Hybridization of Hemp Fiber and Recycled-Carbon Fiber in Polypropylene Composites

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Abstract: In recent years there has been a substantial growth in the use of natural fiber reinforced composite in more advanced applications. However, high strength applications require high mechanical properties. Hybridization of natural fibers with synthetic fibers is an effective method of increasing the field of application and mechanical properties. The effects of hybridizing hemp (Cannabis sativa L.) fiber with recycled-carbon fiber were investigated in this study to determine the trends in mechanical properties resulting from varied weight fractions. Characterization of void content was accomplished using micro computed tomography (micro-CT). Through hybridizing hemp fiber and recycled carbon fiber in a polypropylene thermoplastic, a new class of high performance, low cost composites were demonstrated for injection molding applications. This study showcased a 10–15% increase in tensile strength after the reinforcement of recycled-carbon fiber with hemp fiber. A 30–35% increase was observed in the flexure strength after the reinforcement of recycled-carbon fiber with hemp fiber. Impact strength also had an increase of 35–40% for hemp fiber reinforced recycled-carbon fiber polypropylene composites.

Keywords: bio-sustainability; recycled-carbon fiber; hemp fiber; thermoplastic composite

1. Introduction

Composite materials are gaining more preference as a choice of material in several industrial applications. They help in achieving the desired mechanical properties by combining different materials in a skillful way. In general composites reinforced with lignocellulosic long fibers such as hemp, flax, jute, cotton possess high strength-to-weight and high stiffness-to-weight ratios, which makes them useful in aerospace and automotive applications [1]. Natural fiber composites are found to have higher strengths than wood composites and a few plastics, which improves their opportunity as materials of choice for competing applications in the future. Despite their suitable properties, natural fibers lack thermal stability, exhibit strength degradation, water absorption and poor impact properties. To overcome these properties researchers have turned their focus towards studying the effect of hybridizing natural fibers with synthetic fibers [2]. Hybrid composites consist of an amalgamation of two or more fibers in a polymer matrix.

Hybrid composites have comprehensive applications in the engineering field due to low cost, high strength to weight ratio and ease of manufacturing [3]. A different blend of mechanical properties such as stiffness, ductility, and strength can be achieved through hybridization which cannot be achieved by single fiber reinforced composites. A natural fiber reinforced hybrid composite possess a good strength and stiffness value near to glass fiber reinforced composites [4]. Hemp fiber is one of the economical and promptly available natural bast fibers which has attracted attention of several researchers. Much less literature is available on the use of short hemp fibers as reinforcement. [5]. Several researchers used industrial hemp fiber to reinforce biopolymers such as cellulose acetate and...
found that composites prepared by extrusion followed by injection molding exhibited high flexural strength and stiffness [6].

Carbon fiber has already gained significant acceptance in automotive and aerospace industries. Carbon fiber is an expensive material and out of reach for many industries. Therefore, recycling carbon fibers and mixing them with thermoplastic offers an affordable alternative [7]. Reducing waste and reusing materials with high-embedded energy and recycled in an energy-efficient manner is environmentally and socially desirable. Recycled-carbon fiber retains many of its inherent advanced properties, even though it has been reclaimed from waste. In addition, the price of recycled carbon fiber is at least half than that of virgin carbon fiber [8].

Several researchers examined the flexure behavior of short recycled carbon fibers (rCF) which were mixed with flax fiber in the PLA matrix. Experimental data showed that the flexural properties increased with higher rCF content, with the maximum being a flexural modulus of approximately 14 GPa and flexural strength of 203 MPa [9].

There are many different possible fiber combinations for high stiffness applications. Several researchers examined the hybrid effect for flax and rCF with thermoset resin [10]. There is lack of research for the impact of hybrid effect on thermoplastic composites which use rCF blended with natural fibers [10,11]. In this study, hemp fibers were hybridized with recycled carbon fiber keeping the polypropylene matrix constant. Polypropylene was used since it can save on material cost as well as can be used as regrind without compromising the performance of the resin [12]. The total environmental impact of polypropylene is less than traditional materials in life cycle analysis [12]. As an initial investigation this study only considered a few combinations to observe potential trends. The hybridization of hemp and recycled carbon fiber offers a good potential for developing high stiffness composites for various automotive applications while simultaneously incorporating bio-based materials into a product.

