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Lightweight alternatives to glass fiber/epoxy sheet molding compound composites: A feasibility study

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Abstract

The focus of this study is to (i) understand the effect of the fiber type and content on the mechanical properties of sheet-molding compounds composites and (ii) investigate possible lightweight alternatives to glass fibers-sheet molding compound composites. Glass fiber and basalt fibers are used to make sheet-molding compound composites and the mechanical performance are determined as a function of the fiber type and content. In addition, cellulose nanocrystals are used to enhance the properties of the sheet-molding compound resin system. The possibility of lightweighting the basalt fiber/epoxy and glass fiber/epoxy sheet-molding compound composites is explored by replacing a portion of the fibers, i.e. 12–16 wt%, with a small amount cellulose nanocrystals, i.e. 1–2 wt%. No significant difference was found between the basalt fiber/epoxy and glass fiber/epoxy sheet-molding compound composites in terms of mechanical and impact properties. When cellulose nanocrystals were added to the composites, the properties of glass fiber/epoxy sheet-molding compound composites were enhanced while those of basalt fiber/epoxy sheet-molding compound composites deteriorated.

Keywords

Cellulose nanomaterials, sheet-molding compounds, polymer-matrix composites, lightweighting, mechanical properties

Introduction

Increasing the fuel economy in the US transportation sector has become a vital part of US policy to ascertain its energy security and decrease the CO_2 emission. Lightweight vehicles have been identified as a promising approach to increase the fuel efficiency as 10% reduction in the vehicle weight can result in 6–8% increase in fuel efficiency.[1] One approach towards light weighting is use of fiber-reinforced polymer composites instead of metallic components in vehicles.[1]

Life-cycle assessments have shown that use of glass fiber-reinforced polymer (GFRP) composites in automobiles results in lower greenhouse gas emission from cradle to grave compared to conventional materials.[2,3] Moreover, use of natural fibers as reinforcement in polymer composites is gaining attention in the recent years to reduce the environmental impacts. Especially, basalt fibers (BFs) have advantages over other natural fibers (e.g. sisal, kenaf, hemp, flax) because of their superior hygrothermal stability, high strength, modulus and elongation at break, chemical and thermal stability (in the range of –200 to 600–800°C), good thermal insulation, resistance to most weather conditions, easy processability and ecofriendly and economical cost.[4–6] Comparative studies report a better or equal performance of BFRP to GFRP composites.[7–10] In addition, multiscale composites (nano/micro) with BF and carbon nanotubes have also been studied in the past.[11,12]

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Although polymer composites are considered as lightweight materials, especially in auto industries, there is a demand to reduce their weight even more. One approach towards reducing the weight of composites without compromising mechanical performance is to replace the heavier materials in a composite with lighter and stronger ones.[13–15]

Sheet-molding compounds (SMCs),[16] which generally consist of short GFs impregnated between two layers of thermosetting resin, are the precursor materials for automotive applications. SMCs manufacturing method allows for high-volume production and excellent part reproducibility with high specific strength and stiffness and desired surface finish in a cost-effective way due to the low labor requirements and minimum industry scrap. As shown in a prior study by the authors, addition of 1–1.5 wt% cellulose nanocrystals (CNCs) in 25 wt% GF/epoxy SMC composites can result in composites with tensile and flexural properties equal to those of 35 wt% GF/epoxy SMC composites and 8% lower density.[17]

CNCs are cellulose-based, spindle-shaped nanoparticles (3–20 nm in width and 50–500 nm in length) that can be extracted from trees and plants by acid hydrolyses.[18–20] Their low density (1.6 g/cm^3), high aspect ratio (10–100) and surface area, tensile strength of 3 GPa, elastic modulus of 110–220 GPa, surfaces with accessible hydroxyl side groups and their low toxicity make CNC an ideal reinforcement either as a coating on GF[21,22] or as a dispersion in the polymer matrix[22] for polymer composites. Although there are several approaches for achieving a homogeneous dispersion of CNC in epoxy, such as use of waterborne epoxies,[23] solvent exchange methods[24,25] and chemical modification of CNC surfaces,[26,27] achieving a homogeneous dispersion of CNC in epoxy matrices in high-volume production is still a challenge. A factor of uncertainty is the future costs and availability of large volumes of CNCs. CNCs are now produced globally; the production volumes are increasing and prices are decreasing.[28] With continued adoption of CNC in composites and other applications, it will drive the cost of CNC downward as more efficient larger volume production facilities come on line to meet the increased demand for CNCs. A recent study has predicted that future purchase costs of CNC will trend downward to around $7200/ton ($3.60/lb) for optimized industrial-scale production.[29]

In this study, we compare the BF to GF in SMC composites in terms of the mechanical performance and density; and we explore whether the properties of the epoxy resin can be adjusted upon addition of CNCs so that the performance of the GF/epoxy and/or BF/epoxy composites can be enhanced. Furthermore, the possibility of lightweighting of the GF and BF SMC composites by replacing portion of the fibers with a smaller portion of CNC is also investigated. The CNC were introduced in the SMC manufacturing line as a dispersion within the epoxy resin (abbreviated as CNC-epoxy) for the fiber/epoxy formulations with the maximum GF and BF content, which was determined experimentally. The amount of CNC added to the epoxy matrix was in the range of 1.4–2 wt% and was determined based on theoretical calculations by imposing the condition that the composites with CNC should have the same specific modulus as those without. The mechanical performance of both GF/epoxy and BF/epoxy SMC composites was investigated based on the fiber type and content, i.e. GF and BF, and on the content of CNC ranging from 0 to 2 wt%.

Methodology

Two different approaches were used for making lightweight SMC composites: (i) replace GF with BF and (ii) replace a portion of GF or BF with CNC. In the second approach, in order to determine the amount of GF or BF that can be removed and the amount of CNC to be added in the SMC composites, a design criterion based on specific modulus (e.g. modulus divided by density) was applied. Specifically, the SMC composites with CNC should have the same specific modulus as those composites without. Two CNC concentrations of 1.4 and 2 wt% in the epoxy resin were used, according to our prior study,[30] and the density, modulus and strength of the CNC-epoxy resin system, determined experimentally, are presented in Table 1. Then, the reduced amount of GF or BF was calculated using an iteration approach, i.e. the modulus of the neat epoxy was substituted with that of the CNC-epoxy and then the amount of fiber was determined so that the specific modulus of the lightweight composite is equal to that of the composite containing the maximum fiber content and no CNC.

The modulus of the corresponding composites was calculated using equation (1), which is applicable for composites with discontinuous fibers randomly distributed[31]

$$E_{Composite} = \frac{3}{8}E_{11} + \frac{5}{8}E_{22} \quad (1)$$

where, E_{11} and E_{22} are the longitudinal and transverse modulus, respectively, of composites containing discontinuous aligned fibers and determined using Halpin - Tsai micromechanical model described in equations (2) and (3) below

$$E_{11} = E_m\ \ 1 + 2\frac{l_f}{d_f}\ \ {}_L V_f\ \ /\psi 1 \qquad {}_L V_f$$
$$E_{22} = E_m\ \ 1 + 2\ \ {}_T V_f\ /\psi 1 \qquad {}_L V_f \quad (2)$$

Table 1. Density and modulus of the fibers and the resin systems used in the composites.

