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In-line variable spreading of carbon fibre/thermoplastic pre-preg tapes for application in automatic tape placement

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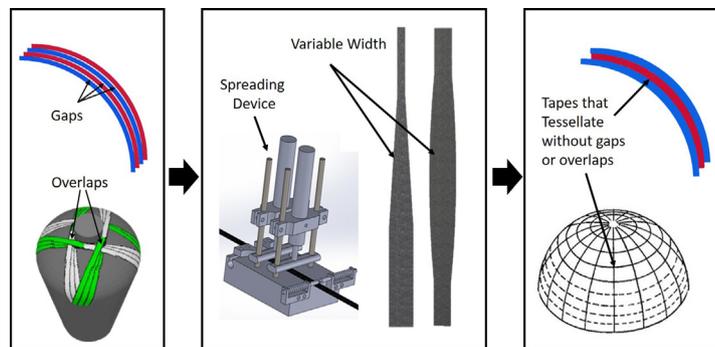
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HIGHLIGHTS

- Design of device is proposed which can in-situ spread carbon fibre/PEEK pre-preg tapes with use of automated tape placement.
- Spreading could be a solution to eliminate gaps and overlaps in variable angle tow laminates and doubly curved structures.
- Spreading appears to reduce void content of pre-preg tapes.
- Spreading increases the degree of crystallinity of carbon fibre/PEEK pre-preg tapes.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigates a device for in-line continuous spreading of carbon fibre/thermoplastic pre-preg tape for potential application in the Laser-Assisted Automatic Tape Placement (LATP) laminate manufacturing process. The spreading device allows variable tape width to be achieved locally during lay-up. Integration of this device in the LATP process would remove gap and overlap manufacturing defects in variable angle tow (VAT) laminates and complex curvature components. During trials different width tapes were produced using the novel spreading device. Three different width increases were investigated, viz. 15%, 30% and 45%, and were compared with as-received tape. Initial trials indicate that it is possible to achieve a tape width increase of 62%. Preliminary characterisation tests show that the spreading process does not adversely affect the properties of the tapes. Physical properties including cross-sectional area, fibre volume fraction and void content remain similar to as-received tape. Furthermore, differential scanning calorimetry data show that levels of crystallinity increase due to spreading, improving related mechanical properties.

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1. Introduction

There is an increasing demand to produce composite components using out-of-autoclave methods that are both time and cost efficient [1]. Laser-Assisted Automatic Tape Placement (LATP) in-situ consolidation of carbon fibre/thermoplastic pre-preg tapes is an out-of-autoclave

process that can produce high performance composite structures. LATP allows greater control of fibre orientation, and it also enables Variable Angle Tow (VAT) laminates to be produced. In VAT layers, fibres are orientated in curvilinear paths so as to improve stress distributions and align with desired load paths, providing excellent performance without increasing weight [2–6]. However, VAT laminates are prone to manufacturing defects such as fibre wrinkling, fibre pull-up, gaps and overlaps [7]. Fibre wrinkling and fibre pull-up are avoided by manufacturing steered laminates with a sufficiently large radius, that

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may limit performance. Gaps are caused by offsetting the reference path of consecutive tapes resulting in misalignment of their edges. Overlaps are created by not fully offsetting the start point of each tape deposition, resulting in neighbouring tapes overlapping. Several authors completed studies to investigate the effect of gaps on the mechanical properties of a constant stiffness laminate made by Automated Fibre Placement (AFP). It was found that gaps reduce both laminate strength and average strain to failure [8].

A previous study completed by Clancy et al. [9] shows that it is possible to manufacture VAT laminates with carbon fibre polyether ether ketone (CF/PEEK) pre-preg tapes without incurring fibre wrinkling or buckling issues for a minimum steering radius of 400 mm on a tape width of 6 mm. Notably, this value is smaller than that reported for carbon fibre/epoxy tape using ATP/AFP processes, which is 508 mm [8], and in doing so, offers potential for significant performance enhancement by allowing greater levels of load redistribution. Fibre reinforced thermoplastics, such as CF/PEEK, have several other benefits compared to thermoset systems, such as in-situ consolidation, recyclability, excellent fracture toughness and eliminating the need for frozen storage [1]. In addition, our proposed method of in-situ spreading of CF/PEEK pre-preg tapes during the LAMP process, which involves adding a spreading step before the consolidation process, can eliminate gaps and overlaps, providing the prospect of tessellated tapes within a layer. By using an additional compaction roller and heated platen, the width of the incoming tape can increase to fill the corresponding gap between successive neighbouring steered tapes. Processing variables such as pressure, heat and rate can be altered to vary the width as the tape is steered. Tape spreading also gives the ability to manufacture wrinkle-free doubly-curved surfaces such as domes, which can be used in applications such as aircraft noses and nacelles. For such structures, tapes need to be wider at the base than the pole, as shown in Fig. 1. Varying tape width can be beneficial as it produces doubly-curved surfaces without gaps, overlaps or fibre cuts, resulting in an overall more efficient structure with improved geometric tolerances. Furthermore, spreading of CF/PEEK tapes, due to conservation of volume, allows tailoring of thickness distributions to meet specific performance requirements, including tuning thickness and fibre orientation independently of each other. Tuning thickness by spreading is beneficial as it minimises stress and strain concentrations due to local fibre cuts (ply drops), improving both strength and damage tolerance properties.

Several studies have previously investigated the spreading of dry fibre tows to manufacture thinner pre-preg tapes. This process has involved dry tows that were spread by using several rollers; the tows were subsequently impregnated with resin by passing them through a resin bath. Wilson [10] derived a relationship showing that the width of a spread tow depends on the lateral distance and angle between two rollers. However, the expression developed indicates that the

width of the spread dry tow does not depend on the tension applied. However, Irfan et al. [11] completed a further study that showed that tension applied to the dry tow does affect the amount of tow spreading. They also completed a number of experimental studies, where dry fibre tows were passed through several different roller configurations to identify the set-up that provides the greatest amount of spreading. A schematic diagram of their test set-up is shown in Fig. 2, which includes the ability to vary length and angle between rods.

Related work includes numerous studies [12–15] that investigated the deformation of viscous pre-preg tapes while pressure is applied. In particular, the effect of processing conditions, temperature as well as pressure, from autoclave and automated tape placement on thermoplastic pre-preg tapes was studied. Increasing temperature makes the resin increasingly viscous and then various levels of pressure consolidate the tape layers together. However, applying a downward pressure to a viscous pre-preg tape also creates a transverse squeeze flow effect in both resin and fibres, which deforms the tapes by increasing width and decreasing thickness. Squeeze flow describes the shear deformation of viscous materials and the effects of which are often measured using rheometry [16]. When constrained by fibres the flow mechanisms become anisotropic and transverse squeeze flow describes the main mechanism which allows resin to coalesce across laminae, resulting in good interfacial bonding. However, excessive flow may induce resin or fibre migration and adversely affect mechanical properties, dimensions and integrity of the final product [12]. Therefore, the effects that different temperatures and pressures, have on squeeze flow of pre-preg tapes is an important consideration. Transverse squeeze flow is also the main mechanism by which width and thickness change during spreading of pre-preg tapes. Many of the previous studies investigating squeeze flow of pre-preg tapes completed a static analysis, where a stationary pre-preg tape is compressed and gradually heated between two platens. These studies analysed the influence that parameters such as pressure, temperature, time, fibre content and fibre orientation have on squeeze flow characteristics, namely geometrical changes in width and thickness and also fibre dispersion. Previous studies [12–14] conclude that squeeze flow only occurs perpendicular to fibre direction, as the fibres are stiffer than the viscous matrix. Two of these studies [12,13] show that fibre content prevents squeeze flow, due to the constraint of relatively stiff fibres. Additionally, one of the studies [13] showed that fibre orientations affects squeeze flow. When two 0° plies are adjacent to each other, transverse flow perpendicular to the fibres is unrestricted. However, when a 0° ply is stacked on top of a 90° ply, squeeze flow is restricted in both plies as fibres are perpendicular to each other confining the viscous resin.

