

Delft University of Technology

Influence of repass treatment on carbon fibre-reinforced PEEK composites manufactured using laser-assisted automatic tape placement

Chanteli, Angeliki; Bandaru, Aswani Kumar; Peeters, Daniël; O'Higgins, Ronan M.; Weaver, Paul M.

DOI 10.1016/j.compstruct.2020.112539 **Publication date**

2020 **Document Version** Accepted author manuscript

Published in **Composite Structures**

Citation (APA)

Chanteli, A., Bandaru, A. K., Peeters, D., O'Higgins, R. M., & Weaver, P. M. (2020). Influence of repass treatment on carbon fibre-reinforced PEEK composites manufactured using laser-assisted automatic tape placement. Composite Structures, 248, Article 112539. https://doi.org/10.1016/j.compstruct.2020.112539

Important note

To cite this publication, please use the final published version (if applicable). Please check the document version above.

Copyright Other than for strictly personal use, it is not permitted to download, forward or distribute the text or part of it, without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license such as Creative Commons.

Takedown policy

Please contact us and provide details if you believe this document breaches copyrights. We will remove access to the work immediately and investigate your claim.

Influence of repass treatment on carbon fibre-reinforced PEEK composites manufactured using laser-assisted automatic tape placement

Angeliki Chanteli^a, Aswani Kumar Bandaru^a, Daniël Peeters^b, Ronan M. O' Higgins^a, Paul M. Weaver^a

^a Bernal Institute, School of Engineering, University of Limerick, Ireland

^bFaculty of Aerospace Engineering, Delft University of Technology, Kluyverweg 1, Delft 2629HS, The Netherlands

ABSTRACT

Laser-assisted automated tape placement (LATP) in-situ consolidation of thermoplastic composites offers an efficient solution for production of large, high performance composite structures. The present work investigates laser repass treatment as a method of enhancing surface roughness and mechanical properties of LATP processed CF/PEEK laminates. Three laminate stacking sequences were studied, *viz.* $[0^\circ]_{16}$, $[-45^\circ/0^\circ/45^\circ/90^\circ]_{2s}$ and $[\pm 45^\circ]_{4s}$. Four repass treatments were performed, including single, double, perpendicular and tool-side repasses. The effect of these repasses on interlaminar shear strength (ILSS), open-hole compression (OHC), in-plane shear (IPS), density, fibre volume fraction, void content and surface roughness was investigated. Surface roughness tests confirmed the improvement of the surface finish due to repass treatment. However, insignificant differences in ILSS values were observed across all repass treatments, but a single laser repass did appear to improve the OHC and IPS performance. Autoclave treated samples were used as a benchmark to compare with LATP processed laminate properties.

Keywords: Carbon Fibre/PEEK; Mechanical properties; Automated fibre placement (AFP); Annealing

1. INTRODUCTION

Laser-Assisted Automatic Tape Placement (LATP) is considered as an advanced in-situ consolidation technique for rapid processing of thermoplastic composite structures with minimal material waste. The demand for out-of-autoclave automated manufacturing techniques is increasing, in particular for the commercial aerospace sector in which the LATP technique could serve as a state-of-the-art manufacturing solution for the continuous production of large and optimised structures with reduced time and overall manufacturing cost. By using LATP, CF reinforced PEEK tapes have shown improved fracture toughness [1, 2] in comparison with similar laminates using an autoclave, [1]. Recent studies from the University of Limerick [3, 4] used CF/PEEK prepreg tapes to successfully manufacture a representative skin-stiffener of an

aeroplane wingbox and an integrally stiffened, unitised wingbox demonstrator. The latter structure included a number of innovations, including laser-assisted in-situ bonding of the stiffeners during manufacture [5, 6] and variable stiffness panels to increase panel buckling load, achieved by fibre-reinforced thermoplastic tape steering [7].