2. Materials and Methods

The materials used in this study for developing hybridized composites were maleic anhydride, recycled-carbon, and hemp fiber in a polypropylene matrix. The materials were processed into a composite by using a Leistriz co-rotating twin-screw extruder Type Mic 18/GI-40D and pelletized with a Scheer Bay pelletizer model BT25. Once the materials were processed they were injected using a Technoplas hydraulic injection molder (Technoplas Inc, SIM 5050A, USA). After injection, a wide range of mechanical testing was performed to determine the mechanical abilities of the hybrid composites. Further analysis of void formation was also done using SEM and micro-CT.

2.1. Fibers Description

The recycled-carbon fiber used in this study was supplied by ELG Carbon Fiber Ltd. The recycled-carbon fibers came already chopped and were on average 6.3 mm in length and 7 µm in diameter. A published fiber density of 1.40 g/cm³ was used [11].

Hemp fibers used in this study were provided by Sunstrand LLC. Fibers came already chopped and were on average 6.35 mm in length and 20 µm in diameter. A published fiber density of 1.48 g/cm³ for hemp was used [12]. Hemp fibers had a tensile modulus of 30 GPa and a tensile strength of 300 MPa.

2.2. Polymers Used

The polymer used throughout this study was polypropylene (PP78151E) supplied by ExxonMobil Chemical. The density of polypropylene used was 0.90 g/cm³. The flexure modulus and tensile strength were 2.068 GPa and 34.40 MPa, respectively. Polybond 3200 was used as the maleic anhydride compatibilizer. The density was 0.91 g/cm³ and melt flow rate was 115 g/10 min.
2.3. Materials Processing

Hemp and recycled-carbon fibers were compounded with polypropylene (PP) and a coupling agent, maleic anhydride grafted PP (MA-g-PP/MAPP), using a co-rotating twin-screw extruder and then subsequently pelletized with a pelletizer. Maleic anhydride surface treatments were tested at 2% weight concentrations. The mechanical properties of the materials were analyzed in accordance with ASTM standards. The PP was dried for 24 h at 80 °C. The moisture content of the PP was tested using a MAX 4000XL moisture analyzer and showed 0.001% moisture content prior to processing operations. Hemp and recycled-carbon fibers were dried at 80 °C for 12 h and moisture content was below 0.1%. After drying, the materials were mixed according to the material weight fractions with formulations used shown in Tables 1 and 2.

| Sample | Hemp Fiber | rCF-Chopped Fiber | Matrix-PP | Treatment-MAPP |
|--------|------------|-------------------|----------|----------------|
| 1      | 20         | 10                | 70       | None           |
| 2      | 10         | 20                | 70       | None           |
| 3      | 20         | 10                | 68       | 2              |
| 4      | 10         | 20                | 68       | 2              |

† rCF: Recycled carbon fiber. PP: Polypropylene. MAPP: Maleic anhydride.

Table 2. Samples with constant fiber wt % and matrix PP †.

| Sample | Fiber  | Matrix-PP |
|--------|--------|-----------|
| 1      | Hemp-30| 70        |
| 2      | rCF-30 | 70        |

† rCF: Recycled carbon fiber. PP: Polypropylene.

The materials were compounded using a Leistritz co-rotating twin-screw extruder Type Mic18/Gl-40D and pelletized with a SCHEER BAY pelletizer model BT25 (Bay Plastics Machinery, Michigan, USA). The compounding of materials was carried out at 250 rpm screw speed with a barrel profile temperature from feed throat to nozzle of 149, 171, 177, 182, 188, and 188 °C for each section respectively. Test samples were dried again and then injected into a mold using a 50-ton Technoplast hydraulic injection molder model SIM-5080. The injection molding processing conditions for samples were an injection pressure of 20.7 MPa with a temperature profile from nozzle to feed throat of 193, 199, 199, 186, and 188 °C, for each section respectively. A time of 25 s was given for the specimens to cool under pressure before opening the mold.

