The Effects of Multi-walled CNT in Bamboo/Glass Fibre Hybrid Composites: Tensile and Flexural Properties

Ariff F. M. Nor,[a,c] Mohamed T. H. Sultan,[a,b,c,]* Mohammad Jawaid,[b] Abd R. Abu Talib,[c] Ahmad M. R. Azmi,[a,c] Ahmad S. Harmaen,[d] and Ainun Z. M. Asa'ari [d]

Recently, polymer nanocomposites have been fabricated using carbon nanotubes (CNTs) as reinforcement nanofillers. However, the effect of incorporating CNT/polymer into hybrid composites with natural fibre is not clear. This study investigated the effect of using multi-walled carbon nanotube material (MWCNT) as the nanofiller on the tensile and flexural properties of bamboo/glass fibre hybrid composites. Composites containing various weight fractions of CNTs (0.1 wt.%, 0.3 wt.%, 0.5 wt.%, and 1.0 wt.%) were compared with the control hybrid composites. The hybrid composites were prepared with epoxy resin. The experimental results revealed an increase in the tensile strength of the composites with the addition of up to 0.5 wt.% CNTs (+7.7% over the control hybrid). However, beyond this value, *i.e.*, with 1.0 wt.% CNT additives, the composite strength showed a remarkable decrease (-36.8% compared with the control hybrid). Moreover, introducing CNTs into hybrid composites resulted reduced the flexural properties with increasing weight fractions as low as 8.45% compared with the controls. In sum, the tensile properties increased with the addition of up to 0.5 wt.% CNTs, but there was a decrease in the flexural properties.

Keywords: Bamboo; Tensile; Flexural; Hybrid; CNT; Nanofillers

Contact information: a: Aerospace Manufacturing Research Centre (AMRC), Faculty of Engineering, Universiti Putra Malaysia (UPM), 43400 UPM Serdang, Selangor Darul Ehsan, Malaysia; b: Laboratory of Biocomposite Technology, Institute of Tropical Forestry and Forest Products (INTROP), UPM, 43400 UPM Serdang, Selangor Darul Ehsan, Malaysia; c: Department of Aerospace Engineering, Faculty of Engineering, UPM, 43400 UPM Serdang, Selangor Darul Ehsan, Malaysia; d: Laboratory of Biopolymer and Derivatives, INTROP, UPM, 43400 UPM Serdang, Selangor Darul Ehsan, Malaysia;
** Corresponding author: thariq@upm.edu.my*

INTRODUCTION

Composite materials are broadly used as part of the structural industry and allow engineers to overcome traditional disadvantages when using the materials individually. Enhanced properties are acquired when the materials are used as composites, and they do not disintegrate or merge completely when used together. New advanced materials are applied broadly, especially in advanced structures (Carlsson *et al.* 2014). The structural properties of hybrid composites have also improved.

Fibres are divided into three types—natural, synthetic, or a mix of both—and they can be defined in a single matrix made up of two or more fibres (Nunna *et al.* 2012). Researchers are actively exploring the benefits of hybrid composites to increase their usage, especially those involving natural fibre. The mechanical properties of hybrid composites are better than those containing only natural fibres (Thwe and Liao 2003). The additional reinforcement of synthetic fibres in natural fibre composites strengthens the mechanical properties of the hybrid (Sanjay *et al.* 2015).

Bamboo fibre (BF) is a natural fibre that is widely available around the world. The bamboo tree, one of the fastest growing plants on earth, is found in Asia, America, Africa, and several European countries. Bamboo is traditionally used by rural residents in household and structural applications, such as long houses, bridges, and handicraft items.

