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Technical Note

Remanufacturing of Woven Carbon Fibre Fabric Production Waste into High Performance Aligned Discontinuous Fibre Composites

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Abstract: The composites industry generates considerable volumes of waste in a wide variety of forms, from the production of by-products to end-of-life parts. This paper focuses on the remanufacturing of dry fibre off-cuts, produced during the composite fabric weaving process, into highly aligned discontinuous fibre prepreg tapes with High Performance Discontinuous Fibre (HiPerDiF) technology. Unidirectional laminate specimens are prepared using various combinations of fibre lengths and tested in tension, obtaining a stiffness of 80 GPa, a strength of 800 MPa, and a failure strain of 1%. Several applications are envisaged for the produced tape: adhesive film, feedstock for filament winding, and tow for weaved fabrics. This work demonstrates the possibility to extract value from what is currently considered manufacturing waste.

Keywords: recycling; aligned discontinuous fibre composites; waste management; sustainability

1. Introduction

The management and utilisation of waste is of paramount importance in the current composite industry scenario, due to the wide variety and volume of both production waste, generated during manufacturing, and post-consumer waste, in the form of end-of-life parts and components. Composite recycling generally comprises a two-step process: fibre reclamation (i.e., its recovery from the matrix), and remanufacturing into a valuable material [1–3].

To be able to retrieve the fibres, the highly reticulated matrix is degraded by subjecting it to elevated temperature (i.e., pyrolysis or chemical reagents). The pyrolysis of thermosetting matrices is typically conducted at between 450 °C and 700 °C in an oven [4,5], in a fluidized bed [6], or with the aid of microwaves [7]. Chemical reclamation methods use reactive, solution-based processes to break down the matrix material (e.g., acid digestion, solvolysis, and supercritical fluid solvolysis). Acid digestion uses a variety of highly corrosive reactant systems (e.g., nitric acid [8,9], sulphuric acid [10], acetic acid [11,12], and some alkalis [13]), to degrade the thermoplastic matrix at low temperatures (<150 °C). Solvolysis methods use less hazardous solvents and catalysts, but suitable reactivity is achieved by operating at higher temperatures (200–300 °C) to afford a suitable reactivity for a specific polymer [14]. Supercritical solvolysis methods employ higher temperatures (350–440 °C) and pressures (20–30 MPa) to increase reaction rates and degradation efficiencies, and provide reactivity in non-hazardous solvents without catalysts [15–17]. Optimised fibre reclamation methods do not alter the stiffness of the fibres and cause a minimal loss of strength; however, these methods usually cause the loss of fibre sizing, the formation of surface residues, and changes in fibre surface chemistry that compromise fibre/matrix adhesion [1–3]. Reclaimed carbon fibres (rCF) are generally available in a filamentised, randomly oriented, low-density-packing (fluffy) form. A remanufacturing process, able to produce high performance recycled composite, should ideally be able to realign them into a high fibre volume fraction composite.

While there are efficient industrial scale setups for fibre reclamation [18–21], remanufacturing methods capable of producing high performance recycled composites are still rare. The most immediate way to remanufacture rCF is to use indirect moulding techniques, such as injection moulding, bulk moulding compound (BMC), and Sheet Moulding Compound (SMC) compression. The literature indicates that the mechanical performance of BMC/SMC, made using rCF, is higher than those available for SMC made using glass fibres [3,22] and comparable properties to virgin carbon fibres have been reported in the literature [3]. Discontinuous and long rCF can be processed into nonwoven mats through processes analogous to wet paper making [23,24], GF chopped strand mat production [25], or carding, cross-lapping, and needling, and can be subsequently impregnated with virgin matrix in liquid [26] or film [27] formats. Nonwoven fabrics can also be used as intermediate materials to obtain, through a spinning process, yarns or sleeves, as demonstrated in the processes developed by Miyake et al. [28], Cherif et al. [29–31] and by the Institut für Verbundwerkstoffe [32,33]. Finally, techniques for the alignment of short rCF are available, (e.g., the centrifugal fibre alignment drum [22] developed at the University of Nottingham and the Tailorable Feedstock for Forming (TUFF) developed by the University of Delaware [34–36]). One of the technologies that has the potential for a commercial scale set-up is the HiPerDiF (High Performance Discontinuous Fibre) method [37].

