



Demanufacturing and remanufacturing of thermoplastic composites: a hot and cold-peel feasibility study for circularity

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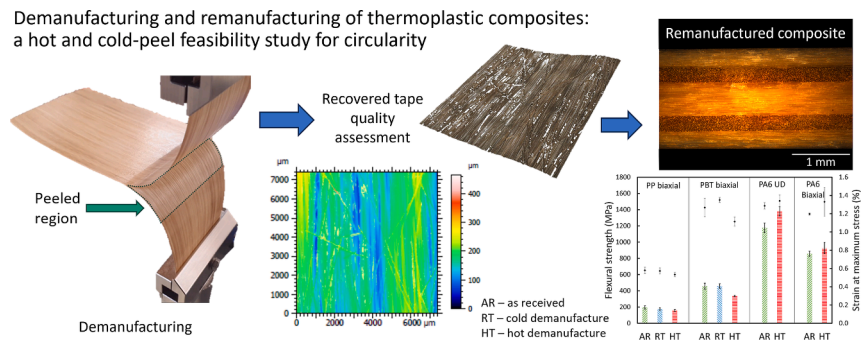
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HIGHLIGHTS

- Local heating enabled easy demanufacturing of UD thermoplastic tape plies.
- Peeling UD thermoplastic tapes can be achieved with low forces and high rates.
- Recovery at elevated temperatures affects surface quality and fibre alignment.
- Recovered tape plies were successfully remanufactured into composites.
- Remanufacturing achieves close to virgin mechanical properties.

GRAPHICAL ABSTRACT



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ABSTRACT

Composite materials are increasingly used in structural applications where lightweighting can reduce energy requirements, particularly in the transport and the aerospace sectors. However, there is a pressing need to improve the circularity of composite materials, to reduce waste and production energy. Thermoplastic composites offer a low-energy route to recovery by ply separation and reconstitution, rather than by pyrolysis or other destructive downcycling processes.

This study investigates the potential for recovery and re-use of continuous fibre reinforced thermoplastic tapes. Two-ply tapes are compression moulded and then separated using either a room temperature or hot-peel approach, analogous to the reversal of a typical tape laying process. The surface quality and fibre distribution of the recovered tapes is recorded, with the tapes subsequently reprocessed into composite panels. A comparison of mechanical properties of the reprocessed panels is carried out relative to panels manufactured from as-received tape equivalents.

Peel of the thermoplastic tapes was achieved with ease and low forces. However, recovery deteriorated surface quality and fibre alignment. Recovery of the tapes led to comparable composite properties, although a small reduction in flexural strength was observed. This was likely as a result of changes in ply distribution, an increase in voids and loss of fibre alignment.

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1. Introduction

Fibre reinforced thermoplastic (FRTTP) composites are becoming increasingly attractive structural materials due in part to their potential for a sustainable use cycle [1,2]. While thermoset composites (TSC) rely on an irreversible cross-linking reaction during manufacturing, FRTTP composites do not and so re-use of the constituent polymer matrix and reinforcement materials at end of product life is, at least in principle, achievable. However, an approach for complete recovery and re-use is not yet realised. Typically, the means of recovery for all fibre reinforced composites is to undertake an energy-intensive process of fibre/matrix separation such as by thermal or chemical means (pyrolysis or solvolysis, respectively). This process results in some reduction in fibre length and constituent properties, as well as destruction of the matrix [3–10].

Such an approach is normally necessary for TSC, where the matrix ordinarily cannot be reprocessed. Whilst some less aggressive processes for separation are in development, such as through the use of tailored matrix materials that can be irreversibly de-cross linked to recover resin components [11,12], the core of the process for thermosets remains to separate fibre and matrix for individual recovery.

The opportunity for FRTTP composites is recovery through simple heating and re-moulding. In this case, the separation of the thermoplastic matrix from the long fibre reinforcement is not necessary, since the matrix is not chemically cross-linked and can be remelted. This results in fewer recycling steps as there is no need to reprocess and recombine the fibres with a fresh matrix. Hence the matrix is retained and the fibre surfaces are not exposed to the air or to excessive temperatures that can cause damage, particularly a problem with glass fibres [13]. While this could be achieved by simple shredding and bulk moulding [14], there is also the opportunity to preserve fibre lengths by using more sophisticated processes.

While simple in concept, the realisation of heating and re-moulding continuous fibre TP composites is a complex process. In fabric reinforced composites for example, the fabric may have experienced shear during forming. This means that pressing the part back into a flat sheet or directly into another component is not trivial, since the tow movement is difficult to invert. Explorations of reverse-forming, where the FRTTP component is heated then flattened to sheet form, noted that wrinkling occurred in components that had undergone yarn slippage during initial manufacture [15,16].

In the context of the automated tape placement (ATP) process, reclaiming the fibre reinforced thermoplastic matrix tape by layer-wise separation is more accessible. In this process the reinforcement is utilised such that the fibres remain as straight as possible, in order to carry load more effectively. In principle, a component produced by ATP will be supported through-life by a digital twin containing information on the tape geometry, orientation and end position [17]. With this knowledge it is easy to envisage the application of a peeling force to the tape, reversing the deposition path, in order to recover the tape. This may require the thermoplastic matrix to either fail along the interface, or perhaps to melt so as to enable the peeling ply to separate from the substrate, permitting subsequent rewinding onto a spool. This is in essence a reversal of the original tape laying process, as suggested in Janssen's patent [18].

Practical application of this approach requires an understanding of the loads, temperatures and rates required or achievable for separation of layers, as well as the impact of recovery on the quality of the tape material. Process-induced degradation of polymers is a complex phenomenon which is highly dependent on temperature, on shear and shear rate, on polymer species and on some additives [19,20]. Typically recycling processes can see a ~ 5–20 % reduction in properties, though this would include any in-life degradation [21]. To preserve matrix properties, a balance must be struck in the demanufacturing process between temperatures high enough to minimise shear and low enough to minimise thermal degradation.

A number of studies have considered the recovery of polypropylene (PP), polyamide (PA) and Polyphenylene sulphide (PPS) tapes using

ambient or low temperature wedge driven separation [22] and peel processes [23,24]. Ultrasonic separation has also been considered [25]. These studies suggest that fibre orientation is better conserved when peeling at low temperatures while elevated temperature improves conformation with a roller during the peeling process, giving improved process control and avoiding fibre kinking (fibre breakage in ply bending). Drum roller guides are sometimes used to support peeling and prevent failure during testing of thin composites, even in thermoset systems [26].

Previous work by the authors on *iso*-thermal, small-scale peeling demonstrated that separation could be achieved with relative ease at elevated temperatures, with the surface character of the recovered material being dependent on the temperature [27]. Peeling at temperatures above the matrix melting point resulted in a relatively smooth surface through cohesive failure of the matrix. Peeling was also shown to be achievable below the melting point. This resulted in a rougher surface finish through increasingly adhesive failure of the fibre–matrix interface.

In this follow-up study, the focus is now on demonstrating the viability of a continuous, non-isothermal demanufacturing process with localised heating, as would have to be the case in a dedicated industrial facility. The quality of recovered plies and the subsequent performance of composites remanufactured using these recovered plies was investigated. Consolidated two-ply laminates were peeled using a line heat source and fixed load. Room temperature (RT) peeling was also attempted for comparison. The morphology of the recovered plies was studied before they were then reconsolidated, without further reprocessing, into six-ply laminates. The mechanical performance of composites manufactured entirely from peeled plies was measured and compared to identical composites manufactured from virgin material. Thermoplastic tapes composed of two different fibres and three different polymer matrices were investigated and their behaviours compared.

2. Materials and methods

2.1. Materials

Three distinct, unidirectionally reinforced (UD) thermoplastic tapes were utilised in this study: polyamide 6 (PA6)/carbon fibre (SGL Group, 45 % V_f carbon, ~250 gsm), polypropylene (PP)/Glass (Comfil, 75 wt% glass, ~900 gsm) and Polybutylene terephthalate (PBT)/Glass (Comfil, 67 wt% glass, ~900 gsm). All three polymer matrices are semi-crystalline. These three different fibre–matrix combinations were selected to identify potential behavioural differences due to the polymer, reinforcing fibre or volume fraction.

PTFE coated glass fabric (Cytec, FF03) was used as a release film in moulding. Self-adhesive polyimide (PI) tape (ULT013, Vital-Parts) was used to secure thermocouple cables and other components during high temperature operations. Aluminium foil (Multifoil, 0.15 mm thick) was used as a shim material in moulding.

2.2. Composite manufacture

The UD tapes were dried overnight in a vacuum oven (Medline scientific OV-11) at 50 °C before moulding. The press that was used in 2.2.1 and 2.2.2 was a 4" diameter cylinder upstroke press driven by an Enerpac P802 manual hydraulic pump. The platens were 250 mm x 250 mm in size, electrically heated using cartridge heating elements controlled by Eurotherm controllers and cooled using water flow through channels in the tool.

2.2.1. Two-ply peel specimens

Two-ply UD laminates were produced by compression moulding dried, as-received tapes between flat steel plates lined with PTFE-coated fabric release film. Pieces of the PTFE film were used between plies to create separated ends for demanufacturing through peeling. Five fine-

tipped 0.3 mm K-type thermocouples (RS PRO) were placed between the layers of a PA6 specimen and affixed to the flat plate with PI tape to avoid movement during moulding, in order to record the temperature during subsequent peeling.

The assembled stacks were introduced to the manually activated hydraulic press at temperature (PP – 190 °C, PA6 – 225 °C, PBT – 230 °C) and the platens were closed to firm contact (resistance felt on the hydraulic lever) for 10 mins to allow time to melt. An initial pressure of 20 bar (corresponding to ~ 1.65 tonnes) was applied for a few seconds, and then removed and re-applied quickly three times as breathe cycles, before finally holding at 20 bar for a further 10 min. Platens were water-cooled at a rate of ~ 7 °C/min and the stacks were removed from the press once the temperature was below 50 °C. Thicknesses of the 2-ply composites were consistent across the moulding. Positions were measured across examples of each of the material types, with results as follows (n = 15): PP – 0.50 ± 0.01 mm; PBT – 0.48 ± 0.01 mm; PA6 – 0.39 ± 0.01 mm; where the ± value is 1 standard deviation.

2.2.2. Composite panels

Six-ply composites were produced from both as-received tapes and tapes that were recovered after demanufacturing through peeling. The tapes were cut into 80 mm x 80 mm squares and compression moulded in a three-part (base, fixed picture frame and top with matched male punch) 80 mm x 80 mm cavity tool lined with PTFE-coated fabric release film. The initial intent was to produce UD panels, however the PP and PBT UD panels had very poor transverse strength. As a result, some transverse stability was incorporated by using a [0,90,0]_s layup. The PA6 was pressed both into UD and the transverse stabilised configurations [0]₆ and [0,90,0]_s. PA6 panels were nominally 1 mm thick, while the PP and PBT panels were nominally 1.5 mm thick. The moulding process (the temperature and pressure cycle) was the same as described for the peel specimens in 2.2.1.