2.4. Tensile Test

The tensile properties of the samples, strength and modulus were measured using an Instron universal tester model 5567. Tensile testing was performed according to ASTM D638, using an MTS Extensometer model 632.11B-20. Five specimens were tested for each test sample. The specimens were tested in tension at a crosshead speed of 5 mm/min with a 2 kN load cell. They were 3.92 mm in thickness and 9.96 mm in width.

2.5. Flexure Test

Flexural properties of the samples, strength and modulus were tested using a three-point bend testing, as specified in ASTM D790, using an Instron 5567 load frame. The speed of the crosshead was calculated according to the guidelines specified in the standard, which resulted in 1.36 mm/min. Five specimens were tested for each test sample. A load cell of 2 kN was used in the test setup.
The support span used for the testing was 51 mm in length. The specimens were 3.2 mm in thickness and 12.7 mm in width.

2.6. Impact Test

The composite materials were tested for their impact properties using a Tinius Olsen Model Impact 104 Izod impact tester with a pendulum energy of 2.7548 J. The impact testing was carried out in accordance with ASTM standard D256. A minimum of five specimens were tested for each sample. The samples were 10.93 mm wide at the notch and 3.18 mm in depth.

2.7. Scanning Electron Microscope (SEM)

Fractured-test specimens were mounted on cylindrical aluminum mounts with colloidal silver paste (Structure Probe Inc., West Chester PA, USA) for view of the fractured surface and then coated with a conductive layer of gold using a Cressington 108 auto sputter coater (Ted Pella Inc., Redding CA, USA).

2.8. Micro-CT

The General Electric (GE) v|x micro CT equipment enabled nondestructive evaluation of internal structure. Micro-CT refers to micro computed tomography. A 240 kV micro focus X-ray computed tomography system with an additional 180 kV submicron X-ray tube. A high-contrast digital flat panel detector was used for the greatest possible versatility. Void content and fiber concentrations of the fractured surface was observed using the micro-CT setup.

3. Results and Discussion

3.1. Tensile Test

The analysis of variance (ANOVA) carried out for fibers and treatments and the interaction between them was significant \( p \leq 0.05 \) for the main effects and the interaction. Recycled-carbon fiber has significant difference when added in proportions of more than 10% in hybrid composites. Figure 1 shows the comparison and interaction between the mean values of tensile strengths of fibers and treatments. The letters a, b, c, d, and e show significant differences between treatments based on ANOVA results. The reinforcement of recycled carbon fiber in hemp fiber improved the tensile strength by 5–15%. A significant 30–35% increase in the tensile strength of hemp fiber was observed after the addition of the compatibilizer (Figure 1). Recycled carbon fiber shows a higher tensile strength than that of hemp fiber when blended with the resin. Other researchers conducted similar studies where different fibers such as flax, jute and glass were compounded with polypropylene at different weight fractions [13]. Similarly, to the current results, tensile strength decreased as the amount of flax fiber bundles was increased, this reduction being more drastic when MAPP modifier in the matrix was not used. MAPP-modified composites exhibited better mechanical properties than the unmodified ones since MAPP maleic anhydride group can bond with both flax and glass fibers, resulting in improved interfacial adhesion between matrix and both types of fibers [14]. Maleic anhydride can bond more effectively with rCF than hemp fibers as the OH group of the fibers reacts with the anhydride group and maleic acid group of MAPP forming a hydrogen bonding and ester linkage [7]. Research studies on recycled carbon fiber composites have shown similar test results when compared with the current study as well. The reinforcement of recycled-carbon fiber in the polypropylene matrix has increased the tensile and flexure properties significantly [15].

For tensile modulus, the ANOVA indicated a significant effect of fiber, treatment and their interaction \( p \leq 0.05 \). Recycled-carbon fiber when hybridized with hemp fiber composites does not show significant difference when treated with compatibilizer. Figure 2 shows the comparison and interaction between the mean values of tensile modulus of the fibers. A significant 10–15% increase in the tensile modulus of hemp fiber can be observed after the addition of the compatibilizer.
A strong adhesion between the fiber and the resin can be observed in the top left corner of Figure 3b, (Figure 2). Letters a, b, c, d, and e show significant differences between treatments based on ANOVA results. Recycled carbon fiber composite exhibits a higher tensile modulus than hemp fiber composite. The modulus in all hybrid composites observed a decreased trend when compared to the recycled carbon fiber composite. Another study also indicated similar trends [16]. MAPP-modified hybrid composites have also been shown to have a higher modulus than unmodified hybrid composite ones [14].