Material	(g/cm^3)	E (GPa)		Tensile strength (MPa)	
GF	2.54[a]	75[a]		4100	150
BF	2.75[a]	87[a]		4500	500
Epoxy	1.15	3.0	0.3	64.9	4.2
1.4CNC-epoxy	1.15	4.4	0.5 [17]	30.2	5.8
2CNC-epoxy	1.15	4.7	0.3 [17]	40.3	3.3

GF: glass fiber; BF: basalt fiber; CNC: cellulose nanocrystal.
[a]Manufacturer's data.

$$L = \frac{E_f}{E_m} \quad 1 \Big/ \left(\frac{E_f}{E_m} + 2\frac{l_f}{d_f} \right)$$
$$T = \frac{E_f}{E_m} \quad 1 \Big/ \left(\frac{E_f}{E_m} + 2 \right) \qquad (3)$$

where l_f is length, d_f is diameter, V_f is volume fraction, E_f is the elastic modulus of the fiber (GF or BF) and E_m is the elastic modulus of matrix (either epoxy or CNC-epoxy). V_f, V_m and $_c$ are the volume fractions of the fibers and of the matrix and the density of the composite, respectively, and are calculated using equations (4) and (5)

$$V_f = \frac{w_f/\rho_f}{(w_f/\rho_f) + (w_m/\rho_m)}, \quad V_m = 1 \quad V_f \qquad (4)$$

$$_c = \frac{1}{(w_f/\rho_f) + (w_m/\rho_m)} \qquad (5)$$

where w_f and w_m are mass fractions and $_f$ and $_m$ are density of the fiber and matrix, respectively.

Experimental details

Materials

Multi-end roving BFs (TEX 4800, single filament diameter of 10 1 μm) were provided by Mafic Ireland (Kells, County Meath, Ireland) and multi-end roving glass fibers ME1510 (TEX 4800, single filament diameter of 10 1 μm) were received from Owens Corning (Oak Brook, IL, US). Both BF and GF were used in the manufacturing of the SMC as received. The resin used in the SMC line was a bicomponent epoxy resin consisting of diglycidyl ether of Bisphenol-A epoxy and 556 slow polyamide hardener, by US Composites (Wes Palm Beach, FL). The average length of the BF and GF rovings cut in the SMC line was of 25 0.5 mm. Fumed silica (Aerosil-Cabosil supplied by US composites, 5–50 nm particle size) was also used as the thickening agent in the SMC resin. As received freeze-dried powder of sulfonated CNCs, manufactured by the USDA Forest Service-Forest Products Laboratory, Madison, WI, USA, were purchased from the Process Development Center at the University of Maine. The individual CNC particles have an average width and length of 7 2 nm and 134 54 nm, respectively.[32] Figure 1 shows a scanning electron microscopy (SEM) micrograph of the received freeze-dried CNC powder.

Preparation of the CNC-resin dispersion

The resin mixture was prepared in a two-step process: (i) The CNC were dispersed in the hardener using sonication and (ii) the CNC-hardener suspension was mixed with the epoxy, as described by Peng et al.[24] The hardener was selected as the dispersing medium for CNC due to its lower viscosity 400 cP compared to the 19,000 cP viscosity of the epoxy. The desired CNC amount of 1.4 and 2 wt% with respect to the resin was slowly added into 500 g of hardener, mechanically stirred using a small impeller and finally sonicated (UIP500hd heilscher ultrasonic processor, 34 mm probe diameter, amplitude of 90) for 5 min. The temperature during sonication was kept below 50°C with the help of an ice-water bath and visual inspection was employed to monitor the quality of the sonication. Next, 60 g fumed silica thickening agent was mixed with 1000 g epoxy by mechanical stirring at ambient temperature, but no sonication was used as the mixture was too viscous. Based on a set of trial experiments, we found this amount of fumed silica provides enough viscosity for SMC production (e.g. 25,000 cP). Finally, the CNC-hardener suspension was added to fumed silica-epoxy mixture and mechanically stirred for 5 min until a homogenous mixture was achieved. An epoxy-to-hardener ratio of 2:1 wt%, as suggested by the supplier, was used. The prepared resin mixture was used in the SMC line within 10 min of its preparation to prevent gelation (gel time of 35–40 min) and to ensure that the viscosity was still low so that the resin could sufficiently wet the fibers.[22]

Fabrication of SMC composites

The naming scheme for the GF(BF)/epoxy SMC composites is nGF(BF)/mCNC-epoxy, where n is the fiber wt% and m is the CNC wt% in the composite. Initially, in order to determine the maximum fiber content in the SMC, composites with 50, 60, 65 and 70 wt% GF or BF were produced. Then, GF and BF SMC composites with 60 wt% fiber were chosen for the lightweighting study as dry spots were seen in the SMC composites with higher fiber content. For comparison purposes,

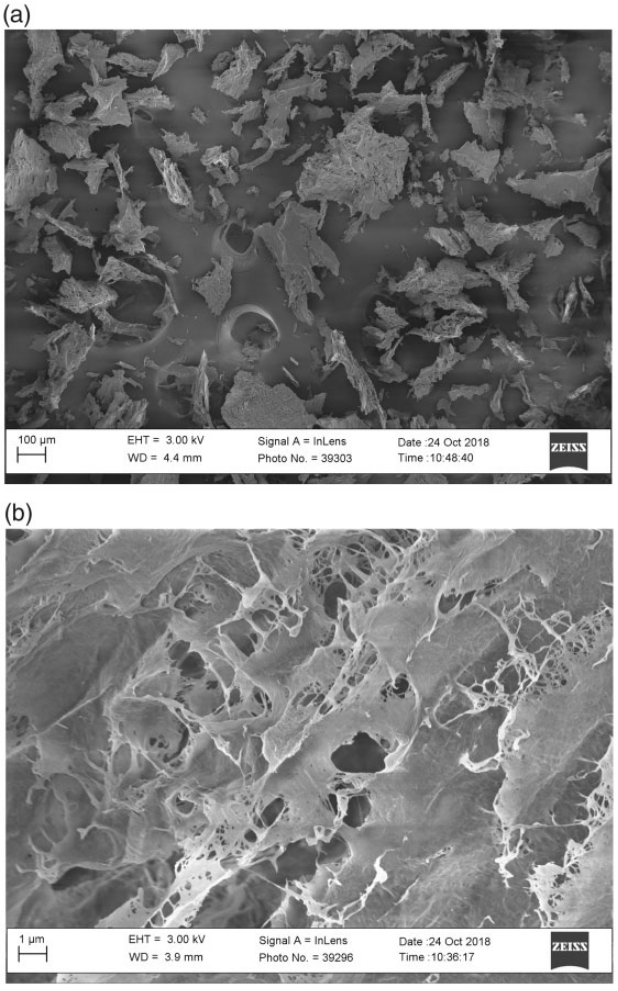

Figure 1. SEM micrograph of the freeze-dried CNC used in this study. SEM: scanning electron microscopy; CNC: cellulose nanocrystal.

CNC was also added to 60 wt% GF/epoxy and BF/epoxy SMC by using the CNC-epoxy dispersion prepared as described earlier. The SMC sheets of different fiber and CNC content were manufactured using a Finn and Fram SMC line at Georgia Tech. The basic difference between this SMC line and the heavy-duty industrial ones is the width of the SMC materials (0.3 m vs 0.9–1.5 m) indicating that the knowledge gained on the processing-structure-property of the resulting SMC composites can have industrial relevance.