2.3. Demanufacturing of 2-ply specimens

2.3.1. Room temperature peel

To produce demanufactured plies for re-consolidation into test composite panels (as described in section 2.2.2), 80 mm wide two-ply specimens (produced as described in section 2.2.1) were peeled apart. This was performed for the PP and PBT specimens by a simple manual method. The unbonded edges of the two-ply specimens were attached to clamps, with a mass attached to the lower one for a total of approximately 1 kg. This mass was then placed on a bench to provide a stable base for the mass to rest on. The upper clamp was then manually raised to cause the specimen to peel, with the lower mass providing up to 1 kg of peeling force (after which it would lift off from the bench). This process is illustrated in Fig. 1. It was not possible to perform peeling at room temperature with the PA6 specimens as the specimens kinked and broke before peeling could occur.

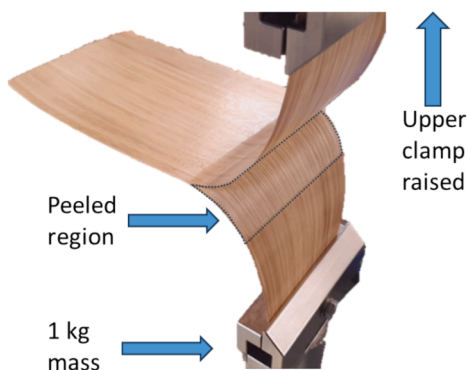


Fig. 1. Photograph showing the manual, room temperature peeling process applied to a PBT tape.

To quantify the load required to peel the PP and PBT tapes at room temperature, controlled tests were performed using an Instron 5581 testing frame. To avoid edge effects, the two-ply specimens were cut to 25 mm wide to fit the wedge grips and had ~ 160 mm of bonded length. Tests were performed using a 1 kN load cell and a cross-head speed of 100 mm/min.

2.3.2. High temperature peel

Demanufacturing was also carried out on the two-ply specimens by peeling using high temperature (HT) at the bond line to enable separation. This application of localised heating was selected for two reasons. First, it represents a similar heating approach to that used in an automated fibre placement manufacturing process. It can be considered that demanufacturing could be achieved by reversal of this process, and so it would be appropriate to approximate this method. Secondly, previous studies that considered heated peeling in a temperature-controlled chamber (i.e. global heating) caused problems in maintaining the integrity of the specimens [27].

A similar clamping setup was used to the RT setup, but the additional weight at the bottom was removed and instead just the lower clamp (weighing ~ 450 g) was left hanging free. Localised heating was applied to the bond line at the end of the bonded region using a heat gun with a wide nozzle (Bosch GHG 660 LCD Professional), held approximately 1 cm from the specimen. It was determined that heat needed to be applied to the back of the peel point rather than the front, since the application of heat directly into the joint caused it to collapse and peeling did not occur. The applied heat and the ~ 450 g clamp weight were sufficient to cause peeling in all of the 80 mm wide tape types; PP, PBT and PA6. A thermal camera (FLIR C5) was used to record the approximate temperature during the test.

The thermal camera only provided information on the surface temperature of the specimens and not the true temperature at the bond line. To obtain additional information on the temperature in the bond line, one specimen was equipped with a line of five thermocouples embedded between the plies. Data was collected by a multi-channel logger (Pico-log) at a 10 Hz sampling rate.

2.3.3. Surface assessment

Surface assessment was made of the tape specimens: the surface in contact with the tooling after initial two-ply moulding; the separated cold peeled surface; and the separated hot peeled surfaces. An Alicona G5 focus variation metrology system using a 20x lens was used to generate a point cloud data set. The two-ply moulded specimens were measured over an area of (10 x 10) mm². The more undulating peeled specimens were measured over a (5 x 5) mm² area. This data was then processed by using Mountains software (Digital Surf) [28], applying ISO 25178 settings with cross sectional height graphs generated to show the height change in several positions along the specimens and to generate surface roughness (S_a and S_q) values.

2.3.4. Thermal analysis – Differential scanning calorimetry

Thermal analysis was performed on a TA Instruments DSC2500 differential scanning calorimeter (DSC) operating under nitrogen. Specimens were heated at 10 °C/min up to 260 °C (PA6 and PBT) or to 220 °C (PP). They were then cooled at 50 °C/min back to room temperature and allowed to equilibrate. Subsequently they were subjected to a second heating cycle at 10 °C/min up to 260 °C (PA6 and PBT) or to 220 °C (PP). Melting point(s) were determined from the second run using TRIOS software (TA Instruments Trios v5.1.1.46572). Three repeats were performed for each specimen type.

2.3.5. Scanning electron microscopy (SEM)

SEM images were obtained using a Hitachi TM4000Plus II Tabletop SEM. Specimens were assessed without coatings using the low vacuum, charge-up reduction setting. Images were taken using a mixed detector setting combining secondary electron (SE) and backscattering (BSE)

detectors to provide topographic images enhanced by density contrast. Accelerating voltage was 10 kV, with a working distance of approximately 6 mm.

2.4. Characterisation of 6-ply composites

Specimens were cut to 34 mm long x 5 mm wide for torsion and to 70 mm x 15 mm for bending and burn off tests. Flexural tests have been previously employed for the observation of matrix degradation[29], and burn off tests are commonly used to determine fibre volume fractions. Specimens were cut to size from the six-ply composite panels using waterjet, following the pattern shown in Fig. 2, keeping away from the edges of the panel to avoid unwanted effects due to small amounts of flow (flash) near the edges of the mould. The test specimens were vacuum dried at 50 °C prior to testing.

2.4.1 Volume fraction

Fibre volume fraction measurements were undertaken on the six-ply composites (section 2.2.2) using a burn off method. After vacuum drying at 50 °C, the dimensions of the centre specimen from each composite panel were taken (values of thickness and width were determined using a micrometer and callipers, respectively) and their weight measured on a five-point balance, in order to determine the composite density (ρ_c). Since specimens included carbon fibre, the thermal cycle was selected to minimise fibre loss. Autoignition and peak mass-loss temperatures for PP, PBT and PA6 were obtained from an Office of Aviation Research report [30] and are provided in Table 1. This indicated a suitable burn off cycle loading at 350 °C and raising to 450 °C, with minimal char expected. Literature data suggested that mass loss rates for T300 carbon fibre should be relatively low at these temperatures [31]. Based on the literature data, an estimated loss value of 0.0016 %/s at 450 °C was determined for the carbon fibres during the burn off test and accounted for in the volume fraction calculations. Mass measurements were taken after holding for an hour at 450 °C and again after a further 30 min to ensure complete combustion of the matrix. Fibre volume fraction (V_f), matrix volume fraction (V_m) and void volume fraction (V_v) were determined as:

$$V_f = \left(\frac{M_f}{M_c} \right) \left(\frac{\rho_c}{\rho_f} \right)$$

$$V_m = \left(\frac{M_c - M_f}{M_c} \right) \left(\frac{\rho_c}{\rho_m} \right)$$

$$V_v = 1 - V_f - V_m$$

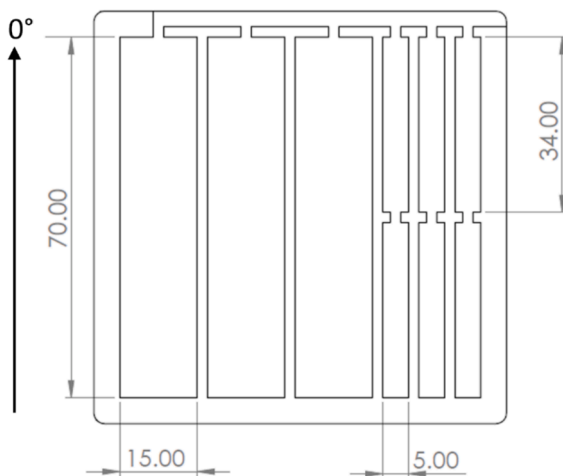


Fig. 2. Waterjet cutting layout used to section six-ply composite panels into specimens for torsion, bending and burn off tests. Fibre direction of 0° indicated with an arrow.

Table 1

Ignition data for polymer matrix materials.

Polymer	Autoignition (°C)	Peak mass-loss (°C)	Char (g/g)
Polypropylene (PP)	367	424	0
Polybutylene terephthalate (PBT)	382	407	0.07
Polyamide 6 (PA6)	432	454	0.02

following the guidance in ASTM D3171-15. Here M_c is the composite mass before burning, M_f is the fibre mass after burning, ρ_c is the composite density (determined by geometry and mass), ρ_f is the fibre density and ρ_m is the matrix density. Average density values used for the volume fraction calculations were taken from literature and are listed in Table 2. Cooling rates for the PBT composites were around 0.14 Ks⁻¹. According to the degree of crystallinity data provided in [32], this indicated that the PBT matrix would exhibit a density of 1.33 gcm⁻³.

2.4.2. Torsion testing

Dynamic mechanical analysis (DMA) is a technique that has been successfully used for the characterisation of matrix degradation[35]. Specimens 34 mm long x 5 mm wide were placed into the rectangular bar fixture of a rheometer (Anton Paar MCR302) with a free length of 22 mm. Firstly, suitable conditions for torsion were determined by using a frequency sweep. Subsequently, the specimens were subjected to an oscillating shear strain amplitude of 0.01 % in torsion at a frequency of 1 Hz while heating up to melt temperature following the schedule in Table 3, slowing the temperature ramp to 5 °C/min for greater resolution in the main region of interest. Values of storage modulus (G'), loss modulus (G'') and tan δ were calculated by the RheoCompass software. Three specimens were tested for each sample type.

2.4.3. Flexural testing

Flexural testing was performed using an Instron 5581 testing frame with a three-point bend fixture and a 1 kN load cell. Span and test rates were selected in accordance with BS EN ISO 14125, with the glass fibre composites taken to be Class III with a nominal 1.5 mm thickness and the carbon fibre composites taken to be Class IV with a nominal 1 mm thickness. Values of thickness and width were determined using a micrometer and callipers, respectively. Values of elastic modulus, E , and ultimate strength, σ_{UTS} , were determined as the gradient of the stress/strain graph between the limits of 0.05 % and 0.25 % strain, and as the maximum flexural stress, respectively. The sample size for each test was 3 specimens.

3. Results

Data from this study is hosted on Mendeley and can be found at: <https://doi.org/10.17632/v9jbs6d757.1>. Statistical analysis of the data was performed using Minitab version 21.1 (64-bit).

3.1. Demanufacturing assessment of 2-ply specimens

3.1.1. Room temperature peel

RT peel forces measured using the Instron (section 2.3.1) are reported as a function of grip displacement in Fig. 3, for the two-ply PP and

Table 2

Density values used for volume fraction calculations.

Material	Density value (gcm ⁻³)
Glass fibre	2.545 [33]
Carbon fibre	1.76 [31]
Polypropylene (PP)	0.905 [34]
Polybutylene terephthalate (PBT)	1.33 [32]
Polyamide 6 (PA6)	1.13 [34]

Table 3

Heating rates used for torsion in rheometer.