Wang & Gutowski [14] investigated the elimination of gaps and overlaps in laminates produced using ATP. During tape lay-up, laps and gaps result from inherent machine and human inaccuracies, as well as by the inability of the tape to conform to complex geometries. Their study examines whether these flaws can be removed during processing by transverse flow processes during consolidation. Samples of thermoplastic composites were compressed in a static testing device, and deformation measured. A pressure of 1.4 MPa based on the mould dimensions, and a consolidation time of 10 min were used for all experiments at the manufacturer's recommended consolidation temperature of 390°C . Experimental results were also compared with mathematical models using the flow of a power law fluid to predict transverse shear flow. Findings from their study showed that it is plausible that transverse shear flow could be used to fill gaps between adjacent tows. However, it is more complicated to eliminate overlaps using transverse shear flow. Analysis also showed that shear flow is time dependent, initially an increase in the consolidation time gives an increase in the maximum allowable overlap or gap, but that the rate of increase diminishes with time. Due to the nature of the flow, transverse spreading initially occurs very rapidly but soon reduces, indicating that while processing with ATP, slower lay-down speeds could result in increased spreading. Wang & Gutowski [14] also show that shear within the interior of the

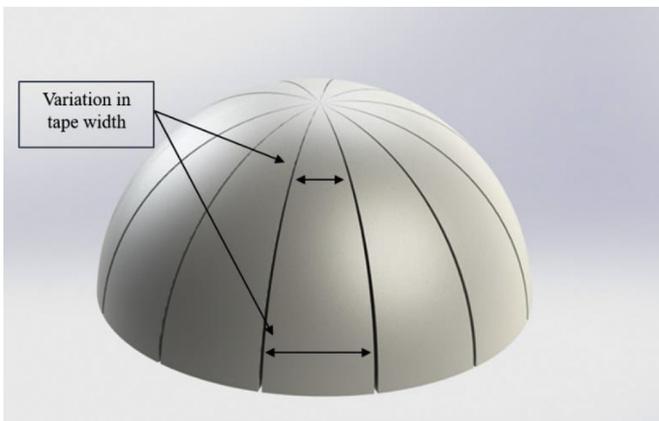


Fig. 1. Example of how segment of dome varies in width from base to tip.

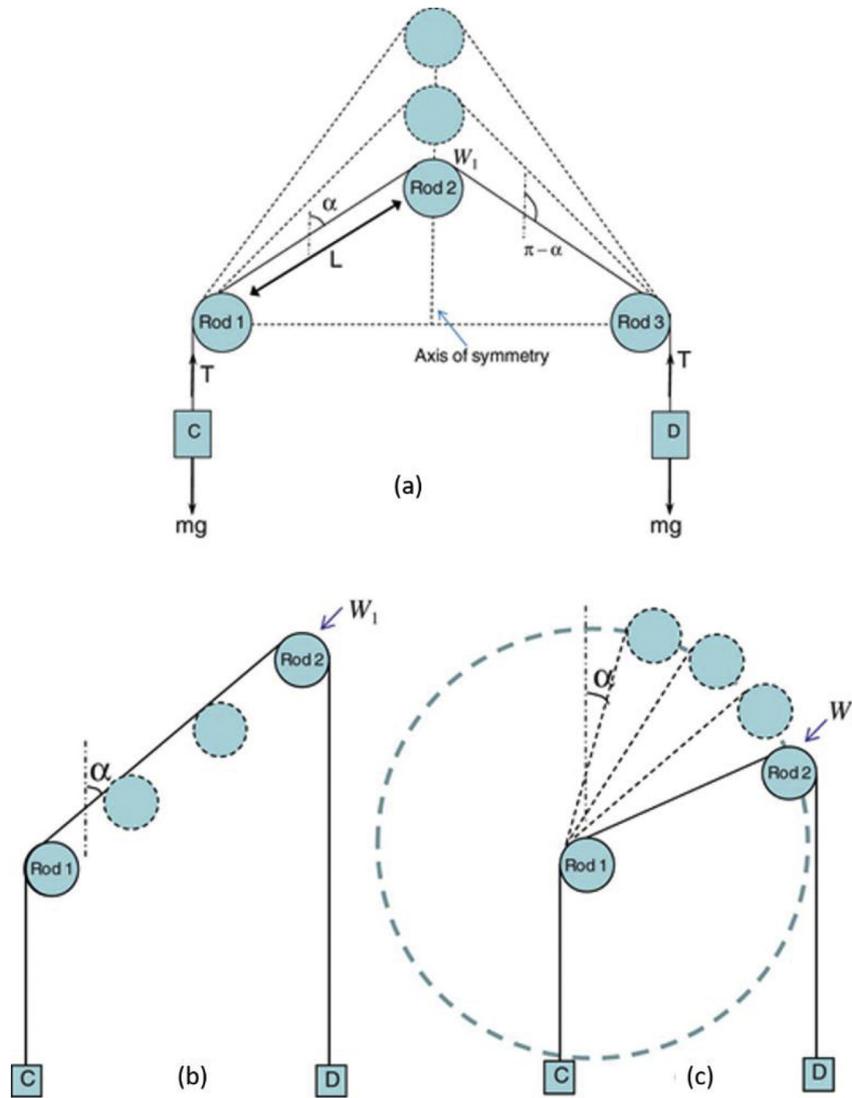


Fig. 2. (a) Varying length & angle between rods, (b) varying length between rods, (c) varying angle between rods [11].

composite layer occurs and that deformation is not on the upper and lower surfaces of the pre-preg tape. Modelling, which is in good agreement with prediction of elimination of gaps, shows that narrower tapes produce a larger increase in width than wider tapes when using the same consolidation pressure, as expected since the downward force exerts over a smaller area.

The majority of these studies carried out steady-state experiments, where tapes were held static during testing, whereby pre-preg tapes were compressed and gradually heated between two platens and resulting deformation measured, similarly to that shown in Fig. 3. This type of experiment is more representative of autoclave processing and not ATP, where tapes are rapidly heated and placed into position with a constantly moving head. One particular study [15] investigated squeeze flow of carbon fibre/PEEK tapes for the application of ATP. Deformation of tape was analysed after being processed by an ATP head with varying degrees of temperature (370 °C, 385 °C, 400 °C), consolidation force (10 kg, 25 kg, 40 kg), laydown speed (16.5 mm/s, 33.5 mm/s, 50 mm/s) and fibre angle of the substrate (0°, 45°, 90°). They found that temperature had little effect on squeeze flow as viscosity changed insignificantly once the melt temperature was reached. Pressure imparted by consolidation roller and fibre angle were found to have greatest influence. In the case of a 40 kg consolidation force and 0° fibre angle

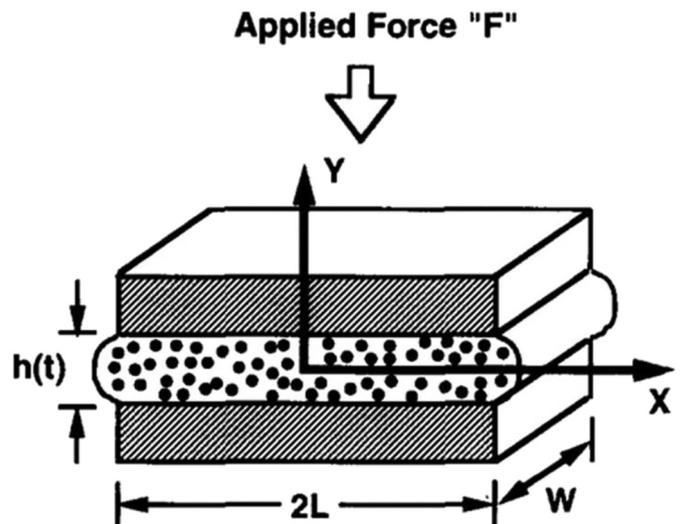


Fig. 3. Example of steady state experimental set up to measure squeeze flow [13].

substrate, a 48% average increase in width was observed. However, the authors do note that this consolidation force is 'moderately high' in comparison to usual ATP processing parameters.

