



Article

Sustainable Sandwich Composites Manufactured from Recycled Carbon Fibers, Flax Fibers/PP Skins, and Recycled PET Core

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Abstract: European union end of life vehicle directive mandates the use of more sustainable/recyclable materials in automotive industries. Thermoplastics matrix-based composites allow recyclability of composites at the end of life; however, their processing technology is more challenging than thermoset composites. Manufacturing process and mechanical testing of sustainable sandwich composite made from sustainable materials: flax, recycled carbon fiber, polypropylene, and recycled PET foam are presented in this article. High pressure compression molding with adhesive thermoplastic polymer film was used for manufacturing sandwich composite skin. The recycled PET foam core was integrated/joined with the skin using a thermoplastics adhesive film. A three-point bending test was conducted to compare the flexural properties. The results show that such sustainable sandwich composites will be an excellent material for truck side panel to operate in adverse wind/storm conditions. The sustainable sandwich composite can potentially be an excellent candidate for the fabrication of light-duty, lightweight, and low-cost engineering structures in automotive industry to meet the EU end of life requirements.

Keywords: polypropylene; recycled carbon fiber; flax; sustainable; mechanical property; sandwich composite



Citation: Jiang, Q.; Chen, G.; Kumar, A.; Mills, A.; Jani, K.; Rajamohan, V.; Venugopal, B.; Rahatekar, S. Sustainable Sandwich Composites Manufactured from Recycled Carbon Fibers, Flax Fibers/PP Skins, and Recycled PET Core. *J. Compos. Sci.* **2021**, *5*, 2. <https://dx.doi.org/10.3390/jcs5010002>

Received: 10 October 2020

Accepted: 20 December 2020

Published: 23 December 2020

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1. Introduction

In the transportation sector, since the number of fossil fuel-powered vehicles projected in this decade is environmentally unsustainable, there is a rapid movement of automotive manufacturers towards hybrid propulsion and electrification to substitute non-renewable sources of energy, that account for approximately 96% of the fuel used for propulsion. The electric vehicles have battery limitations to achieve a long range. Hence, the electric vehicle manufacturers are exploring strategies to increase the driving range of the electric vehicles. One such strategy is to design and develop lightweight sandwich composites made of green alternatives: low-cost natural materials which are promising candidates that could rope in recyclability and sustainability in tandem to the enhancement of performance [1–4].

Recently, several researchers have reported sandwich composites manufactured from natural, renewable or recycled materials [5–19]. These include balsa wood [5–8], hardwood [9], bamboo [10], rubber from waste tires, foams of mushroom, PET, and PVC, and honeycombs of flax/LLDPE and PP. The face skins used in these studies encompassed a combination of synthetic fiber (glass fiber) and natural fiber (jute and flax) reinforced thermoplastics and thermosets.

A substantial fraction of scientific literature on sandwich composites relies on thermoset matrices [5,11–16]. While it is almost impossible to recycle thermosets, fibers embedded in them can be recycled by machining the composite before subjecting them to pyrolysis and chemical treatment to burn off the thermoset resins, the result being a discontinuous fibrous material with degraded mechanical properties [20–24]. Therefore, recyclable thermoplastic resins and natural materials with structural properties comparable to those of glass fibers were utilized in our research to retrofit the component of sustainability in composite manufacturing.

Thermoplastic matrices have been successfully used in the manufacturing of sandwich composites. A sandwich composite manufactured using flax reinforced Elium thermoplastic resin skin and balsa wood core displayed comparable fatigue performance and better interfacial strength than sandwich composites made of glass fiber reinforced polymer (GFRP)/foam core [8]. In another study, Elium was used to impregnate glass fibers to manufacture the spar cap of a wind turbine blade. Their prototype was using recycled glass fibers which showed similar tensile strength and a small 12% stiffness reduction compared to virgin glass fibers composites. However, the recycling of thermoplastic composites can be economically viable only when recycled fibers substitute virgin fibers in the supply chain by virtue of resale of recycled fibers [23].

The thermoplastic sandwich composites show an excellent combination of sustainability/recyclability and high impact resistance. The impact responses of a sustainable sandwich composites comprising of Glass fiber/PP skin with LLDPE honeycomb cores were studied before [17]. Interestingly, these cores were manufactured from thermoplastic material; they were also reinforced by natural fibers to increase the properties of the core section. They reported higher peak loads and energy absorption in continuous fiber reinforced composites and their Finite Element Analysis (FEA) modeling showed a good agreement with their experimental results [18,19]. Materials, such as PP, PE, and the flax fiber-based composites, offer an excellent combination of sustainability and vibrational damping for interior automotive component. Numerical and experimental investigation of two sandwich composites made from a recyclable DIAB Divinyl-cell F90 (PVC) core with different skins (flax/PE and Glass fiber/PP) proved that natural fiber-based composite generated more damping than glass fiber composite, making them suitable for automotive interiors [17–19]. Trucks for food transportation require a good combination of damping and mechanical properties which were satisfied by sandwich made of glass/PP skin and PET foam [25]. Other shock-absorbing cores, such as recycled PET foam (Armacell GR100), made from post-consumer products have been used in the market and could replace virgin PET foams and multi-ply panels [26].