Polymer	Ramp 10 °C/min	Ramp 5 °C/min
PP	25 – 120 °C	120 – 190 °C
PBT	25 – 140 °C	140 – 230 °C
PA6	25 – 140 °C	140 – 230 °C

PBT tapes. Forces start negative as a result of the clamping procedure, which led to the bending of the peel specimens pushing away the grips at the start of the test as the spring load tries to straighten the fibres. Although RT peel of the PA6 was attempted, it was not possible as tape failure occurred before peeling due to kinking and fibre failure at the joint. Example peel curves for these attempts are provided in the

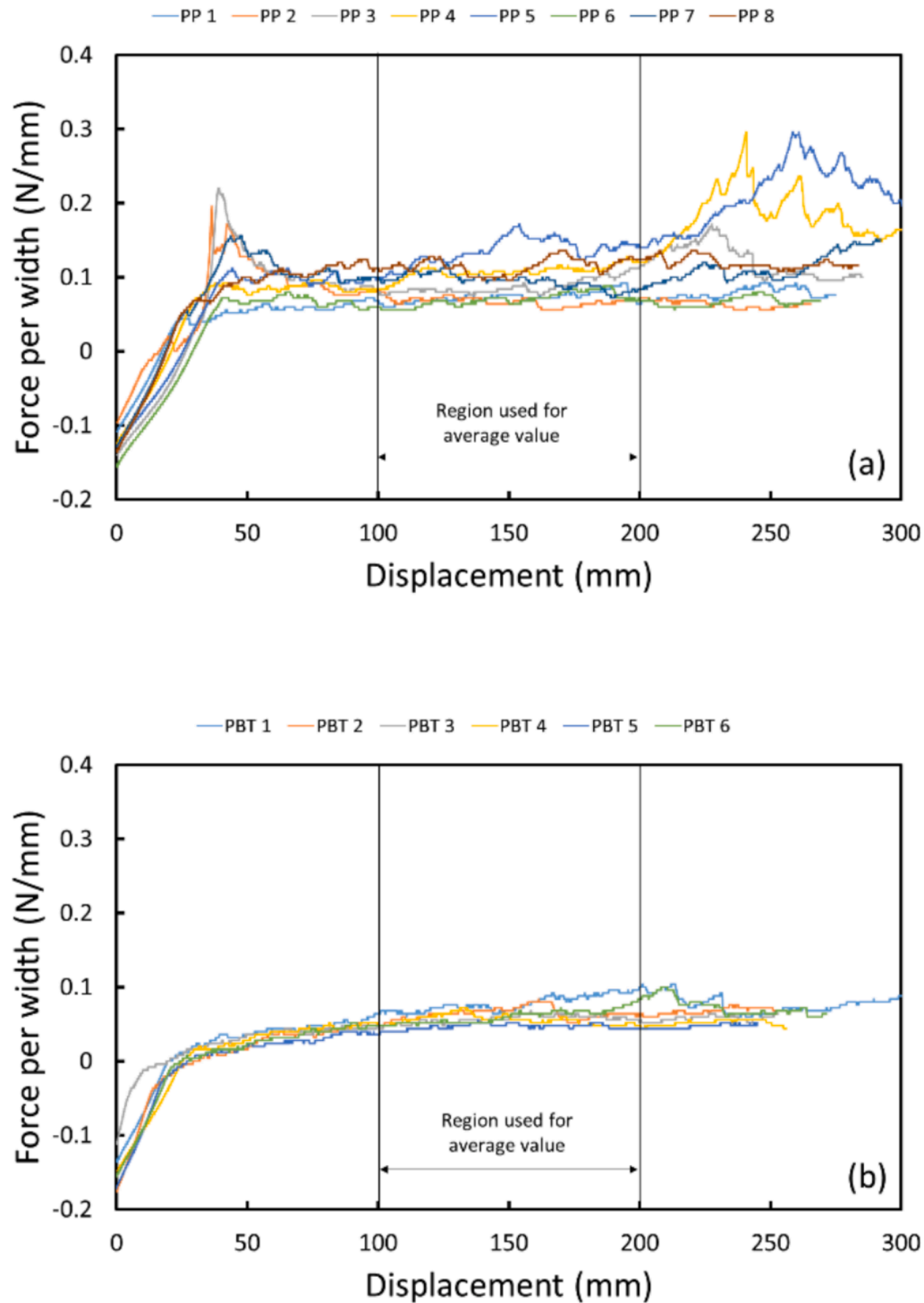


Fig. 3. Force vs displacement room temperature peel data for 25 mm wide PP (a) and PBT (b) two-ply specimens, with the range indicated over which an average force measurement was taken. Curves PP1-PP8 and PBT1-PBT6 represent repeat tests.

supplementary data (Fig. S1). In terms of both the shape of the force–displacement curves and the magnitudes, the behaviours observed for the PP and PBT specimens were similar to that observed for wedge peel testing by Zhang and co-workers [36]. The load values averaged across a displacement range of 100 mm to 200 mm, as indicated in Fig. 3, were (0.09 ± 0.02) N/mm for the PP specimens, and (0.06 ± 0.01) N/mm for the PBT specimens, where $\pm$ refers to the standard deviation. Around half of the PP specimens exhibited a load peak just prior to the start of peeling, suggesting a potential difference in peel initiation behaviour between the PP and PBT specimens. Peaks in the later region of the data, at 150 mm and beyond, are a geometric effect associated with uneven peeling, visible as the peel line transitioned away from perpendicular to the fibre axis.

3.1.2. High temperature peel

Preliminary attempts at hot peeling helped to determine that, for all specimens, the ~ 450 g mass of the lower clamp was sufficient to induce peeling of the two-ply specimens with the application of hot air to the peel line. Since the specimens were 80 mm wide, this corresponds to a required peel force of ≤ 0.055 N/mm. As soon as the point of peel reached a sufficient temperature the peel progressed continuously, moving peeled tape away from the heated zone and exposing new material to the heated peel front.

The positions of the embedded thermocouples for the instrumented peel are shown in Fig. 4a. The data recorded for the embedded thermocouples during the course of peeling is provided in Fig. 4b, alongside the thermal camera data. An example of the thermal camera data is provided in Fig. 4c. The thermal camera data was taken as the highest value for the image; for example in Fig. 4c it was 252°C . There was no common trigger for the camera and thermocouples, so image time stamps were used to correlate the time axis approximately. The lowest peak temperatures within the bond line were recorded by thermocouples TC2 and TC3 as $161\text{--}163^\circ\text{C}$, while at a similar time the thermal imaging camera indicated an external surface temperature behind the bond region of $214\text{--}223^\circ\text{C}$. The true bond line temperature is assumed to lie between these values.

The same heat gun arrangement was used in all the HT peel tests carried out in this study; thermocouples were not embedded in other trials in order to preserve the tape interface for subsequent

remanufacturing. Images from the thermal imaging camera suggest that similar temperatures were achieved, albeit with some variability due to the set-up and the limitations of using IR cameras on high emissivity materials. Although it is experimentally challenging to obtain precise measurements of temperature at the peel point heated with a hot air gun, the steady state nature of the obtained peel, as well as the successful outcome of the trials, suggest that the process is likely to self-regulate to reach a sufficient peel temperature in all cases. It is assumed that, as the two layers peel apart, new material is exposed to the heat; the system peels more quickly if there is excess heat, and more slowly if there is insufficient heat, quickly returning to a steady state. The typical peel rates observed in these trials were in the region of 5 mm s^{-1} .

3.1.3. Surface assessment

The surfaces of the peeled tapes were characterised using the Alicona G5, and topographical information was obtained as height maps for each specimen. The overall S_a (arithmetic mean height) and S_q (standard deviation of heights) values for the set of specimens are provided in Table 4.

The as-moulded tape specimen shows relatively little variation in surface height. Both RT and HT peeled specimens show greater variation in surface height, with fibre bundles as well as loose fibres disrupted by the peeling being visible in the scan. Much larger S_q values were observed for the hot-peeled specimens. Qualitatively, the hot peeled

Table 4

Surface roughness values S_a and S_q for the different single ply specimens as-received, and after peeling at room temperature (RT) and high temperature (HT).

S_a (mm)	PP	PBT	PA6
As received	8.23	9.72	2.67
After RT peel	45.60	26.50	–
After HT peel	92.67	57.67	45.13
S_q (mm)	PP	PBT	PA6
As received	10.50	13.20	3.60
After RT peel	61.30	37.20	–
After HT peel	126.09	135.52	54.38

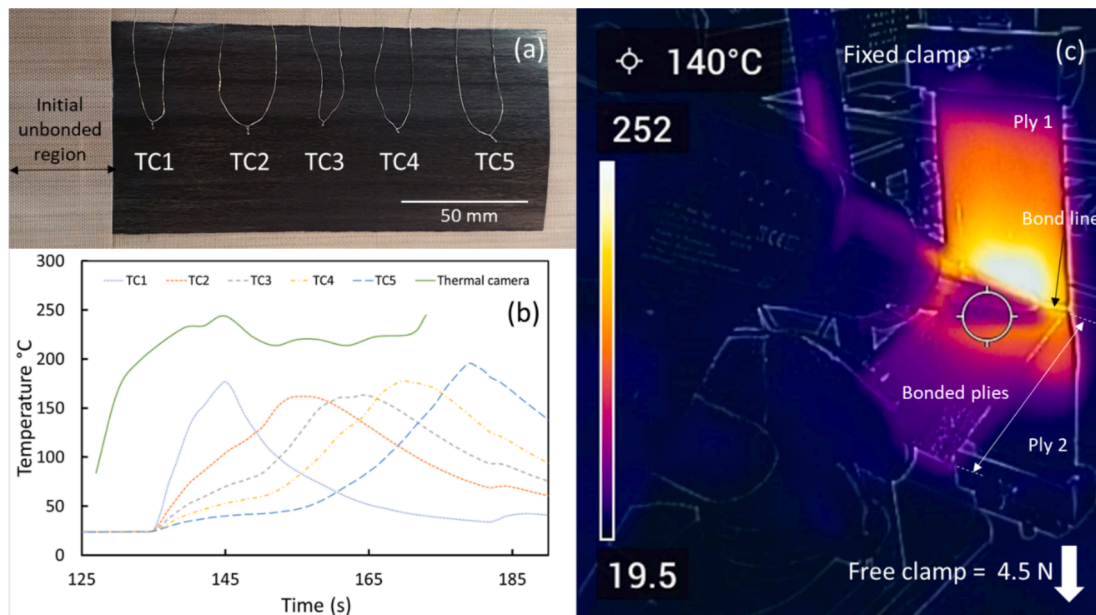


Fig. 4. (a) position of the thermocouples embedded between the tape layers prior to moulding; (b) temperature profile during hot PA6/carbon peeling as recorded by the thermal camera and the embedded thermocouples along the length of the specimen (thermal camera values taken as the highest value for the image); (c) thermal image of the peel process.



Fig. 5. Backlit image of a HT peeled PA6 tape qualitatively showing splitting and fibre waviness caused by the peeling process.

specimens also showed more splitting and waviness than the cold-peeled specimens (see Fig. 5). The observed surface roughness values for the HT PA6 peel are substantially higher than were observed in the small-scale isothermal testing in previous work ($S_a \sim 10$, $S_q \sim 15$). This suggests that cooling rate after peel may be important to achieving a good surface finish.

3.1.4. Differential scanning calorimetry

Melting point temperatures for the polymer matrix materials were determined by DSC. They were measured on specimens taken from tapes after 2-ply moulding, and on the tapes recovered after heated peeling of the 2-ply specimens. In this way the properties of the matrix are investigated before and after short-term exposure to high temperature peeling. The results are provided in Table 5. The values obtained are consistent with literature expectation[34], with no statistically significant difference in the values ($P \leq 0.05$) as a result of the high-temperature peeling.