There is abundant bamboo in Malaysia, which gives many opportunities for its application in daily life. However, the lack of technology available for processing raw, fresh bamboo into ready-to-use fibres slows down its development and application, especially during composite fabrication (Suhaily *et al.* 2013). Bamboo can be processed similarly to wood material; bamboo powder can be used as a filler in polymer composites. Like other organic fillers made from agricultural industrial residue, bamboo powder filler has been studied for its application in wider fields. In natural fibre selection, bamboo fibre is selected due to its high strength-to-weight ratio and has attracted research attention for use in composites (Khalil *et al.* 2012). Moreover, bamboo species are easily obtained throughout tropical climate countries, including Malaysia (Shah *et al.* 2016).

The tensile and flexural properties of composites made by reinforcing natural fibre into a polyester resin matrix have been investigated. The tensile strength, tensile modulus, flexural strength, and flexural modulus are better than when vakka, sisal, jute, coir, and banana fibre are used as reinforcement (Rao *et al.* 2010; Prasad and Rao 2011; Biswas *et al.* 2013). Different ratios of bamboo powder have been compounded with polyester and fabricated using a compression moulding technique. Composites with 25 wt.% bamboo powder possess the highest strength, while the impact test shows the highest value at 20 wt.% bamboo powder (Leha *et al.* 2014). Compared with neat epoxy, the inclusion of bamboo fibres increases the tensile and flexural strength of composites (Wong *et al.* 2010). Different bamboo species including *Gigantochloa scortechinii* (GS), *Gigantochloa levis* (GL), *Dendrocalamus asper* (DA), and *Dendrocalamus pendulus* (DP) have been investigated, and the DA species exhibits the best mechanical and physical properties (Zakikhani *et al.* 2017).

For the past 10 years, a great expansion in the research of polymer nanocomposites has been made due to the high potential of using advanced materials in technology applications (Kurahatti *et al.* 2010; De Volder *et al.* 2013). Polymer nanocomposites are made up of a polymer matrix combined with a range of filler materials and have at least one in the nanometer range (for example, one, two, or three-dimensional). On the other hand, carbonaceous nanofillers bring better structural and functional properties because of their high aspect ratio, electrical properties, and mechanical strength (Lee *et al.* 2008; Behabtu *et al.* 2013; Guo *et al.* 2014).

The main problem with nanocomposites is the dispersion of the nanofillers. A better dispersion of the nanofillers contributes to transferring the stress, hence increasing the overall performance (Mei *et al.* 2016).

An investigation that involved only 0.1 to 0.2 wt.% CNTs in the epoxy matrix revealed an improvement in the tensile strength compared with neat epoxy (Gojny *et al.* 2004; Rahmanian *et al.* 2014). The Young's modulus values are doubled and quadrupled for nanocomposites with 1 wt.% and 4 wt.% CNT, respectively, compared with the neat over-aged epoxy (Allaoui *et al.* 2002). Nanofillers also strengthen synthetic fibre hybrid composites. The hybridisation of glass/epoxy laminates with MWCNTs leads to an increase of up to 30% for its natural frequencies, toughness, and flexural strength (Khashaba 2015). Moreover, improvements of as much as 12% have been reported in the strength and stiffness of the epoxy by introducing MWCNTs into carbon fibre reinforced composites, hence increasing the overall tensile properties (Tehrani *et al.* 2013).

In addition to nanofillers, there are different types of conventional filler, *e.g.*, aluminium oxide, silicon carbide, red mud, and copper slag, which have been tested for their effects on the mechanical properties of natural fibre reinforced epoxy composites. These composites typically exhibit lower tensile strengths than those with pure natural fibre epoxy composites (Yu *et al.* 2012).

There is limited research on this type of material. Thus, this study investigated the effect of carbon nanotube viscosity on bamboo/glass fibre hybrid composites and determined the mechanical properties of the hybridization.

EXPERIMENTAL

Bamboo Fibre

Bambusa vulgaris was harvested from Raub, Pahang, in Peninsular Malaysia. A pruning sealer called Tree Wound liquid was applied to the cross-section of each bamboo culm to prevent the loss of moisture during delivery. In the laboratory, the bamboo culms were split into smaller strips, approximately 1 m in length and 50 mm in width, before passing through a chipper machine. Small bamboo chips from the chipper machine were oven-dried for 24 h at 80 °C. To obtain bamboo powder, the dried bamboo chips were crushed in a flaker machine and sieved according to size. The bamboo powder ranged from 250 µm to 500 µm in size.

