Investigation of flexural properties of epoxy composite by utilizing graphene nanofillers and natural hemp fibre reinforcement

Shankar A Hallad[1,2], Sharanabasava V Ganachari[3], Manzoore Elahi M Soudagar[4,5], NR Banapurmath[1,2], Anand M Hunashyal[6], Islam MR Fattah[7], Fayaz Hussain[8], Muhammad A Mujtaba[9], Asif Afzal[5,10], Mohammad S Kabir[11] and Ashraf Elfasakhany[12]

1Center for Material Science, KLE Technological University, Hubballi, Karnataka, India
2Department of Mechanical Engineering, KLE Technological University, Hubballi, Karnataka, India
3School of Advanced Sciences, KLE Technological University, Hubballi, Karnataka, India
4Department of Mechanical Engineering, School of Technology, Glocal University, Delhi-Yamunotri Marg, Uttar Pradesh, India
5Department of Mechanical Engineering and University Centre for Research & Development, Chandigarh University, Mohali, Punjab, India 6Department of Civil Engineering, KLE Technology University, Hubballi, Karnataka, India
7Centre for Green Technology, School of Civil and Environmental Engineering, Faculty of Engineering and IT, University of Technology Sydney, Ultimo, New South Wales, Australia
8Modeling Evolutionary Algorithms Simulation and Artificial Intelligence, Faculty of Electrical & Electronics Engineering, Ton Duc Thang University, Ho Chi Minh City, Vietnam
9Department of Mechanical Engineering, Faculty of Engineering, University of Malaya, Kuala Lumpur, Malaysia
10Department of Mechanical Engineering, P. A. College of Engineering (Affiliated to Visvesvaraya Technological University, Belgavi), Mangaluru, India 11Institute for Frontier Materials, Deakin University, Waurn Ponds, VIC, Australia
12Mechanical Engineering Department, College of Engineering, Taif University, Taif, Saudi Arabia

Corresponding author:
Mohammad S Kabir, Institute for Frontier Materials, Deakin University, Waurn Ponds, 75 Pigdons Road, VIC 3216, Australia.
Email: mohammad.kabir@deakin.edu.au

Abstract
This study aims to determine the optimum reinforcement required to attain the best combination of flexural strength of modified green composites (graphene oxide + hemp fibre reinforced epoxy composites) for potential use in structural applications. An attempt was also made for the combination of graphene and hemp fibres to enhance load-bearing ability. The infusion of hemp and graphene was made by the weight of the base matrix (epoxy composite). Results showed that graphene reinforcement at 0.4 wt.% of matrix showed load-sustaining capacity of 0.76 kN or 760 MPa. In the case of hemp fibre reinforcement at 0.2 wt.% of the matrix, infusion showed enhanced load-bearing ability (0.79 kN or 790 MPa). However, the combination of graphene (0.1 wt.% graphene nanofillers) and hemp (5 wt.% hemp fibre) indicated a load-sustaining ability of 0.425 kN or 425 MPa, whereas maximum deflection was observed for specimen with hemp 7.5 % + graphene 0.2 % with 1.9 mm. Graphene addition to the modified composites in combination with natural fibres showed promising results in enhancing the mechanical properties under study. Moreover, graphene-modified com-posites exhibited higher thermal resistance compared to natural fibre reinforced composites. However, when nanofiller rein-forcement exceeded a threshold value, the composites exhibited reduced flexural strength as a result of nanofiller agglomeration.

Keywords
Graphene nanoparticles, hemp fibres, three-point loading, flexure strength, thermogravimetric analysis (TGA)

Introduction

Established as a 'super material', graphene has been utilized as a reinforcing phase in metals and composites since the last two decades,[1–7] because of its unique mechanical and physical properties, especially in terms of high surface area. It was reported that graphene is more resilient than steel, lighter than aluminium and has better conductivity than copper.[8] In the field of polymer composites, graphene has been widely used as a potential nanofiller material to improve the mechanical properties of polymer-based

composites at very low loading conditions.[9,10] However, there are still challenges that need to be addressed while reinforcing graphene in the polymer matrix.[9,10] The problems encountered during the inclusion of graphene are (i) Homogeneous dispersion in the holding matrix and (ii) interfacial adhesion of the fillers with the polymer chain of the holding matrix.[9,10]

As such, nanofiller implementation at different proportions in modifying the mechanical properties of composite has been reported in the literature.[11] Moreover, nanofillers are highly preferred over conventional micro fillers as they can impart improved mechanical properties, such as strength and ductility to polymer matrices in realizing advanced composites.[12] This could be attributed to the particle size effect which aids in rendering strong interfacial bonding between the filler and the holding matrix in terms of physical and chemical properties. However, the combination of the organic and inorganic fillers in tailoring the properties of modified composites is scantily reported and needs to be explored for their efficacy in the reinforcement of the composites. The intention of selecting the nanofillers is their ability to stop the crack formation at the nano level, thus avoiding the development of a micro-level crack in the polymer composites intended for structural applications.[13,14]

In addition, the step towards the development of more versatile polymer-based materials due to rising environmental concerns has led to increasing demand for natural-organic fillers to obtain green composites.[15] However, previous research[15] has reported that green composites limit the reinforcement phase to less than or equal to 40 wt.%, which could potentially hinder the further development of polymer-based composites. The combination of inorganic and organic nanofiller readily solves this problem as the dispersion of inorganic fillers can increase the natural filler dosage beyond the mentioned level. A two-fold advantage of tailored strength and environmentally sound green composite can thus be achieved. The dispersion of these inorganic fillers not only enhances the strength of the natural reinforcements instead renders the developed composite into a multifunctional composite. The increasing viscosity resulting from the infusion of micro-scale fillers can be greatly nullified by using nanoscale fillers. The infusion of Diamond-Like Carbon (DLC) materials such as graphene could replace aviation metal structures in the future. Graphene, as mentioned before, is known for its exceptional thermal, mechanical and electrical properties.[1–7] The high surface area and excellent mechanical/electrical/thermal properties of graphene make it an ideal reinforcement phase for polymer composites.