As well as eliminating gaps in VAT laminates, spreading can also have the beneficial effect of improving mechanical properties of conventional constant fibre angle laminates. Spreading of fibres and matrix creates more consistent fibre dispersion, reducing resin-rich areas. As volume is conserved, spreading pre-preg makes thinner plies. It is noteworthy that thinner plies have less load transferred to free edges, thereby increasing interlaminar shear strength. Thin laminae were also reported to suppress both micro-cracking and delamination damage [17]. Thinner plies also provide more choice in optimising laminate structure, as more layers are required for a given laminate thickness. Sih et al. [18] completed a study where laminates were manufactured with different numbers of layers but the same total thickness, and where the thin-ply laminate comprises laminae five times thinner than the thick laminate. Results showed that the thin-ply laminate displays higher strain to failure and greater tensile strength for a quasi-isotropic layup as it was capable of delaying the onset of micro cracking and delamination. Additionally, the laminate with thinner layers improved fatigue performance showing lower levels of micro-cracking and greater residual strength after 50,000 cycles at 60% of the ultimate tensile strength. This study highlights that spreading can improve the performance of constant fibre angle laminates.

1.1. Motivation of this study

The motivation for this study originates from previous work by Zucco et al. [19], who investigated the effect of variable angle tows in a composite wingbox (Fig. 4(a)). VAT plies were introduced into the unsupported skin between stringers to redistribute bending loads to supported (stringer) regions to delay the onset of buckling. An experimental static test validated finite element models. The study successfully showed that the buckling load for the VAT wingbox increases by 14% compared to a constant fibre angle wingbox. However, gaps between neighbouring steered tows on the wingbox occurred due to the mismatch of steering radius between the inner and outer edge of steered tapes (Fig. 4(b)). These gaps resulted in steps between tapes, creating potential stress concentrations. Eliminating gaps could yield a further increase in the performance of the wingbox. Incorporating a spreading process into the LAMP head mechanism before the tapes are consolidated into place would be highly beneficial in this case, as shown in Fig. 5. The current study focuses on the development and feasibility of a tape spreading device, which can be integrated into an LAMP head. The spreading device comprises a pneumatic actuator, compaction shoe, a guide and a heated platen. The platen is heated up close to the melt temperature of PEEK, thus reducing the viscosity of the CF/PEEK tape. The CF/PEEK tape is then compressed by the compaction shoe, which is controlled by a pressure regulator. The pressure is

gradually increased to widen CF/PEEK tape by an amount corresponding to the changing gap between neighbouring steered tapes. A more in-depth description of this process is given in Section 2.

The work presented here advances preliminary research by Clancy et al. [20], investigating the spreading of CF/PEEK pre-preg tapes by using a new, additional step in the LAMP manufacturing process. The novelty of this study is the ability to continuously spread carbon fibre/PEEK pre-preg tapes with a device that can be integrated into an ATP head. To the best of the authors' knowledge, no previous work has been published which can continuously vary the width of carbon fibre/PEEK pre-preg tape with a separate device to the ATP compaction roller.

In this study, a geometrical study examines how spreading benefits steering capability. Initial characterisation tests examine the effect of spreading on the quality of the pre-preg tapes, for 0% (original), 15%, 30%, and 45% increases in width. Geometrical analysis, optical microscopy, scanning electron microscopy and differential scanning calorimetry characterise the quality of spread tapes. Section 2 describes applications where spreading could be utilized, specifically in the area of tow steering or VAT laminates. Section 3 gives details of the tape spreading process and how parameters are varied to vary the width of pre-preg tapes. Section 4 gives details of the experimental testing used to analyse does the spreading process negatively affect the quality of the pre-preg tapes. Results and discussions are presented in section 5 and finally conclusions are outlined in section 6.

2. Applications of spreading

To eliminate tape overlapping in steered plies, the location of the starting point of a neighbouring tape needs to be offset by the width of the tape in the horizontal and vertical direction (Fig. 6 (a) & 6 (b)). However, this step creates a discontinuity (i.e. gap) between centres of arc between first and second tape positions, which manifests itself as a gap between consecutive tapes (Fig. 6 (c)). The maximum gap between neighbouring steered tapes depends on the width of the tape itself. This relationship is illustrated in Fig. 6 (c), by offsetting the tape by one width in the y-direction and x-direction prevents the tape from overlapping. However, this action moves the centre point of the steering arc a distance given by the square root of twice the width squared in the 45° direction. The maximum width of a gap between adjacent steered tapes is the resultant vector minus the width of the tape, calculated by

$$\text{Max Gap width} = \sqrt{x^2 + y^2} - \text{Tape width} \quad (1)$$

The maximum percentage by which tapes need to be spread to eliminate the gap is the resultant vector minus the width of the tape, which is 41.4% of the initial starting width of the tape. As the tape is steered, the width of the tape can be increased gradually by spreading as it is consolidated into place. Once it reaches the location of the maximum

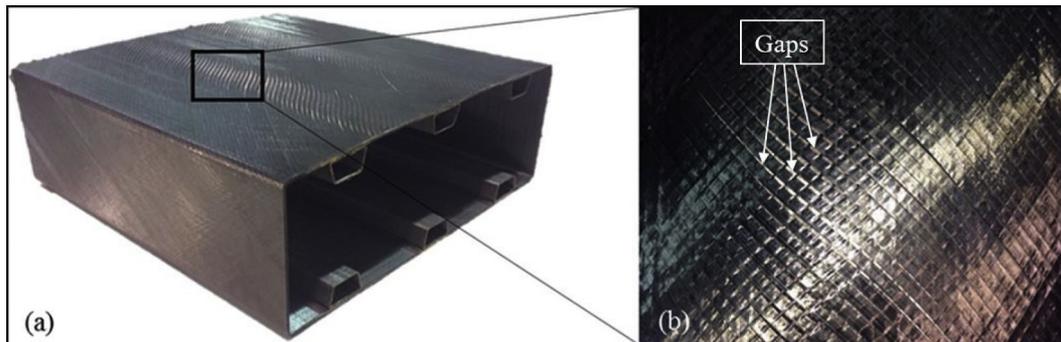


Fig. 4. (a) Composite wingbox with variable angle tow, (b) highlighting steered pattern with gaps.

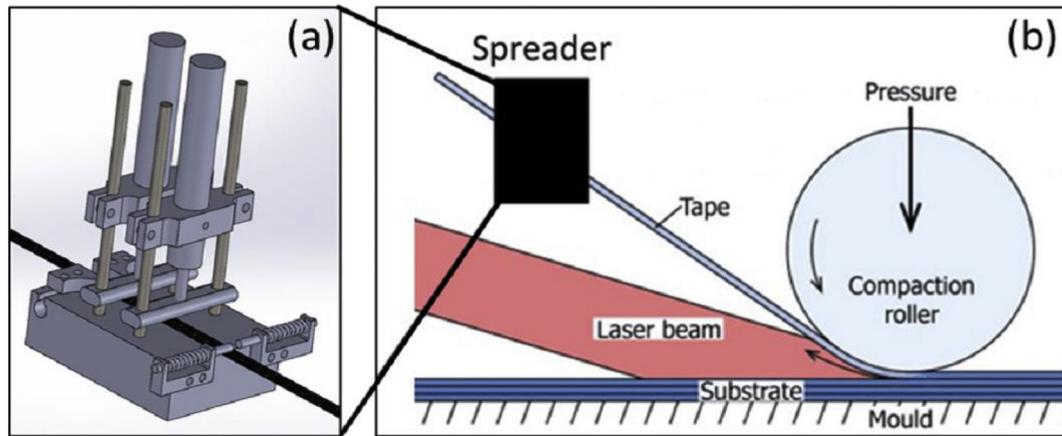


Fig. 5. (a) Spreading device, (b) spreading device incorporated into the LAMP machine.

gap, the width can be decreased gradually, resulting in steered tapes that tessellate. Other examples of the benefit of tape spreading application to doubly-curved surfaces, such as engine nacelles, are given in [20].