3.1.5. SEM

SEM was performed on the peeled surfaces of the different 2-ply composites after cold-peeling and hot-peeling. These are provided for PP (Fig. 6) and PBT (Fig. 7). It was not possible to cold-peel the PA6 and so only hot-peel results are shown (Fig. 8).

It is readily apparent that the PP and PBT materials have poor interfacial adhesion, with the cold peel and hot peel showing almost clean fibre surfaces in both cases. Conversely the interfacial adhesion for the PA6 appears to be much better, with good fibre coverage. Cold peeling results in fracturing and spalling of polymer for both the PP and PBT, while hot peeling results in splitting and potentially surface tension or crystallisation driven pin-holing. Conversely, the PA6 shows a smooth matrix covering after hot peeling.

3.2. Mechanical performance of manufactured and remanufactured composites

All six-ply panels were well consolidated, although some flashing at the edges was apparent. Longitudinal cross-sectional images are provided in the supplementary data (Figs. S2 and S3).

Table 5

Melting point onset and peak temperatures obtained from differential scanning calorimetry on moulded and on peeled specimens, demonstrating no statistically significant ($P \leq 0.05$) difference in the melting behaviour as a result of high-temperature peeling.

Specimen ($n = 3$)	Onset ($^{\circ}\text{C}$)	Peak ($^{\circ}\text{C}$)
	Average	Average
PP 2-ply moulded	151.7 ± 3.4	158.7 ± 1.6
PP hot peeled	149.9 ± 3.0	157.9 ± 1.2
PBT 2-ply moulded	n/a	220.3 ± 0.5
PBT hot peeled	n/a	220.7 ± 0.1
PA6 2-ply moulded	210.6 ± 0.5	219.0 ± 0.4
PA6 hot peeled	210.8 ± 0.3	218.5 ± 0.2

3.2.1 vol. fraction

The results of the assessment of fibre volume fraction of the six-ply composites, determined by burn off, are provided in Table 6. Where more than one composite panel of the same type has been tested, the n value, average and standard error have been included in the table.

The measured fibre volume fractions of the PA6/carbon are slightly higher than the nominal tape volume fraction, with low void content. The exception is the UD PA6 HT peel specimen, which reported a notably higher fibre volume fraction. The PP and PBT specimens all showed considerably higher volume fraction than the original tape ($\sim 50\%$), although the specimens that were reconsolidated after peeling appeared to have slightly lower fibre volume fractions in comparison to the as-received versions.

There appeared to be a greater degree of fibre packing in the PP specimens, when compared to the PBT. The PP and PBT specimens also had higher voids than the PA6 specimens. The PBT specimens reconsolidated after peeling appeared to have more voids than those made using the as-received tape.

3.2.2. Torsional stiffness

The real and imaginary parts of the shear modulus of the six-ply composites, measured at 1 Hz as a function of temperature, are reproduced in Fig. 9 (a – d). Measurements are provided for specimens that are manufactured from as-received tape, and for specimens remanufactured from both room temperature peeled (where available) and hot-peeled tapes. In the unidirectional specimens the axis of torsion corresponds with the longitudinal fibre axis. In the biaxial specimens the axis of torsion corresponds with the dominant fibre direction (0°), and the other fibre direction is perpendicular.

All three systems show the same general trend: the storage moduli fall with increasing temperature, at first relatively slowly, and then more dramatically as the melting temperature is approached and the specimen delaminates. The loss moduli exhibit a small rise with increasing temperature before a more substantial fall at the melt temperatures. In the PBT and PA6 systems there is a minor peak in the loss factor tangent, $\tan \delta$, in the region of ~ 50 to 75°C , corresponding approximately with the glass transition temperatures[32,37]. $\tan \delta$ peaks become dominant at $\sim 170^{\circ}\text{C}$ for the PP system, and above 230°C for the PBT and PA6 systems, associated with the melting of the crystal structures and breakdown of the specimen. In all cases these peaks are $\sim 10^{\circ}\text{C}$ above the melting points identified by DSC (Table 5).

The introduction of a demanufacturing stage may introduce damage to the fibres in the form of fractures or misalignment, and to the matrix and fibre–matrix interface in the form of polymer degradation. Whereas fibre damage is likely to manifest as a decrease in the elastic properties, matrix and interface degradation may be more easily visible in the loss factor tangent.

In the PP/glass system, the hot demanufacturing stage appears to produce a reduction in G' across the temperature range, as well as a broadening of the $\tan \delta$ peak associated with the melt relative to the as-received manufactured specimen. The response of the RT peeled remanufactured specimen is instead very similar to that of the as-received

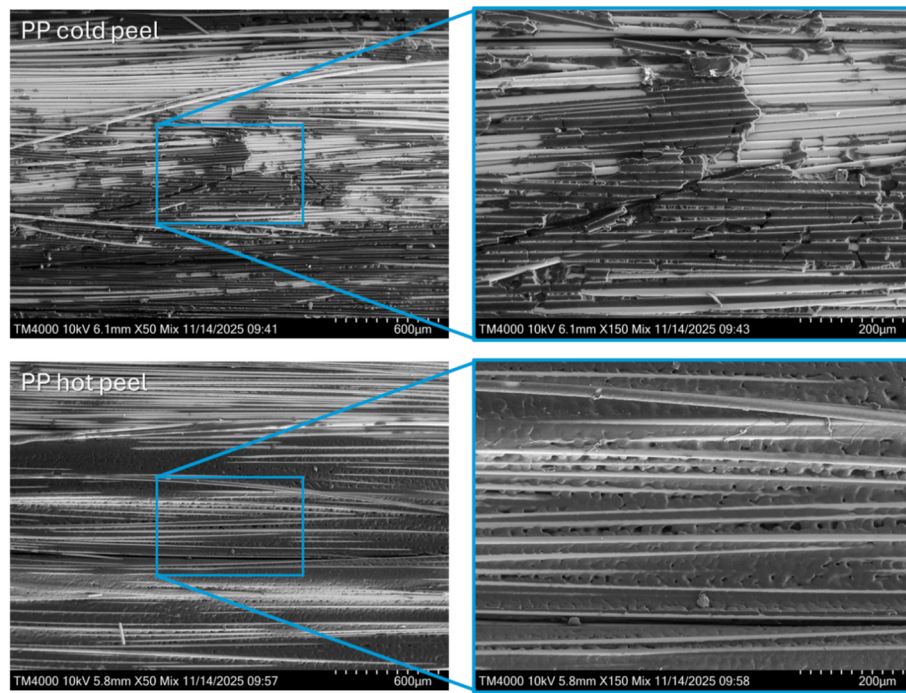


Fig. 6. SEM images of the surfaces of the 2-ply PP composites after being subjected to cold and hot-peeling.

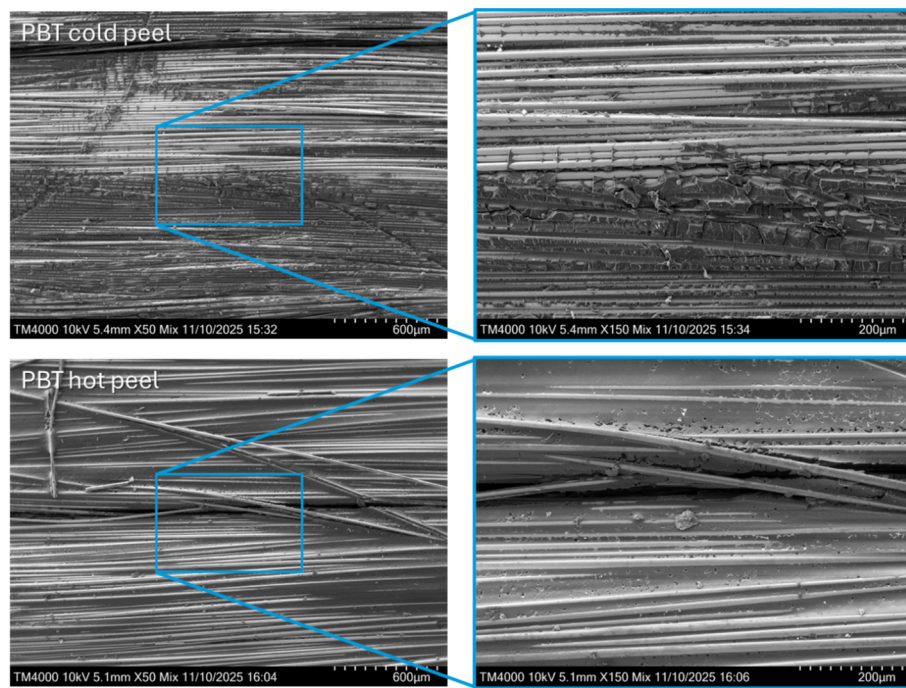


Fig. 7. SEM images of the surfaces of the 2-ply PBT composites after being subjected to cold and hot-peeling.

manufactured specimen. This suggests that the high temperature peel may have had a detrimental effect on the PP matrix, or caused a disruption or damage to the fibres, something that did not occur with the room temperature peel.

The PBT/glass system exhibits a response that appears to show little effect due to the demanufacturing step, whether at high temperature or room temperature.

The PA6/carbon system could only be demanufactured hot. The UD system shows a result that is at first slightly surprising: the elastic G' is higher in the remanufactured specimen than in the as-received

manufactured, however it has been shown to have a slightly higher volume fraction (Table 6). The biaxial PA6/carbon did not show a difference between the shear properties of the remanufactured and the as-received manufactured specimens.

The room temperature (25 °C) measurements of the real, elastic part of the shear modulus are shown in Fig. 10, to enable a more detailed comparison of the effect of the demanufacturing steps on the standard stiffness performance in each of the specimens.

In general, the differences in modulus as a result of the demanufacturing step are relatively minor, with remanufactured specimens

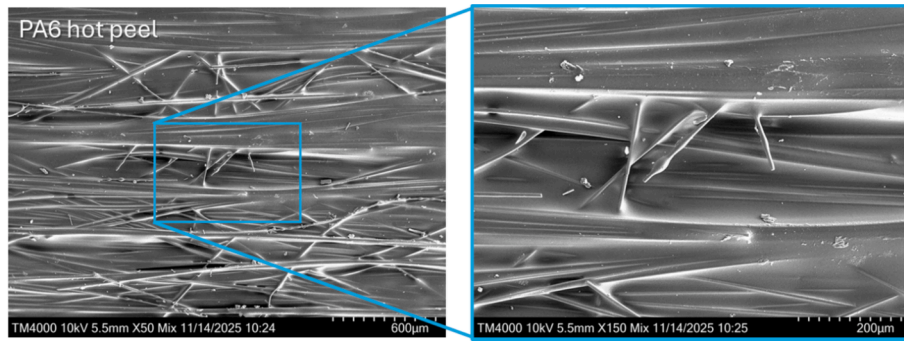


Fig. 8. SEM images of the surface of the 2-ply PA6 composites after being subjected to hot-peeling.

Table 6

Volume fraction assessment measurements on moulded six-ply laminates, either manufactured from as-received materials or remanufactured from peeled plies. Where multiple measurements are made, n is the number of repeats and the standard error is given as ($\pm$).