This work focuses on the usage of graphene in combination with hemp fibres (natural organic fillers) to reinforce the epoxy composites intended for structural applications. Hemp fibres are one of the ideal reinforcement for polymer composites among the class of organic fillers.[16–18] Furthermore, hemp fibres find reinforcement applications because of their flexibility, density (231 kg/m^3) and surface area (321–600 m^2/g).[19] Although research on hybrid reinforcement in enhancing the mechanical properties such as tensile and flexural properties of modified composites has been reported, the combination of hemp and graphene in influencing the mechanical properties of the modified composites is hardly found in the literature. From the flexural test, it was found that modified composite beams showed enhanced performance against conventional specimens. The improvement in properties could be attributed due to the usage of graphene in combination with natural fillers/fibres.

Experimental

The raw materials used in this study include plain epoxy resin, curing agent, hemp fibre and graphene nanofiller. The characteristics of the specimen utilized for the experiment work are mentioned in Table 1. The physical and chemical properties of the epoxy resin are tabulated in Table 2. The physical and chemical properties of the graphene nanofiller are tabulated in Table 3. The physical and chemical properties of the hemp fibres utilized for this work are mentioned in Table 4. The test specimens that have been used for mechanical testing, that is, plain composite, a combination of hemp fibre and/or graphene nanofiller with plain composite have been provided in Table 5. Since the dispersal of nanoparticles posed a challenging issue in the highly viscous epoxy resin, suitable

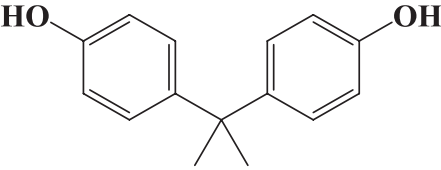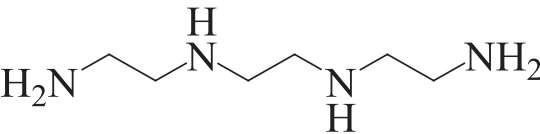Table 1. Specimen characteristic for experimentation.

Specimen Characteristic	Specifics
Dimension	$40 \times 12 \times 6 \times 10^{-9}$ m^3
Pristine Epoxy resin (Bisphenol A chemical structure)	PE

Curing agent (Tri-ethylene-tetramine)	K-6

Hemp fibre (HF) composition	0.1–10 wt.% by epoxy resin
Graphene (GE) composition	0.1–0.4 wt.% by epoxy resin

Table 2. Physical and chemical properties of the epoxy resin.

Characteristic Property	Inferences
SNS Part Number	31200000LP X 0002
Type of Product	Epoxy Resin with Hardener
Brand	Lapox
Model No	L-12/K-6
Temperature (K)	373 K
Minimum Curing Time	900–1800 sec at 373K
Shear Strength	1.4 kg.mm/min
Pot Life	108000–216000 sec at 293 K
Viscosity	9000–12000 mPa.s at 298 K

Table 3. Physical and chemical properties of the graphene nanofiller.

Sourced from	United Nanotech, India
Bulk Density	0.24 g/cc
Diameter average X & Y Dimensions	5–10 micron
Thickness average Z dimension	3–6 nm
Carbon purity	> 99%
Number of layers	Average number of layer 3–6
Surface area	150 m^2/g
Thermal conductivity	400 $Wm^{-1}k^{-1}$

Table 4. Physical and chemical properties of Hemp fibres.

Physical Property	
Diameter	3 μm
Length	0.005 m
Tensile strength	4100 GPa
Tensile modulus	169 GPa
Colour	Brown
Suppliers	Acrotex, India
Chemical constituents	
Pectin	0.89%
Cellulose	75%
Waxes and oils	2.5%
Hemicelluloses	19%
Lignin	3%

solvents like acetone were utilized. The viscosity of the epoxy at 298KmPas as per ASTM D2196 was 9000–12,000 cps. The hydrophilic nature of the natural fibre acted as a resistance for the surface adhesion between the matrix and fillers. Hence, sonication duration of 5400 s was performed hand layup technique, which was employed for casting all the specimens in this study. Several predefined volumes of HFs and GE were infused into the epoxy matrix as shown in Table 5. The graphene used in this work is procured with purity greater than 95 wt.%. The epoxy resin reinforced was in the range of 0.1–0.4 wt.% of the epoxy resin. The associated curing agent was added as 10 % of the weight of epoxy resin. The modified composition was filled in aluminium moulds. The crystallization temperature or the glass transition temperature of the matrix initiates at 301K due to epoxy being a thermosetting resin. Curing was carried for 24 h at room temperature.

A destructive test was employed to determine the flexural strength of the modified composites. The test specimen of dimension 40 × 12 × 6 × 10^{-9} m^3 was subjected to flexure test, that is, a three-point loading as illustrated in Figure 1 according to ASTM D2344M standard.[20] The mould used for casting the specimens and the specimens cast for flexural testing are depicted in Figure 2(a) and (b). To obtain data accuracy, five replicas of each composition were subjected to the flexure test. A closed feedback hydraulic machine was utilized for experimenting. The technical specifications of the closed-loop hydraulic testing machine are mentioned in Table 6. Further analysis of the test specimens included both thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). Thermogravimetric analysis (TGA) of polymers was conducted to measure weight changes as a function of temperature and time. The thermal degradation of the test specimens in this study was recorded by the instrument Universal V4.5A TA SDT Q600 at a temperature of 283 K/60s in a range of temperatures between 298 and 873 K. Differential scanning calorimetry (DSC) was employed to investigate the response of polymers to heating to