Carbon Nanotubes

CNTs are used as nanofillers for reinforcement. There are three types of CNTs available in the market nowadays: single-walled carbon nanotubes (SWCNTs), double-walled carbon nanotubes (DWCNTs), and multi-walled carbon nanotubes (MWCNTs). In this study, MWCNTs were used to modify the epoxy resin. The dimensions were 6 to 9 nm outer diameter and 5 µm length, with 96% purity (ZKK Sdn. Bhd, Selangor, Malaysia). The material was stored in an airtight container with an oxygen absorber sheet to prevent its oxidation.

Epoxy

Smooth-On EpoxAmite 100 epoxy resin and Smooth-On EpoxAmite 103 slow hardener were purchased from Mecha Solve Engineering (Selangor, Malaysia). The density of the epoxy is 1.1 g/cm^3, and its tensile strength is 35 to 100 MPa (Holbery and Houston 2006). The epoxy and hardener were mixed in a ratio by weight of 100:28.4, according to the datasheet provided by the supplier. It had a cure time ranging from 20 to 24 h. To conserve the shelf life of the epoxy resin, the materials were maintained in a controlled-temperature environment of 23 °C. To achieve the high strength and solid cure properties, both the hardener and resin were measured and properly mixed.

Methodology

The MWCNTs were supplied in agglomerated form to reduce the net surface energy. Therefore, both mechanical and chemical approaches had been made to untangle the agglomeration. Prior to experiments, the MWCNTs were immersed in 150 mL of acetone (dimethyl ketone, 2-propanone; Friendemann Schmidt Chemical, Parkwood, Australia) and stirred at 5000 rpm for 1 h by a shear mixer to disperse the MWCNTs (Rathore *et al.* 2016). A high shear mixer was used as the mechanical approach to separate nanotubes from each other; meanwhile acetone is an organic solvent as the chemical approach that wetted the hydrophobic carbon nanotubes and resulting less self-attraction in bundles and ropes for CNTs (Vaisman *et al.* 2006). Acetone has demonstrated positive improvement on Vicker's hardness and flexural strength properties on CNT/epoxy as the solvent for dispersing CNTs compared to other organic solvents (Lau *et al.* 2005). Next, the epoxy/MWCNTs/acetone suspension was stirred for another hour at 10000 rpm and placed a vacuum oven for 2 h until the acetone had completely evaporated. The leftover suspension was mixed with the bamboo fibre, followed by the hand lay-up technique with 2 layers of glass fibre (one layer for each top and bottom) and compression moulding, with 0%, 0.1%, 0.3%, 0.5%, and 1.0% MWCNTs weight fractions as parameters. The glass fibre used was E-type and obtained in roving form (ZKK Sdn. Bhd, Selangor, Malaysia). The density of the glass fibres was 2.58 g/cm^3. One layer of woven glass fibre was embedded each at the top and bottom part to produce a "sandwich" composite structure. Glass fibres were utilised for these hybridisations to enhance strength and water resistance of well-known low strength and poor water resistance natural fibre in composites (Jarukumjorn and Suppakarn 2009). Therefore, the influences of glass fibre in these hybridisations were not considered. The procedure is shown in Fig. 1, and the composition of each specimen is shown in Table 1. The laminated specimens were kept at room temperature for 24 h. The samples were then post-cured at 80 °C for 2 h (Kumar *et al.* 2015). Samples for the tensile and flexural tests were cut from the composites using a CNC cutter.