![Figure 1. Comparison of the tensile strength for hemp and rCF composites between: Sample 1—hemp fiber 20% + rCF 10% + PP 70%, Sample 2—hemp fiber 10% + rCF 20% + PP 70%, Sample 3—hemp fiber 20% + rCF 10% + PP 68% MAPP 2%, Sample 4—hemp fiber 10% + rCF 20% + PP 68% MAPP 2%, and Sample 5—(hemp or rCF) fiber 30% + PP 70%.

![Figure 2. Comparison of the tensile modulus for hemp and rCF composites between: Sample 1—hemp fiber 20% + rCF 10% + PP 70%, Sample 2—hemp fiber 10% + rCF 20% + PP 70%, Sample 3—hemp fiber 20% + rCF 10% + PP 68% MAPP 2%, Sample 4—hemp fiber 10% + rCF 20% + PP 68% MAPP 2%, and Sample 5—(hemp or rCF) fiber 30% + PP 70%.

To support these results an SEM study was conducted. The following images portray the fiber polymer bonding between different fibers which help to study the characteristics and nature of the fibers. Figure 3 shows the comparison of SEM images for all the fibers and composites. Though fiber pullout can be spotted in Figure 3a there still exists a strong adhesion between the interface of recycled-carbon fiber and the polypropylene resin. This tends to improve the tensile strength for the composites. This combination has no voids which supports the fact of high mechanical properties. A strong adhesion between the fiber and the resin can be observed in the top left corner of Figure 3b,
which is evidence as to why the bond is strong. It can be observed that there are no polypropylene particles stuck on the hemp fibers which reduces the stress concentration giving it high strength. The tensile strength is high and after the addition of the compatibilizer it can be observed that there is a remarkable increase in the tensile properties. Researchers used SEM for studying flax fiber blended with polypropylene resin. The results showed a clean fiber surface indicating poor wettability and lack of adhesion, which agreed with the tensile results. It was also observed that fibers were unevenly distributed throughout the matrix due to differences in characteristics between flax fiber and the matrix [15].

![SEM images](image)

**Figure 3.** Comparison of SEM images for (a) rCF-30% PP-70%, and (b) hemp-30% PP-70%.

This current study also shows similar results and agrees with the strength and modulus values of other composites reported [15]. By comparing with the specimen without the coupling agent, a significant difference in fracture morphology was observed [15]. At high magnification, the fiber surfaces appeared rough and surrounded by a layer of polymer. All these indicated that a good adhesion was achieved. Degree of polymer coverage on the pulled-out fibers and their adhesion with the host matrix depends on the coupling agent type and loading [15]. Plastic deformation of the host matrix was also very different from that of the sample without coupling agent and again the appearance varied with the type of coupling agent. Each coupling agent exhibited a somewhat different improvement in strength [17].

### 3.2. Flexure Test Results

The ANOVA indicated a significant effect of fibers and treatments, and the interaction between them ($p \leq 0.05$). Recycled-carbon fiber when hybridized with hemp fiber reinforced composite materials has significant difference between interactions. Figure 4 shows the comparison of flexure strength for hemp fiber reinforced composites. The letters a, b, c, d, and e show significant differences between treatments based on ANOVA results. The reinforcement of recycled-carbon fiber in hemp fiber improved the flexure strength by 30–33%. A significant 20% increase in the flexure strength of hemp fiber composite was observed after the addition of the compatibilizer. Recycled-carbon fiber shows high flexure strength values when compared with hemp fiber. Other researchers investigated different mechanical properties of hemp and flax fibers. The results showcased that addition of flax and hemp fibers to PP matrix increased the flexure strengths when compared with PP. MAPP treatment increased the flexure strength values significantly. A marked increase was also seen after the addition of the fibers in a polymeric matrix [18].