The potential of natural fiber-based thermoplastic composites to be used in low and medium loaded structures have been proved in recent literature. Nevertheless, in order to be used for high-performance applications, hybridization must be adopted [27]. In this context, carbon fibers offer high performance, lightweight, and have decent mechanical properties even after the end of life of their aerospace and automotive structures. Due to the embedded energy and high cost of carbon fibers, it is economically valuable to extract carbon fiber and reuse it for high-performance structures. Hybrid carbon fiber/flax reinforced composites have been researched in the last decade, and, in a review paper, it was concluded that stacking sequences and individual fiber percentages can influence one property positively and another property undesirably [26]. The review highlighted a study that used an autoclave-manufactured recycled carbon fiber/flax composites to demonstrate that the incorporation of flax in a carbon fiber reinforced polymer (CFRP) composite enhanced impact performance and vibrational damping at the cost of a reduction in bending and tensile properties, besides significantly diminishing the overall composite cost. It is a noteworthy observation that the energy-intensive and time-consuming autoclave process with high equipment capital offsets the benefits of using recycled carbon fiber and flax [27].

Therefore, to ameliorate the reduction of bending and tensile properties observed in previous research, fully recyclable sandwich composites from recycled carbon fiber-based thermoplastic (polypropylene) skin and recycled thermoplastic foam core (PET foam) have been formulated in this study. The material combination was aimed at offering excellent mechanical properties, economic viability, light-weighting, lower environmental footprint, and 100% recyclability by virtue of thermoplastic resin and core. Furthermore, skins are hybrid in nature, comprised of external layers of recycled carbon fibers and internal layers of flax fibers. Such carbon-flax-carbon configuration provides better flexural properties than the flax-carbon-flax combination [27]. It will also offer reduced moisture absorption by shielding the flax from moisture ingress. Additionally, a case study has been done regarding the use of such composites as a truck side panel as a potential application in the transport sector. These fully recyclable sandwich composites will find excellent application in automotive truck panels and potentially in the mass transport (railways and bus transport) industries.

2. Materials and Methods

2.1. Materials and Panel Design

A non-woven mat of recycled carbon fiber was supplied by ELG Carbon Fiber Ltd. (ELG Carbon Fiber Ltd., Coseley, West Midlands WV14 8XR, UK; density 1370 kg/m³) The recyclable fiber was obtained by chopping carbon fiber components end of their life. The pyrolysis process was used to remove the thermoset resin. After pyrolysis, the recycled carbon fibers were compressed into randomly oriented fiber mat. Flax fiber (unidirectional fabric; density 1320 kg/m³) were utilized as reinforcements for the composite skin. Polypropylene (1 mm PP sheet; source: SIMONA PP-H natural) with a melting point of 161 °C and recrystallization temperature around 122 °C, elastic modulus of 1.2 GPa, and density of 950 kg/m³ served as the matrix. A recycled 10-mm thick PET foam manufactured from recycled plastic bottles (Armacell Company, Mars St, Oldham OL9 6LY, UK) with a shear modulus of 18 MPa and a density of 100 kg/m³ was used as the core. A polyester-based copolymer adhesive film BEMIS-5256 (Bemis Associates, Units 3, 5 Turnpike Cl, Grantham, UK) with a low glue line temperature (148 °C to 181 °C) was used to enhance the bonding of the sandwich structure.

Two types of samples were manufactured, each with a different skin configuration. As shown in Figure 1, the skins were designed as follows: (1) four layers of recycled carbon fiber and (2) two layers of recycled carbon fibers (RCF) as the outer layers and four layers of flax fibers as the inner layers. Both sample panels were fabricated, at the size of 150 mm × 100 mm, to be able to fit inside the compression mold before being machined to three-point bending specifications.

2.2. Manufacturing of the Sandwich Panel

Compression molding was used to manufacture the sandwich panels in primarily two stages. First, each of the composite skins was compression molded separately (as shown in Figure 1c). All the materials were positioned into the mold according to the designed sequence, wherein PP sheets were inserted between the fiber plies to ensure a complete infiltration of the fibers. A mold temperature of 168 °C, which is slightly above the PP melting point (confirmed based on the DSC results below, Figure 2), was applied to facilitate the flow of molten PP. A pressure of 50 bar (75 kN on sample) was applied after the temperature reached 180 °C and dwelled for about 10 min to consolidate the PP and augment the fiber wet-out. Once the relative position between the mold plates was stabilized, composite skins with constant thickness were obtained. The mold was gradually air quenched (1 °C/min) to 110 °C that allowed the recrystallization of PP (according to the DSC test result, 122 °C, Figure 2). Based on the DSC study of PP samples (Figure 2), the recrystallization temperature was found in the range of 115 °C to 124 °C. Afterwards, the compression molded skins were subjected to rapid water cooling to room temperature.

