.48	183	2.06	16.7	0.48
1.0%MCC-FG	318	38.76	7.72	0.67	184	15.87	21.2	0.60
2.0%MCC-FG	384	25.36	9.71	0.50	149	4.54	19.4	1.06

4. Conclusions

This work investigated the effects of GNP and MCC reinforcements on the tensile and flexural properties of epoxy composites, carbon fiber composites, and glass fiber composites. First, the effects on the mechanical properties of the epoxy resin reinforced with 0.1, 0.5, 1.0, 2.0, 3.0 wt.% GNP and 0.5, 1.0, 1.5, 2.0, 2.5, 3.0 wt% MCC were studied. The results revealed that GNP and MCC could improve both tensile and flexural properties of the epoxy significantly. Specifically, with 0.1 wt.% GNP, the tensile strength and flexural strength were improved by 10% and 21%, respectively, compar ed to those of EP. At 2.0 wt%, a substantial increase in the flexural modulus by 65%. The incorporation of 2.0 wt% MCC significantly enhanced the tensile strength, tensile modulus, and flexural modulus by 11%, 11%, and 46%, respectively. Also, at 1.0 wt%, the flexural strength was improved by 26% compared to that of EP. Based on their performance, the 0.1% GNP, 2.0% GNP, 1.0% MCC, and 2.0% MCC, were chosen for further investigation on their effects on the mechanical properties of CF and FG composites.

For the carbon fiber composites, the addition of both GNP and MCC resulted in the improved mechanical properties. Particularly, the 0.1% GNP enhanced the tensile strength by 14% and tensile modulus by 32% compared to those of the CF. The flexural strength of the composites increased as the GNP content increased. Specifically, at 2.0 wt%, the flexural strength was improved by 36%. As for MCC, a significant decrease in tensile strength was observed as the MCC increased. However, the tensile modulus increased as the MCC content increased. For 2.0% MCC-CF, the tensile modulus was 37% greater than that of the CF. In the case of flexural properties, the MCC had negative effects on both flexural strength and modulus.

For the fiberglass composites, GNP and MCC generally had negative impacts on both tensile and flexural properties. Only 0.1% GNP-FG and 1.0% MCC-FG showed slight improvements in the flexural strength by 8.0% and the flexural modulus by 4%, respectively. This was likely attributed to a variety of reasons such as poor dispersion and poor interfacing between the GNP, MCC and the FG.

This study provides valuable insights into the reinforcement mechanisms of GNP and MCC in different polymer matrix composites.

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