3. Tape spreading process

3.1. Tape spreading device

The benefits of tape spreading can only be realised by the development and incorporation of a tape spreading device onto an LAMP head to allow active spreading prior to laydown and in-situ consolidation. This section describes such a tape spreading device developed by the authors, which has recently been filed for a patent [21]. An initial concept for spreading CF/PEEK tapes is shown in Fig. 7, which consists of a tape spreader equipped with a heated platen, two compaction stages and two pneumatic actuators (Fig. 7 (b)). A pneumatic regulator,

variable speed pull-through rig and temperature controller are also included in the experimental set-up (Fig. 7 (a)). The CF/PEEK tape is attached to the pull-through rig, which draws the tape through the tape spreader. The temperature of the platen is set close to the melt temperature of PEEK, ensuring that the viscosity/stiffness of the PEEK material decreases when in contact with the plate. Simultaneously, the compaction shoes apply pressure to the heated tapes, compressing them. This compression squeezes matrix and fibres in the through-thickness direction causing their lateral dispersion by squeeze flow processes, which manifests as increased tape width. The extent of spreading can be varied by modifying relevant processing parameters of the spreading device, such as temperature and pressure. Temperature can be increased or decreased to change the viscosity of PEEK accordingly, varying the amount of spreading that occurs. The pressure applied to the pneumatic actuators can be varied to apply different levels of compaction force to the tapes. The rate of tape deposition is controlled by the pull-through rig that has a variable speed controller. The number of passes is dictated by passing the CF/PEEK tape through the spreader multiple times. Finally, the tape spreader compaction shoes are interchangeable, allowing the use of either a rotating or stationary roller, or a flat compaction shoe with different surface areas to apply pressure to the tape. Fig. 7 (c) shows the cross section of three different compaction shoes used, 1 a stationary roller, 2 a flat compaction shoe and 3 a flat compaction shoe with larger surface area. Using compaction shoes with different surface areas gives variability in the downward pressure applied to the tapes. Larger surface areas also allow the tape a longer time to gradually change width, while heat is also given more time to transfer from the heated surfaces to the tapes.

3.2. Proof of concept

Initial testing was carried out to examine whether it is possible to spread CF/PEEK tapes. Different parameters were investigated to determine the effect each had on tape spreading. After a preliminary testing study, it was evident that parameters such as temperature and pressure have the most significant influence on the quality of spread CF/PEEK tapes. If too much heat or pressure is applied, there is a possibility that the tape deconsolidates. Higher processing temperatures reduce the viscosity of PEEK to a point where it becomes too weak (low shear strength) to remain intact. Too high a compaction pressure results in increased friction that can cause the tape to split. Conversely, insufficient temperature and pressure results in no spreading occurring. However, initial processing trials yielded optimal processing parameters, resulting in CF/PEEK tape spreading being achieved. A Vernier calliper was used to measure tape width before and after the spreading process. Measurements show that the maximum amount of spreading achieved was 62%,

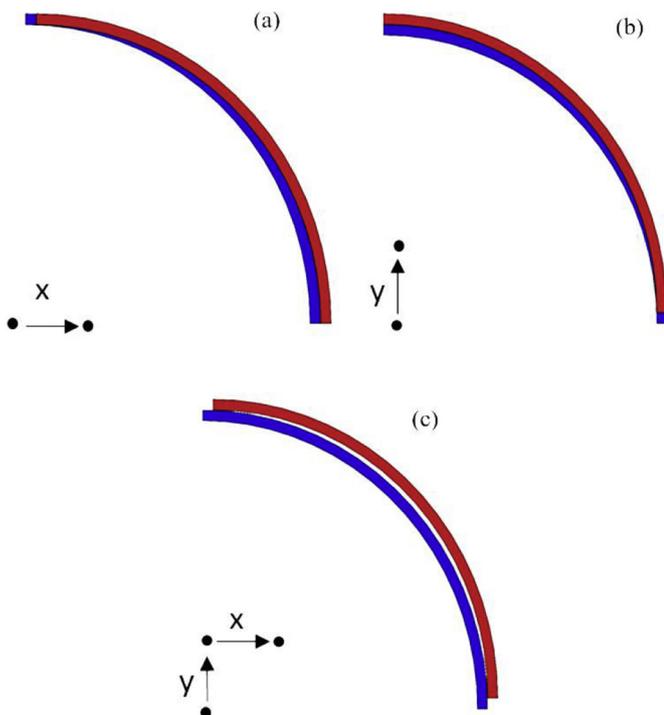


Fig. 6. Examples of offsetting constant width tows.

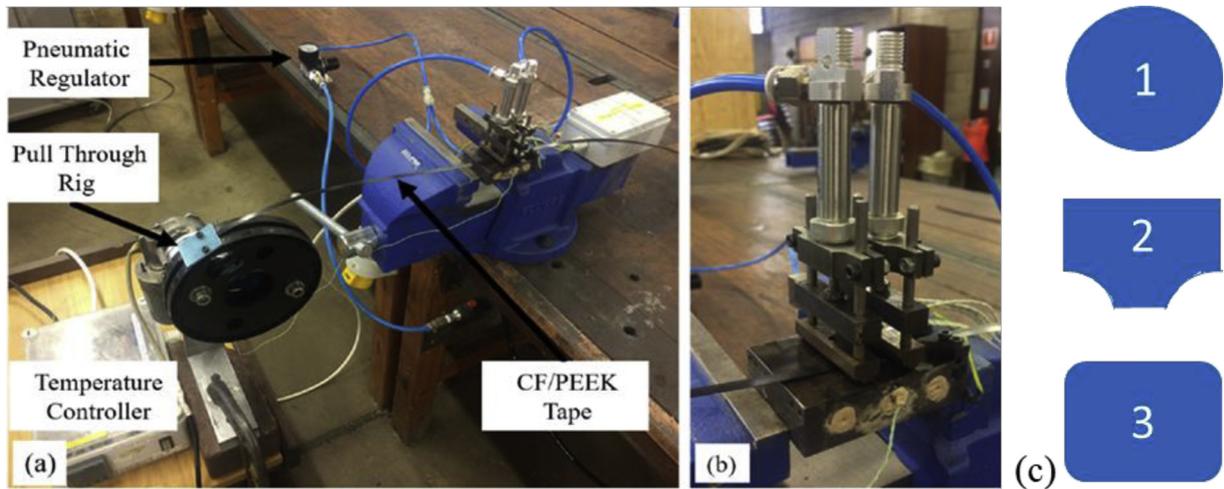


Fig. 7. (a) Experimental set up for spreading CF/PEEK, (b) Tape spreader, (c) different compaction shoe geometry profiles

as shown in Fig. 8. The initial nominal width increases from 6.35 mm to 10.3 mm after passing tape through the spreading device; there is no evidence of splitting or fibre pull-out, and surface roughness appears to be the same as before spreading.

The ability to increase the width of CF/PEEK tapes by 62% indicates that the current device has the capability to eliminate gaps between neighbouring steered tapes, as highlighted in section 2, where it was shown that a 41.4% width increase is required to fill the maximum gap between neighbouring steered tapes. In addition, the current device is capable of spreading tape sufficiently to facilitate the manufacture of doubly-curved surfaces, such as engine nacelles, where a case study in [20] indicated a maximum tape width increase of 37% was required.