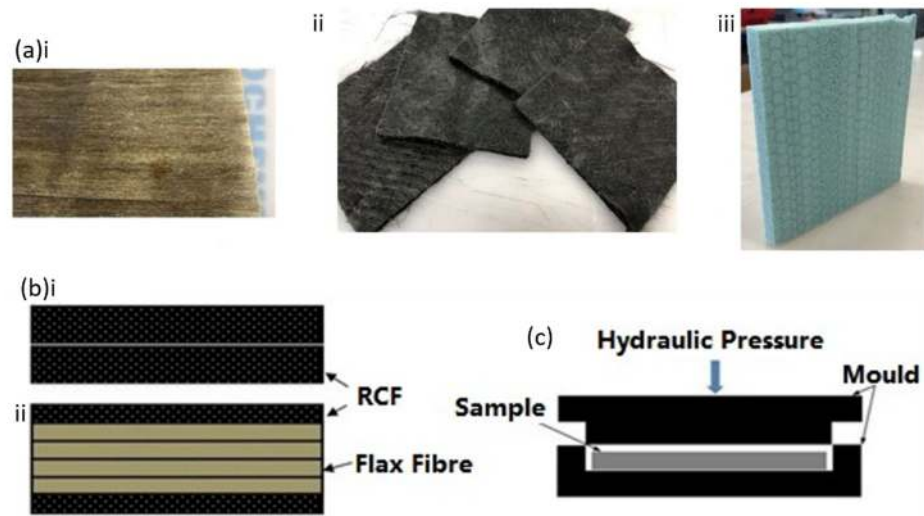


Figure 1. (a) Materials: (i) flax fibers; (ii) recycled carbon fiber mat; (iii) recycled PET foam; (b) skin configurations: (i) recycled carbon fiber (RCF) skin (S1); (ii) hybrid skin of RCF/Flax fibers (S2); (c) compression molding.

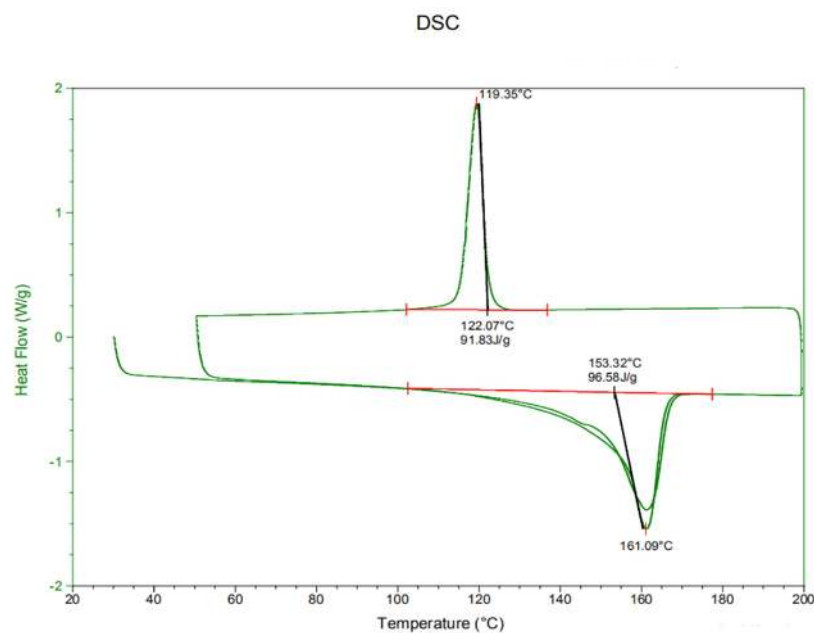


Figure 2. DSC result of Polypropylene via heat-cool-heat cycle.

The second stage was to manufacture the sandwich panel. After producing the skins, they were bonded to the recycled PET foam using 0.1 mm bond line of BEMIS 5256 thermoplastic film. The stack was then placed into the same mold with a temperature set at 160 °C (actual mold temperature at 152 °C) and pressure of 4 bar (6 kN) suggested by the film manufacturer. The pressure was applied after the temperature attained the target value and held constant for 2 min to ensure the bonding of skin and core (mediated by BEMIS-5256 thermoplastic film). After the thermoplastic fusion bonding, the sandwich composite assembly was slowly cooled to 110 °C to allow the recrystallization of PP, followed by water cooling to room temperature. The resultant samples are shown in Figure 3. The fiber volume fraction of both samples was calculated through the weight fraction and the density of the materials and was found to be 20% (s1) and 30% (s2); thus, the skin modulus was also calculated to be 15 GPa (s1) and 19.4 GPa (s2). The thickness of each pure RCF skin was around 2.8 mm, and that of hybrid skin was 3.4 mm.

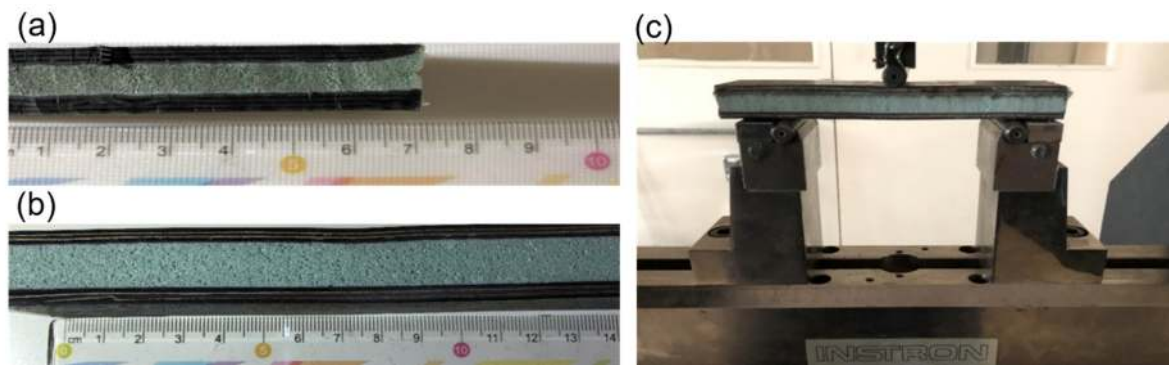


Figure 3. (a) Pure RCF skin sample 1; (b) hybrid skin sample 2; (c) three-point bending test of samples.