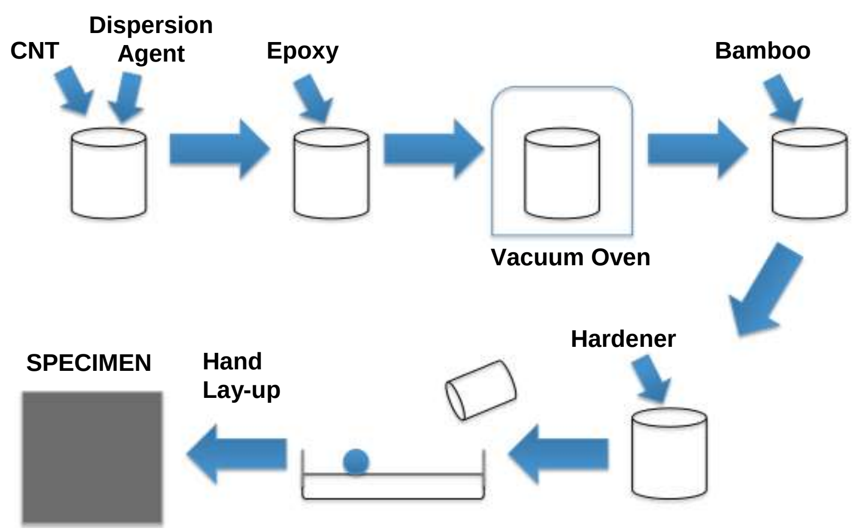

Fig. 1. Schematic of composite fabrication

Table 1. Specimen Composition

Sample	Bamboo Fibre (% w/w)	CNTs (% w/w)	Notes
A	0	0.0	Control (Overall)
B	0	0.1	Control (w/o BF)
C	10	0.0	Control (Hybrid)
D	30	0.0	-
E	10	0.1	-
F	10	0.3	-
G	10	0.5	-
H	10	1.0	-

Tensile and Flexural Characterisation

Each sample was fabricated into five specimens for repeatability and accurate results. The specimens underwent tensile tests using a Shimadzu AG-IS ultimate testing machine (Selangor, Malaysia), in accordance with ASTM D3039-14 (2014). The machine has a capacity of 50 kN, a maximum speed of 1000 mm/min, and a vertical test space of 1150 mm. The speed used for all configurations was 2 mm/min. The specimens were marked and clamped at 50 mm from the top and bottom. The flexural tests were performed on an Instron 4204 flexural testing machine (Selangor, Malaysia) using the 3-point bending method (ASTM D790-17 2017). The span-to-depth ratio of the samples was 16:1. The testing machine has a capacity of 50 kN, a maximum speed of 500 mm/min, and a vertical test space of 1170 mm. The speed used for all the configurations was 5 mm/min.

RESULTS AND DISCUSSION

Tensile Properties

The representative tensile stress-strain curves for each specimen are shown in Fig. 2. While all hybrid composites displayed an elastic tensile behaviour, there were linear deformations at first, and non-linear behaviour occurred thereafter. Pure epoxy showed the most brittle nature followed by 1.0 wt.% CNTs as caused by the poor dispersion described below.