The presence of polypropylene treated with maleic anhydride in the composite matrix has been associated with a significant increase in flexural strength, while paradoxically has also been associated with a decrease in the flexural modulus. Elastic deformation is experienced in the compression stage and not in the tensile stage which supports the fact of low modulus over strength [19]. Other researchers
Sustainability 2019, 11, x FOR PEER REVIEW 8 of 13

Figure 5. Comparison of the flexure modulus for hemp and rCF composites between: Sample 1—hemp fiber 20% + rCF 10% + PP 70%, Sample 2—hemp fiber 10% + rCF 20% + PP 70%, Sample 3—hemp fiber 10% + rCF 20% + PP 70%, Sample 4—hemp fiber 10% + rCF 20% + PP 68% MAPP 2%, Sample 5—(hemp or rCF) fiber 30% + PP 70%.

The ANOVA results indicated fibers, treatments, and the interaction between them was significant (p < 0.05). Recycled carbon fiber does not show significant differences in flexural modulus compared to hemp fibers, while the addition of the compatibilizer, MAPP, increases the flexural modulus of the composite. The reinforcement of recycled-carbon fiber composites with PP matrix has higher flexural strength and modulus than hemp fiber [7]. Other researchers investigated different methods to recycle carbon fiber and use it in different industrial applications. The study also showed a very strong bonding between CF and PP resin which leads to high flexure properties [21].

Flexure Modulus (GPa)

(a) (b) (c) (d) (e) (f)

Samples 1 2 3 4 5

Flexure Modulus (GPa)

(a) (b) (c) (d) (e) (f)

Samples 1 2 3 4 5

Flexure Modulus (GPa)

(a) (b) (c) (d) (e) (f)

Samples 1 2 3 4 5

Flexure Modulus (GPa)

(a) (b) (c) (d) (e) (f)

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Flexure Modulus (Gpa)

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Flexure Modulus (Gpa)

(a) (b) (c) (d) (e) (f)

Samples 1 2 3 4 5

Flexure Modulus (Gpa)

(a) (b) (c) (d) (e) (f)
To support these flexure results, micro-CT was carried out for the flexure samples to investigate void formation. Figure 6 portrays the comparison between different fibers. Figure 6a–c shows the micro-CT for fractured surface of hemp fiber composites. The composite shows a clean and pristine surface with no evidence of large void formation which leads to high flexure properties. The absence of void formation also leads to high mechanical property trends. Figure 6d,e, shows the micro-CT for the fractured surface of the hemp fiber composites treated with MAPP. The composite shows random orientation of fiber for both hemp and rCF. A study was conducted wherein glass fibers were reinforced with PP and the failure strain analysis was investigated. Micro-CT scans were also conducted which reveal a strong heterogeneity of the microstructure, in terms of fiber orientation. In other work, short-glass fibers followed in-plane distributions of orientation around a preferential orientation in shell and core layers of the injected composite plates, with preferential orientation parallel to the injection flow direction (IFD) in shell layers and highly angled with respect to IFD in core layer [23].

Figure 6. Comparison of micro-CT between (a) hemp-30% PP-70%, (b) hemp-10% PP-70% rCF-20%, (c) hemp-10% PP-68% MAPP-2% rCF-20%, (d) hemp-20% PP-70% rCF-10%, and (e) hemp-20% PP-68% MAPP-2% rCF-10%.
3.3. Impact Test Results

The ANOVA results indicated a significant effect of fibers and treatments, and the interaction between them ($p \leq 0.05$). Recycled-carbon fiber does not show significant differences between interactions in samples two and three as well as one and five for hemp fiber composites. Figure 7 shows the comparison and interaction between the mean values of impact strengths of all the fibers. The letters a, b, c, d, and e show significant differences between treatments based on ANOVA results. A 30–35% increase was observed in the impact strength of hemp fiber after the maleic anhydride treatment. The impact strength for recycled-carbon fiber is the lowest as compared with that of hemp fiber. Hemp fibers show the highest impact strength. Hemp fiber has very high shear strength compared with that of recycled-carbon fiber making it difficult to break. Other researchers investigated the effect of recycled-carbon fiber composites on different mechanical properties. The results showed that the dispersion of the reinforcements/fillers has a strong influence on mechanical and morphological properties of composites [23]. The impact strength decreased as the percentage of recycled-carbon fiber increased which agrees with the results of the current study. In addition, other researchers investigated the effect of reinforcement of hemp fiber composites on mechanical properties [24]. The results showed that the bonding between the hemp fiber and the resin was very strong which was attributed to the good adhesion between fibers and the matrix. It was worth noticing that the interactions between the PP matrix and the hydroxyl groups on cellulosic fibers were enhanced as the fiber weight percentage increased. Finally, other researchers investigated the impact properties for different composite materials. Higher impact strength values for many natural fibers such as flax, hemp, and sisal when compared with other synthetic fibers such as carbon fiber as weight fraction increased were reported [25].