2.3. Mechanical Characterization

The sample was machined to three-point bending test specifications. The width was twice the thickness as per ASTM D7249/7250 and ASTM C393 [28–30]. The test was conducted in Instron 5500R machine (Instron, Norwood, MA, USA) with a 5 kN load cell at a speed of 1 mm/min. The sample was placed on a roller support having a span of 100 mm and the load head located in the middle of the sample. A laser extensometer with reflective tapes was used to measure the maximum deflection. In total, six samples were tested, with three samples from each configuration, as shown in Figure 3. The deformation behavior of the sandwich composites was predicted by employing Equations (1)–(3) given below:

$$D = k_s \frac{PS^3}{EI} + k_c \frac{PSc}{bGh^2}, \quad (1)$$

$$EI = \left(\frac{bt^3}{6} + \frac{bth^2}{2} \right) E_s + \frac{bc^3}{12} E_c, \quad (2)$$

$$\sigma = \frac{3PdS}{b(a^3 - c^3)}, \quad (3)$$

where, D is the deflection under load, b is the sample width, t is the face layer thickness, h is the distance between center lines of the upper and lower face skins, c is the core thickness, d is the sample thickness, S is the supporting span, σ is the flexural strength, P is the maximum load applied, EI is the flexural stiffness (rigidity), E_s and E_c are the Young's modulus of skin and core, respectively, k_s and k_c are coefficients of bending and shear, respectively, and G is the core shear modulus [31,32].

These equations were used to calculate the deflection, flexural rigidity, and flexural strength based on suitable assumptions and material properties [31,32], followed by experimentation. The experimental test results were fed into Equation (3) to evaluate the flexural strength. Usually, the modulus of the recycled carbon fiber is considered as 1/4 of the properties of virgin carbon fiber. Hence, it was taken as the same value as flax fiber at 50 GPa. The skin modulus was calculated through the rule of mixtures using the fiber volume fraction. The Young's Modulus of the core was quite small and was neglected during the computation. Thus, the deflection was related to skin flexural rigidity and core shear effect only. The bending factor K_s was 1/48, and shear factor K_c was 1/4, which was suitable for three-point bending criteria (middle load condition) [31,32]. The predicted deflection and the experimental deflection are displayed in the following section. The sandwich modulus was calculated from Equation (4) (same technique as that of the flexural modulus) using the maximum slope of the load versus deflection plot:

$$E_{sandwich} = \frac{PS^3}{4Db(d^3 - c^3)} \quad (4)$$

The composite design module of Ansys was also used to model different configurations of sandwich beams: RCF/PET foam and hybrid/PET foam. For the sake of numerical efficiency and having reasonable computation time, the following assumptions were made: (1) Core is homogeneous and isotropic. This allowed applying shell elements to the model. (2) Perfect bonding is present between core and skins. (3) Skins are stiff enough so that the distortion of cells of the core does not disrupt the skin's in-plane deformation. The stacking sequence of skins used is illustrated in Figure 1b. Properties of skins and cores used in the FEA models can be found in Table 1.

Table 1. Materials properties used for Finite Element Analysis (FEA).

Type of Material	E1 (GPa)	E2 (GPa)	G12 (GPa)	G13 (GPa)	G23 (GPa)	μ_{12}	Density (kg/m ³)
RCF skin	14.96	1.49	0.52	5.72	0.50	0.4	1034
Hybrid skin	21.84	1.70	0.6	8.07	0.61	0.41	1085
PET foam	2.5	2.5	0.018	0.018	0.018	0.43	1380

The models were subjected to two boundary conditions: Simple support at the ends and force at the center of the beams. Three-point bending analysis was performed to obtain deformation responses at different load levels and recorded together for load-displacement plots.

3. Results and Discussion

3.1. SEM Characterization of Bonding Region

The bonding between the skin and the foam core in the sandwich composites is a critical issue in manufacturing. In this study, thermoplastic copolymer film was used to enable fusion bonding between the skin and the core. Figure 4a clearly illustrates three distinct regions: skin (left), adhesive fusion-bonded layer (middle), and the foam core (right), which appear to be bonded seamlessly without any void formation.

Figure 4b illustrates a high resolution and magnified image of the bonding of skin and the thermoplastic film. This image also does not show any indication of finer voids/gaps. Hence, it was inferred that the thermoplastic film is an excellent candidate for thermoplastic fusion bonding the skin and core in a sandwich composite.