In the SMC line, the fiber rovings are pulled through a set of cutting rolls, chopped to bundles of 25.4 mm length and scattered randomly on the lower resin layer, before the upper layer of resin is deposited. A polyethylene carrier film is used to support and carry the resin layers through the SMC line. The resin amount is controlled by doctor blade system and the fiber length and content are controlled by the rotational speed of the cutters and speed of the conveyor belts respectively. Six fiber rovings were used for both the GF and BF but processing settings, i.e. belt speed and the doctor blade height, varied with the fiber type and amount. The fiber/resin sandwich structure passes then through a set of compaction rollers to remove trapped air and ensure full impregnation by the resin. The operation of the line stops every 10 min and the resin mixture is replaced with a new batch to ensure that the resin viscosity remains within its minimum range to facilitate GF (BF) impregnation during compounding, but still sufficiently high to avoid resin leakage from the carrier film.[33] The SMC is collected as a continuous sheet with dimensions of 1.8 m, 254 mm and 3–8 mm (thicker for higher fiber contents). It is noted that the fibers are in the form of fiber bundles (with a length of 25 mm and a diameter in the range of 0.3–1 mm) and we expect that they remain straight when the bundles are away from the surface and edges of the charge (molded part).[34]

Next, the SMC roll is conditioned at ambient temperature for 10 min (set time) to allow the compound viscosity to reach a maturation state where the viscosity is sufficiently high to allow handling and sufficiently low to allow molding of the compound. For every composite made, three SMC layers were stacked on top of one another, placed in a $420 \times 280 \times \sim 5$ mm rectangular mold and hot-pressed using a Wabash V50-1818-2TMX Hydraulic heated press. The molding took place at 400 kPa and 100°C for 1 h, followed by post-curing at 120°C for 2 h. Vacuum was applied during the curing process. The closing speed of the hot press was kept constant at 7 cm/s (the maximum speed) for all the batches. It is expected that the closing speed would not significantly impact the resin flow pattern[31] and thus the properties of the SMC composites as the charge used was thin and completely covered the mold surface. After curing, the plates remained at ambient temperature for 48 h prior to cutting the testing coupons to prevent any potential plastic deformation during handling/testing. Test coupons were cut from the plates using a waterjet (MAXIEM 1515).

Characterization techniques

Water displacement method was used to measure the specific density according to ASTM D-792. Each density value reported is an average of at least 11 measurements. The void content of the composites was measured using acid digestion test according to ASTM D3171. Nitric acid 70% was used to dissolve the epoxy matrix of the composites at 80°C for 24 h. Then, the remaining fibers were thoroughly washed with distilled water, dried at 50°C for 6 h and weighed to measure the fiber mass fraction and void content.

The void content for the composites was calculated according to the following equation

$$V_{void} = 100 \times (\ _{exp,wd} \quad _{exp,ad})/\rho_{exp,wd} \qquad (6)$$

where $_{exp,wd}$ and $_{exp,ad}$ are the measured density from water displacement and acid digestion methods, respectively. $_{exp,ad}$ is determined by substituting the measured fiber mass fraction and the known densities of the fiber and matrix in equation (5). Each data point for the void content of a composite with a specific composition is an average of at least six measurements.

The tensile properties of the SMC composites were determined according to ASTM D638 using an Instron 5982 equipped with 100 kN load cell for dogbone specimens with a gauge length of 57 mm, width of 13.1 mm and thickness of 5 mm. An extensometer, Instron 2630-106, with a gauge length of 25 mm was used to record the axial strain. The modulus was calculated between the axial strain values of 0.05% and 0.2%. Samples that fractured at the grip section were removed and not included in the analysis. The flexural properties were measured based on three-point bending tests performed using an Instron 33R 4466 equipped with 10 kN load cell according to ASTM D790-02 with a support span of 50 mm and thickness of 5 mm at a displacement rate of 2.15 mm/min. Each tensile and flexural data point is an average of at least 10 measurements. The impact energy was measured using Charpy on non-notched rectangular samples with a support span of 43 mm, width of for 12.7 mm and thickness of 5 mm, using an Instron SI series pendulum impact tester with a maximum impact head of 406.7 J (300 ft-lbf) according to ISO179. Each impact data point is an average of at least 10 measurements.

A Phenom G2 Pro (Phenom-World BV) and a LEO1530 SEM at acceleration of 5 and 3 kV, respectively, were used to study the fracture surface of the SMC composite failed in tensile testing. A plasma sputter (Ted Pella Inc.) was used to apply gold coating on the surface of the samples prior to SEM imaging to minimize charging effect.

The quality of fiber wetting by the epoxy resin with or without CNC was assessed based on contact angle measurements. The measurements were made using a Ramé–Hart Goniometer (model 500-U1) at ambient conditions. Static contact angles were obtained by dispensing 4 μL of neat epoxy and 1.4 CNC-epoxy onto a fiber mat. Each fiber mat was composed of eight strands (pieces of roving used in the SMC line) of tightly packed fibers (short glass or BFs), which were held tight and flat against a glass slide. To adhere the mats to the glass slides, deionized water was placed at the interface and the edges were taped. The mats were allowed to dry at room temperature for 24 h before performing the measurements. The peripheral program DROPimageAdvanced™ was used to capture the images and measure the contact angles.

To assess whether or not the effects of CNC additions on specific properties were statistically significant, statistical analysis using one-way analysis of variance (ANOVA) with a level of significance of 5% (i.e. 95% level of confidence) was completed. The single-factor (CNC effect) ANOVA was carried out between GF or BF composites with CNC to the epoxy resin to those of 60GF/epoxy or BF/epoxy with no CNC. When the p value is larger than 0.001 and the F ratio ($F/F_{Critical}$) is less than 1, the difference between the mean values is considered negligible.

Results and discussion

Glass fibers vs basalt fibers: Effect of fiber type and content on the mechanical properties of SMC composites

The tensile, flexural and impact properties of SMC composites as a function of fiber type, i.e. GF vs BF, and fiber content varied from 50 to 70 wt% and are presented in Figure 2. As expected, the properties increase with fiber content for both fiber types. A fiber content of 60 wt% is determined as the upper bound for both the GF and BF SMC composites. In case of BF SMC, the plates made with 70 wt% BF, contained dry spots, indicating that there was not enough resin to fully wet the fibers. Composite plates with 70 wt% GF SMC were free of defects and contained significantly fewer dry spots. However, considering the statistical standard deviation indicating that there was no enhancement of the mechanical properties upon use of 70 wt% GF instead of 60 wt%, 60 wt% was also the upper bound for the GF content. It is noted that the large statistical standard deviation observed in the results is likely due to fiber-rich regions at the center of the SMC composite plates. This is the result of the outward flow of the resin due to the pressure during the compaction in SMC manufacturing process and/or the hot press.[33]

As shown in Figure 2, there is no clear trend on the effect of the fiber type, as the property enhancement is similar for the same fiber content and type, i.e. GF or BF. A similar trend, that is BF and GF SMC composites exhibit similar tensile, flexural and impact properties at a lower fiber content of 25 wt%, was reported in a prior study by the authors.[30] In addition, considering that BFs have higher tensile modulus and strength but higher density, as reported in Table 1, one can claim that the BF and GF can be used interchangeably as far as the enhancement of the mechanical properties is