![Figure 7. Comparison of impact strength for hemp and rCF composites between: Sample 1—hemp fiber 20% + rCF 10% + PP 70%, Sample 2—hemp fiber 10% + rCF 20% + PP 70%, Sample 3—hemp fiber 20% + rCF 10% + PP 68% MAPP 2%, Sample 4—hemp fiber 10% + rCF 20% + PP 68% MAPP 2%, and Sample 5—(hemp or rCF) fiber 30% + PP 70%.](image)

To support these results an SEM study was conducted. The following images portray the fiber polymer bonding between different fibers and help to study the characteristics and nature of the fibers. Figure 8 shows the SEM comparison between hemp and recycled carbon fiber composites. It can be observed in Figure 8a that the polymer–fiber bonding is relatively weaker resulting in low impact properties. Debonding takes place internally in the specimen, which results in poor strength of the impact test. The impact strength is decreased due to the non-uniform size of filler material (recycled-carbon fiber) and its distribution over the surface occupying the over edges of the matrix. Fiber pullout can be easily spotted. Voids can be observed which supports the fact of low impact
strength. Recycled carbon fiber has low shear strength and hence is brittle in nature. Therefore, it can be observed that the impact strength of recycled-carbon fiber is lower than hemp fiber. It can be observed from the Figure 8b that the polymer fiber bonding for hemp fiber is relatively stronger than recycled carbon fiber resulting in high impact properties. Strong adhesion between the interface of fiber and matrix can be observed in the image. The SEM image clearly portrays no occurrences of fiber pulled out in the specimen, leading to high impact properties. High impact strength plays a key role in aerospace and automotive applications. Figure 9a,b shows the comparison of SEM study for hemp fiber reinforced-rCF composites with and without the treatment of the compatibilizer. It can be observed from Figure 9a that the polymer fiber bonding is relatively stronger than the untreated fibers resulting in high impact properties. Although fiber pullout can be spotted, it can be observed that after the maleic anhydride treatment the covalent bond between the fiber and polymer was strong. Hybridization of recycled fiber with hemp fiber has improved the impact strength.

**Figure 8.** Comparison of SEM between (a) rCF-30% PP-70% and (b) hemp fiber-30% PP-70%.

**Figure 9.** Comparison of SEM between (a) SEM study for hemp fiber-20% rCF-10% PP-70% and (b) hemp fiber-20% rCF-10% PP-68% MAPP-2%.

Other researchers investigated different properties of hemp fibers and their composites. The results were similar to the current study, which proves that hemp fiber has high shear strength and high impact properties. Both the natural fibers are hydrophilic in nature and contain many fibrils which tend to bond and entangle with the hydrogen bonds in the matrix [26]. Researchers investigated different fiber reinforced composites and the results showed low impact properties for recycled carbon fiber with increasing weight fractions [27]. The results were also in agreement with the current study which proves the fact that rCF has low impact properties when compared with other hybrid composites with increasing natural fibers content.
4. Conclusions