3.2. Three-Point Bending Test Results

The load versus displacement plot is illustrated in Figure 5. The blue curve illustrates the experimental data obtained during the three-point bend test (raw test data could be found through the link in Supplementary Materials). The red line is indicative the predicted results from Equations (1) and (2). The grey line with green dots for each data point is FEA analysis result that predicted the deflection with changing load. The experimental results (blue) of both sample 1 and 2 show a small toe region before the curve increases linearly. This is mentioned in ASTM D7250, and the effect is attributed to the samples 'adjusting' themselves to fit into the load condition. A correction was made on the load versus displacement plot to eliminate this effect by inserting the black dotted line. The intersection of this correction and the x -axis was considered as the point of zero deflection. Equation (1) is the Timoshenko beam model that was utilized to predict the deflection within the 'elastic region' before the prediction had intersected with the results at such a point could be considered as where plastic deformation initiated. It provides an ideally prediction very close to the static FEA results with slightly lower gradient than actual results but without attenuation when approaching the intersection point (where failure initiate) that can used to predict the mechanical property (via maximum load taken) of the sandwich composites when a good deflection (estimation of the intersection) assumption is made.

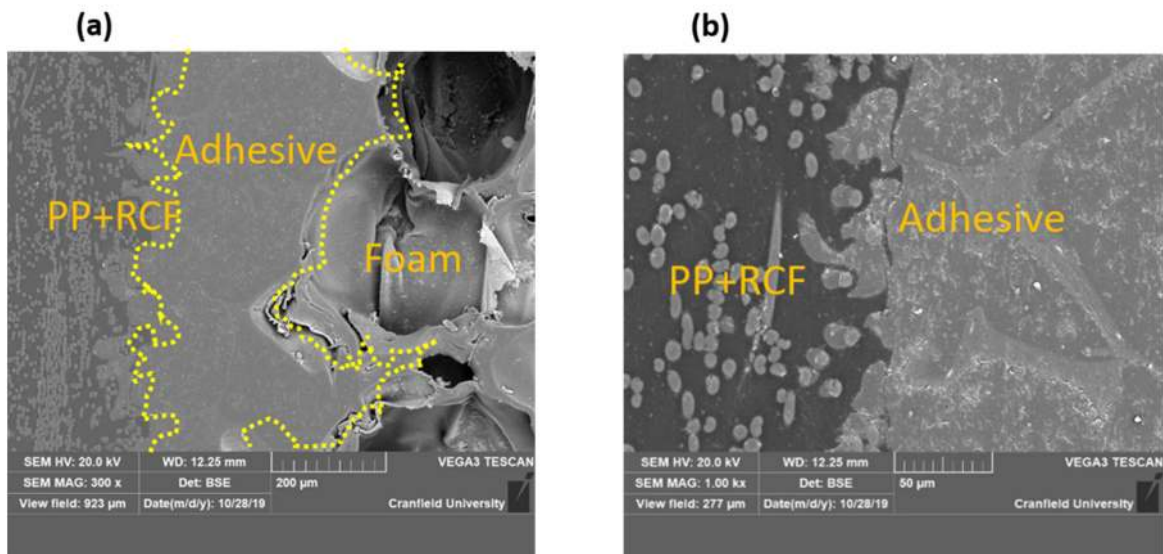


Figure 4. (a) Adhesive region of pure RCF sample. (b) Magnified image.

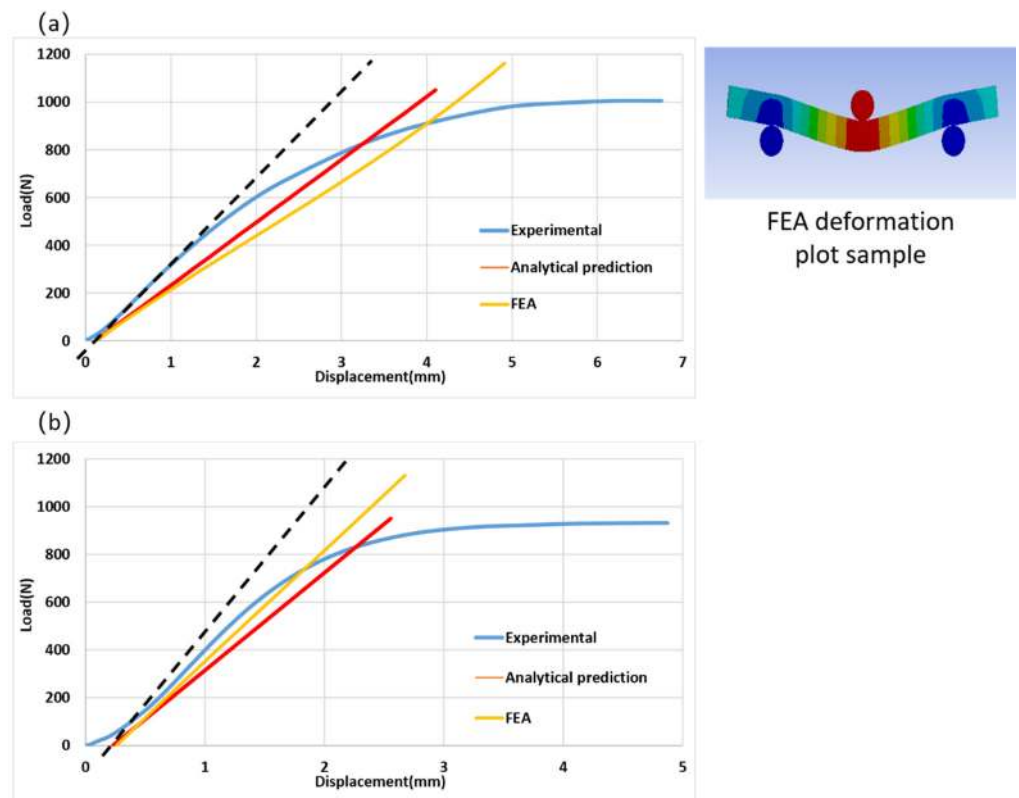


Figure 5. (a) Load versus displacement of sample 1 (RCF); (b) load versus displacement of sample 2 (mixed).