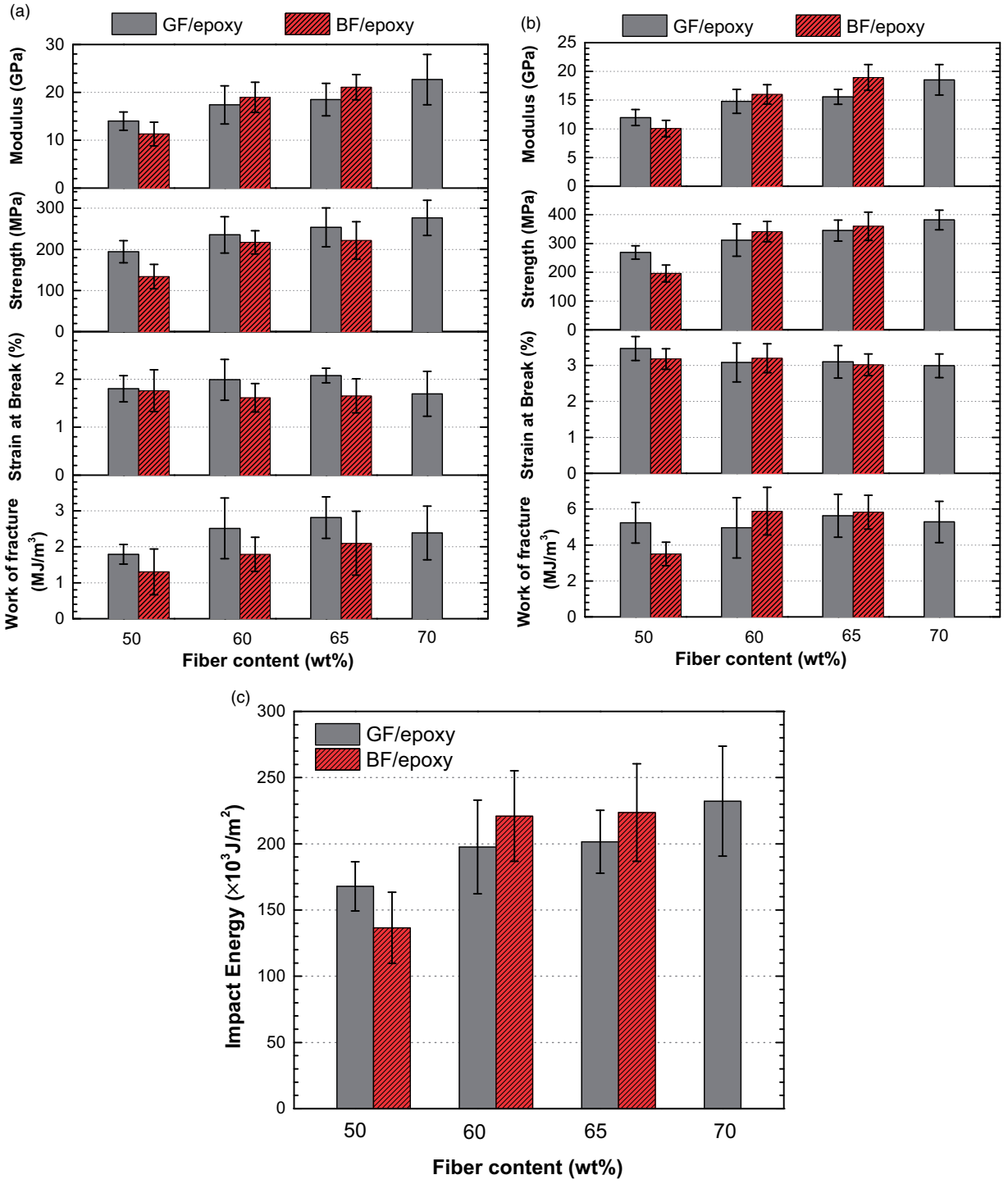

Figure 2. Mechanical properties of fiber/epoxy SMC composites as a function of the fiber type and content: (a) Tensile, (b) flexural and (c) impact properties. Error bars are 1 standard deviation.

concerned. Other fiber features including cost, sustainability and/or recyclability should be considered during the fiber selection process.

In summary, although theoretically lightweight composites can be obtained by replacing GF with BF, based on the results presented, it becomes clear that this possibility is rather slim. The reason is that the BF/epoxy composites exhibit similar properties with GF/epoxy composites at the same fiber content and are heavier. A more appropriate surface treatment of the BF may

enhance the interfacial interactions with the epoxy resin and yield the expected property enhancement including lightweighting.

Effect of CNC on the properties of GF/epoxy and BF/epoxy SMC composites

As reported by the authors,[17] addition of CNC in GF/epoxy SMC composites resulted in 8–10% weight reduction, achieved by reducing the GF amount from 35 wt% to 25 wt% in GF/epoxy and adding 1–1.5 wt% CNC into the epoxy resin formulation. The explanation for the enhanced properties of the GF/epoxy composites upon addition of CNC is that CNC increase significantly the modulus of the epoxy resin, as shown in Table 1, which allows for reduction of the GF content. Also shown, the strength of the epoxy significantly reduces upon addition of CNC; however, as discussed above, the property of interest as the guideline was the modulus and the specific modulus in particular.

The hypothesis in this study is that higher weight reduction can be achieved in composites that contain higher amount of either GF or BF. Considering that the maximum fiber content in fiber/epoxy SMC composites is 60 wt%, as discussed above, equations (1) to (5) were employed to determine the amount of GF or BF that can be replaced by CNC with no compromise in specific tensile modulus.

The tensile modulus and density of the composites as a function of the fiber and CNC content calculated using equations (1) to (5) along with the specific modulus, defined as the modulus-to-density ratio, are presented in Table 2. As shown, the fiber content for both GF and BF can be reduced from 60 wt% to either 48 wt% or 44 wt% upon addition of 0.9 wt% or 1.1 wt% respectively, and the first step without compromising the specific modulus. These theoretical calculations were used as guidelines, the first step, in designing and manufacturing of lightweight composites. It is noted that the CNC were added into the resin mixture used in the SMC line.

The tensile and flexural properties including modulus, strength, elongation at break and work of fracture, as well as the impact strength of both the GF/epoxy and BF/epoxy composites with or without CNC, are shown in Figure 3. The following observations are made. First, there is no clear trend in any of the properties determined, when it comes to comparison of GF and BF at 60 wt% fiber content irrelevant of whether the epoxy resin contains CNC or not. Also, addition of 0.6 wt% CNC does not affect the properties of fiber/epoxy composites containing 60 wt%. When the fiber amount is reduced from 60 wt% to 48 or 44 wt% and 0.9 or 1.1 wt% of CNC is added, the tensile modulus reduces with no significant decrease in the tensile strength, elongation at break and work of fracture in case of GF/epoxy composites. On the contrary, in case of BF/epoxy composites all tensile properties, especially the strength, deteriorate significantly. As the strength is dictated by the stress transfer ability across the fiber/matrix interface, it becomes clear that non-favorable interfacial interactions between the BF and the CNC/epoxy resin exist.

Table 2. Estimated modulus, density and specific modulus according to equations (1) to (5), for GF/epoxy and BF/epoxy SMC composites as a function of the fiber and CNC content.

Composite	E (GPa) from equations (1) to (5)	(g/cm^3)	$E_{specific}$
60GF/epoxy	14.38	1.71	8.40
48GF/0.9CNC-epoxy	13.23	1.56	8.48
44GF/1.1CNC-epoxy	12.73	1.51	8.41
60BF/epoxy	14.99	1.76	8.48
48BF/0.9CNC-epoxy	13.41	1.58	8.47
44BF/1.1CNC-epoxy	13.17	1.54	8.52

GF: glass fiber; BF: basalt fiber; CNC: cellulose nanocrystal; SMC: sheet-molding compound.

In terms of flexural modulus, a similar decrease is observed for both the GF/epoxy and BF/epoxy composites upon partial replacement of the fibers by the CNC; whereas the other flexural properties including strength, strain at break and work of fracture slightly decrease. The impact energy also drops upon reduction of the fiber content with larger decrease observed again in the case of the BF/epoxy composites. The decrease of the flexural properties and impact strength upon addition of CNC is higher in the case of the BF composites.

Considering that the fibers have different density and the design requirement was to create composites of the same specific tensile modulus, the rest of the specific properties are determined in order to validate the hypothesis of lightweighting. In particular, for specific properties, the tensile and flexural properties and impact strength of the fiber/epoxy SMC composites as a function of the fiber type and content and the CNC content are shown in Figure 4. Accounting for the experimental error, there is no difference among the properties of the various composites. In order to determine whether the results are statistically significant, analysis of variance (ANOVA) was performed. The results for the GF/epoxy and BF/epoxy composites are presented in Tables 3 and 4, respectively.