Table 5. Test specimen details.

| Sample	Specimen Constituent/Composite	Reinforcement proportion by wt.% of epoxy resin
1	Plain Epoxy- (PE)	0
2	PE + Graphene (G1)	0.1
3	PE + Graphene (G2)	0.2
4	PE + Graphene (G3)	0.3
5	PE + Graphene (G4)	0.4
6	PE + HF (H1)	0.1
7	PE + HF (H2)	0.2
8	PE + HF (H3)	0.3
9	PE + HF (H4)	0.4
10	PE + Graphene+ HF (GH1)	Hemp 2.5% + Graphene 0.1%
11	PE + Graphene+ HF (GH2)	Hemp 2.5% + Graphene 0.2%
12	PE + Graphene+ HF (GH3)	Hemp 5% + Graphene 0.1%
13	PE + Graphene+ HF (GH4)	Hemp 5% + Graphene 0.2%
14	PE + Graphene+ HF (GH5)	Hemp 7.5% + Graphene 0.1%
15	PE + Graphene+ HF (GH6)	Hemp 7.5% + Graphene 0.2%
16	PE + Graphene+ HF (GH7)	Hemp 10% + Graphene 0.1%
17	PE + Graphene+ HF (GH8)	Hemp 10% + Graphene 0.2%
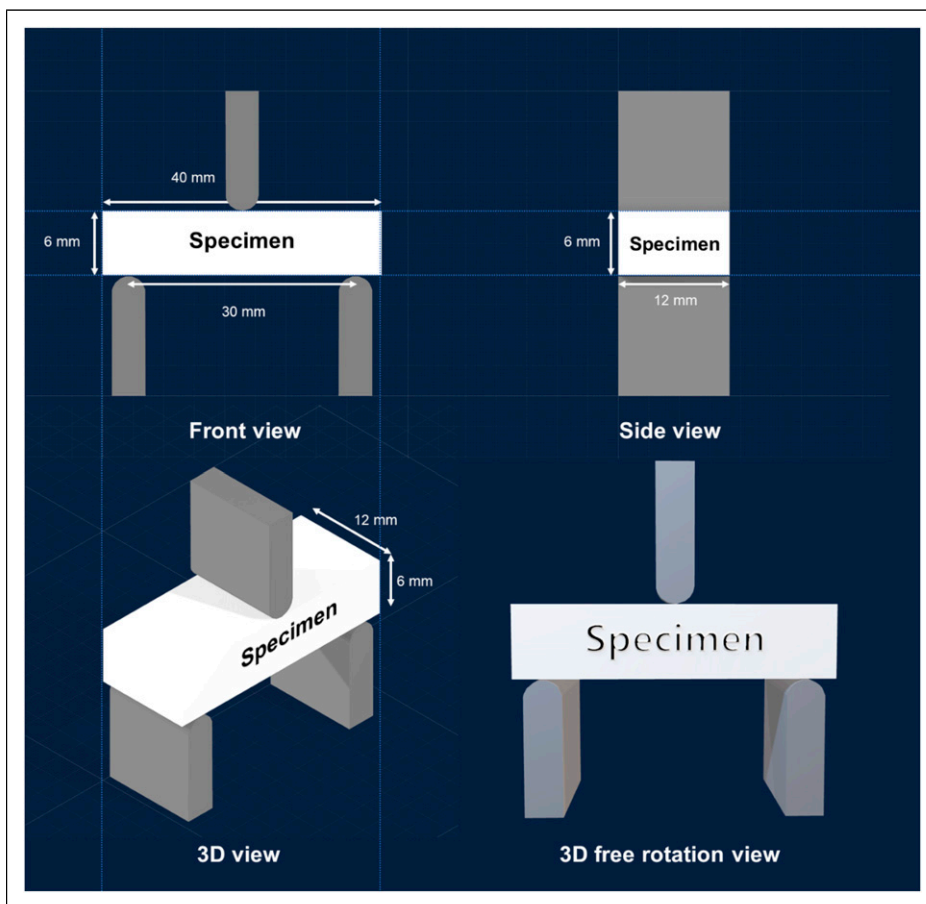
Figure 1. Three-dimensional Graphical representation of three-point load set-up with the specimen.

determine the melting of a crystalline polymer or the glass transition and was conducted by Universal V4.5A TA SDT Q600 Instruments to determine the reaction and glass transition temperature. The exothermic reaction for the composite was noticed at 597.2 K with an energy release rate of 11.65J/g, whereas the endothermic reaction was obtained at 347.3 K.
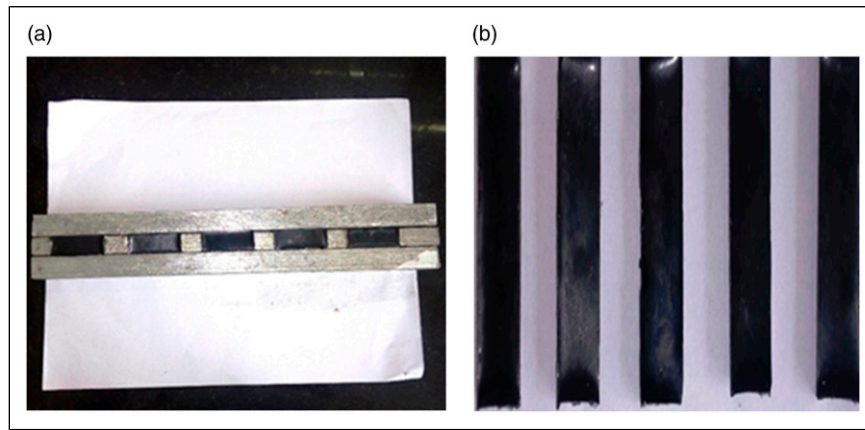
Figure 2. (a) Mould used for casting the specimens. (b) Specimens subjected to flexural testing.