4. Experimental testing

4.1. Manufacturing test samples

Spreading is a process that alters the physical properties of CF/PEEK tapes. As such, it is essential to investigate whether spreading negatively affects the quality of tapes. Teijin CF/PEEK tapes (carbon fibre (Tenax®-E HTS45 24 K)/PEEK) were used to manufacture test samples. The tapes, supplied by Teijin, used in this study were produced using solvent

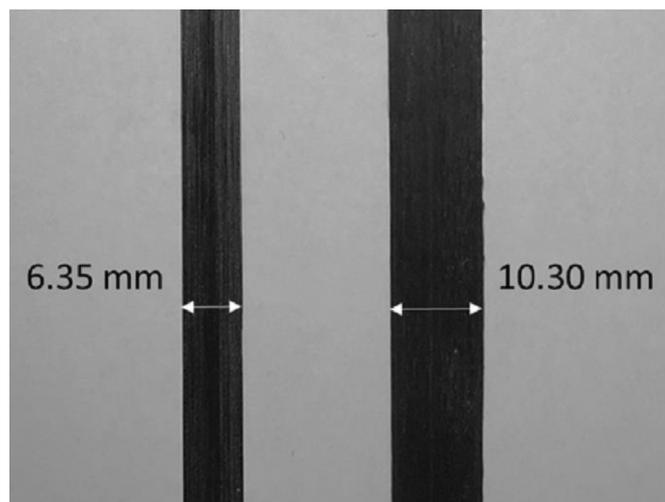


Fig. 8. Comparison of as-received and spread CF/PEEK tape.

impregnation, whereby dry fibres are passed through a bath of a solution of PEEK and a solvent [22]. The fibres are wetted with the solution, and the solvent evaporated during a drying process, leaving a carbon fibre/PEEK pre-preg tape. Three different sample groups were produced based on percentage increase in width of the tape and then compared to as-received tape; these were 15%, 30% and 45% spread tapes as well as a 0% (as-received tape) control. For this study the width of CF/PEEK tapes was increased by only varying the pressure applied to the tape by the spreading rig, other variables such as temperature, pull-through rate and compaction shoe geometry remained constant. The CF/PEEK tape was passed through the heated tape spreader and clamped to the pull-through rig. Actuation of both compaction shoes was achieved using a pneumatic regulator. The CF/PEEK tape was then pulled through the spreader automatically by the pull-through rig at a constant rate. For each test group, 2 m of tape was spread and samples were extracted at different locations. To achieve the three sample groups, the pneumatic regulator was adjusted to change the pressure of the compaction shoe, which resulted in different amounts of spreading. A type 3 compaction shoe (Fig. 7 (c)) was used to spread these samples. Three test groups were successfully produced, where examples of the change in width are shown in Fig. 9, along with an as-received sample. The processing parameters used are shown in Table 1, the number of passes refers to the amount of times the tape was passed through the spreading device, with 1/2 meaning it was passed through once with only one compaction shoe engaged. Numbers 1 and 2 refer to the tape passing through either once or twice respectively with both compaction shoes down. The rate of 1 m/min is slow for commercial LAMP processing, however this study's aim is to verify whether the concept of spreading is viable and does not adversely affect the quality of the pre-preg tapes. Future work will investigate faster lay-down rates. One added variable to consider is the additional heat generated due to friction between the tape and the heated surfaces at faster rates. An investigation will be required to determine suitable temperatures for a corresponding laydown rate, so that optimum tape spreading can be achieved without damaging the tape. Alternative heating methods may also have to be considered for tape spreading at higher rates.

4.2. Characterisation tests

Four characterisation methods assessed whether the spreading process adversely affects quality of CF/PEEK tapes. Methods undertaken include geometrical analysis, optical microscopy, Scanning Electron Microscopy (SEM) and Differential Scanning Calorimetry (DSC).

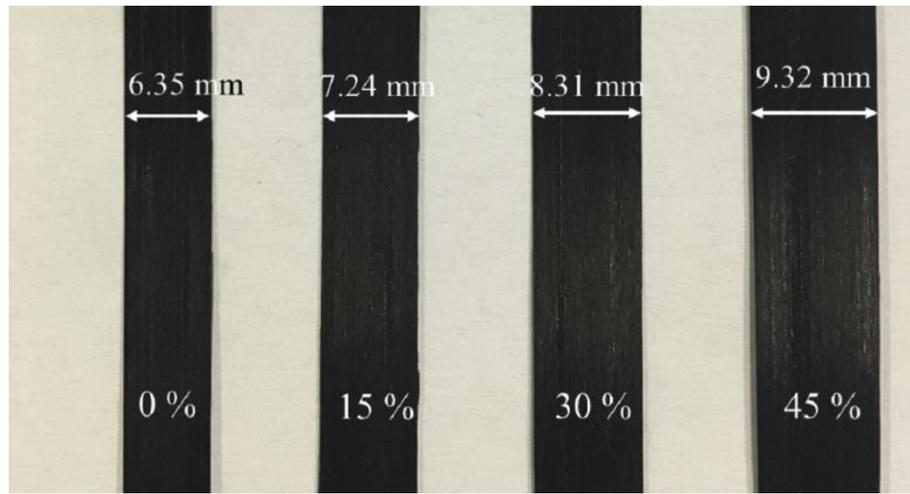


Fig. 9. Four sample groups; 0%, 15%, 30% & 45% showing difference in width.

Geometrical analysis involved measuring the width and thickness of CF/PEEK tapes after they were spread; comparison was subsequently made with the as-received tape (0%). Ten measurements of width and thickness were taken at intervals of 20 cm along the 2 m length of spread tape. A Mitutoyo Series 500 Vernier callipers with a resolution of 0.01 mm was used to measure the width of the samples. A Mitutoyo Series 293 μm with a resolution of 0.001 mm was used to measure the thickness of the tapes. Cross-Sectional Area (CSA) was also calculated to examine if the spreading process affects the volume of tapes.

Samples were extracted from original and spread tapes and were mounted in epoxy, then ground and polished. Microscopy, along with image capture, enabled examination of the effect spreading has on the quality of the CF/PEEK tapes. Three samples were taken from each of the four sample groups (0%, 15%, 30% and 45% spread tapes). Image processing software was used to examine whether spreading altered the composition of the tapes. Fibre volume fraction and void content were measured for spread samples and compared to original tapes.

A Hitachi SU-70 scanning electron microscope (SEM) was used to examine the samples. Three samples were extracted from each of the four sample groups. Samples were covered in a gold speckle to prevent PEEK from gathering charge, which reduces the quality of images captured. SEM was used to visually determine whether spreading affected the alignment of fibres, also to identify any defects caused by spreading such as fibre breakage or pull-out.

Differential scanning calorimetry (DSC) determined whether spreading affects the degree of crystallinity of CF/PEEK tapes. The degree of crystallinity of PEEK is a vital characteristic as it influences important mechanical properties including yield stress, elastic modulus and impact resistance [23]. The crystallinity of the CF/PEEK tapes was measured using a Netzsch DSC 214 Polyma. Two samples were extracted and analysed from each of the test groups. Samples of 10 ± 1 mg were placed inside an aluminium crucible and placed in the calorimeter.

The calorimeter operated with a nitrogen flow of 40 ml/min. A heating rate and cooling rate of $10^\circ\text{C}/\text{min}$ were used up to a maximum temperature of 350°C .

5. Results & discussions

5.1. Geometrical analysis

Geometrical measurements are shown in Table 2, including values for mean and standard deviation. Geometrical analysis gives a clear response of CF/PEEK tapes after spreading, as the width increases, there is a proportional decrease in thickness. CSA appears to remain constant for all sample groups, except for the 15% spread group, which shows a 5% increase. It is not clear why this difference occurred, but the CSA has not changed significantly for the 30% and 45% spread tapes, signifying that no significant voids, tears or fibre separation occurred due to the spreading process. A possible explanation for the CSA increase in the 15% sample group is due the CF/PEEK being produced using solvent impregnation. Tapes produced using solvent impregnation can vary in quality, large slits along the length of fibres as well as large internal voids along the length of fibres can occur. These slits or voids are caused by poor wetting of the fibres in the manufacturing process. The 15% group may have had a large internal void before being spread, which would result in an increase in CSA compared to the as-received tapes. This explanation is further supported by results from optical microscopy discussed in section 5.2.