Specimen type	Fibre volume fraction, V_f	Matrix volume fraction, V_m	Void volume fraction, V_v
PP as-received (n = 3) manuf.	67.1 ($\pm$ 0.9)	29.2 ($\pm$ 0.7)	3.6 ($\pm$ 0.2)
PP RT peel remanufactured	64.7	32.4	2.8
PP HT peel remanufactured	64.0	32.9	3.2
PBT as-received (n = 2) manuf.	59.1 ($\pm$ 0.4)	39.7 ($\pm$ 0.4)	1.2 ($\pm$ 0.1)
PBT RT peel remanufactured	57.1	40.1	2.8
PBT HT peel remanufactured	56.0	41.3	2.6
PA6 UD as-received manuf.	48.8	49.9	1.2
PA6 UD HT peel remanuf.	53.1	45.2	1.7
PA6 biaxial as-received manuf.	48.0	50.3	1.8
PA6 biaxial HT peel remanuf.	48.4	50.9	0.7

exhibiting a similar or slightly reduced modulus compared to the as-received manufactured specimens, as expected from thermo-oxidative mechanisms of degradation inducing primarily chain scission in the matrices [38–40]. In addition, PA6 is also susceptible to hydrolysis; pre-drying may help to reduce this, but was not employed here. The exception is the remanufactured PA6 UD system. Here the specimens remanufactured from HT peel tape are uncharacteristically stiffer than those manufactured from as-received tape. When measured at room temperature, the stiffness of HT tapes is 5.29 GPa, and of AR tapes 4.64 GPa, suggesting that the demanufacturing and remanufacturing leads to an increase in stiffness of $14\% \pm 7\%$. The V_f increased from 48.8 % to 53.1 %; using either an inverse rule of mixtures approach [41] or a composite cylinder model [42] to estimate the shear modulus produces a relative increase in shear modulus of between 7 % and 9 % when using an indicative shear moduli of 1 GPa and 10 GPa for the PA6 and carbon fibres, respectively. Allowing for errors, the increased V_f as a result of matrix squeeze-out during the remanufacturing step accounts for a substantial part of the increase in shear modulus.

3.2.3. Three-point bend

In order to examine the effect of demanufacturing on the strength of composites, measurements of flexural strength and strain evaluated from the 3-point bend tests of the six-ply composites are shown in

Fig. 11. The flexural strength of the PP system indicates a loss in strength after peeling and remanufacturing, though it is not a statistically significant difference ($P \leq 0.05$). The PBT system shows a statistically significant drop in strength as a result of hot-peel ($26\% \pm 8\%$), but no significant change as a result of the RT peel. The reduced strength may be as a result of slightly lower volume fractions and of the greater degree of disruption of fibre orientation that occurs during hot peel. This is consistent with the observed changes in surface roughness (S_a) (Table 4) and the apparent fibre orientation disruption (Fig. 5).

The PA6 remanufactured specimens instead show an apparent increase in flexural strength relative to the as-received manufactured, more so for the case of the UD ($15\% \pm 7\%$) than for the biaxial specimens (not statistically different). This is consistent with the observed increase in shear modulus for the UD ($14\% \pm 7\%$), and most likely predominantly due to the 4.3 % increase in fibre volume fraction (Table 6). The biaxial panels had only a very small difference in shear modulus and an almost identical fibre volume fraction, whether remanufactured or not. However, the level of voids was reduced in the remanufactured specimens.

Measurements of the flexural modulus were obtained from the same flexural experiments, and are shown in Fig. 12. In these flexural tests the direction of the fibres in the UD specimens corresponds with the beam axis, and the biaxial specimens have additionally fibres running along the width of the specimens.

The flexural moduli of the PP specimens are reduced by the demanufacturing stage, whether HT or RT; the loss is around 10 % in both cases. The reduction of modulus is also observable for the PBT specimens, but is only statistically significant for the HT peel ($15\% \pm 5\%$ drop). The PA6 biaxial specimens experienced no significant change, however the PA6/carbon UD specimens exhibited a remanufactured modulus that is $11\% \pm 4\%$ higher than that of the as-received manufactured specimens. This increase is similar to what could be observed in torsion in Fig. 10, where the PA6/carbon UD specimen showed a $14\% \pm 7\%$ increase, and hence is attributed primarily to the increase in fibre volume fraction (Table 6).

The mode of failure in the tests was different depending on the material, but was similar for the virgin and recovered tapes. Examples of the failure modes for the different materials are provided in Fig. 13. The PP specimens failed primarily through delamination and buckling, the PBT failed through extensive delamination and the PA6 failed locally through thickness. These modes of failure are in keeping with the poor interfacial strength for the PP and PBT and the good interfacial strength for the PA6 observed in Figs. 6–8.

4. Discussion

4.1. The demanufacturing process

Section 3.1.1 Room temperature peel has shown that both PP and PBT prepreps that have been moulded together can be peeled at room

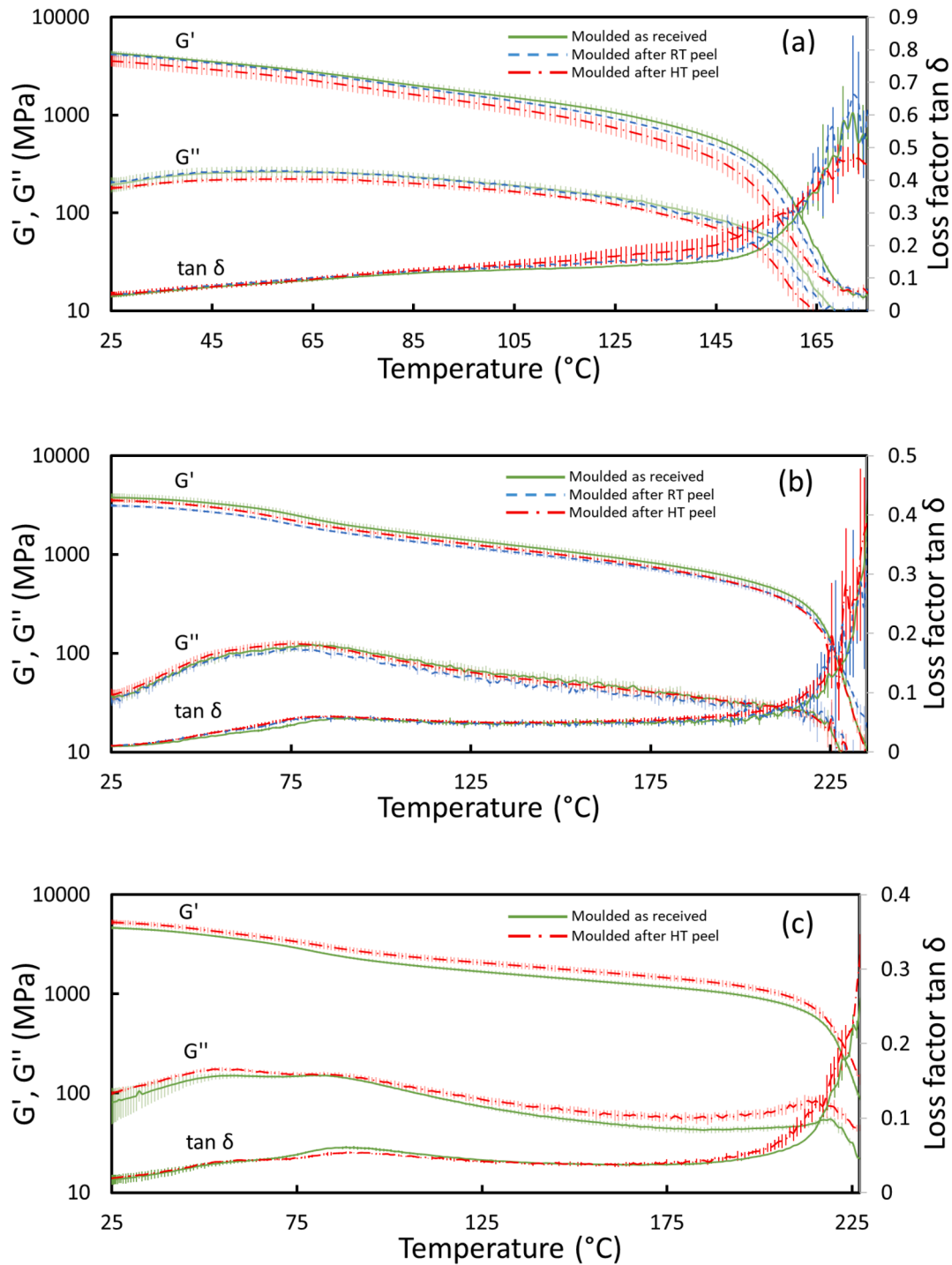


Fig. 9. Storage and loss shear moduli (left abscissa), and loss factor tangent (right abscissa) as a function of temperature, measured in torsion at 1 Hz for the six-ply composites. Specimens were produced from as-received tapes or from plies demanufactured at either room temperature (RT) or high temperature (HT) peel: (a) PP/glass; (b) PBT/glass; (c) UD PA6/carbon; (d) Biaxial PA6/carbon.

temperature, repeatably and with low forces. The peel forces of 0.09 N/mm for the PP/glass and of 0.06 N/mm for the PBT/glass are very low, being of a similar magnitude to those observed for typical adhesive tapes [43]. SEM imaging of the peeled surfaces (Figs. 6 and 7) indicates that there is very little adhesion between the fibres and the matrix. This poor interface is commensurate with the very low peel forces that were observed. Other studies have measured higher peel forces, on the order of 0.3 N/mm for a self-reinforced UD PP tape [44] and 3 N/mm for a

woven glass/PP composite [45]. The woven glass/PP study reports that the glass uses an acryl-silane coupling agent to improve fibre/matrix adhesion.

The inability to peel the PA6 specimens at room temperature in this study may be as a result of the improved fibre/matrix bonding observed in the SEM in Fig. 8, requiring greater peeling forces. In the setup that was used, at room temperature the fibres kinked and broke before peeling could occur. A drum-supported peeling, as might be used

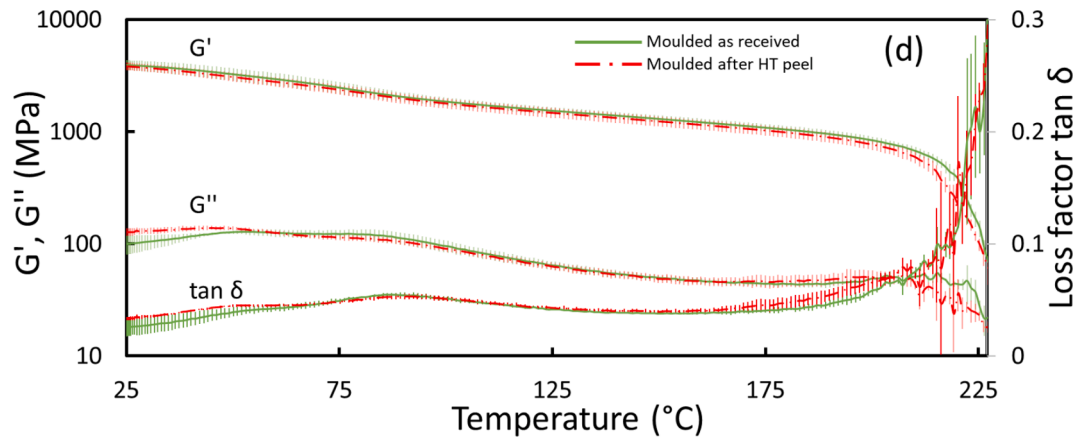


Fig. 9. (continued).

