Resource-friendly carbon fiber composites: combining production waste with virgin feedstock

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Abstract Reclaimed carbon fiber materials were studied in this paper with the aim of improving virgin fiber feedstock usage. Both processing and mechanical properties were investigated. The compaction response showed lower fiber volume fractions in reclaimed fiber materials than the virgin continuous reinforcement from which it was reclaimed. In addition, localized high-strain regions were observed during consolidation of the dry fiber and mechanical loading of cured laminates. These vulnerable failure points were mitigated by incorporating virgin continuous fiber feedstock into the laminate. A knockdown in mechanical properties was observed, however classical laminated plate theory identified a planar stiffness drop of 3.5 GPa for every 10% increase in reclaimed carbon fiber content in a continuous fiber laminate. Increased feedstock usage by combining both virgin and reclaimed carbon fibers was shown to be a viable option to implement more resource efficient, but heavier, composite structures.

Keywords Composite recycling, Sustainable manufacture, Polymer matrix composites, Liquid composite molding, Laminate analysis, Mechanical testing

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Introduction

A substantial new market for composite materials is emerging in the automotive sector as vehicle manufacturers look to reduce the environmental footprint of their cars through light-weighting. Carbon fiber-reinforced plastics (CFRP) are a viable option to replace metals due to their high stiffness-and strength-to-weight ratios, and successfully used in a number of high-performance industries, such as aerospace and motorsport. Thermosetting polymer matrices are often used in these applications because of their advantageous mechanical/physical properties and ease of processing. However, due to the cross-linked network of thermoset polymers, end-of-life components cannot simply be melted and recycled/reformed.

Production scrap presents an opportunity for the composites sector to do more with existing materials. If production waste can be minimized or re-used, the resource and material impact of composite components can be improved. Granted, manufacturing waste volumes are fabrication method and sector dependent, with feedstock waste ranging from 7 to 46% in R&D enterprises, and large aerospace companies estimating 30 to 50% of production materials become scrap. Landfilling this high-value resource as waste is both damaging to the environment and costly. Disposing of composite waste by landfilling can cost approximately £0.20 per kg and virgin carbon fiber production is estimated to be the single largest contributor to component cost. Recycling carbon fiber leads to a potential impact reduction in climate change by 78%, resources by 84%, ecosystem quality by 82%, and human health factors by 65%.

In-light of the clear economic and environmental benefits of recycling composite materials, significant research efforts have developed processes to recover in-process scrap and end-of-life CFRP components. The recycling technologies remove the matrix from the collected waste using a variety of processes, including mechanical grinding, thermal pyrolysis – such as the popular fluidized bed oxidation process, and chemical solvolysis. After removing the matrix using certain processes, the recovered fibers retain their strength and stiffness at a filament level. However, fibers reclaimed from dry manufacturing waste are even closer to their virgin feedstock and do not require a matrix removal step.

Re-manufacturing the recovered fibers into composites laminates presents a unique set of challenges compared to the virgin feedstock. The reclaimed products have lower mechanical properties, with reported strength reductions up...
to 85% and stiffness reductions up to 45%. The reduction in properties at the laminate level arises from the loss of alignment of reprocessed short fibers, larger pieces of reclaimed product acting as a failure initiation site, and low fiber volume fractions. Random fiber mats experience less fiber nesting during consolidation due to less alignment within the fiber bed. Techniques to align short fibers to a much higher degree than conventional methods have been proposed, and until the recycled product is improved, it will be limited to non-critical applications.

A current barrier to adopting reclaimed fiber composites is the lack of an established market for the recycled product. Reclaimed materials suffer from poor fiber alignment and unacceptably low fiber volume contents required for structural components. The aim of this study was to investigate the potential to incorporate a commercially available reclaimed carbon fiber material into structural composites. The mechanical properties and re-manufacturing of virgin and reclaimed carbon fiber products were investigated. A resource efficient composite laminate where the waste feedstock is incorporated back into the parent waste generating virgin fiber laminate is proposed to continue structural light-weighting with high-value carbon fiber.

**Experimental methods**

**Raw materials**

Commercially available virgin and reclaimed carbon fiber materials were investigated in this study. Type 62 RECATEX non-woven complex with an areal density of 200 g/m² from SGL Automotive Carbon Fibers (ACF) was used as the reclaimed product. This mat is created from dry fiber production waste that is shredded, carded, and then sewn to improve product handling using similar process to non-crimp fabric materials. A picture of the reclaimed fiber mat is shown in Fig. 1. A preferred fiber direction parallel to the sewing yarn is visible, but fibers are oriented in multiple directions at various lengths and sewing yarns from the original NCF is also present. Reported anisotropy in the reclaimed fiber material is 2.4 times higher modulus and 2.3 times higher strength parallel to the sewing direction. A virgin fiber counterpart, also from SGL ACF, SIGRATEX C U320-0/ST was supplied as a unidirectional non-crimp fabric (NCF) with an areal density of 320 g/m², and is shown alongside the reclaimed fiber mat in Fig. 1. The NCF material included a powder binder.