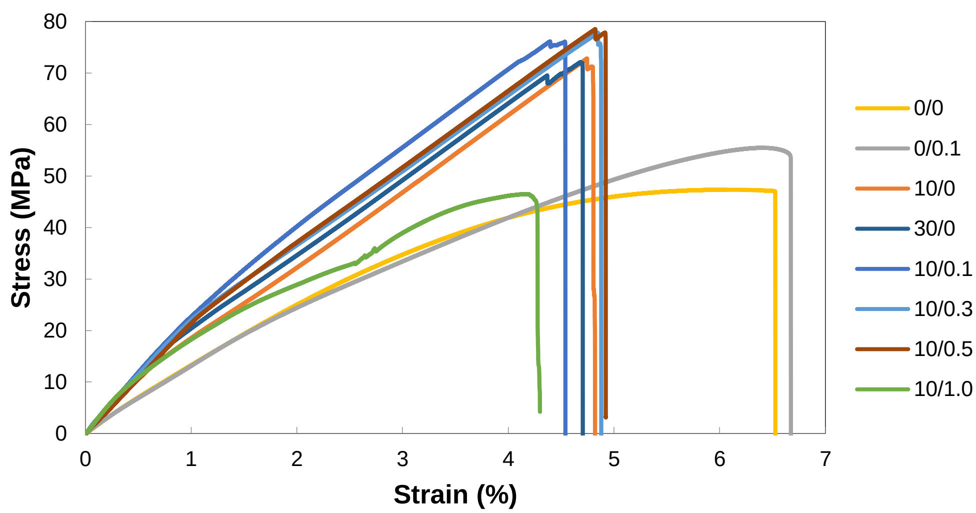

Fig. 2. Tensile stress-strain curves of bamboo/glass fibre hybrid composites enhanced with CNTs

The tensile strength and tensile modulus values of the bamboo/glass fibre hybrid composites enhanced with difference percentages of CNT modified epoxy are shown in Table 2 and Fig. 3. The tensile strength and modulus for the hybrid composites were affected by the addition and the amount of the CNTs. Figure 3 shows that the tensile strength of added CNTs (0.1 wt.%) on pure epoxy and 10% BF noticeably increased by 16.4% and 1.7%, respectively. The tensile strength also showed an incremental increase as the CNT content increased, up to 0.5 wt.% CNTs. Increments in the CNT content of 0.1, 0.3, and 0.5 wt.% presented better tensile strength (by 1.7%, 4.7%, and 7.7%

respectively) compared with the hybrid composites without CNTs (10% BF / 0% CNTs). This result suggested that the high strength property of the CNTs, such as the large interfacial area caused by its nanosize dimension, reduced the interfacial stress concentration. Hence, the stress transfer from the epoxy to the MWCNT resulted in better mechanical properties of the hybrid composites.

Table 2. Tensile Properties of Bamboo/Glass Fibres Hybrid Composites Enhanced with CNTs

Specimen	Tensile Strength (MPa)	Tensile Modulus (GPa)
0% BF/ 0% CNT	47.584 ± 0.786	1.422 ± 0.034
0% BF/ 0.1% CNT	55.364 ± 2.031	1.459 ± 0.095
10% BF/ 0% CNT	74.038 ± 18.831	2.620 ± 0.298
30% BF/ 0% CNT	72.677 ± 3.377	2.405 ± 0.173
10% BF/ 0.1% CNT	75.318 ± 3.318	2.334 ± 0.158
10% BF/ 0.3% CNT	77.540 ± 2.121	1.928 ± 0.146
10% BF/ 0.5% CNT	79.763 ± 3.253	1.866 ± 0.236
10% BF/1.0% CNT	46.797 ± 4.722	2.413 ± 0.294

In contrast, the tensile strength decreased with 1.0 wt.% CNT content, by as much as 36.8%. The lower tensile strength with higher CNT content occurred due to the poor dispersion of the CNTs in the epoxy resin. A high content of CNTs caused poor dispersion of the CNTs in the epoxy resin. This led to the formation of CNT agglomerated regions due to the entanglements of the CNTs. These may act as stress concentration points, which is region of weakness lead to composite failure (Ma and Kim 2011). Hence, this decreased the tensile strength. The poor dispersion of the nanofillers is the main factor that results in not wetting and/or impregnation of the fibre, which compromises the mechanical strength (Ajayan *et al.* 2006; Tarfaoui *et al.* 2016; Tehrani *et al*. 2013).