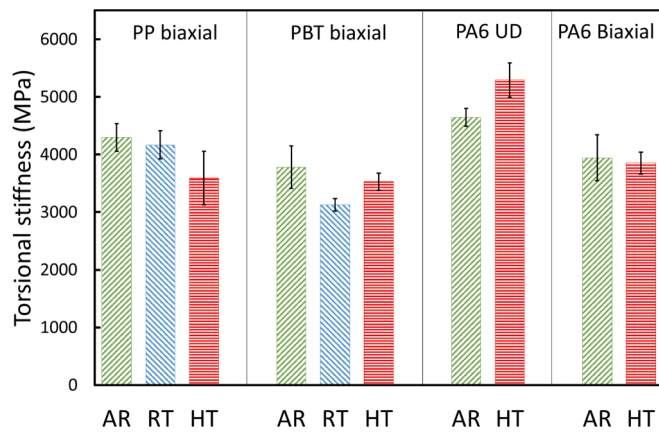


Fig. 10. Dynamic torsional stiffness measured at room temperature and 1 Hz for specimens manufactured from as-received tapes (AR) and remanufactured from both room temperature peeled (RT) and hot-peeled (HT) tapes.

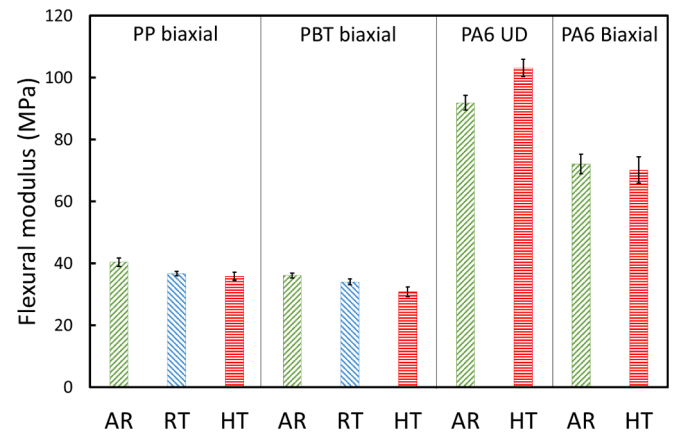


Fig. 12. Flexural modulus for each of the specimen types subjected to 3-point bending. Specimens manufactured from as-received tapes (AR) and remanufactured from both room temperature peeled (RT) and hot-peeled (HT) tapes.

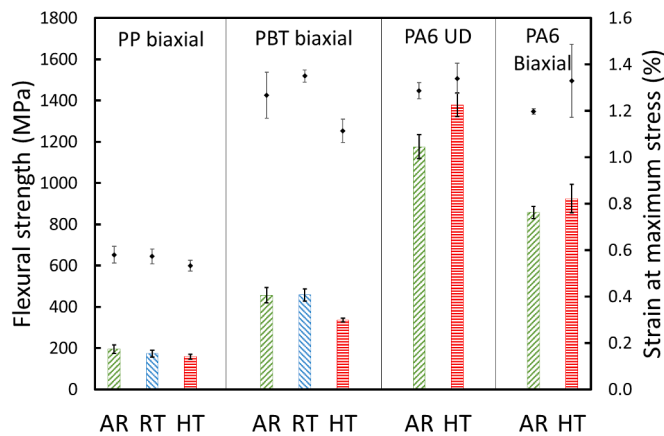


Fig. 11. Flexural strength (bars, left ordinate) and strain at maximum flexural stress (dots, right ordinate) for each of the specimen types subjected to 3-point bending. Specimens manufactured from as-received tapes (AR) and remanufactured from both room temperature peeled (RT) and hot-peeled (HT) tapes.

industrially when demanufacturing a component, should reduce the likelihood of fibre kinking and may facilitate room temperature peeling. This will be considered in future testing.

In section 3.1.2 High temperature peeling has been explored in all three systems. Although a relatively simple hot-peel set-up was used, the

application of heat proved to be a facile means of separating the ply layers. The use of a line heat source, applied to the back face of the two-ply specimen bond region under modest load (4.5 N), resulted in relatively rapid and controlled debonding. The rate of debonding at this load level appeared limited only by the rate at which heat could be applied to the zone, but this is only a qualitative observation. This suggests that this recovery process could be implemented at high speed in an automated process. This method would fit a force-controlled system, as opposed to displacement-controlled. The force control would help to prevent or limit fibre damage, and the peel rate would be indirectly controlled by the application of heat and help to prevent overheating and matrix damage – as more heat is applied to the peel zone, the peel rate accelerates and moves material away from this heat zone. Looped control would need to be established between the heating and rewinding speed, and an acceptable temperature window of operation determined.

Specific measurement of the temperature at the peel zone is difficult to achieve, as highlighted by the differences in temperature readings seen between IR camera and thermocouple techniques in Fig. 4. In a previous study of the behaviour of the same PA6 tape, DSC was used to determine that the earliest onset of melting is around 180 °C, with two melting peaks at 217 °C and 225 °C [27]. In that same study, miniature peel tests also suggested that significant peel forces, equivalent to > 1.6 N/mm, would be required for peeling to occur below around 192 °C [27]. Given that the current test has a peel force of ≤ 0.055 N/mm, this would suggest that the thermocouple readings of 161–163 °C are much below the true temperature at the peel line. Despite the small size of the thermocouples and the 10 Hz sample rate, it is likely that the residence

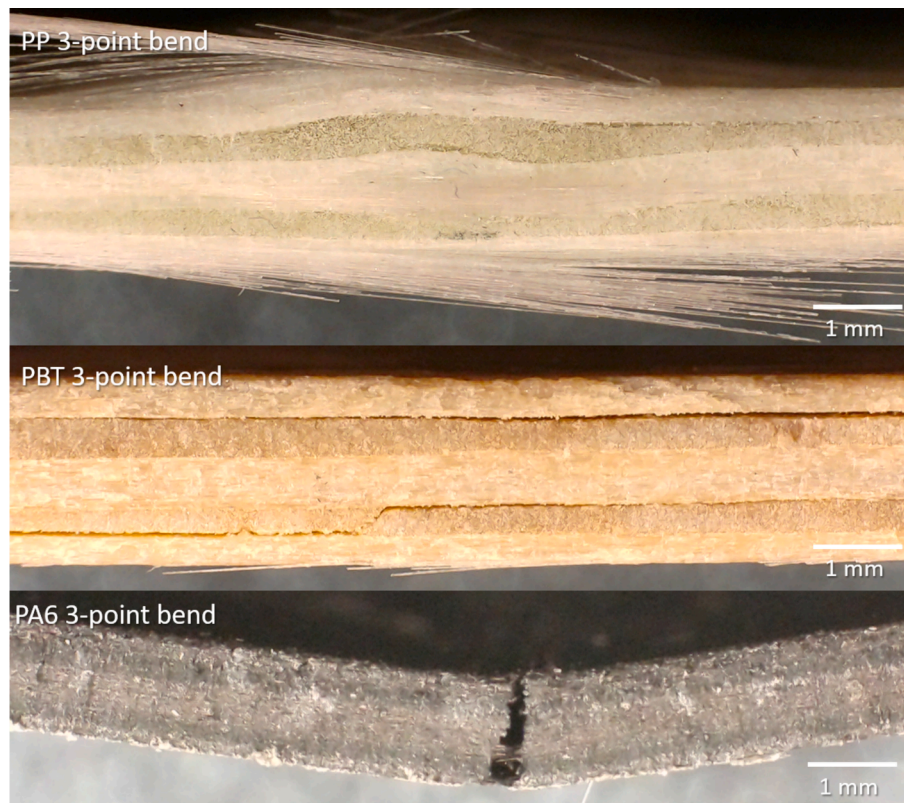


Fig. 13. Example failure modes for the 3-point bend specimens. PP (top) demonstrated delamination and buckling, PBT (middle) experienced extensive delamination and PA6 (bottom) failed through thickness locally. Scale bars are approximate.

time in the peel zone is too short for the true peel temperature to be recorded. Conversely, prior miniature peel tests also showed peel occurring freely with very low forces of 0.02 N/mm above about 212 °C. This suggests that the back side temperature recorded by the thermal imaging camera is likely to be higher than the temperature at the peel point.

The transverse thermal conductivity of carbon fibre is $\sim 2 \text{ W m}^{-1} \text{ K}^{-1}$ [46], and the thermal conductivity of the PA6 matrix is $\sim 0.27 \text{ W m}^{-1} \text{ K}^{-1}$ [47]. Individual plies are $\sim 0.2 \text{ mm}$ in thickness, and the heat transfer coefficient for a heat gun at 600 K [48] is $200\text{--}300 \text{ W m}^{-2} \text{ K}^{-1}$, depending on the geometry of the surface. The Biot number should therefore lie between 0.02 and 0.2, which would suggest that the temperature field through the thickness of the ply can be considered as nearly uniform. This suggests that an automated demanufacturing process should be able to heat the peel point through the thickness of one ply without excessive degradation of the layer of polymer between the surface and the peel point. This is, however, a UD tape material, and the longitudinal conductivity for carbon is higher, around $7 \text{ W m}^{-1} \text{ K}^{-1}$. As a result, fibres are likely conducting heat away from the site and providing a degree of thermal shielding to the peel site. This particular anisotropic thermal behaviour of carbon fibre is exploited in heat shielding materials [49], and could become a significant factor in considering peeling process control for thicker composite materials.

The capacity of plies to bend without excessive fibre damage during peeling is undoubtedly an important aspect of successful demanufacturing. Ply bending stiffness and strength are influenced by both material property effects (properties of the fibres and properties of the matrix polymer at the temperature of interest) and by geometric effects (ply thickness and fibre distribution, and importantly, the bending radius). In this study it was not possible to control each of these variables individually, but the successful peel of PP/carbon and PBT/carbon, at

both room temperature and close to the melt temperature, and of PA6/glass, close to the melt temperature only, suggests that there is a relatively large window of opportunity. The inability to peel PA6/glass at room temperature is unlikely to be due to one factor alone, since all plies were of similar thickness, and since both PA6 and PBT are below T_g at room temperature but PP is not. Failure strains are generally larger for glass fibres than for carbon fibres at comparable dimensions [50], so this is also not likely to be the cause. Differences in the matrix behaviour, in terms of the room temperature crystallinity and resulting properties, and in the fibre–matrix interface might contribute to explain this observation.

As well as consideration of the actual peeling process, successful implementation of a demanufacturing process also requires a means of creating the initial delamination crack. Several studies have considered methods of doing so, from simple impact to shearing, ultrasonic cutting and the use of a pre-installed delamination initiator, such as an embedded expanding material [24,25,51]. Further, it is necessary to consider the potential for peel deviation between plies, that might result in a thickening or thinning of the recovered material. This is unlikely to occur in woven or UD structures, since there are continuous fibres in the sub-ply along the axis of the peel. However, most components produced with UD tape will have different orientations between plies. Mitigating this issue may require enhanced temperature control [27] and/or mechanical restriction of sub-ply such as a wedge [18,22].