**Test matrix**

An experimental campaign was performed to measure the compaction response of the dry reinforcements and the mechanical properties of the resulting composites. The test matrix is presented in Table 1. Plies of reclaimed carbon fiber were designated by an R before the material direction – where 0° is the roll direction. Hybrid laminates of reclaimed mat and virgin continuous material were also investigated. The total fiber volume fraction of the composite laminates was measured by acid digestion or pyrolysis, and the density of the fiber is required for these calculations. The density of the virgin carbon fiber was taken as 1.8 g/cm³ from the material datasheet. The reclaimed fiber mat is reportedly composed of 75% carbon fiber, 11% glass fiber, 11% polyester fiber, and 3% binder. The exact type of glass and polyester fiber were unknown therefore density values were assumed to be 2.5 g/cm³ and 1.3 g/cm³, respectively. The binder was unknown, however assuming a low molecular weight polymer with a 1.2 g/cm³ density yields a representative density of 1.7 g/cm³ for the reclaimed carbon fiber mat. The resulting fiber volume fraction is then calculated as follows:

![Figure 1 Reclaimed carbon fiber mat (left) and unidirectional carbon fiber non-crimp fabric (right)](image)

![Table 1 Lay-up sequences and laminates investigated in this study](chart)
where $m$ is the measured mass of the lay-up; $h$ is the lay-up thickness; and $A$ is the surface area.

Consolidation measurements
The compaction response of the reinforcement relates the mold closure forces needed during manufacture to achieve the desired fiber volume fraction in the final composite article. The compaction response is typically measured between two flat plates by recording the normal force and displacement of the moving plate. Each 100 mm × 100 mm sample was loaded up to 5 MPa in a 100 kN electromechanical Zwick test machine at a compaction rate of 1 mm/min to avoid any fiber relaxation. The applied load was measured using the load indicator of the test machine; the displacement was an average measurement from a pair of Micro-Epsilon ILD1700-100 laser-optical displacement sensors. Figure 2 shows the compaction measurement fixture.

In order to investigate the distribution of the applied pressure, a number of compaction experiments were conducted with a Tekscan 7501 pressure mapping sensor, as shown in Fig. 2. The sensor is formed of an array of capacitive pressure sensors spaced at 1 mm × 1 mm over an area of 88.1 mm × 100.1 mm. The sensor was taped to the fixture base plate and the sensor thickness was subtracted from the height when calculating specimen thickness according to Eq. (1).

Sample manufacture
Composite laminates were manufactured using the reclaimed mat and virgin unidirectional carbon fiber materials. Laminates were made at a variety of pressures to achieve different fiber volume fractions using liquid composite molding techniques.

Mechanical property evaluation
Tensile and compressive properties of virgin, reclaimed, and hybrid laminates were measured to capture the 1–1 and 2–2 properties of the materials.

Tension
The tensile tests were performed in accordance with the ASTM D3039-14 standard using a 100 kN Zwick test machine with mechanical wedge grips. A Dantec Digital Image Correlation (DIC) system was used to capture the surface strain distribution during testing. Test specimens were cut using a waterjet. Virgin carbon fiber specimens had the rectangular 25 mm × 250 mm dimensions recommended in the ASTM D3039-14 test standard, as shown in Fig. 3(a).
Rectangular reclaimed fiber material coupons failed in the grip section, therefore a waisted width geometry specimen was used to ensure the specimen failed in the gauge section. The ASTM D638-14 test standard for unfilled plastics was adapted to have the same 25 mm wide gauge section as the continuous fiber laminates, as shown in Fig. 3(b). The waisted geometry had a rectangular 25 mm x 75 mm rectangular section where sample statin was measured. The waisted specimen alleviated the stress concentrations observed in the grip section and, the reclaimed fiber specimens failed in the constant cross-section section (see Fig. 3(b)).

Compression

Compression tests were performed according to the ASTM D695 test standard with an Imperial College of Science, Technology and Medicine (ICSTM) test fixture. Compressive UD test coupon dimensions were 30 mm x 115 mm. Compressive tests were performed using a 250kN Instron test machine. An Imetrum video gauge system was used to capture the compressive strain during testing.

Laminated plate analysis of hybrid reclaimed-virgin fiber composites

A model to describe the mechanical properties of a hybrid reclaimed-virgin fiber laminate was created using classical laminated plate theory (CLPT). The model was written in MATLAB using the mechanical property inputs shown in Table 2. Most of the material properties were measured through this study; however, the in-plane shear properties of the virgin carbon fiber were acquired from Ref. 26, who used the exact same material as in this study. The in-plane shear properties of the reclaimed carbon mat were obtained from Ref. 27, who tested a discontinuous carbon fiber preform with 6 mm fiber length and similar fiber volume fraction to the reclaimed fiber mat used in this study.