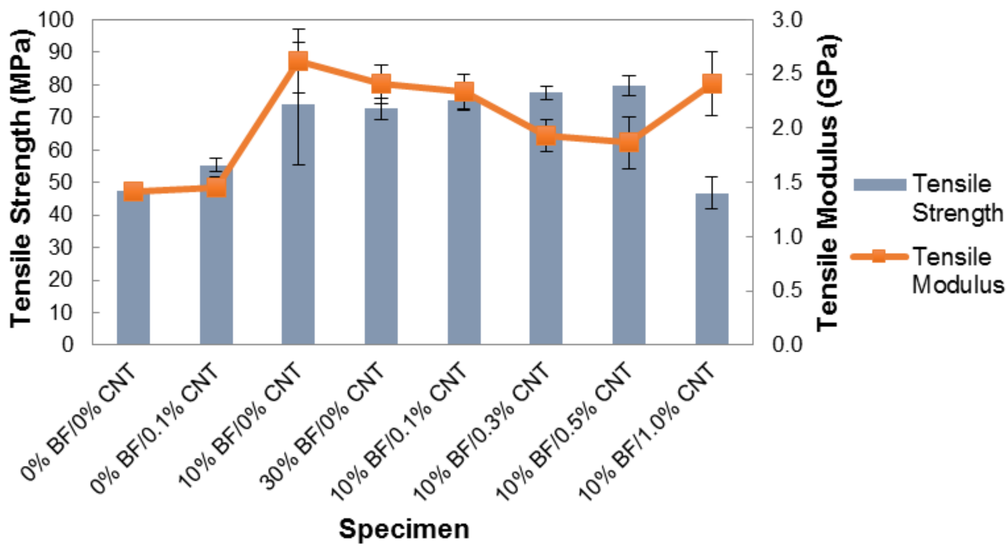

Fig. 3. Tensile modulus and tensile strength of bamboo/glass fibre hybrid composites enhanced with CNTs

Morphology analysis was conducted to observe the dispersion of CNTs in the hybrid composites. Different CNTs weight percentages, *i.e.* 0.5 wt.% (optimum content) and 1.0 wt.% (high content) were observed by Field Emission Scanning Electron Microscope (FE-SEM). Figure 4 shows the fracture surface (induced by tensile) morphology of the hybrids. As depicted in FE-SEM pictures, Fig. 4(a) for optimum content of CNTs revealed nanofillers' full dispersion was achieved. Therefore, fibre pull-out and matrix cracking were the main failures for this type of hybrids as the strength of the hybrid was enhanced. Meanwhile, with increasing of CNTs content (1.0 wt.%) indicates some agglomerates still exist even dispersion agent and mechanism had been applied to the CNTs as in Fig. 4(b). Agglomeration reduced the strength of the hybrid as discussed before.