Table 6. Technical specifications of the closed loop hydraulic testing machine.

| Sl. No	Facilities for tests	Specification	Test conducted
1.	Load frame testing machine	Capacity-10 kN, Least count = 0.01 kN, Displacement = 0.00001 m FSR = ±1 count, Accuracy = ±0.1% Operating condition = 10 to 45 °C, Power supply = 230 V, 50 Hz, AC mains Strain rate = 0.00005 m/60 s Make: Fine testing machines Model: TKG 1000	Three-point test on beams
2.	LVDTs and Displacement indicating units	Least count = 0.00001 m, Range = ±0.025 m Linearity and accuracy = ±1 digit Operating temp = 283.15 to 323.15	
3.	Strain measuring system	K Range-0 to 1999 micro-strains Nominal strain = 50 μm/m Input resistance >1000 V Output resistance = 1000 V Gauge factor = 1.5 to 5 Zero-point deviation = 5%	
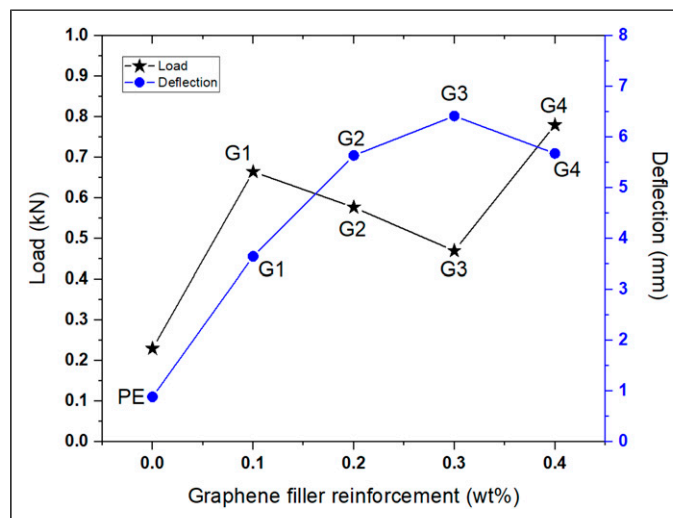
Figure 3. Load-Deflection curves of graphene filler modified composites (wt. % w.r.t. to epoxy).

Results and discussions

Flexure behaviour

The three-point bend test/flexure test was carried out to determine the optimum reinforcement composition of the GE/HFs/ GE+HFs-modified composites for obtaining improved load-sustaining ability. A flexure bend test was also carried out for pristine epoxy (PE) resin to compare the control specimen with the reinforced specimens. The load-deflection curves for graphene-modified composites are depicted in Figure 3, which reveals that the load-sustaining ability of specimen G4 is highest when compared to any other composition and ~70 % higher than G3. Moreover, the load-sustaining ability of graphene-reinforced composites is almost three times higher than the pristine epoxy sample (PE). The maximum deflection is observed for specimen G3. Therefore, in terms of graphene addition to the epoxy matrix for enhancing flexural strength, 0.4 wt.% reinforced composite can be said to possess improved properties. A similar trend indicating improved mechanical properties with increasing graphene filler reinforcement was also noticed by Prasob et al.[21] It was reported that using graphene and its derivatives, significantly increased the mechanical properties of polymer matrix composites reinforced with natural lignocellulosic fibres. However, Prasob et al.[21] also reported a reduction in mechanical properties of the natural lignocellulosic fibre reinforced polymer matrix composites when graphene filler reinforcement exceeded 11 wt.% owing to agglomeration of the fillers.

The load-deflection curves of hemp fibre (HF) modified composites are illustrated in Figure 4, which revealed that the load-sustaining ability of specimen H2 is highest when compared to any other composition and better than plain epoxy sample by ~3.5 times. This increase in the load-sustaining ability can be attributed to the polar groups responsible for enhancing molecular bonding between the plain epoxy (PE) + HF modified composites as filler was increased from 0.1
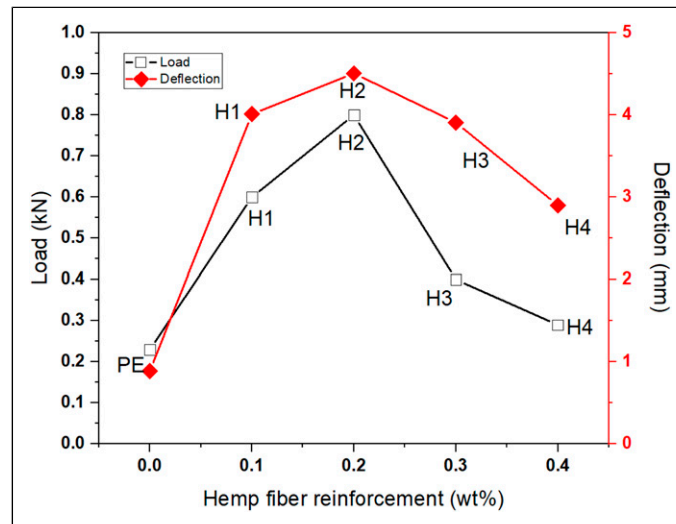

Figure 4. Load-Deflection curves of hemp fiber (HF) modified composites (wt. % w.r.t. to epoxy).
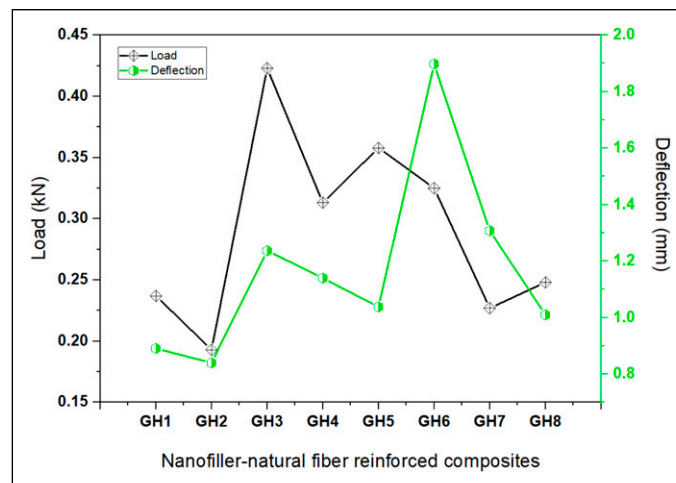