The HiPerDiF method, shown in Figure 1, is a patented manufacturing process, invented at the University of Bristol, to produce highly aligned discontinuous fibre tapes [38]. The construction of the third generation of the HiPerDiF machine, funded by an EPSRC project (EP/P027393/1) and capable of high throughput (kg/hour quantities of prepreg) with full instrumentation, is currently nearing completion in the National Composites Centre (NCC) in Bristol.

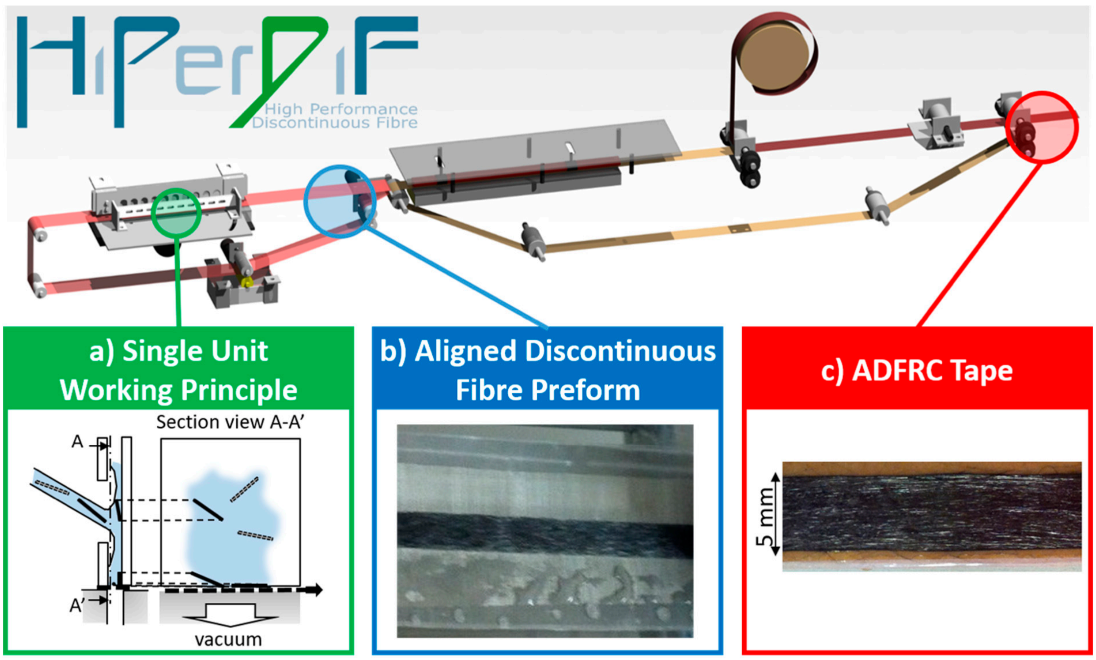

Figure 1. Schematic of the HiPerDiF alignment process.

Discontinuous fibres, with lengths between 1 and 15 mm, are dispersed in a low viscosity aqueous carrier [39] and sprayed through a nozzle towards an array of thinly spaced parallel plates, as shown in

Figure 1a. The momentum change induced by the impact of the fibres on the furthermost plate causes them to align parallel to the direction of the gap between the plates. The suspension falls onto a moving conveyer mesh belt where the water is extracted by suction without affecting the fibre alignment, shown in Figure 1b. The fibres are then dried completely with the aid of infrared heating and coupled with a resin film, with the aid of heat and pressure, to obtain the prepreg, shown in Figure 1c.