An increase in standard deviation of the width of the CF/PEEK tapes is shown, indicating the tolerance of the tape width increase with spreading. The width tolerance increase could be a result of the significant temperature difference between the spreading rig and ambient (room) temperature. The spreading rig is not insulated which causes fluctuations in the surface temperature of the heated platen, leading to fluctuations in the viscosity of PEEK, increasing width tolerance.

Table 1

Processing parameters used to produce spread tape groups.

Width increase (%)	Actuator Pressure (bar)	No. of passes	Platen Temperature ($^\circ\text{C}$)	Rate (m/min)
15%	2	1/2	370	1
30%	2.5	1	370	1
45%	4.0	1	370	1
62%	4.0	2	370	1

Table 2

Results from geometrical analysis.

Sample Group	Width (mm)	Thickness (mm)	C.S.A (mm^2)
0%	6.34 ± 0.026	0.166 ± 0.006	1.05 ± 0.042
15%	7.24 ± 0.059	0.153 ± 0.002	1.11 ± 0.008
30%	8.31 ± 0.046	0.123 ± 0.003	1.02 ± 0.026
45%	9.32 ± 0.082	0.111 ± 0.004	1.04 ± 0.039

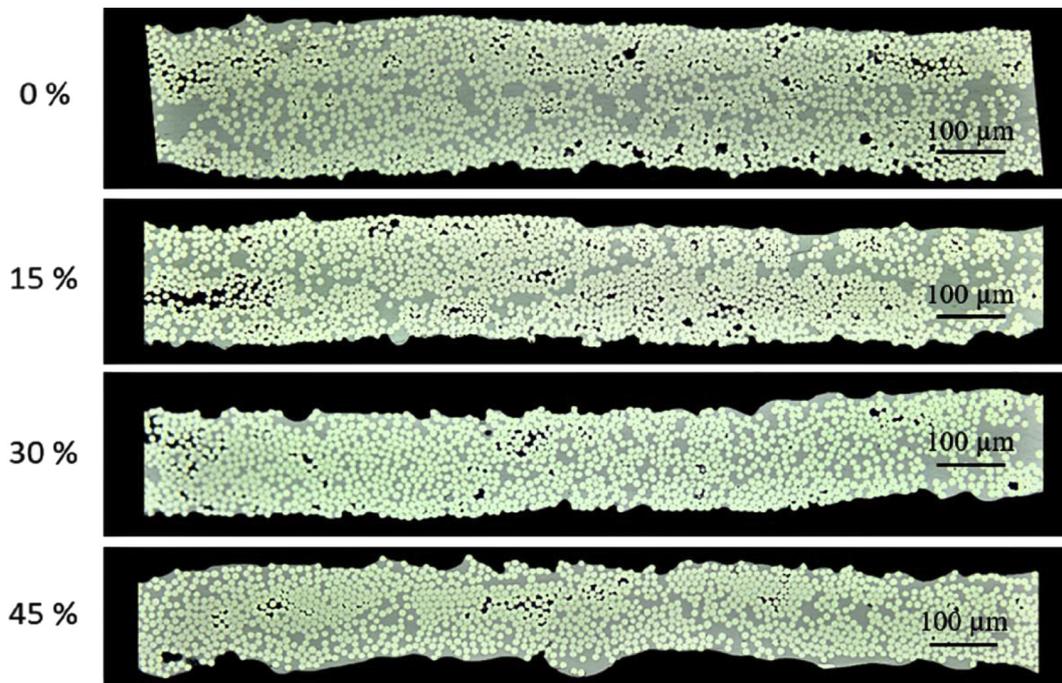


Fig. 10. Optical microscopy images comparing spread tapes.

5.2. Optical microscopy

Optical micrographs are shown in Fig. 10, comparing as-received (0%) tape to spread tape with 15%, 30% and 45% increase in width. The quality of the tapes does not appear to be greatly affected. There are no obvious visual defects such as tears or splitting of the tapes. Conversely, when the fibre dispersion is analysed, the spread tapes appear to have an improved fibre dispersion consistency. There appear to be less resin rich areas and fibre dominated areas in the spread groups when compared to the as-received group.

Surface smoothness is similar for all micrographs except for the 45% sample, which appears to have increased amounts of undulations. However, this effect could be due to a local defect in the tape before it was spread. These undulations may be eliminated when the spread tapes are processed using the LATP head, as it would then be heated above its melt temperature and compacted into place.

Micrographs were also analysed using image processing software to investigate whether fibre or void content changed by spreading with results shown in Table 3. Fibre content experiences an increase for the spread sample groups, where it is most for the 15% and 30% groups while the 45% samples experience a smaller increase. This increase in fibre content, occurs due to improved fibre dispersion as there are smaller (less) resin pockets in the spread tapes in comparison to original tapes. It is worth remarking that this increase in fibre content is a localised effect and not reflective of the fibre content of the whole tape, as the micrographs only focus on a section of the CF/PEEK tapes and not the whole tape.

Table 3
Results from image processing of optical micrographs.

Sample Group	Fibre Content (%)	Void Content (%)
0%	59.6 ± 2.0	3.4 ± 0.72
15%	63.1 ± 4.3	3.7 ± 0.93
30%	62.2 ± 3.3	2.8 ± 1.25
45%	61.7 ± 2.3	1.4 ± 0.60

When void content values were analysed, a decrease was observed for the 30% and 45% sample groups. This decrease may have been caused by voids being compressed during the spreading process so reducing their size and therefore overall void content, noting a similar mechanism of void reduction has been reported previously [24,25]. The 15% sample group has a marginally larger void content, possibly due to a large internal void along the length of the fibres, an example of which is shown in Fig. 10. This defect may not actually arise as a result of the spreading process. The large void may have been caused by the solvent impregnation process used to manufacture the CF/PEEK pre-preg tapes, as discussed in section 5.1.

5.3. Scanning Electron Microscopy (SEM)

Results from SEM were inconclusive. Images obtained from SEM are shown in Figs. 11 & 12. From analysing the SEM images, it is evident that all sample groups (including 0%) vary in quality. Good quality areas of each sample groups are shown in Fig. 11, which show no signs of loose fibres or large voids and appear to have an appropriate quantity of resin on the upper surfaces, which is necessary to achieve a good bond with subsequent layers. Fig. 12 highlights examples of areas from the four sample groups with defects evident. These examples of fibre pull-out and fibre breakage would be expected to reduce the performance of the CF/PEEK tapes. Both sides of the pre-preg tapes were analysed, as one side would have been in contact with the heated platen and the other in contact with the compaction shoes. Visually, no difference was observed between either side.

From analysing SEM images, it is not clear whether the spreading process adversely affects the quality of CF/PEEK tapes. Defects were observed in spread tapes but were also observed in as-received tapes. It is not possible to say whether the spreading process caused these defects or whether they were already present on the tapes before spreading occurred. Further testing would be required to examine if spreading created the damage to fibres. A comparison of tensile properties, at a laminate level, of spread and as-received tapes would reveal if the spreading process reduces strength and stiffness of the CF/PEEK Tapes, which can form the basis of future work.