In particular, the tensile, flexural and impact specific properties for 60GF/epoxy and 60BF/epoxy are not affected by the addition of 0.6 wt% CNC in the epoxy resin of the SMC composites. For 48GF/0.9CNC-epoxy and 44GF/1.1CNC-epoxy SMC composites, the tensile, flexural and impact-specific properties are

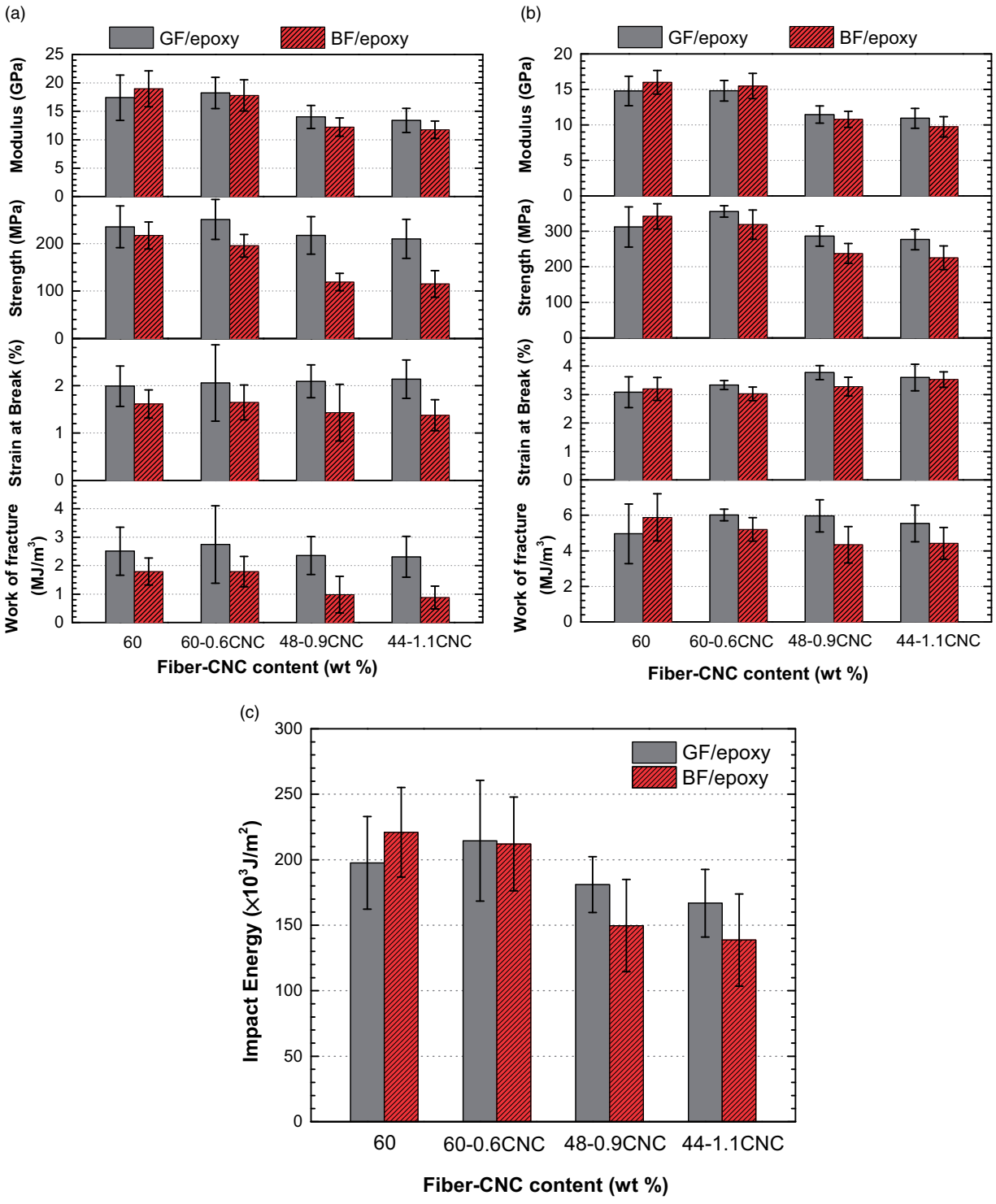

Figure 3. Mechanical properties of fiber/CNC-epoxy SMC composites as a function of the fiber type and content and CNC concentration: (a) Tensile, (b) flexural and (c) impact properties. Error bars are 1 standard deviation. CNC: cellulose nanocrystal; SMC: sheet-molding compound.

comparable to each other. Interestingly, several of the specific properties are similar to 60GF/epoxy, despite the lower GF content, suggesting that the CNC additions to epoxy resin has a positive effect on specific properties. The enhancement of the modulus of GF/CNC-epoxy composites is expected to be the result of the increase in the apparent modulus of the matrix (CNC-epoxy) due to the stiffening effect of the CNC.