Figure 5. Load-Deflection curves of GE + HFs altered composites subjected to a three-point loading (wt. % w.r.t. to epoxy).

wt.% to 0.2 wt.%, resulting in strengthened interfacial adhesion. However, a decrease in load-sustaining ability was noticed as hemp fibre reinforcements exceeded 0.2 wt.%. A similar decrease in flexure strength with increasing hemp fibre reinforcement was reported by Singh et al.[22] It was reported that the addition of hemp fibres, that is, 30 wt.% in the polymer matrix (equal proportions of virgin and recycled HDPE), reduced the flexure strength by ~5 MPa. Moreover, a

reduction of tensile strength was also observed when hemp fibre reinforcement increased from 10 wt.% to 30 wt.%. Singh et al.[22] attributed the reduction in strength to fibre delamination and poor interfacial adhesion between hemp fibres and the polymer matrix.

Load-deflection curves of natural fibre-nano filler modified composites subjected to the flexural test are depicted in Figure 5. From the graph (Figure 5), it can be observed that for a reinforcement combination of plain epoxy (PE) + 5 wt.%hemp + 0.1 wt.% graphene nanofillers, that is, sample GH3 (Table 1), the modified composites have shown improved load-sustaining ability compared to the other GH-samples. When reinforcement exceeded 0.2 wt.% of graphene filler and 5 wt.% hemp fibre, the modified composites exhibited diminishing drifts in the strength facets (refer to load curve in Figure 5) as a result of filler agglomeration. The agglomeration at specific spots initiates critical crack growth leading to failure of the modified composites. Hence, two peaks can be observed in the load-deflection curve in Figure 5, that is, the peak (0.42 kN or 420 MPa) in the load curve exhibited by specimen GH3 (Hemp 5% + Graphene 0.1%) and the peak (1.9 mm) in the deflection curve exhibited by sample GH6 (Hemp 7.5% + Graphene 0.2%). It can be deduced that the load-sustaining ability of sample

GH3 is better than neat epoxy showing an increase of ~83 %. While the deflection of sample GH6 is two times better than neat epoxy. This could be attributed to the length of the hemp fibre, that is, increased fibre length helps in tolerating more stress than shorter fibres resulting in high strength.

In the case of filler reinforcement greater or less than 0.2 wt.%, no change in mechanical properties was observed. This could be due to the break in the linkage of polymer chains. The proportion of 5 wt.% hemps and 0.1 wt.% of graphene nanofillers (GH3) showed maximum ultimate load (Figure 5). This can be attributed to random reinforcement obtained by natural fibre and nanoparticles which provide additional toughness for modified composites.[23] However, the deflection of composite GH3 was low. From Figure 5, the optimal combination of properties is observed for composite GH6 in terms of load-sustaining ability and deflection. Table 7 enlists the properties of the best-modified composites from each group and compared them with neat epoxy/pristine epoxy.

The initial increase in the load-sustaining ability of the modified composites can be attributed to the load transferability of the fillers to the holding matrix by providing more surface area.[24] In general, it can be overviewed that among

Table 7. Critical load and deflection values of specific composites.

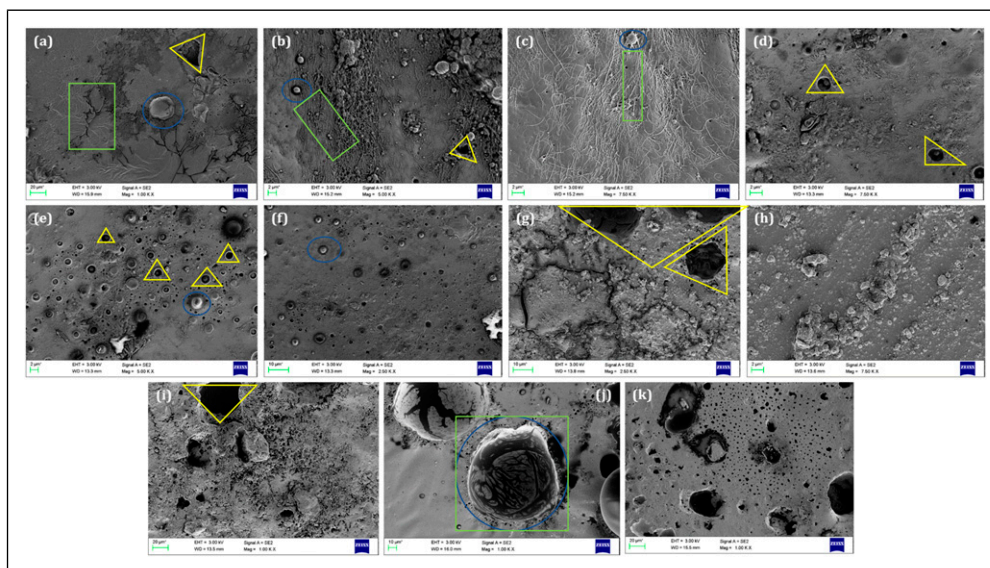| Serial no.	Case Composition	Critical load in (kN)	Maximum Deflection in (m)
I	Pristine epoxy (PE)	0.23	0.00089
2	PE + GE (G3)	0.78	0.0064
3	PE + HFs (H2)	0.80	0.0045
4	PE + GE + HFs (GH6)	0.33	0.0019