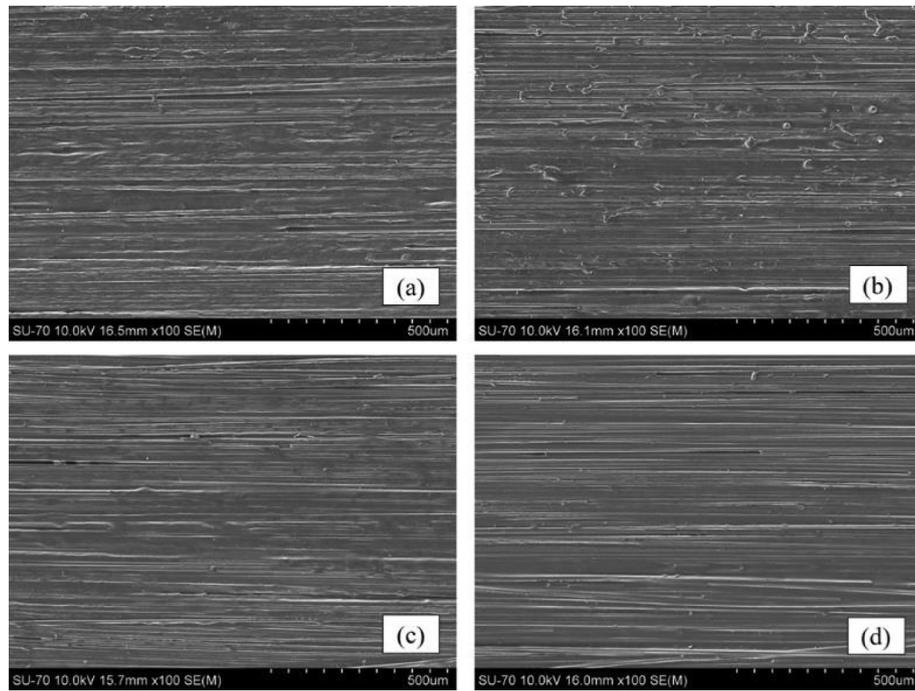


Fig. 11. SEM images obtained (a): 0%, (b) 15%, (c) 30%, (d) 45%.

5.4. Differential Scanning Calorimetry (DSC)

It is clear from the results that the spreading process increases the degree of crystallinity. Curves from DSC characterisation are shown in Fig. 13; enthalpy of cold crystallisation and degree of crystallinity are presented. The spreading process provides a heat treatment that essentially anneals PEEK, resulting in a change of crystallinity. There is a relationship between width and crystallinity, a larger increase in width results in a larger increase in crystallinity. There are two potential

reasons for this relationship. The first may be due to the width of the CF/PEEK tapes been varied by varying pressure of the spreading rig. Previous studies show that an increase in pressure during melting of PEEK increases crystallinity [26,27]. The increased pressure of spreading assists with alignment and packing of polymer chains of PEEK, resulting in an increased crystalline structure. The second reason is that, as the width increases, the thickness decreases since the tape is only heated from one side. As the tape becomes thinner, it achieves a uniform heat distribution through the thickness. This uniform heat distribution

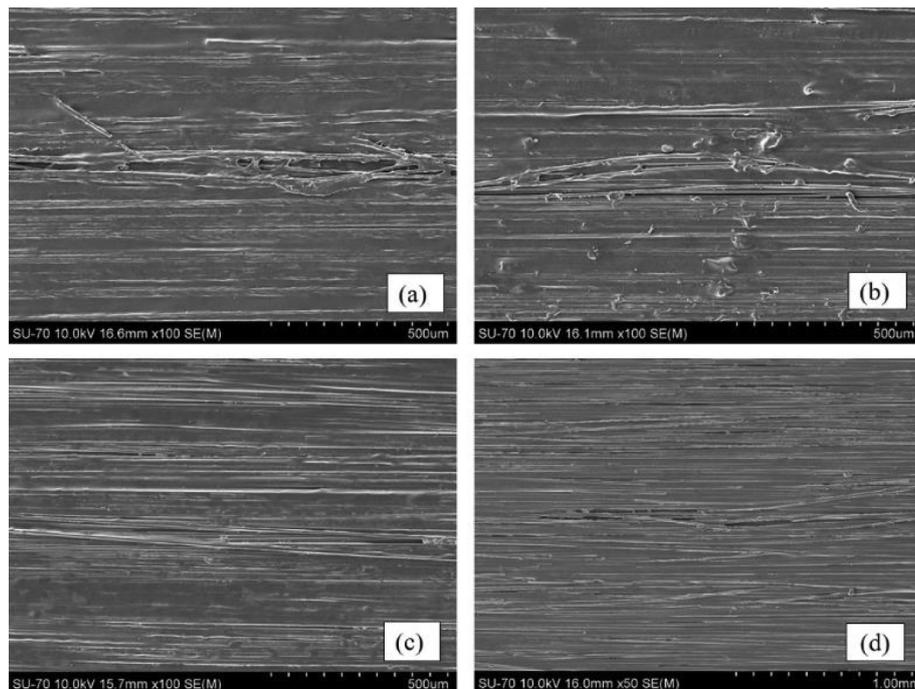


Fig. 12. Defects observed in CF/PEEK Tapes (a): 0%, (b) 15%, (c) 30%, (d) 45%.

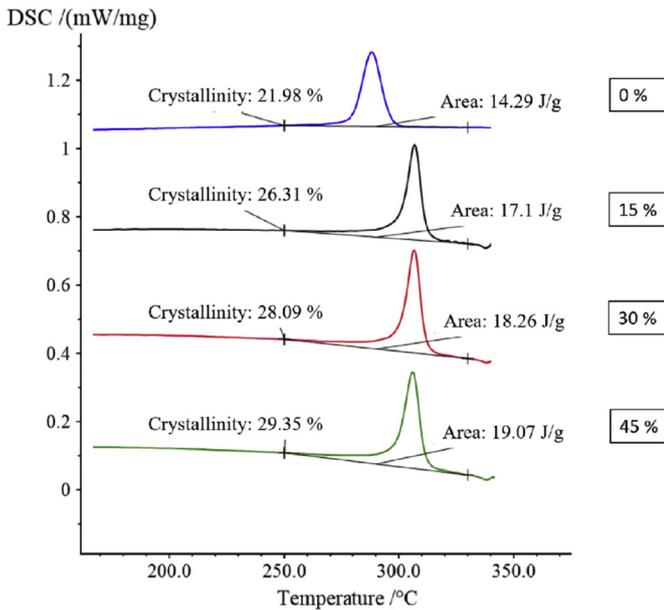


Fig. 13. Results of Crystallinity obtained from DSC.

results in the entire tape width receiving the annealing treatment from the spreading device, causing an increase in the degree of crystallinity. The increase in crystallinity is beneficial as it may potentially improve mechanical properties such as modulus and yield strength [26,27]. The quality of the crystal structure has not yet been analysed to examine if they are composed of large or small spherulites. Future work will investigate the crystal quality achieved by heat treatment during the spreading process.

In summary, results from the four characterisation methods show there is strong evidence that the spreading process does not adversely affect the mechanical properties of CF/PEEK tape. Geometrical tolerances are only marginally affected, while fibre volume fraction, void content and fibre dispersion remain similar to as-received tape. Defects on the top and bottom surfaces do not appear to occur with higher frequency or severity. Finally, crystallinity levels increase as a result of spreading and further work will investigate quality levels. Future work will investigate whether spreading adversely affects mechanical properties at a laminate level. Laminates will be manufactured using spread tape, and coupons extracted. Tests including tensile, flexure, combined loading compression, interlaminar shear and in-plane shear will be completed. Results from these tests will then be compared with laminates manufactured from original tape to see the effect of spread tapes at a laminate level.

6. Conclusion

An innovative device has been developed to enable the width of CF/PEEK pre-preg tape to be varied (spread) as part of an in-line process within an LAMP head. Initial trials show that controlled spreading of CF/PEEK pre-preg tapes is possible. The current spreading device, which is under further development, is capable of increasing the width of a CF/PEEK tape by 62%. The ability to vary the width would be advantageous in manufacturing steered laminates without gaps and to manufacture doubly-curved surfaces without fibre cuts or gaps. Elimination of gaps and fibre cuts in such components has the potential to increase their structural efficiency. Preliminary characterisation tests show that the spreading process does not detrimentally affect the properties of CF/PEEK pre-preg tapes. Properties such as cross-sectional area,

fibre volume fraction and void content remain similar to as-received tape. Furthermore, differential scanning calorimetry shows that crystallinity increases due to spreading, which is beneficial as it may potentially improve mechanical properties such as elastic modulus and strength. Future work will focus on completing mechanical characterisation to examine whether spreading affects the structural performance of CF/PEEK at a laminate level, which will include tensile, flexure, combined loading compression, interlaminar shear and in-plane shear. Once shown that spreading does not adversely affect the quality of pre-preg tapes or the mechanical properties of laminates manufactured from spread tapes, work will focus on implementing the device onto an LAMP processing head. Matters such as programming the spreading device to compact and spread tapes to comply with gaps in VAT laminates will be investigated. Also, work will be completed to optimise the laydown rate to achieve efficient manufacturing throughput without compromising structural performance.