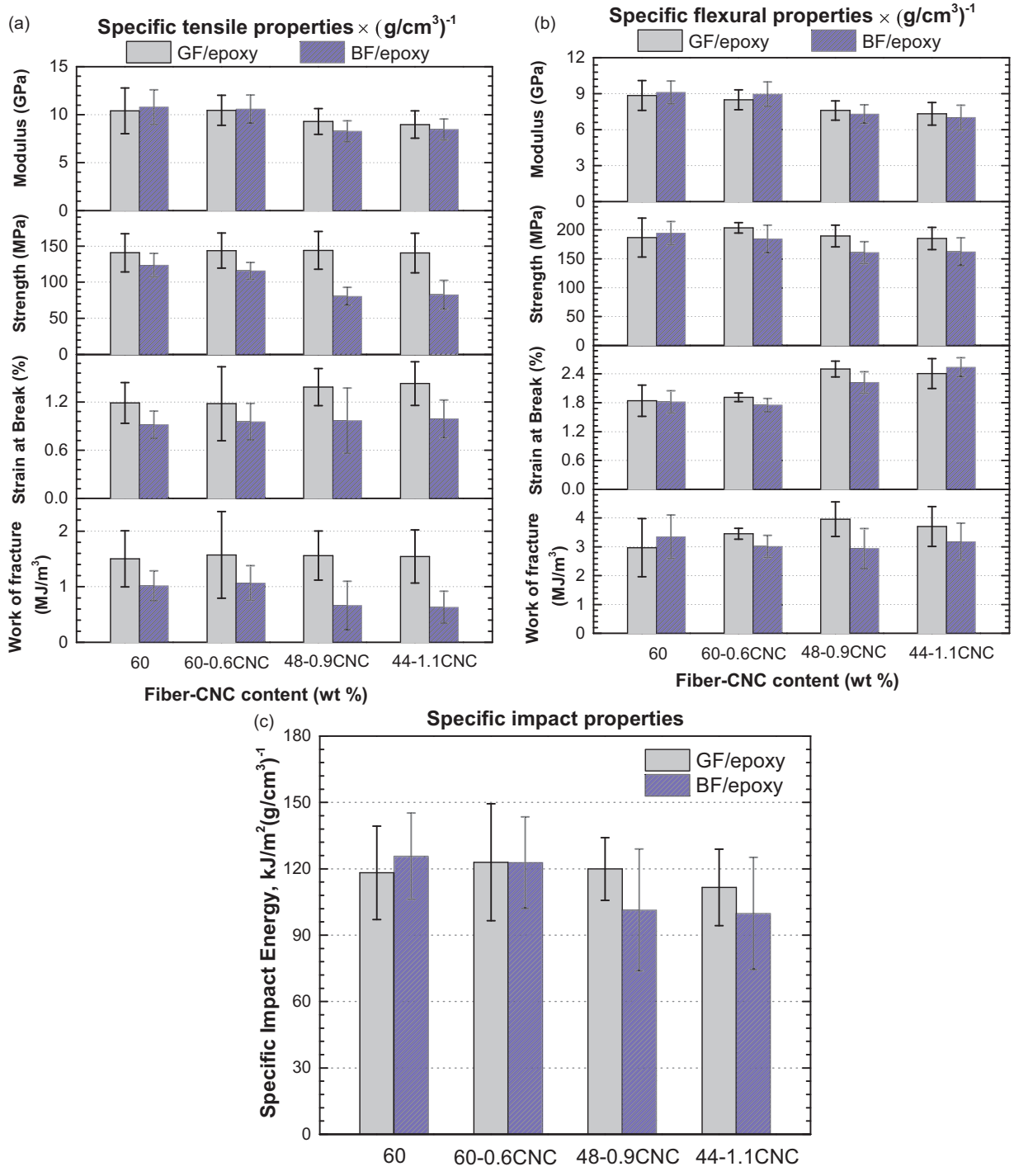

Figure 4. Specific (a) tensile, (b) flexural and (c) impact properties of GF/CNC-epoxy and BF/CNC-epoxy SMC composites in light weighting study. Error bars are 1 standard deviation. GF: glass fiber; BF: basalt fiber; CNC: cellulose nanocrystal; SMC: sheet-molding compound.

Also, increases in strength may possibly result from a stronger GF-matrix adhesion and hence, a more efficient stress transfer ability across the GF/CNC-epoxy interface.[35] It is also noted that the strength of the epoxy resin decreases upon addition of CNC, as reported in Table 1, which indicates that the strength enhancement in the composites is due to positive interfacial interactions. This hypothesis is supported by micrographs of the corresponding fracture surfaces, as shown by representative SEM images in Figures 5 and 6, which indicate the increase of adhesion between the GF and epoxy in presence of CNC.

Table 3. ANOVA test results for mechanical performance between specific properties of 60GF/epoxy and *n*GF/*m*CNC-epoxy SMC composites.

Sample	Sum of squares	*p* Value	*F* ratio	Sum of squares	*p* Value	*F* ratio
	Tensile modulus			Tensile strength		
60GF/0.6CNC-epoxy	0.01	>0.001	6×10^{4}	36.16	>0.001	0.01
48GF/0.9CNC-epoxy	4.78	>0.001	0.33	43.11	>0.001	0.01
44GF/1.1CNC-epoxy	7.68	>0.001	0.49	0.27	>0.001	8×10^{5}
	Flexural modulus			Flexural strength		
60GF/0.6CNC-epoxy	0.59	>0.001	0.12	1392.3	>0.001	0.59
48GF/0.9CNC-epoxy	11.46	>0.001	2.21	9.65	>0.001	3×10^{3}
44GF/1.1CNC-epoxy	11.46	>0.001	2.21	9.65	>0.001	3×10^{3}
	Impact energy					
60GF/0.6CNC-epoxy	118.65	>0.001	0.04			
48GF/0.9CNC-epoxy	16.33	>0.001	0.01			
44GF/1.1CNC-epoxy	226.4	>0.001	0.14			

ANOVA: analysis of variance; *F* ratio: the ratio of $F/F_{Critical}$; GF: glass fiber; BF: basalt fiber; CNC: cellulose nanocrystal; SMC: sheet-molding compound.

Table 4. ANOVA test results for mechanical performance between specific properties of 60BF/epoxy and *n*BF/*m*CNC-epoxy SMC composites.

Sample	Sum of squares	*p* Value	*F* ratio	Sum of squares	*p* Value	*F* ratio
	Tensile modulus			Tensile strength		
60BF/0.6CNC-epoxy	0.19	>0.001	0.01	285.81	>0.001	0.31
48BF/0.9CNC-epoxy	31.85	=0.001	3.25	9247.26	<0.001	10.08
44BF/1.1CNC-epoxy	27.37	=0.002	2.78	8326.03	<0.001	5.71
	Flexural modulus			Flexural strength		
60BF/0.6CNC-epoxy	0.10	>0.001	0.02	482.66	>0.001	0.22
48BF/0.9CNC-epoxy	14.72	<0.001	4.36	5081.43	=0.002	2.96
44BF/1.1CNC-epoxy	19.75	<0.001	4.44	4660	=0.005	2.12
	Impact energy					
60BF/0.6CNC-epoxy	48.93	>0.001	0.03			
48BF/0.9CNC-epoxy	3254.38	>0.001	1.32			
44BF/1.1CNC-epoxy	3514.49	>0.001	1.59			

ANOVA: analysis of variance; *F* ratio: the ratio of $F/F_{Critical}$; GF: glass fiber; BF: basalt fiber; CNC: cellulose nanocrystal; SMC: sheet-molding compound.

For 48BF/0.9CNC-epoxy and 44BF/1.1CNC-epoxy SMC composites, the tensile, flexural and impact-specific properties are comparable to each other. However, in contrast to the GF system, several of the specific properties were lower than the corresponding properties of the 60BF/epoxy. In this case, the property enhancement of the matrix due to the addition of CNC did not offset the deterioration of properties due to the decrease of the fiber content. It is possible that the interactions between the BF and the CNC are not as favorable and the wetting of the BF by the CNC-modified epoxy resin is not as complete as in the case of GF. This hypothesis is confirmed comparing the void content of the various composites. As shown in Table 5, the void content of 44BF/0.9CNC-epoxy and 44BF/1.1CNC-epoxy is higher (14–19%) compared to that of corresponding GF/CNC-epoxy composites (9%). The void content in our study is similar to Le et al.'s[34] findings on the strong dependence of the void content to applied pressure during compression molding, where 10% void content was reported for 400 kPa (the pressure used in our study). Figure 7 represents the micrograph of thickness sides of a BF/epoxy composite sample showing the voids.