Hybridizing synthetic fibers with natural fibers has been found to be an effective method for improving the mechanical properties of a composite. In addition to the gains in mechanical properties, which resulted from hybridizing synthetic fibers with natural fibers, it was also concluded that the use of recycled carbon fiber as a reinforcement improved flexure and tensile properties significantly. The reinforcement of recycled carbon fiber reduced the mechanical variability which is evident in the micro-CT images. Micro-CT showed improvement in the orientation of the fibers which helped to reduce mechanical variability having a very strong effect on the mechanical properties. The treatment of natural fibers with maleic anhydride played a major role in improving the interfacial adhesion between fibers and matrix thus enhancing the mechanical properties. An increase of 35–40% was observed in the tensile strength of the composites, whereas an increase of 30% was observed in the flexure properties of the composites after the treatment. Impact properties showed an increase of 10–15% after the treatment of the compatibilizer. From a composite designer’s point of view hemp fiber composites can be used for applications with reasonably high tensile and flexure strengths, whereas they would be the best choice for applications which require high impact strengths. Hemp fiber is coarse in nature due to the arrangement of its constituents (pectin, lignin and hemi-cellulose) which promote higher impact strength. Further expansion of usage of more sustainable and bio-based polymers could also be taken into consideration for future work. The end goal of this research was to provide a means for natural fibers to expand into various lightweight and sustainable design applications. Through hybridizing natural bast fibers and recycled carbon fibers in a polyolefin thermoplastic, a new class of high-performance composites can be demonstrated for a cluster of different applications.

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References
1. Elkhaoulani, A.; Arrakhiz, F.; Benmoussa, K.; Bouhfid, R.; Qaiss, A. Mechanical and thermal properties of polymer composite based on natural fibers: Moroccan hemp fibers/polypropylene. Mater. Des. 2013, 49, 203–208. [CrossRef]
2. Mochane, M.J.; Mokhena, T.C.; Mokothu, T.H.; Mtibe, A.; Sadiku, E.R.; Ray, S.S.; Ibrahim, I.D.; Daramola, O.O. Recent progress on natural fiber hybrid composites for advanced applications: A review. Express Polym. Lett. 2019, 13, 159–198. [CrossRef]
3. Gohil, P.; Patel, K.; Chaudhary, V. Natural fiber-reinforced polymer composites: A comprehensive study on machining characteristics of hemp fiber-reinforced composites. In Biomass, Biopolymer-Based Materials, and Bioenergy; Elsevier: Amsterdam, The Netherlands, 2019; pp. 25–50.
4. Pickering, K.L.; Efendy, M.A.; Le, T.M. A review of recent developments in natural fibre composites and their mechanical performance. Compos. Part A Appl. Sci. Manuf. 2016, 83, 98–112. [CrossRef]
5. Panthapulakkal, S.; Sain, M. Injection-molded short hemp fiber/glass fiber-reinforced polypropylene hybrid composites—Mechanical, water absorption and thermal properties. J. Appl. Polym. Sci. 2007, 103, 2432–2441. [CrossRef]
6. Vignon, M.; Dupeyre, D.; Garcia-Jaldon, C. Morphological characterization of steam-exploded hemp fibers and their utilization in polypropylene-based composites. Bioresour. Technol. 1996, 58, 203–215. [CrossRef]
7. Akonda, M.; Lawrence, C.; Weager, B. Recycled carbon fibre-reinforced polypropylene thermoplastic composites. Compos. Part A Appl. Sci. Manuf. 2012, 43, 79–86. [CrossRef]
8. Schinner, G.; Brandt, J.; Richter, H. Recycling carbon-fiber-reinforced thermoplastic composites. J. Thermoplast. Compos. Mater. 1996, 9, 239–245. [CrossRef]
9. Tse, B.; Yu, X.; Gong, H.; Soutis, C. Flexural Properties of Wet-Laid Hybrid Nonwoven Recycled Carbon and Flax Fibre Composites in Poly-Lactic Acid Matrix. *Aerospace* **2018**, *5*, 120. [CrossRef]

10. Pickering, S.J. Recycling technologies for thermoset composite materials—Current status. *Compos. Part A Appl. Sci. Manuf.* **2006**, *37*, 1206–1215. [CrossRef]

11. Bachmann, J.; Wiedemann, M.; Wierach, P. Flexural mechanical properties of hybrid epoxy composites reinforced with nonwoven made of flux fibres and recycled carbon fibres. *Aerospace* **2018**, *5*, 107. [CrossRef]