The HiPerDiF method has been used to remanufacture a variety of reclaimed fibres into high performance recycled composites, including those based on different carbon fibres, glass fibres [40], and natural fibres [27]. Moreover, the HiPerDiF method is key to the closed-loop recycling of composite materials with both thermosetting [41] and thermoplastic [42,43] matrices. Furthermore, the HiPerDiF technology can accommodate a variety of feedstocks, capable of simultaneously processing fibres of different lengths that have been reclaimed using different processes, as demonstrated through a quality control and property assurance methodology developed in house [44].

With their commitment to promote a more sustainable and zero-waste composites industry, Microtex Composites [45] supported a University of Bristol Masters student's research project aimed at developing efficient remanufacturing and reuse strategies for the dry fibre off-cuts produced during the composite fabric weaving process. The objectives were the evaluation of the possibility to remanufacture those off-cuts into a highly aligned discontinuous fibre prepreg tape with the HiPerDiF technology, and the characterisation of the mechanical behaviour of the resulting composite.

2. Experimental Work

The weaving process employed by Microtex Composites SRL generates a small volume of dry carbon fibre wastes when the tows, which are stitched at their extremities to help the weaving/looming process, are trimmed to remove the stitching yarn; i.e., the white thread shown in Figure 2.

Figure 2. Carbon fibre waste produced from the stitching process.

In this study, 24k MR60H carbon fibres (manufactured by Mitsubishi Rayon Co., Ltd., Tokyo, Japan) were used. According to the manufacturer's data sheet, the fibres (5 μm diameter, density of 1.81 g/cm^3) were sized for epoxy-based resin systems and had a stiffness of 290 GPa, a strength of 5.68 GPa, and an elongation to failure of 1.9%.

As the received fibres had an average length of 60 mm, to make them compatible with the HiPerDiF process, they were comminuted, after manually removing the stitch yarns, using an in-house rotating blade system, in three different nominal lengths—3 mm, 4.5 mm, and 6 mm. The real fibre length distribution, measured through microscopy image analysis, is shown in Figure 3.

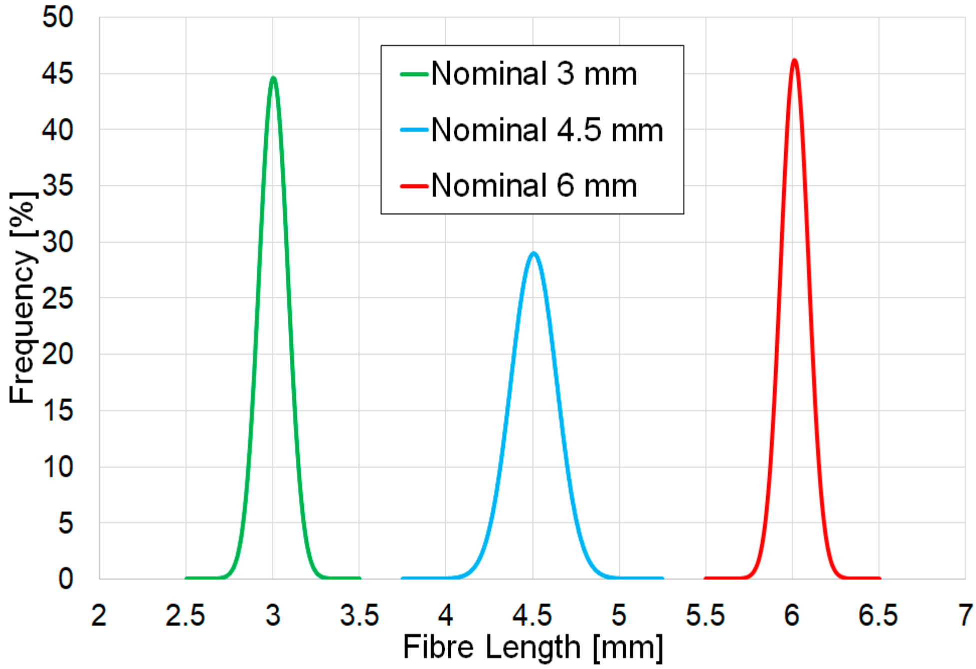

Figure 3. Fibre length distribution.