Figure 6. SEM images of composite (a) GH1, (b) GH2, (c) GH3, (d) GH4, (e) GH5, (f) GH6, (g) GH7, (h) GH8, (i) H2, (j) G3, (k) G4 [graphene filler-marked by a blue circle, hemp fibres-marked by a green rectangle, voids-marked by yellow triangles].

the race between organic and inorganic reinforcement, graphene leads the reinforcement effect in enhancing the mechanical aspects. This could be attributed due to the presence of graphene that relieves stress deposition by providing more surface area. However, it can also be noted from Figure 3 that reinforcement of fillers increased the mechanical properties only to a certain extent. Further reinforcement resulted in a decrease in properties, which was also reported by Prasob et al.[21] and Singleton et al.[25] For loading beyond 0.2 wt. % of GE and 7.5 wt. % of HF, the decreasing drifts of the strength characteristics were observed as the reason for this could be the reduction in polymer linkage connectivity because of increased wetting of the nanofiller and natural fibre surface to the polymer chains thereby enhancing the viscosity and reducing the polymer chain mobility.[26]

SEM characterization

The bonding between the fillers and polymer matrix was investigated by using SEM. The shining particles indicate the presence of graphene as graphene reflects light during SEM characterization, whereas the thread-like entities confirm the presence of hemp fibres. The appearance of graphene filler in SEM micrographs is marked by a blue circle, the presence of hemp fibres is marked by a green rectangle and voids are marked by yellow triangles.

Figure 6(a) represents specimen GH1, whereby a continuous linkage of the polymer chain can be observed indicated by a smooth polymer layer. In composite GH2 (Figure 6(b)), a clear distinction between the hemp fibres and the graphene fillers can be observed that led to the formation of the voids marked by the yellow triangle. Composite GH3 (Figure 6(c)) shows both graphene filler and hemp fibre reinforcement as a combined constituent in the shape of letter 'i', indicating the smooth layer of polymer chain mobility leading to the enhancement in the mechanical properties for this composite. Figure 6(d) representing composite GH4 shows agglomeration of the fillers. Figure 6(e) depicts the formation of voids for composite GH5, which has led to the formation of pits and crevices. Figure 6(f) for composite GH6 shows no linkage/bond between graphene fillers and hemp fibres leading to the decline in observed properties. In composite GH7 (Figure 6(g)), excess reinforcement of fillers has led to the decline in the polymer chain mobility depicted by the large holes formed marked by the yellow triangles. Composite GH8 (Figure 6(h)) shows no traces of linkage between fibres and graphene fillers leading to the decline in the observed mechanical properties. Figure 6(i) representing composite H2 shows a good intervention of the natural fibres with the holding matrix leading to the enhancement in mechanical properties. Figure 6(j) representing composite G3 shows shining bubbles of graphene that has led to the increase in deflection of the specimen. Figure 6(k) of representing composite G4 shows polymer linkage with graphene leading to increase in load-bearing ability.

SEM-EDS analyses were performed to verify the presence of metallic species within the constitution of the graphene since it was reported that metallic species influence the mechanical properties of the composites.[27–29] For brevity only, EDS spectrum of graphene filler in composite G3 has been provided in Figure 7. It was found that metallic species were present in the filler in negligible (< 1%) amounts and the main constituents were carbon and oxygen as shown in Figure 7. Hence, these metallic species had no effect on the mechanical properties of composites investigated in this study.

Thermal analysis

Figure 8 shows the thermal degradation of the GH6 composite. The thermal degradation was recorded at a temperature of 283 K/60 s in a range of temperatures between 298 and 873 K. A sample weighing 5.70×10^{-6} kg was used for measurement. The experiments were conducted in a nitrogen atmosphere. The deterioration of the holding matrix was noted at a temperature range of 625.12 K–723 K. The maximum structural decomposition of the epoxy resin was obtained at 673 K. Increased residual patterns were observed for epoxy/graphene+hemp with increasing graphene content when subjected to a temperature of 873 K. Graphene was found to possess more thermal resistance than natural hemp fibre. A weight loss of 93.21 % was recorded due to the destruction of the composite structures. The increase in thermal steadiness can be accredited to the presence of graphene in epoxy resin. The thermal degradation of the natural hemp fibre can be protected by the synthetic
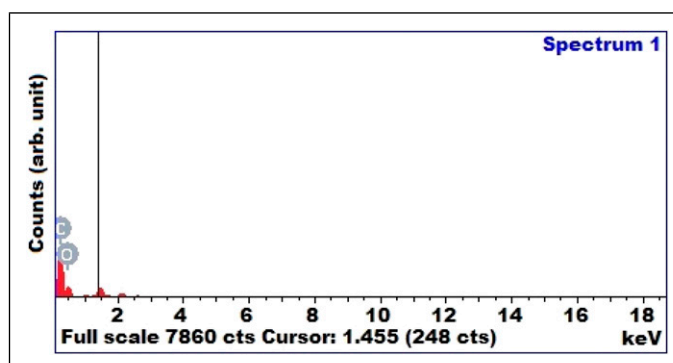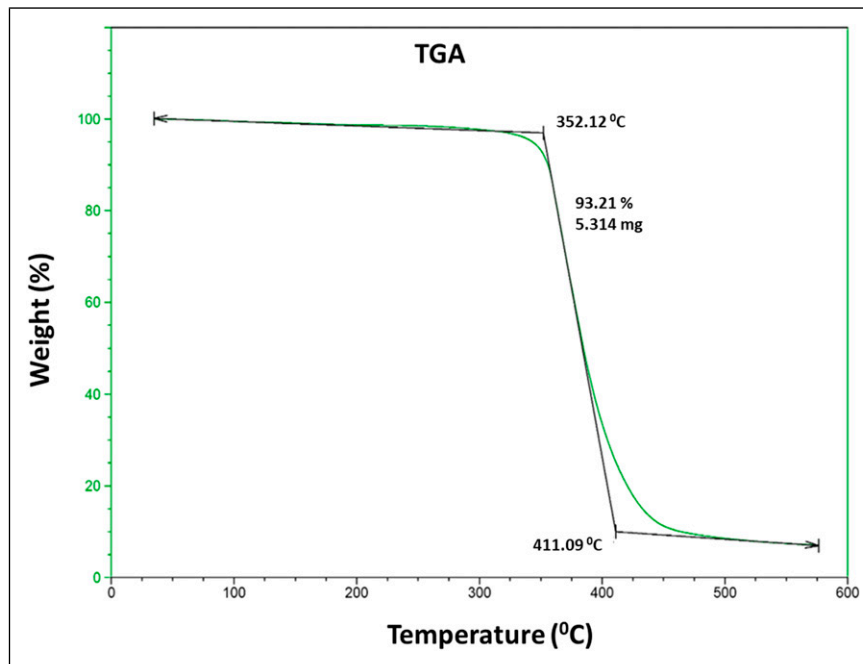