The mechanical properties were averaged by considering the experimental and predictive data and tabulated in Table 2. The flexural strength and sandwich bending modulus were calculated from the average maximum load via Equations (3) and (4). The rigidity was obtained from slope of the linear region of the blue curve in Figure 5 via Equation (1), and the predicted flexural rigidity was estimated by skin modulus and the area moment of inertia.

Table 2. Properties of the sustainable sandwich composites.

Sample Properties	Density (kg/m ³)	Flexural Strength (MPa)	Flexural Rigidity * (Nm ²)	Predicted Flexural Rigidity (Nm ²)	Sandwich Bending Modulus (GPa)
Sample 1 (RCF)	562	76.7 (±2.7)	73.2 (±3.1)	66.4	2.21 (±0.11)
Sample 2 (mix)	551	41.9 (±2.1)	96.2 (±4.8)	119	1.15 (±0.06)

* skin thickness was different for these two configurations.

The skins have different thicknesses (2.8 mm for s1 and 3.4 mm for s2), thus affecting the cross-sectional area largely. While both samples have a similar maximum load (1040 N for s1 and 940N for s2), the actual flexural properties of the mixed skins are much lower than the RCF skins once eliminating the dimension effect, which also reflected in the difference between the calculated and predicted skin flexural rigidity. The RCF skins are about 10% better than prediction, which could come from underestimate of their properties and thus take it into account for the mixed skins; flax could have about 20% properties lower than the estimated value (50 GPa), which might due to the high temperature cycle during the skin manufacturing.

3.3. Failure of the Samples

Core shear failure and skin/core debonding are the most common failure modes in sandwich composites [33,34]. The debonding occurs due to imperfect bonding between skin and core. However, in this study, no debonding between the skin and the core was observed. Instead, shear failure of the foam core was observed in the form of several clefts, as denoted in Figure 6. It indicated that the fusion bonding used in our sandwich composite manufacturing provides sufficient bonding strength between the skin and the foam core, and this method could be an excellent method for bonding sandwich composite.

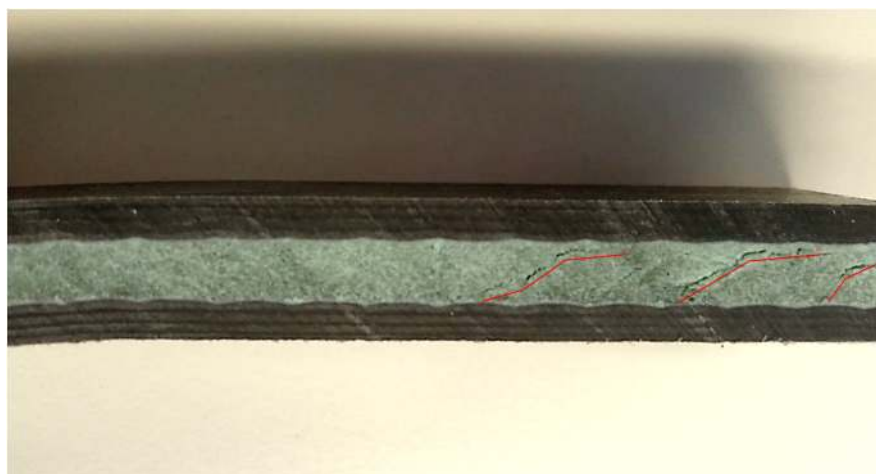


Figure 6. Failure of sample 1 with several clefts.

3.4. Recyclable Sandwich Composites for Truck Side Panels: A Case Study

A case study was conducted to assess the feasibility of employing this sustainable sandwich composite as a side panel of a refrigerated truck (Mercedes Sprinter Luton). The truck schematic can be seen in Figure 7a. Such panels are secondary structures in a refrigerated truck to provide sufficient strength and thermal insulation with a relatively low density. Currently used materials for this purpose are predominantly sandwich composites of GFRP skins and non-renewable/non-recyclable PVC or PU foam core, and some sandwiches also make use of metal skins. The materials currently used in these trucks are not entirely recyclable; thus, sandwich composites obtained in this study shall be conducive in expanding the sustainability.