It is foreseen that, in an industrial environment, it will not be possible to cut the fibres with the accuracy attainable under controlled laboratory conditions; therefore, testing different fibre lengths and distributions allows us to evaluate the effect of fibre length on the material performance. The HiPerDiF machine was used to manufacture five sets of six specimens: three sets with 100% 3, 4.5, and 6 mm long fibres, respectively; one set with 50% of 3 mm and 50% of 6 mm long fibres; one set with 30% of 3 mm, 30% of 6 mm, and 40% of 4.5 mm long fibres. The dry fibre preforms were coupled with a 43 gsm areal weight Skyflex K51 resin film (SKChemicals, Seoul, Republic of Korea), a proprietorially-toughened, bisphenol A-based epoxy resin with a density of 1.21 g/cm^3.

The prepreg tapes obtained from the HiPerDiF process were then laminated in the semi-closed mould and cured in autoclave at 7 bar pressure and a maximum temperature of 135°C for 135 minutes to obtain the specimen with nominal dimensions (length × width × thickness) of 150 mm × 5 mm × 0.3 mm. The fibre volume fraction (V_f) was calculated, through resin burn-off and cross-section image processing, to be 41 ± 2%.

After curing, the specimens were removed from the mould and burrs removed from all edges. Glass fibre/epoxy end-tabs, with a length of 50 mm, were bonded, using Araldite2014 rubber toughened epoxy adhesives (Huntsman, The Woodlands, TX, USA), to all the specimens, leaving a gauge length of 50 mm.

Tensile tests were performed on a servo-electric test machine equipped with a 10 kN load cell (Shimadzu, Japan) at a cross-head displacement speed of 1 mm/min, following ASTM guidelines [46]. The strain was measured with a video extensometer (IMETRUM, Bristol, UK). White dots were applied over a black background allowing for three independent measurements across the specimen with a gauge length of 45 ± 1 mm.

3. Results and Discussion

The representative stress-strain curves, obtained from the tensile test of all of the specimens, are shown in Figure 4.