Figure 7. SEM-EDS of specimen G3.

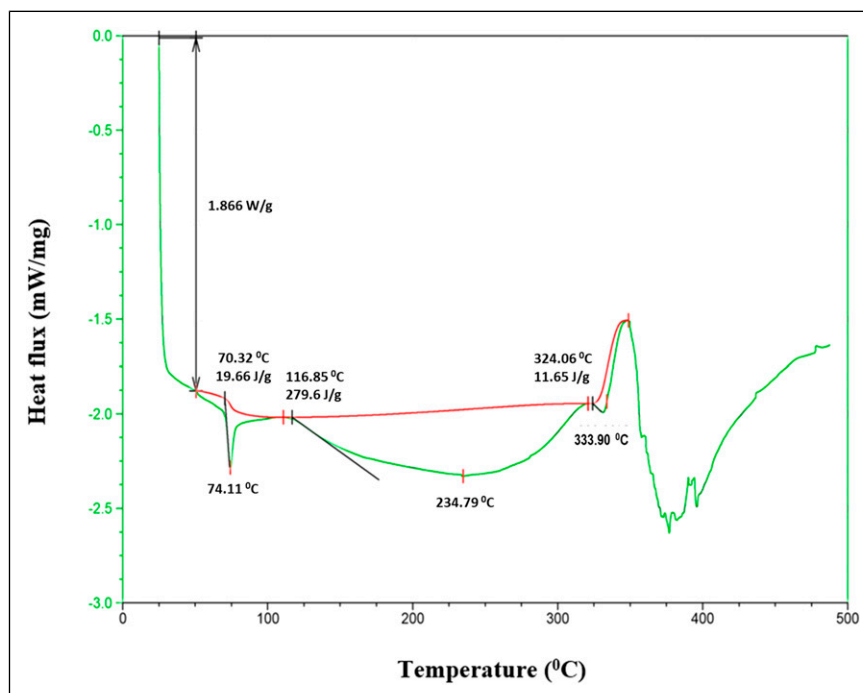**Figure 8.** TGA of epoxy/graphene+Hemp [GH6 composite].

Figure 9. DSC of GH6 composite.

graphene filler. It can be noted from Figure 8 that more energy in terms of heat is required to collapse the interfacial bond between the filler and the medium.

A Universal V4.5A TA SDT Q600 V20.9 Build 20 Instruments differential scanning calorimeter was employed to determine the reaction and glass transition temperature as shown in Figure 9. Readings were taken in an inert nitrogen atmosphere at a heating rate of 283 K/60s. The instrument was calibrated with zinc. It can be observed from the graph that there is an increase in both the initial reaction temperature and an exothermal reaction with the presence of graphene content. It can also be observed that the existence of graphene has enhanced the enthalpy of epoxy cure reaction. The energy absorbed in the total degradation of the epoxy composites structures is from 1.86–11.65 J/0.001 kg. The initial endothermic transition was

observed from 389.85 K–597.06 K due to its melting, followed by an exothermic reaction between 597.06 K and 623 K. The presence of nanofiller has boosted the thermal degradation capacity of modified composites as linked to natural fillers.

Conclusions

The influence of reinforcing pristine epoxy with graphene nanofillers, natural hemp fibres and the combination of nanofiller-natural fibre has been investigated in this study. The desire to curb the hydrophilic nature of natural fibres, which is an undesired property in polymer composite fabrication, led to the combined addition of graphene nanofillers during fabrication. Another important aspect of graphene nanofiller addition to hemp fibre reinforced composites was to increase the thermal resistance of the composites. The graphene/hemp fibre composites were then compared with individual graphene nanofiller modified composites and natural fibre modified composites. It was found that increasing graphene nanofiller reinforcement had a beneficial effect on the load-sustaining ability of the composites. On the contrary, hemp fibre modified composites did not fare well in terms of load-bearing ability when reinforcements exceeded 0.2 wt%. The combined reinforced epoxy composites exhibited lower load-bearing ability compared to their counterparts, which could be mainly attributed to the agglomeration of nanofillers during the fabrication process. Since the combination of the organic (natural fibre) and inorganic (graphene) fillers is an active research area in the field of composites, further investigations, such as homogeneous nanofiller size distribution and or micro filler addition, are required to tailor the reinforcements to attain improved properties in green composites. In addition, investigations in this study will pave the way for future studies to tailor combined reinforcement to produce green composites with improved flexural strength. This material can be utilized for the application, such as aerospace, automotive, construction, marine and corrosion-resistant materials.