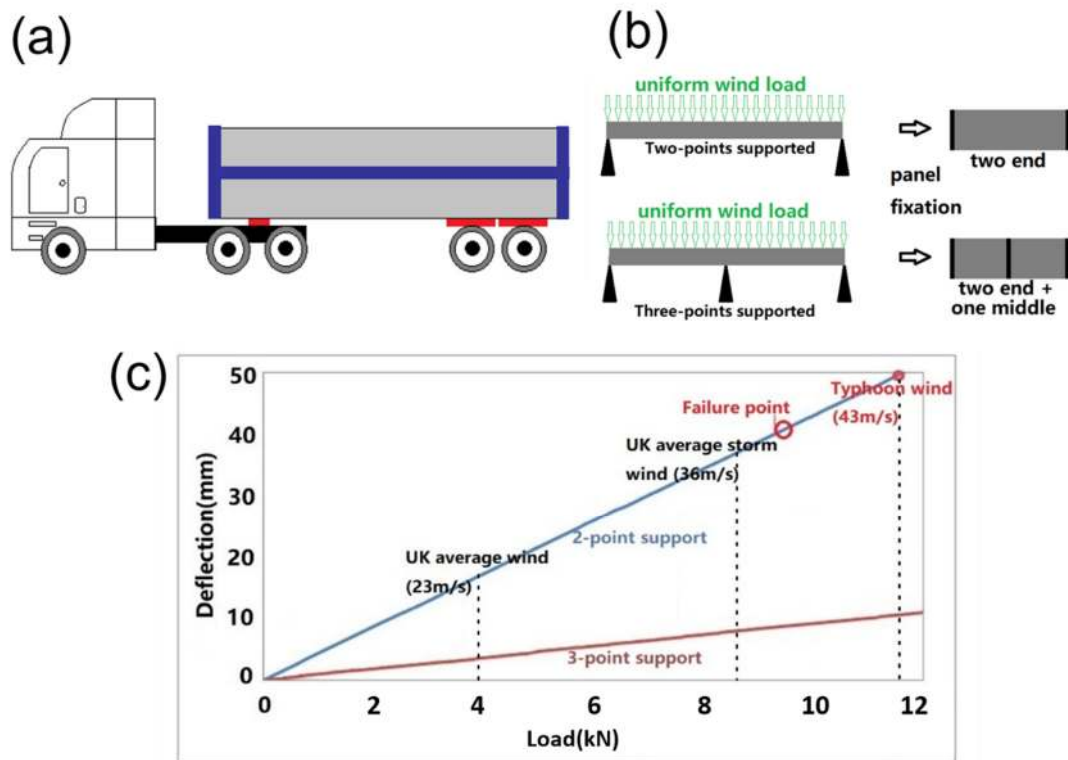


Figure 7. (a) Schematic of a truck with grey side panels and blue supportive frames; (b) wind load and supporting system illustration. (c) Flexural behavior prediction of truck panel with sustainable design.

The truck box size was $4.1 \times 1.9 \times 2.25 \text{ m}^3$ and a truck payload of 1 tonne was assumed. The deflection was predicted from Equation (1) (see Section 2.3, for methodology). Wind loads were estimated to be perpendicular to the side panels and uniformly distributed, as exhibited in Figure 7b. Furthermore, it was assumed that there is no drag on the panels. It can be observed from Figure 5 that experimental deflection of the sandwich composites was 1.5% of the maximum length. Thus, it can be safely deduced that failure would not occur before a deflection of 1% of maximum length.

Two scenarios were taken into consideration, as shown in Figure 7b: (i) The panel is supported by two side edges. (ii) The panel is supported by two side edges and one middle support.

The prediction of the truck side panel behavior under wind can be graphically visualized in Figure 7c. Such a truck panel without middle support could be used in applications exposed to the UK wind weather having an average wind speed of 23 m/s. Besides, the truck panel can survive cornering speeds below 15 m/s. The analysis illustrates that the two-point support (blue line) can also endure the average UK storm speed (36 m/s), shortly beyond which it fails. Moreover, the three-point support (red) offers much less deflection for all wind speeds, even tolerating the wind speed of a typhoon (43 m/s).

CES EduPack software (Granta Design, Cambridge, UK) was utilized to compare the sustainable panel design (pure RCF sandwich) with some popular materials in the market in terms of specific flexural properties and density, as shown in Figure 8. The variables for sandwich panels were controlled at uniform load testing with simple end support; 4 m span length; 5 mm/50 mm/5 mm sandwich design; 35% volume fraction in GFRP skin; and 20% volume fraction in RCF skin. Considering the specific flexural performance of the current design as per the plot in Figure 8, it is evident that the design proposed in this research displays better mechanical properties than most of the candidates in the market.