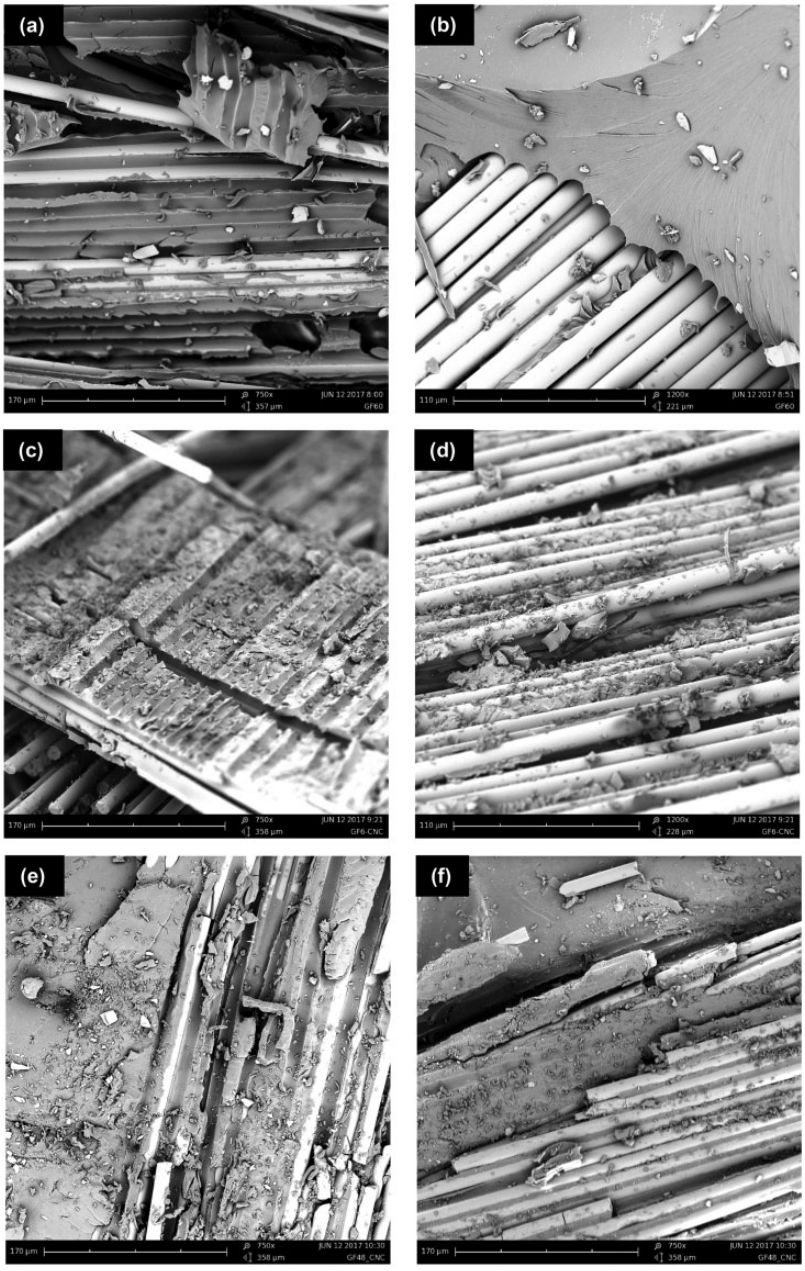

Figure 5. SEM images for tensile fracture surface of (a,b) 60GF/epoxy, (c,d) 60GF/0.6CNC-epoxy and (e,f) 48GF/0.9CNC-epoxy SMC composites. SEM: scanning electron microscopy; GF: glass fiber; CNC: cellulose nanocrystal; SMC: sheet-molding compound.

The interfacial interactions were assessed by contact angle measurements where a drop of the resin with and without CNC was deposited on the fibers. According to the results presented in Table 6, the contact angle is higher for the case of BF and increases for both BF and GF upon addition of CNC in the resin remain higher for BF.

A morphological study of the tensile fracture surface was conducted to investigate possible differences in the failure mechanisms of the GF and BF composites and representative images are shown is Figures 5 and 6, respectively. In GF composites, the main failure mode is fiber debonding in presence or absence of CNC. Clean cavity traces of the pulled-out GF in the matrix were observed. The addition of CNC in the epoxy results in rougher fracture surfaces and more matrix residues on the GF, suggesting an increase in the GF/matrix interfacial adhesion. It is plausible that the presence of CNC in the resin increases the friction between fiber and polymer and thus more energy is

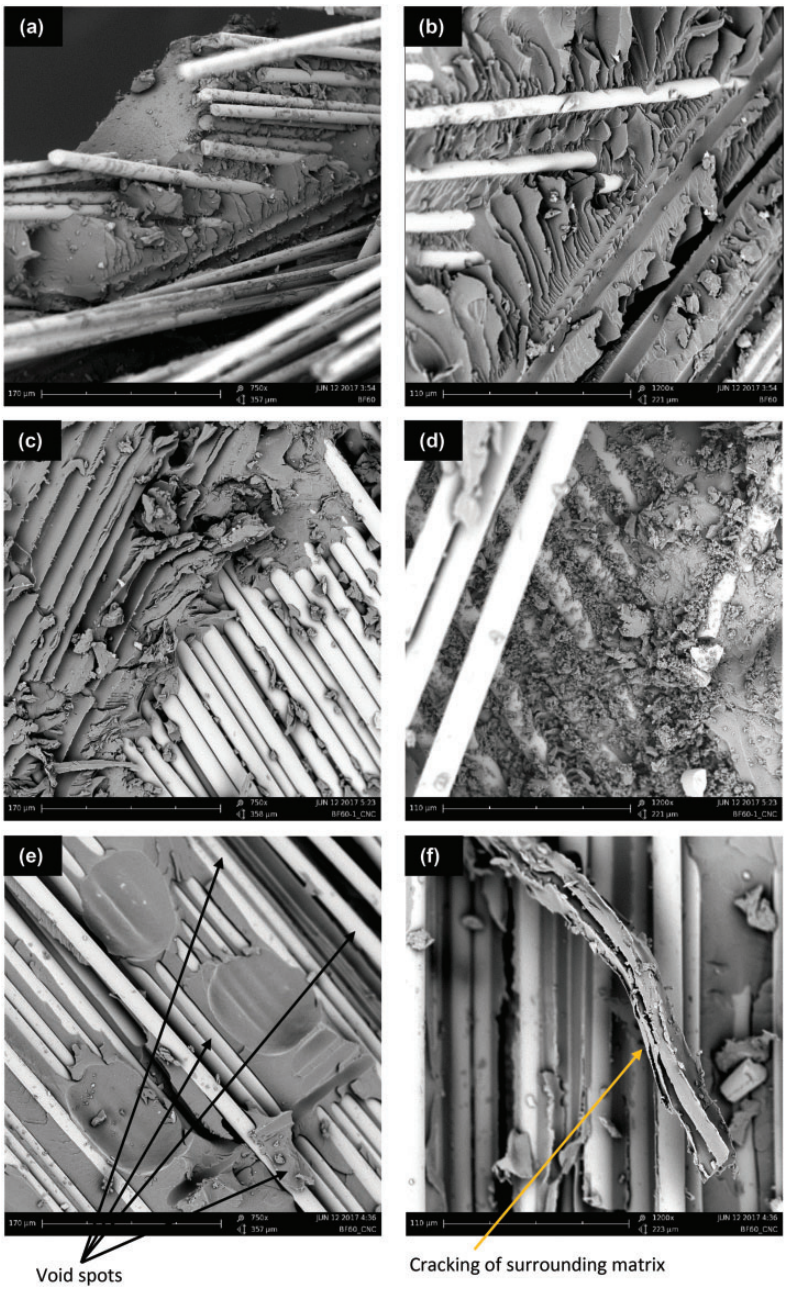

Figure 6. SEM images for tensile fracture surface of (a,b) 60BF/epoxy, (c,d) 60BF/0.6CNC-epoxy and (e,f) 48BF/0.9CNC-epoxy SMC composites. SEM: scanning electron microscopy; BF: basalt fiber; CNC: cellulose nanocrystal; SMC: sheet-molding compound.

required to debond the fiber from the matrix.[36] Toughening mechanisms such as interlocking, restricting GF debonding at the GF/matrix interphase and interfacial crack bridging[37] in the presence of CNC can also contribute to stronger interfacial adhesion. These mechanisms lead to an increase in the absorbed energy in fracture and, thus, improvement in mechanical properties.

The main failure mode in BF composites is fiber pull-out, as seen in Figure 6(a) and (f). In contrast to GF/epoxy composites (Figure 5), the addition of CNC did not appear to improve the BF-resin interfacial adhesion as indicated by (i) smooth fracture surfaces, (ii) clean cavity traces and (iii) pulled-out fibers without any resin residues as shown in Figure 6(b), (c) and (e). In addition, the fracture surface of both 60BF/epoxy and 60BF/0.6CNC-epoxy composites is smooth as shown in Figure 6(a) and (c) indicating similar fracture toughness independent of CNC presence in the matrix. Shear cusps also seem to be smooth and similar in size

Table 5. Theoretical density, measured density and void content.