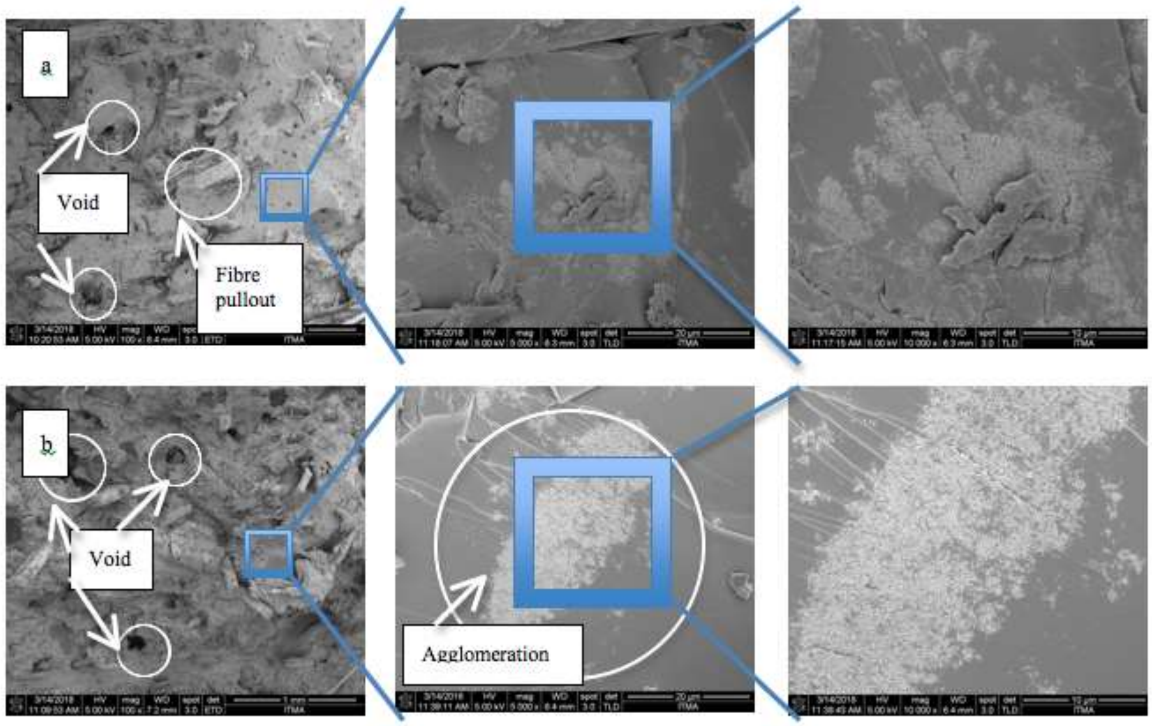

Fig. 4. FE-SEM image of fracture surface of (a) 0.5 wt.% CNTs, and (b) 1.0 wt.% CNTs

For the tensile modulus, added CNTs in the pure epoxy showed a negligible increase, and it decreased when the CNT content increased. The presence of CNTs lowered the tensile modulus of the bamboo/glass fibre hybrid composites. The tensile modulus was decreased by 10.9%, 26.4%, and 28.8% by adding 0.1, 0.3, and 0.5 wt.%, respectively, to the hybrid composites. A value of 10% bamboo/glass fibre hybrid composites, without the presence of CNTs, recorded the highest tensile modulus in this study. However, the tensile modulus increased with 1.0 wt.% CNT content. The trend was irregular, and it was interesting that the tensile modulus increased by 29.3%, compared with 0.5 wt.% CNT content.

Flexural Properties

Flexural properties showed interesting trends because each comparison showed consistent patterns. The graph and table for both the flexural strength and modulus are shown in Table 3 and Fig. 5, respectively. The diffusion of CNTs in neat epoxy and hybrid composites demonstrated lower values in terms of flexural strength and flexural modulus. The presence of 0.1 wt.% CNTs in neat epoxy indicated 1.61% and 8.60% lower flexural strength and flexural modulus, respectively, compared with the neat epoxy. In addition, the flexural strength and flexural modulus values for 10% BF were 14.0% and 4.6%, respectively, higher than 30% BF, and thus, that bamboo weight percentage became the selection for the CNT-added hybrid composites comparison. The matrices did not bind all the higher content of BF completely, and this caused the mechanical properties of the hybrid composites to deteriorate.

Table 3. Flexural Properties of Bamboo/Glass Fibre Hybrid Composites Enhanced with CNTs

Specimen	Flexural Strength (MPa)	Flexural Modulus (GPa)
0% BF/ 0% CNT	71.295 ± 1.411	3.152 ± 0.095
0% BF/ 0.1% CNT	70.146 ± 1.305	2.881 ± 0.068
10% BF/ 0% CNT	155.337 ± 4.783	10.689 ± 0.537
30% BF/ 0% CNT	136.243 ± 4.367	10.220 ± 0.257
10% BF/ 0.1% CNT	132.824 ± 5.601	9.786 ± 0.559
10% BF/ 0.3% CNT	94.599 ± 2.110	9.064 ± 0.952
10% BF/ 0.5% CNT	78.648 ± 11.273	8.342 ± 0.546
10% BF/1.0% CNT	55.550 ± 6.265	8.164 ± 0.664