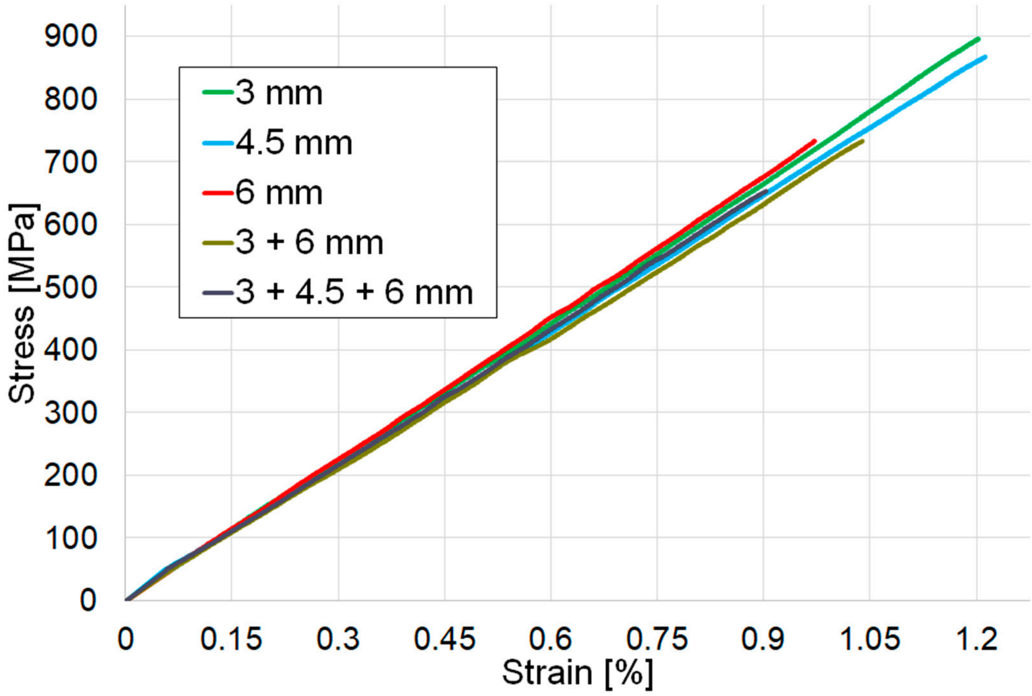

Figure 4. Representative tensile stress-strain curves

All the specimens showed linear elastic tensile behaviour and a brittle failure. The calculated stiffness and failure properties (i.e., strength and failure strain), are summarised in Table 1 and shown in Figure 5.

Table 1. Material property summary.

Fibre Length	Stiffness [GPa]	Strength [MPa]	Failure Strain [%]
3	70 ± 5	841 ± 39	1.19 ± 0.03
3 + 6	66 ± 3	706 ± 42	1.07 ± 0.07
4.5	69 ± 4	769 ± 85	1.11 ± 0.07
3 + 4.5 + 6	70 ± 4	723 ± 74	1.04 ± 0.09
6	81 ± 5	783 ± 102	0.97 ± 0.09

The obtained stiffness values are in line with the theoretical prediction based on the volume fractions of the constituents and the correction factors required to account for fibre misalignment and load transfer efficiency, as explained in [38]. Observing Figure 5a, it is clear that in all but one case, there is no difference between the specimen sets, which is in agreement with the results obtained in [44]. The only sample that displays a statistical difference from the others, using the Kruskal–Wallis one-way analysis of variance, contains 100% 6 mm long fibres. This can be attributed to two factors: The first is that longer fibres allow for a better load transfer [47]. The second factor is manufacturing-related—the spacing between the two parallel plates is maintained while manufacturing all the specimen sets. However, as demonstrated by Huntley et al., through Smoothed Particle Hydrodynamic (SPH) simulation, it is known that a reduction in the plate gap to fibre length ratio improves the fibre alignment level and, therefore, the ultimate mechanical performance of the composite material [48,49].