Conventional CLPT models consider each ply to be of equal thickness and have equivalent primary material properties. In the case of hybrid reclaimed-virgin materials, the ply orientation and material type for every layer was defined by the user through a graphical user interface. The model outputs membrane and bending elastic constants calculated by classical laminate stiffness relations. The laminate strength was calculated using the last-ply-failure method, with complete-ply-failure for plies with the highest failure index at every iteration.

Results and discussion

Fiberbed consolidation

The measured compaction response of the fiber materials are shown up to 3 MPa in Fig. 4. All curves exhibit the typical curvature in fiber volume fraction at lower applied pressures, eventually becoming linear as the fibers nest. The noteworthy observation is that the fiber volume fraction of the reclaimed

| Property                        | Units   | Virgin (νf = 54.6%) | Coefficient of variation (%) | Reclaimed (νf = 13.5%) | Coefficient of variation (%) |
|---------------------------------|---------|---------------------|-----------------------------|------------------------|-----------------------------|
| Tensile strength 1–1, σT11      | MPa     | 1568                | 5.5                         | 185                    | 9.8                         |
| Tensile strength 2–2, σT22      | MPa     | 36.8                | 4.8                         | 98.8                   | 6.9                         |
| Compressive strength 1–1, σC11  | MPa     | 813a                | –                           | 207                    | 3.6                         |
| Compressive strength 2–2, σC22  | MPa     | 160                 | 10                          | 143                    | 12                          |
| Young's modulus 1–1, E11        | GPa     | 119                 | 7.8                         | 17.4                   | 20                          |
| Young's modulus 2–2, E22        | GPa     | 7.42                | 3.3                         | 9.21                   | 14                          |
| Shear strength, τ12             | MPa     | 76.3a               | 1.2                         | 204b                   | 20                          |
| Shear modulus, G12              | GPa     | 2.41a               | 6.9                         | 12.5b                  | 12                          |
| Poisson ratio, ν12              | –       | 0.27                | 7.0                         | 0.39                   | 2.0                         |
| Cured ply thickness             | mm      | 0.37                | 5.4                         | 0.73                   | 6.7                         |

*Material properties from Blok.
*Material properties from Kirupanantham.
fiber mat is less than half of the virgin continuous unidirectional material. The highly aligned fibers that compacted to 65% fiber volume fraction at 1 MPa can only compact to 25% fiber volume fraction after being reclaimed. The hybrid laminate shown in Fig. 4 had a lay-up of [0/R0/0] and a resulting 64% reclaimed carbon fiber content by mass. Accordingly the hybrid fiberbed compaction response is closer to the fully reclaimed fiber mat.

Surface pressure mapping

Figure 5 shows representative pressure distributions from the pressure mapping sensor. The data are displayed as 2D plots for each specimen configuration at average applied pressures of 0.25, 0.5, and 0.75 MPa.

All samples show some variation in the normal pressure distribution during compaction. The reclaimed carbon fiber mat sample showed two areas of higher and lower pressure readings. The virgin unidirectional carbon fiber sample has regular identifiable pressure concentrations along the fiber tows. The hybrid specimen exhibits pressure distribution patterns from both the reclaimed fiber mat and the virgin continuous fiber samples; areas of higher and lower pressure can also be seen in the hybrid sample plot but is less striking than the reclaimed lay-ups.
Mechanical properties
The measured mechanical properties are presented in Table 2. As expected, the strength and modulus in the primary 1–1 direction is far superior in the virgin continuous carbon fiber laminate; however, the strength in the 2–2 direction of the reclaimed carbon fiber mat is greater than the virgin material and the stiffness is comparable. These property relationships will be explored using the CLPT model to evaluate how the reclaimed mat can be incorporated into laminates.

Digital image correlation
The strain distribution during tensile testing was captured using DIC. Figure 6 shows representative engineering tangential strain measurements in the loading direction for two specimens from each laminate type. The strain contours are shown at an average strain of 0.5% in order to analyze the strain variations at a constant specimen elongation. The images shown in Fig. 6 were post-processed with a 9 × 9 local regression smoothing.

The DIC analysis revealed that the reclaimed carbon fiber mat specimens show significantly higher strain variations than the virgin unidirectional fiber laminate. The additional strain variations could be caused by variations in resin or fiber rich regions of the reclaimed fiber materials (as seen in Fig. 5). Increasing the fiber volume fraction of the reclaimed fiber composite reduced the strain variation, as shown in Fig. 6. The highly oriented continuous fibers in unidirectional materials also reduced the variations in laminate strain hybridizing the reclaimed fiber mat with virgin continuous fibers, as shown in Fig. 7. The hybrid laminate of reclaimed carbon fiber mat and virgin unidirectional fiber at 45% fiber volume fraction seems to offer equivalent strain distributions to the virgin unidirectional reference.