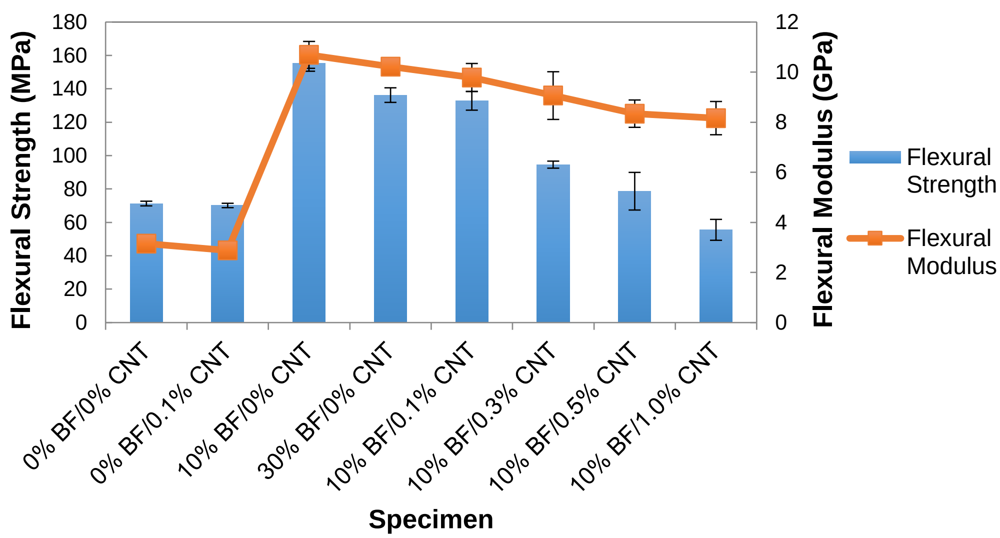

Fig. 5. Flexural modulus and flexural strength of bamboo/glass fibre hybrid composites enhanced with CNTs

Furthermore, the addition of CNTs to the hybrid composites reduced both the flexural strength and the flexural modulus as the content of the CNTs increased. The results were similar to previous research by Sapiai *et al.* (2015); they worked on the flexural properties of kenaf/glass fibre hybrid composites filled with CNTs. The similarities of the research involved the use of natural fibres as fillers and the same testing standards. The flexural strength values for the addition of 0.1 wt.%, 0.3 wt.%, 0.5 wt.%, and 1.0 wt.% CNTs decreased by as much as 14.5%, 39.1%, 49.4%, and 64.2%, respectively, compared to the hybrid control specimen without CNT content. Moreover, decrements of 8.4%, 15.2%, 22.0%, and 23.6% in the flexural modulus were noticeable for 0.1 wt.%, 0.3 wt.%, 0.5 wt.%, and 1.0 wt.% CNTs, compared with hybrid composites without CNTs added (10% BF / 0% CNTs). As the hybrid composites were cured, the higher content of CNTs produced more bendable composites due to the lower interfacial bond between the matrices and the fillers (BF and CNTs). The ineffectiveness of the different shape between BF and CNTs to sustain the stresses transferred from the epoxy contributed to the loss of the composite flexural strength. Further studies need to be conducted to improve the overall properties of the hybrid composites.

CONCLUSIONS

1. The effect of CNT content on the tensile and flexural properties of bamboo/glass fibres hybrid composite was investigated.
2. The addition of CNTs as nanofillers into bamboo/glass fibre hybrid composites enhanced the tensile strength up until 0.5 wt.% CNT by a maximum of 7.7%, and this weakened with further increments in the CNT content at 1.0 wt.% by as much as 36.8% compared with the controls.
3. The flexural properties of hybrid composites decreased a maximum of 64.2% and 23.6% in terms of flexural strength and flexural modulus, respectively, as the CNT content was increased.
4. The promising exploitation of CNTs in a hybrid composite can be achieved if the stress load can be transferred effectively from the epoxy to the nanofillers, and the CNTs are well dispersed.

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