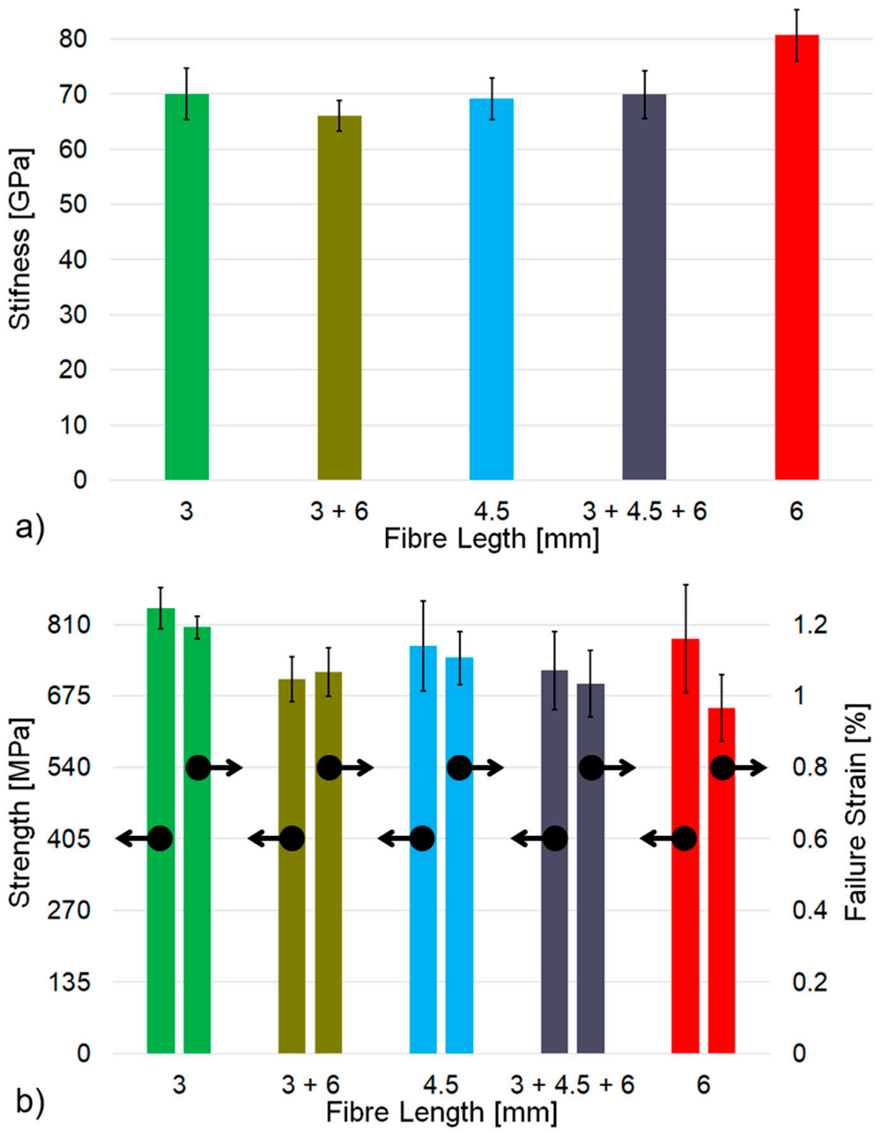

Figure 5. Material property summary: (**a**) Tensile modulus; (**b**) Tensile strength and Failure Strain.

In terms of failure properties (i.e., tensile strength and failure strain), the obtained results are in line with those of previous works conducted on the remanufacturing of reclaimed carbon fibres with the HiPerDiF technology [40]. As observed in [44], there is no direct connection between fibre length and specimen failure, as shown in Figure 5b. Moreover, after performing a Kruskal–Wallis one-way analysis of variance over the whole specimens' populations, it is interesting to observe that there are no statistically significant differences between the specimens with a 4.5 mm average length.

4. Conclusions

Given the mechanical properties that may be attained (i.e., tensile modulus of 80 GPa, tensile strength of 800 MPa, and failure strain of 1%), the recycled material, obtained by remanufacturing the weaving manufacturing waste into a highly aligned discontinuous fibre composite, demonstrates tangible industrial potential. From the perspective of processing, using 6 mm fibres makes it possible to reduce the pre-processing needed to make the waste processable through the HiPerDiF technology.

The third generation of the HiPerDiF machine will be able to produce continuous prepreg tapes, with widths from a $\frac{1}{4}$ to 2 inches (6.35 mm to 50.8 mm), at a production rate of kilogrammes per hour. We envisage that these prepreg tapes could be used directly as reinforced adhesive films to guarantee a constant thickness in the bonding area. Moreover, the ADFRC tape could be a valuable feedstock for filament winding operations (e.g., to produce pressure vessels, bicycles, and other tubular frames and components). Another very interesting potential application is in weaving the tape, which would require pre-impregnation either with a thermoplastic or a thermosetting matrix with no tack at room temperature, into a fabric. Given the low areal weight of the prepreg tape, the weaving process would generate extremely shallow undulations during weaving and forming, and the produced material would behave as a spread fabric, allowing the mechanical performance of the manufactured component to be maximal.

In conclusion, this work demonstrates the possibility to extract value from what is currently considered manufacturing waste, and, through the combined efforts of academia and industry, to implement a circular economy model and achieve the more sustainable production of advanced composites.

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

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