Property calculations
The CLPT model was compared to tensile experiments of virgin fiber quasi-isotropic and hybrid reclaimed-virgin fiber laminates. The results shown in Table 3 indicate the CLPT model can predict the modulus very well for the hybrid and virgin fiber laminates, with strength predictions showing higher disparity.

The error in the virgin fiber quasi-isotropic laminate arises from the failure criteria. In the complete-ply-failure analysis process, when a ply fails the stiffness contribution of the ply is completely nullified. In reality, once a ply fails, its stiffness contribution to the laminate reduces but is not fully eliminated. Therefore, the complete-ply-failure method creates a skewed redistribution of ply stresses after a ply failure in the laminate. Once the first ply has failed, the remaining intact plies are

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Effect of reclaimed fiber content on mechanical properties

The CLPT model was used to design a quasi-isotropic (QI) hybrid laminate using reclaimed fiber mats and continuous virgin fiber plies. The strength and modulus of the resulting laminates are shown in Fig. 9. The model shows the reduction in mechanical properties as a function of increased reclaimed fiber content, with a fully reclaimed carbon fiber mat laminate having 25% of the performance of a fully virgin continuous fiber quasi-isotropic laminate.

The CLPT model suggests that the modulus of a hybrid reclaimed-virgin fiber laminate decreases in a linear manner with each increment in reclaimed fiber content. Using lines of best fit to approximate the performance of a hybrid reclaimed-virgin fiber laminate, every 10% increment in reclaimed fiber content decreased the laminate modulus by approximately 3.5 GPa. The laminate strength also decreases, but in a non-linear manner. Laminates with reclaimed fiber content between 10 and 50% decrease by approximately...
a Reclaimed fiber mat plies are designated by an R before the orientation.

Table 4  Quasi-isotropic lay-ups to increase feedstock usage

| Feedstock usage (%) | Lay-up* | Virgin fiber (%) | Reclaimed fiber (%) |
|---------------------|---------|------------------|---------------------|
| 70                  | [45/0/−45/90] | 100              | 0                   |
| 77                  | [90/0/45/−45/90] | 70              | 30                  |
| 86                  | [90/45/0/45/−45/90] | 48              | 52                  |
| 100                 | [45/0/45/−45/90/R0/−R45/R90] | 32            | 68                  |

40 MPa in strength for every 10% increment in reclaimed fiber content. As the reclaimed fiber content increases above 60%, the hybrid laminate strength decreases by roughly 20 MPa with every 10% increment in reclaimed carbon fiber content.

Quasi-isotropic resource friendly carbon fiber composites

Conclusions

The consolidation and mechanical properties of a commercially available reclaimed carbon fiber mat were investigated. The fiber volume fraction of the reclaimed carbon fiber mat reached a maximum of 35% at 3 MPa, whereas the virgin material would compact to 70% at that pressure. More importantly, at low pressures used for cost-effective manufacture, near 0.1 MPa, the fiber volume fraction of the virgin reinforcement is 55%, compared to 14% for the reclaimed material. Another key difference was the non-uniform compaction response that was observed, leading to weak spots in the reclaimed fiber material during loading.

The low fiber volume fraction and non-uniform load carrying capacity of the reclaimed fiber was overcome by increasing the compaction pressure. The reclaimed carbon fiber mat was consolidated above 2 MPa to increase the fiber volume fraction towards 30%. At this level, reclaimed production waste can be manufactured with equivalent specific strength and stiffness properties to virgin glass fiber composites. This represents a potential market for recycled fiber materials. However, these compaction pressures would require strong/expensive presses for larger components.

An alternative approach to using reclaimed fiber materials was found by blending them with the virgin plies from which they are generated. In the current landscape seeking solutions to improve materials efficiency, this approach increases feedstock use and produces parts that have equivalent load carrying capacity – albeit at a weight penalty, and can be manufactured using existing methods of fabrication. It remains to be seen whether diverting ply cutter waste from landfill by incorporating reclaimed carbon fiber into heavier laminates is more environmentally efficient.

Moving forward, increasing fiber alignment and improving distribution in reclaimed carbon fiber mat products will encourage additional usage. Another approach to increasing feedstock use will arise through the development of zero-waste production processes. Although this will not alleviate end-of-life component recycling, reducing cutting waste will immediately extend feedstock supply and expand opportunities for light-weighting components through more wide-spread use of composite materials.

Data access statement

All data are provided in the results section of this paper.

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Disclosure statement

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