12. Kawasumi, M.; Hasegawa, N.; Kato, M.; Usuki, A.; Okada, A. Preparation and mechanical properties of polypropylene-clay hybrids. *Macromolecules* **1997**, *30*, 6333–6338. [CrossRef]

13. Sanjay, M.; Arpitha, G.; Naik, L.L.; Gopalakrishna, K.; Yogesh, B. Applications of natural fibers and its composites: An overview. *Nat. Resour.* **2016**, *7*, 108. [CrossRef]

14. Lau, K.; Hung, P.; Zhu, M.-H.; Hui, D. Properties of natural fibre composites for structural engineering applications. *Compos. Part B Eng.* **2018**, *136*, 222–233. [CrossRef]

15. Arbelaiz, A.; Fernandez, B.; Ramos, J.; Retegi, A.; Llano-Ponte, R.; Mondragon, I. Mechanical properties of short flax fibre bundle/polypropylene composites: Influence of matrix/fibre modification, fibre content, water uptake and recycling. *Compos. Sci. Technol.* **2005**, *65*, 1582–1592. [CrossRef]

16. Wong, K.; Mohammed, D.S.; Pickering, S.; Brooks, R. Effect of coupling agents on reinforcing potential of recycled carbon fibre for polypropylene composite. *Compos. Sci. Technol.* **2012**, *72*, 835–844. [CrossRef]

17. Etaati, A.; Pather, S.; Fang, Z.; Wang, H. The study of fibre/matrix bond strength in short hemp polypropylene composites from dynamic mechanical analysis. *Compos. Part B Eng.* **2014**, *62*, 19–28. [CrossRef]

18. Li, H.; Sain, M.M. High Stiffness Natural Fiber- Reinforced Hybrid Polypropylene Composites High Stiffness Natural Fiber-Reinforced. *Polym. Plast. Technol. Eng.* **2007**, *42*, 853–862. [CrossRef]

19. Shahzad, A. A study in physical and mechanical properties of hemp fibres. *Adv. Mater. Sci. Eng.* **2013**, 2013. [CrossRef]

20. Adalberto, S.; Junior, R.; Zanchi, C.H.; Carvalho, R.V.D. Flexural strength and modulus of elasticity of different types of resin-based composites. *Braz. Oral Res.* **2007**, *21*, 16–21.

21. Pimenta, S.; Pinho, S.T. Recycling carbon fibre reinforced polymers for structural applications: Technology review and market outlook. *Waste Manag.* **2011**, *31*, 378–392. [CrossRef]

22. Holmes, M. Recycled carbon fiber composites become a reality. *Reinf. Plast.* **2018**, *62*, 148–153. [CrossRef]

23. Notta-Cuvier, D.; Neiri, M.; Lauro, F.; Delille, R.; Chaari, F.; Robache, F.; Haugou, G.; Maalej, Y. Coupled influence of strain rate and heterogeneous fibre orientation on the mechanical behaviour of short-glass-fibre reinforced polypropylene. *Mech. Mater.* **2016**, *100*, 186–197. [CrossRef]

24. Wei, H.; Nagatsuoka, W.; Lee, H.; Ohshima, I.; Sumimoto, K.; Wan, Y.; Takahashi, J. Mechanical properties of carbon fiber paper reinforced thermoplastics using mixed discontinuous recycled carbon fibers. *Adv. Compos. Mater.* **2018**, *27*, 19–34. [CrossRef]

25. Shahzad, A. Hemp fiber and its composites—A review. *J. Compos. Mater.* **2012**, *46*, 973–986. [CrossRef]

26. Safri, S.N.A.; Sultan, M.T.H.; Jawaid, M.; Jayakrishna, K. Impact behaviour of hybrid composites for structural applications: A review. *Compos. Part B Eng.* **2018**, *133*, 112–121. [CrossRef]

27. Park, J.-M.; Quang, S.T.; Hwang, B.-S.; DeVries, K.L. Interfacial evaluation of modified Jute and Hemp fibers/polypolypropylene (PP)-maleic anhydride polypropylene copolymers (PP-MAPP) composites using micromechanical technique and nondestructive acoustic emission. *Compos. Sci. Technol.* **2006**, *66*, 2686–2699. [CrossRef]