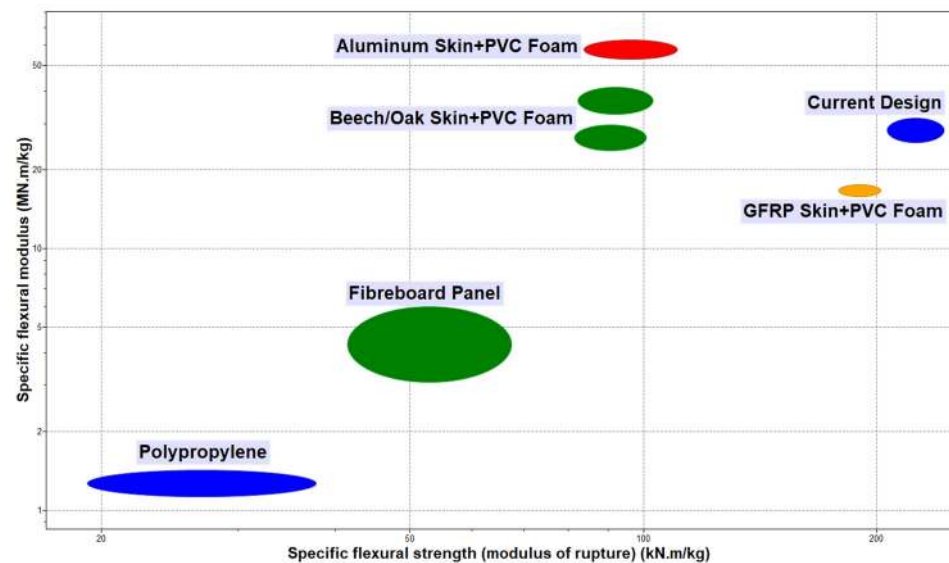


Figure 8. Specific flexural modulus versus specific flexural strength to compare currently used truck panel materials with current sustainable design (source: CES EduPack software).

The design with flax faced two major problems, and the first is the slump in mechanical properties during the high-temperature treatment. Upon comparing the flexural rigidity of the experimental and theoretical investigations, flax lost about 20% of its mechanical property (modulus) after a temperature treatment of 180 °C for 20 min. The second problem is the relatively lower flexural strength of mixed skins. This could be due to the weak affinity of natural fibers with the matrix and its inability to attain 100% performance without surface treatments, as pointed out from previous research. It is also indicated in some studies that high-temperature treatment on natural fibers could reduce their mechanical properties [35,36].

These two problems need to be resolved before the design with flax can be used in potential applications. The manufacturing technique also deserves an upgrade in order to reduce the processing time because more than 70% of the time elapsed in the cooling of the mold to allow PP recrystallization. Alternatively, a stronger thermoplastic could also be selected without the use of natural fibers. The choice of thermoplastics in the sandwich composites makes recycling easier and retains more post-recycling properties when compared to thermoset-based composites [37,38]. The failure mode of the sandwich was a core failure, and skin enhancements would have a limited influence on the overall sandwich properties. In order to have a sizeable increase in the overall properties, a stronger core material with a larger particle size could be utilized. Such cores also involve high-density materials and cause a weight penalty in the final composite. This mandates a trade-off to attain the optimum properties without adding a lot of weight to the sandwich composite.

The recycled PET foam suppliers provide the foams in relatively thin sections (10 mm). The skin thickness used in this work after compression molding was (2.8 mm/3.4 mm). This combination resulted in sandwich composites structure with relatively small core to skin thickness ratio. Such construction is not a common for sandwich composite panels. In future work, we will use thick foams structures from different suppliers to achieve high core to skin thickness ratio.

4. Conclusions

This work was carried out to manufacture a fully recyclable sandwich composites using sustainable materials, like recycled carbon fibers, recycled PET foam, and flax fibers. The sustainable sandwich composites showed very good flexural strength and flexural modulus, as well as superior skin and core bonding. The thermoplastic adhesive film was effective to ensure strong skin core bonding in the sandwich composite. A simple

case study was carried out to show potential use of the composites in truck side panel. The flax fibers showed about 20% reduction in the mechanical properties, potentially due to high-processing temperature required to process high melting point polypropylene matrix. Therefore, future sustainable sandwich designs must compensate for the degradation in the mechanical properties of the sandwich composites. Our sustainable sandwich composites can meet the requirement for secondary structures in engineering applications; further improvements will be needed for its use in high-performance applications.

Supplementary Materials: The Supplementary Materials are available online at <https://www.mdpi.com/2504-477X/5/1/2/s1>.

Author Contributions: Conceptualization, Q.J. and S.R.; Data curation, Q.J. and G.C.; Formal analysis, Q.J. and G.C.; Investigation, Q.J. and G.C.; Methodology, Q.J., G.C., and S.R.; Software, V.R., B.V. and K.J.; Project administration, A.M. and S.R.; Resources, Q.J., G.C., A.K., A.M., K.J., and S.R.; Supervision, A.M. and S.R.; Validation, Q.J. and G.C.; Visualization, Q.J.; Writing—original draft, Q.J. and A.K.; Writing—review & editing, Q.J., A.K., K.J., and S.R. All authors have read and agreed to the published version of the manuscript.

Funding: This research was partial funding by Royal Academy of Engineering, Industrial Academia Partnership Program.

Institutional Review Board Statement: Not applicable, studies not involving human or animals.

Informed Consent Statement: Not applicable, studies not involving human.

Data Availability Statement: Row bending data and one processed file with predictions are available at: <https://data.mendeley.com/datasets/f9vgtm95yb/draft?a=949f5a66-438b-4129-9695-b587dfd28982>.

Acknowledgments: This work is supported by Cranfield University Enhanced Composites and Structures Centre in the UK. Thanks to David Ayre for his help in the DSC testing. The author would like to thank Thibault Hernandez, Jim Hurley, and Ben Hopper for their instruction and help during sample manufacturing and mechanical testing. We would like to acknowledge partial funding from the Royal Academy of Engineering, Industry-Academia Partnership Program for supporting this research work.

Conflicts of Interest: The authors declare no conflict of interest.

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