4.2. Quality of demanufactured tapes

As shown in Table 4, surface assessment of the demanufactured tapes demonstrated that the process of peeling causes a disruption to the initially relatively smooth surface of the material. The HT peel caused a greater increase in the average surface roughness than the RT peel,

though the variability was high in both cases. The SEM images in Figs. 6–8 indicate that roughness increases for the RT peeled tapes is at least partially as a result of adhesive failure and spalling of polymer, creating a stepped surface with areas of glass and areas of polymer. The HT surfaces show undulation and splitting microscopically, which is also readily apparent macroscopically along with splitting and waviness (Fig. 5).

This change in surface character means that direct re-use of the tape in an automated process, reliant on tension control and good quality polymer contact surfaces, might not be possible without some degree of post-processing, or without further improvement to the demanufacturing process. Typical surface roughness values for as-received tapes fall in the region between 5 μm and 35 μm [52,53], somewhat lower than the 25 μm to 95 μm mean range measured for demanufactured tapes in this study. A lower surface roughness is important in achieving rapid and complete bonding, by minimising time to achieve intimate contact [54]. Excessive surface roughness may also contribute to trapping air between plies, leading to potential manufacturing defects. If surface roughness cannot be reduced during demanufacturing, an additional processing step might be necessary, such as the re-pass approach [55]. Alternatively, offline secondary re-processing of the tape may be necessary to improve alignment and to redistribute matrix polymer, for example, by die pultrusion or double belt pressing.

In considering re-use of the tapes, when unwinding larger structures such as pipes and cylinders with ‘endless’ tapes the resulting tape spool would be of good length for re-use and these would be ideal initial targets for this technology. However, in considering smaller parts, with discrete lengths of tape as long as the part is wide, the resulting recovered material would be short lengths of tape. Effective use of this recovery technology would be dependent on finding suitable and economically viable uses for short tape lengths. This is already an area of research as automated tape processes create waste through leftover tape on reels, with research ongoing in patch placement [56–58], patch repair [59,60] and the effects of discontinuities in composite components [61–63].

4.3. Performance of remanufactured composites

While the quality of the recovered tapes does not match those of their as-received equivalents, their use in a manually loaded press moulding demonstrated final composite properties that were generally on a par with those of composites produced from as-received tape.

Torsional testing (Fig. 9 and Fig. 10) was employed as it can help highlight changes to the polymer matrix behaviour. The similarities in behaviour between as-received and peeled specimens, after re-pressing into six-ply composite panels, indicate that in most systems the effect of peeling on the matrix is fairly limited. An apparent reduction in room temperature shear modulus was observed in the hot-peeled PP/glass relative to the as-received, as well as a broadening of the loss factor tangent peak close to melt. This would suggest that a small degree of matrix degradation or fibre–matrix interface degradation, as well as some fibre disruption or damage, is likely to have occurred. SEM indicates that there is already very little matrix–interface strength (Fig. 6). DSC assessment (Table 5) does not indicate a change in melting point (T_m) as a result of the heated peel, but a significant reduction in molecular weight would be needed to impact T_m [64]. This would point to fibre alignment as a primary variable affecting the torsion results.

The PA6/carbon UD remanufactured composite reported a 14 % $\pm$ 7 % larger shear modulus than the as-received manufactured, a factor mostly attributed to an increase in the fibre volume fraction. This is associated with matrix squeeze-out during moulding, something that only occurred noticeably with this particular combination of matrix and compression-moulding lay-up where there are no transverse fibres.

The results observed for flexural testing (Fig. 11 and Fig. 12) provided more insight into the fibre effects. Overall, with the exception of the PA6 UD, there was a trend of slightly lower flexural properties for

the panels that were produced with peeled tapes; up to a 26 % $\pm$ 8 % reduction was observed in the remanufactured HT peel average flexural strength, in comparison to as-received properties. The reasons for this may be in part due to small differences in fibre volume fraction, as well as off-axis fibres observed in the peel surfaces (Figs. 6–8) and macroscopic transverse waviness in the peeled tapes (Fig. 5) creating a degree of off-axis reinforcement. In studies on reforming woven materials, Joustra et al. observed some waviness in their specimens, but found that the effects on resulting flexural properties were relatively small [15]. However, in the present study, there was no woven structure to help mitigate the potential fibre shifting, and higher compaction pressures were applied.

The burn-off measurements showed that there was a small increase in the presence of voids for the re-consolidated PBT specimens, but this was the case both for the RT and HT specimens. The exception to the drop in properties was the UD HT peel for the PA6, which showed an increase in flexural strength consistent with the increase in fibre volume fraction.

5. Conclusions

This study has begun to explore the viability of peeling fibre-reinforced thermoplastic tapes with matrices of PP, PBT and PA6 and fibres of either glass or carbon, in order to facilitate the demanufacture of composite components and produce tapes suitable for remanufacturing. This initial, limited study has demonstrated that peel recovery of UD thermoplastic tapes from 2-ply specimens can be achieved with low forces and potentially high rates, in some cases at room temperature, and in all cases when using a local heating approach. Recovery of the tapes incurs a penalty in tape surface quality and in fibre alignment.

Six-ply composite panels were compression-moulded both from as-received tapes and from demanufactured tapes, for the purpose of evaluating and comparing the mechanical properties. While the study was limited to the assessment of a small number of composite panels, measurements of torsional stiffness suggested that recovery and re-processing of the tapes does not appear to cause major changes in the matrix behaviour of the panels. There was an indication of a small amount of degradation of the PP during the high temperature peel, but the reasons could not be established conclusively. A small knockdown in flexural strength was observed in the re-processed panels relative to panels manufactured from as-received tapes, which was attributed to variations an increase in voids and to the loss of fibre alignment.

Improved fibre orientation and mitigation of potential matrix damage could be addressed in the design of a more controlled demanufacturing set-up, which will be established in our future works.

CRedit authorship contribution statement

Andrew J. Parsons: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Michael S. Johnson:** Writing – review & editing, Methodology. **Samanta Piano:** Writing – review & editing, Supervision, Resources, Methodology. **Davide S.A. De Focatiis:** Writing – review & editing, Visualization, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.compositesa.2026.110072>.

Data availability

Data from this study is hosted on Mendeley and can be found at: DOI: 10.17632/v9jbs6d757.1