References

1. Fan YH, Yu SW, Wang HM, et al. Study on preparation and properties of graphene reinforced epoxy resin composites. *IOP Conf Ser Mater Sci Eng* 2019; 634: 012044.
2. Güler Ö and Bağcı N. A short review on mechanical properties of graphene reinforced metal matrix composites. *J Mat Res Technol* 2020; 9: 6808–6833.
3. Ji X, Qi S, Ahmed R, et al. Graphene-reinforced advanced composite materials. In: *Handbook of Graphene Set*. Hoboken, USA: John Wiley & Sons, Ltd, 2016, pp. 27–89.
4. Liu, Phang IY, Shen L, et al. Morphology and mechanical properties of multiwalled carbon nanotubes reinforced nylon-6 composites. *Macromolecules* 2004; 37: 7214–7222.
5. Mostovoy AS and Yakovlev AV. Reinforcement of epoxy composites with graphite-graphene structures. *Sci Rep* 2019; 9: 16246.
6. Papageorgiou DG, Li Z, Liu M, et al. Mechanisms of mechanical reinforcement by graphene and carbon nanotubes in polymer nanocomposites. *Nanoscale* 2020; 12: 2228–2267.
7. Wong EW, Sheehan PE, and Lieber CM. Nanobeam mechanics: elasticity, strength, and toughness of nanorods and nanotubes. *Science* 1997; 277: 1971–1975.
8. Moniruzzaman M and Winey KI. Polymer nanocomposites containing carbon nanotubes. *Macromolecules* 2006; 39: 5194–5205.
9. Du J and Cheng H-M. The fabrication, properties, and uses of graphene/polymer composites. *Macromol Chem Phys* 2012; 213: 1060–1077.
10. Verma D, Gope PC, Shandilya A, et al. Mechanical-thermal-electrical and morphological properties of graphene reinforced polymer composites: a review. *Trans Ind Inst Met* 2014; 6: 803–816.
11. Šupová M, Martynková GS and Barabaszová K. Effect of nanofillers dispersion in polymer matrices: a review. *Sci Adv Mat* 2011; 3: 1–25.
12. Fu S, Sun Z, Huang P, et al. Some basic aspects of polymer nanocomposites: a critical review. *Nano Mat Sci* 2019; 1: 2–30.
13. Sheheri SZA, Al-Amshany ZM, Sulami QAA, et al. The preparation of carbon nanofillers and their role on the performance of variable polymer nanocomposites. *Des Monomers Polym* 2019; 22: 8–53.
14. Üstün T, Ulus H, Karabulut SE, et al. Evaluating the effectiveness of nanofillers in filament wound carbon/epoxy multiscale composite pipes. *Compos Eng* 2016; 96: 1–6.
15. La Mantia FP and Morreale M. Green composites: a brief review. *Compos Appl Sci Manufact* 2011; 42: 579–588.

16. Dhakal HN and Zhang Z. 3 - The use of hemp fibres as reinforcements in composites. In: Faruk O, and Sain M (eds) *Biofibre Reinforcements in Composite Materials*. Sawston, Cambridge: Woodhead Publishing, 2014, pp. 86–103.

17. Tanasă F, Zănoagă M, Teacă C-A, et al. Modified hemp fibres intended for fibre-reinforced polymer composites used in structural applications—A review. I. Methods of modification. *Polym Compos* 2020; 41: 5–31.

18. Terrones M. Science and technology of the twenty-first century: synthesis, properties, and applications of carbon nanotubes. *Annu Rev Mater Res* 2003; 33: 419–501.

19. Shahzad A. Hemp fibre and its composites – a review. *JCompos Mat* 2012; 46: 973–986.

20. Kotik, Hector G., and Juan E. Perez Ipińa. "Suggested modifications of the ASTM D2344–16 short-beam shear test method to be applied to fiber metal laminates." Journal of Testing and Evaluation 49.2 (2019): 1213–1221.

21. Asoka PP and Mohan S. Viscoelastic and mechanical behaviour of reduced graphene oxide and zirconium dioxide filled jute/epoxy composites at different temperature conditions. *Mat Today Commun* 2019; 19: 252–261.

22. Singh S, Deepak D, Aggarwal L, et al. TENSile and flexural behavior of hemp fibre reinforced virgin-recycled HDPE matrix composites. *Proced Mat Sci* 2014; 6: 1696–1702.

23. Kim Y-P, Kim H-B, Jang M-H, et al. Magnitude- and time-dependence of the effect of treadmill exercise on cell proliferation in the dentate gyrus of rats. *Int J Sports Med* 2003; 24: 114–117.

24. Valadez-Gonzalez A, Cervantes-Uc JM, Olayo R, et al. Effect of fibre surface treatment on the fibre–matrix bond strength of natural fibre reinforced composites. *Compos Eng* 1999; 30: 309–320.

25. Singleton ACN, Baillie CA, Beaumont PWR, et al. On the mechanical properties, deformation and fracture of a natural fibre/recycled polymer composite. *Compos Eng* 2003; 34: 519–526.

26. Espert A, Vilaplana F and Karlsson S. Comparison of water absorption in natural cellulosic fibres from wood and one-year crops in polypropylene composites and its influence on their mechanical properties. *Compos Appl Sci Manuf* 2004; 35: 1267–1276.

27. Li Y, Mai Y-W and Ye L. Sisal fibre and its composites: a review of recent developments. *Compos Sci Technol* 2000; 60: 2037–2055.

28. Ray D, Sarkar BK, Das S, et al. Dynamic mechanical and thermal analysis of vinylester-resin-matrix composites reinforced with untreated and alkali-treated jute fibres. *Compos Sci Technol* 2002; 62: 911–917.

29. Xie Y, Hill CAS, Xiao Z, et al. Silane coupling agents used for natural fibre/polymer composites: a review. *Compos Appl Sci Manuf* 2010; 41: 806–819.