Composite	$_{c,theoretical}$ (g/cm^3)	$_{exp}$ (g/cm^3)		Fiber wt% (acid digestion)		Fiber volume fraction (acid digestion)		$_{exp}$ corrected for fiber % (g/cm^3)		Void content (%)	
60GF/epoxy	1.71	1.67	0.06	69	8	50	9	1.85	0.07	10	0.2
60GF/0.6CNC/epoxy	1.71	1.74	0.06	66	5	46	5	1.80	0.08	3.4	1
48GF/0.9CNC/epoxy	1.56	1.51	0.03	56	5	36	4	1.66	0.06	9.5	1.5
44GF/1.1CNC/epoxy	1.51	1.49	0.05	52	6	33	5	1.61	0.07	8.5	0.5
60BF/epoxy	1.77	1.75	0.07	72	3	52	3	1.97	0.07	12.5	0.5
60BF/0.6CNC/epoxy	1.77	1.73	0.05	71	8	51	9	1.96	0.1	14	1
48BF/0.9CNC/epoxy	1.59	1.48	0.05	55	5	34	4	1.69	0.07	14	1
44BF/1.1CNC/epoxy	1.55	1.39	0.06	53	8	32	7	1.66	0.1	17	4

$_{c,theoretical}$: theoretical density using equation (5) and assuming that the fiber content in the SMC composites is equal to the set value; $_{exp,wd}$: measured density from water displacement method; $_{exp,ad}$: measured density from acid digestion method; GF: glass fiber; BF: basalt fiber; CNC: cellulose nanocrystal; SMC: sheet-molding compound.

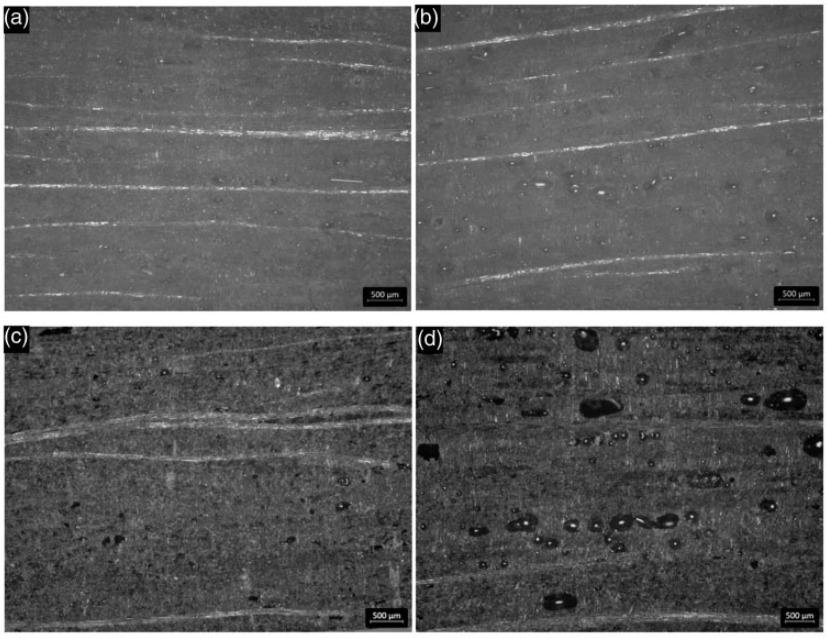

Figure 7. Optical micrographs of the voids in (a) 60GF/epoxy, (b) 48GF/0.9CNC-epoxy, (c) 60BF/epoxy, (d) 48BF/0.9CNC-epoxy (thickness side). GF: glass fiber; BF: basalt fiber; CNC: cellulose nanocrystal; SMC: sheet-molding compound.

Table 6. Contact angle results between fiber rovings and either epoxy or CNC-epoxy.

Fiber	Neat epoxy		1.4CNC-epoxy	
GF	30.71	2.81	35.03	2.23
BF	34.16	1.43	40.87	0.66

GF: glass fiber; BF: basalt fiber; CNC: cellulose nanocrystal.

as shown in Figure 6(b) and (c), suggesting a similar crack growth in BF/epoxy composites with and without CNC. Additionally, more voids in BF/CNC-epoxy composites were observed than in the corresponding GF composites, as seen Figure 6(e). All these observations imply that addition of CNC does not significantly improve the mechanical properties of BF/epoxy SMC composites.

Ramifications of lightweighting approach

In order to validate the design approach and confirm that the target, i.e. making lighter composites without compromising the specific modulus, the experimentally determined modulus was compared to the theoretically predicted one as calculated using equations (1) to (5). It is noted that this time, the value for the fiber content

Table 7. Specific modulus: theoretical predictions (corrected for the actual fiber content) vs experimental results.

Composite	Modulus exp (GPa)	Modulus equations (1) to (5) based on actual fiber % (GPa)	$_{exp}$ based on actual fiber % (g/cm^3)	Specific exp modulus (GPa/g cm ^{3})
60GF/epoxy	17.39 3.99	17	1.85 0.07	9.4 2.16
48GF/0.9CNC/epoxy	14.02 2.03	15.6	1.66 0.06	8.45 1.22
44GF/1.1CNC/epoxy	13.41 2.14	14.8	1.61 0.07	8.33 1.33
60BF/epoxy	18.98 3.16	20	1.97 0.07	9.63 1.6
48BF/0.9CNC/epoxy	12.22 1.62	16	1.69 0.07	7.23 0.96
44BF/1.1CNC/epoxy	11.75 1.54	15.8	1.66 0.1	7.08 0.93

GF: glass fiber; BF: basalt fiber; CNC: cellulose nanocrystal.

used in the calculations was not the one according to the set value at the SMC line (which was used to calculate the theoretical modulus presented in Table 2) but the one that was determined experimentally by the acid-digestion test.

As shown in Table 7, there is strong agreement between the experimental and theoretical moduli for a given composite system in case of GF/epoxy composites. That is, both the experimental and theoretical moduli decrease equally upon addition of CNC and reduction of the GF content. In case of BF composites, the moduli are the same for 60BF/epoxy composite within experimental error, but the theoretical prediction is higher than the experimental value in the composites containing CNC. This deviation reflects the non-favorable interactions between CNC and BF that result in higher void content as discussed above.

Overall, the lightweight benefit becomes clear by comparing the experimental specific modulus, strength and impact energy of 60GF/epoxy composite to those of the lighter composites that contain CNC, i.e. the 48GF/0.9CNC-epoxy and 44GF/1.1CNC-epoxy composites. Considering the overlap of the experimental error, it can be concluded that the lighter GF/CNC-epoxy composites are equivalent in terms of specific properties to the GF/epoxy ones.

Conclusions

The major conclusion of this study is that the tensile, flexural and impact properties of GF/epoxy SMC composites are similar to those of BF/epoxy SMC composites for the same fiber content. Thus, the two different fiber types can be used interchangeably and other criteria such as cost, availability and sustainability should be used in order to identify the appropriate fiber for a specific application. The hypothesis of making lighter composites by replacing GF with the heavier but stronger BF was not validated.

It was demonstrated that lightweight composites can be made by replacing portion of GF in GF/epoxy SMC composites with a small amount of CNC without compromising tensile and flexural properties and impact strength. The interactions between the GF and CNC were found to be positive considering the fact that addition of CNC in epoxy resin resulted in reduction of strength but addition of CNC in GF/epoxy composites resulted in strength increase. Contrary to the GF, the interactions between BF and CNC were not favorable as indicated by the increase of void content of the BF composites upon addition of the CNC and the poor wetting of the BF by the CNC-epoxy resin system. Thus, it became clear that for the given BF used in this study, there is no benefit in adding CNC in the BF/epoxy composites and it is not possible to replace portion of the BF with CNC without significantly compromising the mechanical properties.

Finally, the simple approach of the same specific modulus for designing lighter hybrid composites so that the hybrid GF/CNC-epoxy composites are lighter and exhibit the same specific properties was shown to be very effective. Such an approach can be employed to replace the commonly used ''trial and error'' approach often used in design and manufacturing of composites with tailored properties.

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