References

- [1] G. Gardiner, Recycled thermoplastic composites for production, *Composites World*, (2019), <https://www.compositesworld.com/articles/recycled-thermoplastic-composites-for-production>, Date accessed 08/05/2025.
- [2] M. Richardson, Report says sustainability is driving use of thermoplastics, *Composites in Manufacturing*, (2025), <https://www.composites.media/report-says-sustainability-is-driving-use-of-thermoplastics>, Date accessed 08/05/2025.
- [3] Almushaikeh AM, Alswad SO, Alsuhybani MS, Alotaibi BM, Alarifi IM, Alqahtani NB, et al. Manufacturing of carbon fiber reinforced thermoplastics and its recovery of carbon fiber: a review. *Polym Test* 2023;122:108029. <https://doi.org/10.1016/j.polymertesting.2023.108029>.
- [4] Bernatas R, Dagreou S, Despax-Ferreres A, Barasinski A. Recycling of fiber reinforced composites with a focus on thermoplastic composites. *Cleaner Eng Technol* 2021;5:100272. <https://doi.org/10.1016/j.clet.2021.100272>.
- [5] Jagadeesh P, Mavinkere Rangappa S, Siengchin S, Puttegowda M, Thiagamani SMK, Rajeshkumar G, Hemath Kumar M, Oladijo OP, Fiore V, Moure Cuadrado MM. Sustainable recycling technologies for thermoplastic polymers and their composites: a review of the state of the art. *Polym Compos* 2022;43:5831–62. <https://doi.org/10.1002/pc.27000>.
- [6] Krauklis AE, Karl CW, Gagani AI, Jorgensen JK. Composite material recycling technology—state-of-the-art and sustainable development for the 2020s. *J Compos Sci* 2021;5:28. <https://doi.org/10.3390/jcs5010028>.
- [7] Pakdel E, Kashi S, Varley R, Wang X. Recent progress in recycling carbon fibre reinforced composites and dry carbon fibre wastes. *Resour Conserv Recycl* 2021;166:105340. <https://doi.org/10.1016/j.resconrec.2020.105340>.
- [8] Qureshi J. A review of recycling methods for fibre reinforced polymer composites. *Sustainability* 2022;14:16855. <https://doi.org/10.3390/su142416855>.
- [9] Shehab E, Meirbekov A, Amantayeva A, Tokbolat S. Cost modelling for recycling fiber-reinforced composites: state-of-the-art and future research. *Polymers* 2023;15:150. <https://doi.org/10.3390/polym15010150>.
- [10] Zhang J, Chevali VS, Wang H, Wang C-H. Current status of carbon fibre and carbon fibre composites recycling. *Compos B Eng* 2020;193:108053. <https://doi.org/10.1016/j.compositesb.2020.108053>.
- [11] Shieh P, Zhang W, Husted KEL, Kristufek SL, Xiong B, Lundberg DJ, et al. Cleavable comonomers enable degradable, recyclable thermoset plastics. *Nature* 2020;583:542–7. <https://doi.org/10.1038/s41586-020-2495-2>.
- [12] Ma S, Webster DC. Degradable thermosets based on labile bonds or linkages: a review. *Prog Polym Sci* 2018;76:65–110. <https://doi.org/10.1016/j.progpolymsci.2017.07.008>.
- [13] Jenkins PG, Yang L, Ligat JJ, Thomason JL. Investigation of the strength loss of glass fibre after thermal conditioning. *J Mater Sci* 2015;50:1050–7. <https://doi.org/10.1007/s10853-014-8661-x>.
- [14] Vincent GA, Bruijn TD, Wijskamp S, Rasheed MIA, Van Drongelen M, Akkerman R. Characterisation and improvement of the quality of mixing of recycled thermoplastic composites. *Composites, Part C: Open Access* 2021;4:100108. <https://doi.org/10.1016/j.jcomc.2021.100108>.
- [15] Joustra J, Brans K, Fernandez Villegas I, Sinke J, Teuwen J. Reverse forming thermoplastic composites: Design and process development. *Composites, Part C: Open Access* 2025;16:100550. <https://doi.org/10.1016/j.jcomc.2024.100550>.
- [16] Kiss P, Stadlbauer W, Burgstaller C, Stadler H, Fehrer S, Haeuserer F, et al. In-house recycling of carbon- and glass fibre-reinforced thermoplastic composite laminate waste into high-performance sheet materials. *Compos A Appl Sci Manuf* 2020;139:106110. <https://doi.org/10.1016/j.compositesa.2020.106110>.
- [17] Design and Manufacture of Structural Composites, 1st Edition ed, Woodhead Publishing, 2022.
- [18] H. Janssen, Method And Device For Recycling Thermoplastic Fibre - Reinforced Composite Material, (2020), US 10,723,043.
- [19] Schyns ZOG, Shaver MP. Mechanical recycling of packaging plastics: a review. *Macromol Rapid Commun* 2021;42:2000415. <https://doi.org/10.1002/marc.202000415>.
- [20] Yin S, Tuladhar R, Shi F, Shanks RA, Combe M, Collister T. Mechanical reprocessing of polyolefin waste: a review. *Polym Eng Sci* 2015;55:2899–909. <https://doi.org/10.1002/pen.24182>.
- [21] Biron M. *Thermoplastics and Thermoplastic Composites*. Elsevier; 2007.
- [22] P. Jongebloed, J. Schafaschek, H. Janssen, C. Brecher, Characterization of recycled thermoplastic unidirectional tapes, 6th International Conference on Thermoplastic Composites, (2022) Bremen, Germany.
- [23] R. Grenier, M. Imbert, J. Hohe, F. Balle, M. May, Thermally Assisted Peeling As A High-Quality Disassembly Process For Thermoplastic Tape Wound Composites, 23rd International Conference on Composite Materials, (2023) Belfast, UK.
- [24] Imbert M, Hahn P, Jung M, Balle F, May M. Mechanical laminae separation at room temperature as a high-quality recycling process for laminated composites. *Mater Lett* 2022;306:130964. <https://doi.org/10.1016/j.matlet.2021.130964>.
- [25] Ragupathi B, Bacher MF, Balle F. Separation and reconsolidation of thermoplastic glass fiber composites by power ultrasonics. *Resour Conserv Recycl* 2023;198:107122. <https://doi.org/10.1016/j.resconrec.2023.107122>.
- [26] Michel S, Hauf F, Brunner AJ. Evaluation of a climbing drum laminate peel test to determine the interlaminar mode I fracture toughness of thin CFRP laminates—comparison with the standard mode I DCB test and a mandrel laminate peel test proposed by ESIS TC4. *Front Mater* 2023;10. <https://doi.org/10.3389/fmats.2023.981647>.
- [27] Parsons AJ, Johnson MS, Piano S, De Focatiis DSA. Demanufacturing of thermoplastic composites: small scale experiments exploring ply separation and recovery. *Compos A Appl Sci Manuf* 2025;190:108626. <https://doi.org/10.1016/j.compositesa.2024.108626>.
- [28] Digital Surf. (2025). Mountains software. Version 7.4.9391. www.digitalsurf.com/support/software-updates.
- [29] Fujiyama K, Huang Z-M, Ramakrishna S, Hamada H. Influence of processing conditions on bending property of continuous carbon fiber reinforced PEEK composites. *Compos Sci Technol* 2004;64:2525–34. <https://doi.org/10.1016/j.compscitech.2004.05.014>.
- [30] R.E. Lyon, M.L. Janssens, Polymer Flammability, W. Office of Aviation Research, D. C. 20591, DOT/FAA/AR-05/14.
- [31] Liu L, Huang P, Miao M, Wuyun Q, Li T, Su F. On the oxidative ablation and mechanical properties of T300 carbon fibers at elevated temperatures. *Mater Perform Charact* 2018;7:20170117. <https://doi.org/10.1520/MPC20170117>.
- [32] Stocco A, La Carrubba V, Piccarolo S, Brucato V. The solidification behavior of a PBT/PET blend over a wide range of cooling rate. *J Polym Sci B* 2009;47:799–810. <https://doi.org/10.1002/polb.21687>.
- [33] F.T. Wallenberger, J.C. Watson, H. Li, Glass Fibers, in: D.B. Miracle, S.L. Donaldson, (Eds.), *ASM Handbook Volume 21: Composites*, ASM International, 2001.
- [34] M. Biron, Chapter 4 - Detailed accounts of thermoplastic resins, in: M. Biron, (Ed.), *Thermoplastics and Thermoplastic Composites*, Elsevier, Oxford, 2007, pp. 217–714.
- [35] Afaghi-Khatibi A, Mai Y-W. Characterisation of fibre/matrix interfacial degradation under cyclic fatigue loading using dynamic mechanical analysis. *Compos A Appl Sci Manuf* 2002;33:1585–92. [https://doi.org/10.1016/S1359-835X\(02\)00117-3](https://doi.org/10.1016/S1359-835X(02)00117-3).
- [36] Zhang C, Duan Y, Xiao H, Wang B, Ming Y, Zhu Y, et al. The effects of processing parameters on the wedge peel strength of CF/PEEK laminates manufactured using a laser tape placement process. *Int J Adv Manuf Technol* 2022;120:7251–62. <https://doi.org/10.1007/s00170-022-09181-5>.
- [37] Parodi E, Govaert LE, Peters GWM. Glass transition temperature versus structure of polyamide 6: a flash-DSC study. *Thermochim Acta* 2017;657:110–22. <https://doi.org/10.1016/j.tca.2017.09.021>.
- [38] Esmizadeh E, Tzoganakis C, Mekonnen TH. Degradation behavior of polypropylene during reprocessing and its biocomposites: thermal and oxidative degradation kinetics. *Polymers* 2020;12:1627.
- [39] Montaudo G, Puglisi C, Samperi F. Primary thermal degradation mechanisms of PET and PBT. *Polym Degrad Stab* 1993;42:13–28. [https://doi.org/10.1016/0141-3910\(93\)90021-A](https://doi.org/10.1016/0141-3910(93)90021-A).
- [40] Ren P, Li T, Feng L, Gu X, Jiang M, Duan J, et al. Thermal degradation of polyamide 6: mechanisms, mitigation strategies, and challenges. *Chem Eng Sci* 2025;316:121985. <https://doi.org/10.1016/j.ces.2025.121985>.
- [41] Jones RM. *Mechanics of composite materials*. New York: Hemisphere Publishing Corp; 1975.
- [42] Hashin Z. The elastic moduli of fiber-reinforced materials. *J Appl Mech* 1964;31:223–32. <https://doi.org/10.1115/1.3629590>.
- [43] Rezaee M, Tsai L-C, Haider MI, Yazdi A, Sanatizadeh E, Salowitz NP. Quantitative peel test for thin films/layers based on a coupled parametric and statistical study. *Sci Rep* 2019;9:19805. <https://doi.org/10.1038/s41598-019-55355-9>.
- [44] Alcock B, Cabrera NO, Barkoula N-M, Loos J, Peijs T. Interfacial properties of highly oriented coextruded polypropylene tapes for the creation of recyclable all-polypropylene composites. *J Appl Polym Sci* 2007;104:118–29. <https://doi.org/10.1002/app.24588>.
- [45] Sako N, Matsuoka T, Sakaguchi K. The effect of interface on fracture mechanism of GF/PP composites using O2 plasma treatment. *Compos Interfaces* 1996;4:401–15. <https://doi.org/10.1163/156855497X00235>.
- [46] Ji X, Matsuo S, Sottos NR, Cahill DG. Anisotropic thermal and electrical conductivities of individual polyacrylonitrile-based carbon fibers. *Carbon* 2022;197:1–9. <https://doi.org/10.1016/j.carbon.2022.06.005>.
- [47] Längauer M, Brunnthaller F, Zitzenbacher G, Burgstaller C, Hochenauer C. Modeling of the anisotropic thermal conductivity of fabrics embedded in a thermoplastic matrix system. *Polym Compos* 2021;42:2050–60. <https://doi.org/10.1002/pc.25958>.
- [48] Tahsini AM, Tadayon Mousavi S. Hot air gun identification by inverse heat transfer. *J Space Sci Technol* 2016;8:29–34.

- [49] Craig W. Ohlhorst, Wallace L. Vaughn, P.O. Ransone, H.-T. Tsou, Thermal Conductivity Database of Various Structural Carbon-Carbon Composite Materials, 19970041399, NASA Technical Memorandum 4787.
- [50] Monjon A, Santos P, Valvez S, Reis PNB. Hybridization effects on bending and interlaminar shear strength of composite laminates. *Materials* 2022;15:1302. <https://doi.org/10.3390/ma15041302>.
- [51] Von Freeden J, Erb J, Schleifenbaum M. Separating layer recycling strategy for continuous fiber reinforced thermo-sets based on thermally expanding particles. *Polym Compos* 2022;43:1887–99. <https://doi.org/10.1002/pc.26505>.
- [52] Fereidouni M, Hoa SV. In-situ consolidation of thermoplastic composites by automated fiber placement: characterization of defects. *J Thermoplast Compos Mater* 2025;38:471–511. <https://doi.org/10.1177/08927057241251837>.
- [53] Zaami A, Baran I, Bor TC, Akkerman R. Optical characterization of fiber-reinforced thermoplastic tapes for laser-based composite manufacturing. *Compos A Appl Sci Manuf* 2021;146:106402. <https://doi.org/10.1016/j.compositesa.2021.106402>.
- [54] Yassin K, Hojjati M. Processing of thermoplastic matrix composites through automated fiber placement and tape laying methods: A review. *J Thermoplast Compos Mater* 2018;31:1676–725. <https://doi.org/10.1177/0892705717738305>.
- [55] Shadmehri F, Hoa SV, Fortin-Simpson J, Ghayoor H. Effect of in situ treatment on the quality of flat thermoplastic composite plates made by automated fiber placement (AFP). *Adv Manuf Polym Compos Sci* 2018;4:41–7. <https://doi.org/10.1080/20550340.2018.1444535>.
- [56] Rettenwander T, Fischlschweiger M, Machado M, Steinbichler G, Major Z. Tailored patch placement on a base load carrying laminate: a computational structural optimisation with experimental validation. *Compos Struct* 2014;116:48–54. <https://doi.org/10.1016/j.compstruct.2014.04.028>.
- [57] . Gardiner, Fiber Patch Placement at scale, *Composites World*, (2018), <https://www.compositesworld.com/articles/fiber-patch-placement-at-scale>.
- [58] Coquel M. Efficient additive manufacturing of long fiber composite reinforcements using Fibre Patch Placement, 6th European conference for aeronautics and space sciences (EUCASS). Poland: Kraków; 2015.
- [59] Zhao T, Xu S, Feng Y, Zhang C, Huang Y, Ren X, et al. Towards ultra-fast and high strength structural repair of damaged thermoplastic composites: ultrasonic welding. *Compos B Eng* 2025;298:112385. <https://doi.org/10.1016/j.compositesb.2025.112385>.
- [60] Modi V, Bandaru AK, Ramaswamy K, Kelly C, McCarthy C, Flanagan T, et al. Repair of impacted thermoplastic composite laminates using induction welding. *Polymers* 2023;15:3238. <https://doi.org/10.3390/polym15153238>.
- [61] Kussmaul R, Zogg M, Ermanni P. A failure mechanics and strength optimization study for patched laminates. *Compos Struct* 2019;226:111165. <https://doi.org/10.1016/j.compstruct.2019.111165>.
- [62] Zhang X, Ji C, Chi X, Sun Z, Lei M. Study on the tensile properties and damage analysis of patch laminated composite based on deep neural networks. *Mech Compos Mater* 2025;61:665–82. <https://doi.org/10.1007/s11029-025-10301-x>.
- [63] Joesbury AM, Nagarajan YR, Molinar-Díaz J, Parsons AJ, Qian C, Harper LT. Maximising strength retention for carbon fibre prepreg scraps: a patch-based laminate approach. *J Compos Mater* 2026;00219983261433464. <https://doi.org/10.1177/00219983261433464>.
- [64] Van Der Wal A, Mulder JJ, Thijs HA, Gaymans RJ. Fracture of polypropylene: 1. The effect of molecular weight and temperature at low and high test speed. *Polymer* 1998;39:5467–75. [https://doi.org/10.1016/S0032-3861\(97\)10278-6](https://doi.org/10.1016/S0032-3861(97)10278-6).