Declaration of Competing Interest

None.

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Author contribution statement

Gearóid Clancy: Conceptualization, Methodology, Investigation, Data Curation, Formal analysis, Validation Writing - Original Draft, Visualization. **Daniël Peeters:** Supervision, Visualization, Writing - Review & Editing. **Ronan M O'Higgins:** Conceptualization, Methodology, Visualization, Supervision, Project administration, Resources, Writing - Review & Editing. **Paul M. Weaver:** Conceptualization, Methodology, Visualization, Supervision, Project administration, Resources, Writing - Review & Editing, Funding acquisition.

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.

References

- [1] A.J. Comer, D. Ray, W.O. Obande, D. Jones, J. Lyons, I. Rosca, O' Higgins, R. M., McCarthy, M. A., Mechanical characterisation of carbon fibre-PEEK manufactured by laser-assisted automated-tape-placement and autoclave, *Composites Part A: Applied Science and Manufacturing* Vol. 69 (2015) 10–20.
- [2] B.C. Kim, K. Potter, P.M. Weaver, Continuous tow shearing for manufacturing variable angle tow composites, *Compos. A: Appl. Sci. Manuf.* 43 (8) (2012) 1347–1356.
- [3] M. Rouhi, H. Ghayoor, J. Fortin-Simpson, T.T. Zaccchia, S.V. Hoa, M. Hojjati, Design, manufacturing, and testing of a variable stiffness composite cylinder, *Compos. Struct.* 184 (2018) 146–152.
- [4] C. Wu, Z. Gurdal, J. Starnes, Structural response of compression-loaded, tow-placed, variable stiffness panels, in 43rd AIAA/ASME/ASCE/AHS/ASC structures, structural dynamics, and materials conference, American Institute of Aeronautics and Astronautics. (1512) (2002) <https://doi.org/10.2514/6.2002-1512>.
- [5] M.W. Tosh, D.W. Kelly, On the design, manufacture and testing of trajectorial fibre steering for carbon fibre composite laminates, *Compos. A: Appl. Sci. Manuf.* 31 (10) (2000) 1047–1060.
- [6] B.K. Stanford, C.V. Jutte, K. Chauncey Wu, Aeroelastic benefits of tow steering for composite plates, *Compos. Struct.* 118 (2014) 416–422.
- [7] G.J. Clancy, D. Peeters, V. Oliveri, D. Jones, R. O'Higgins, P.M. Weaver, "Steering of Carbon Fiber/Thermoplastic Pre-Preg Tapes Using Laser-Assisted Tape Placement," 2018 AIAA/ASCE/AHS/ASC Structures, Structural Dynamics, and Materials Conference, American Institute of Aeronautics and Astronautics, 2018.

- [8] D.H.J.A. Lukaszewicz, C. Ward, K.D. Potter, The engineering aspects of automated pre-preg layup: history, present and future, *Compos. Part B* 43 (3) (2012) 997–1009.
- [9] K. Fayazbakhsh, M. Arian Nik, D. Pasini, L. Lessard, Defect layer method to capture effect of gaps and overlaps in variable stiffness laminates made by automated Fiber placement, *Compos. Struct.* 97 (2013) 245–251.
- [10] S.D.R. Wilson, Lateral spreading of fibre tows, *J. Eng. Math.* 32 (1) (1997) 19–26.
- [11] M.S. Irfan, V.R. Machavaram, R.S. Mahendran, N. Shotton-Gale, C.F. Wait, M.A. Paget, M. Hudson, G.F. Fernando, Lateral spreading of a fibre bundle via mechanical means, *J. Compos. Mater.* 46 (3) (2012) 311–330.
- [12] J.A. Goshawk, V.P. Navez, R.S. Jones, Squeezing flow of continuous fibre-reinforced composites, *J. Non-Newtonian Fluid Mech.* 73 (3) (1997) 327–342.
- [13] S.F. Shuler, S.G. Advani, Transverse squeeze flow of concentrated aligned fibers in viscous fluids, *J. Non-Newtonian Fluid Mech.* 65 (1) (1996) 47–74.
- [14] E.L. Wang, T.G. Gutowski, Laps and gaps in thermoplastic composites processing, *Compos. Manuf.* 2 (2) (1991) 69–78.
- [15] X. Brulotte, G., Aspects of In-Situ Consolidation of Thermoplastic Laminates Manufactured by Automated Tape Placement: A Material Deformation Study, McGill University, Montreal, Quebec, 2012.
- [16] J. Engmann, C. Servais, A.S. Burbidge, Squeeze flow theory and applications to rheometry: A review, *J. Non-Newtonian Fluid Mech.* 132 (1) (2005) 1–27.
- [17] F. Ren, Y. Yu, M. Cao, Y. Li, C. Xin, Y. He, Effect of pneumatic spreading on impregnation and fibre fracture of continuous fibre-reinforced thermoplastic composites, *J. Reinf. Plast. Compos.* 36 (21) (2017) 1554–1563.
- [18] S. Sohn, R.Y. Kim, K. Kawabe, S.W. Tsai, Experimental studies of thin-ply laminated composites, *Compos. Sci. Technol.* 67 (6) (2007) 996–1008.
- [19] G. Zucco, V. Oliveri, D. Peeters, R. Telford, G.J. Clancy, C. McHale, M. Rouhi, R. O'Higgins, T.M. Young, P.M. Weaver, "Static Test of a Thermoplastic Composite Wingbox under Shear and Bending Moment," 2018 AIAA/ASCE/AHS/ASC Structures, Structural Dynamics, and Materials Conference, American Institute of Aeronautics and Astronautics, 2018.
- [20] G. Clancy, R.M. O'Higgins, P.M. Weaver, *Spreading of Carbon Fiber/Thermoplastic Pre-preg Tapes*, in 2020 AIAA/ASCE/AHS/ASC Structures, Structural Dynamics, and Materials Conference, American Institute of Aeronautics and Astronautics. (0481) (2020) <https://doi.org/10.2514/6.2020-0481>.
- [21] Clancy, G., O'Higgins, R.M., Weaver, P.M. *Carbon Fibre/Thermoplastic Tape Spreader*, UK Patent application No: 1904264.7, Filed 27th March 2019.
- [22] G. Marsh, Prepregs – raw material for high-performance composites, *Reinf. Plast.* 46 (10) (2002) 24–28.
- [23] F.Y.C. Boey, S.W. Lye, Void reduction in autoclave processing of thermoset composites: part 1: high pressure effects on void reduction, *Composites* 23 (4) (1992) 261–265.
- [24] S. Ranganathan, S.G. Advani, M.A. Lamontia, A non-isothermal process model for consolidation and void reduction during in-situ tow placement of thermoplastic composites, *J. Compos. Mater.* 29 (8) (1995) 1040–1062.
- [25] Y. Kong, J.N. Hay, The measurement of the crystallinity of polymers by DSC, *Polymer* 43 (14) (2002) 3873–3878.
- [26] D. Xi, D. Zhang, J. Tian, J. Lu, Z. Zhou, C. Yuan, X. Liu, S. Long, Y. Huang, R. Huang, Spherulitic growth of poly (ether ether ketone) crystallised at high pressure, *Journal of Macromolecular Science, Part B* 51 (3) (2012) 510–524.
- [27] H.M. Lin, R. Lee, C.H. Liu, J.S. Wu, C.S. Huang, Effects of high pressure on the crystallisation and mechanical properties of carbon-fibre-reinforced poly(ether ether ketone) laminated composites, *Compos. Sci. Technol.* 52 (3) (1994) 407–416.