The quality of the above-mentioned products depends greatly on process parameters utilised during CF/PEEK tape placement. Thermal history significantly affects crystalline morphology of PEEK, particularly the crystallisation rate, nucleation growth which combine to affect matrix dominated mechanical properties [8]. In addition, cold crystallisation phenomena associated with rapid heating and cooling rates from laser assisted processing [9] are responsible for lower levels of crystallinity in some LATP processed structures [10].

Post-consolidation processes, such as annealing heat treatment aims to facilitate the interdiffusion of the polymer chains between layers. Zhang et al [11], experimentally highlighted the change in crystallinity levels within a range of annealing temperatures (342 °C and 347 °C); and noted that cold crystallisation arises from the amorphous PEEK phase during quenching from the melted state. These effects were found to influence matrix-dominated properties including interlaminar shear strength (ILSS) and in-plane shear (IPS). Shadmerhi et al [12], studied the annealing effect of heating repasses on CF/PEEK laminates produced using hot-gas heated ATP. The surface roughness of the laminates reduced after repassing treatment, however, the percentage crystallinity of PEEK also reduced. Hoa et al. [13], reported the influence of repass on ILSS properties for a similar process; the first repass slightly increased the ILSS, but there was no significant increase for the second repass. LATP processing parameters that influence laminate quality are consolidation pressure, laydown speed and tool temperature. Stokes-Griffin et al [14] studied the influence of temperature and placement rate on ILSS values of CF/PEEK composites. They found that ILSS is independent of the process temperature at 100 mm/s up to a temperature of 550 °C, beyond which, ILSS properties degraded.

Heating sources like hot gas and laser, by nature, significantly affect quality of final products. Hot gas torch AFP systems heat the incoming tape and substrate by convection, while laser systems heat by radiation. There is limited research available in the literature [12, 13] on the influence of annealing treatment repass on structural properties, such as ILSS.

The present study considers the influence of laser repass on strength properties and crystallinity content of LATP manufactured CF/PEEK laminates. Three different repass strategies were investigated: a single repass parallel to the fibre direction, a double repass also parallel to the fibre direction and a single repass perpendicular to the fibre direction for $[0^{\circ}]_{16}$ laminates. Quasiisotropic, $[-45^{\circ}/0^{\circ}/45^{\circ}/90^{\circ}]_{2s}$, and angle ply, $[\pm 45^{\circ}]_{4s}$, laminates were laser treated in parallel with 0° direction for the single and double repass strategies. In addition, effects on the first laid ply were investigated with a single laser repass on the tool-side ply. A non-repassed LATP laminate and an autoclave consolidated laminate were used as references for comparison to the above-mentioned laminates. Specimens were collected from the annealed, autoclaved and untreated laminates for open hole compression (OHC), IPS and ILSS testing to assess the effect of annealing on the mechanical performance. In addition, the crystallinity content achieved by laser annealing was investigated using differential scanning calorimetry (DSC). Finally, the surface roughness was also assessed for the $[0^{\circ}]_{16}$ and $[\pm 45^{\circ}]_{4s}$ laminates.

2. MATERIALS AND MANUFACTURE

2.1. Manufacture setup and process parameters

A 6.35 mm wide IMS65/PEEK unidirectional (UD) tape (supplied by Teijin Carbon America, Inc.) was chosen for examination. Laminates with stacking sequences of $[0^{\circ}]_{16}$, $[\pm 45^{\circ}]_{4s}$ and $[-45^{\circ}/0^{\circ}/45^{\circ}/90^{\circ}]_{2s}$ were manufactured using a LATP head (AFPT, GmbH) attached to a Kuka, KR240 L210-2, robot arm (Fig. 1). The LATP system comprises: (i) a connected optical system to a remotely located 3 kW diode-laser heat source (wavelength, λ ~1015 nm), (ii) a tape feed, guidance and cutting system, (iii) a consolidation roller (outer diameter, d_o =80 mm) and (iv) a thermal camera. The optical system spreads the laser beam within a range of spot sizes at the nip point, depending on the focal length of the lens. The optical system used (focal length 200 mm and 250 mm) can process tape widths between 6mm and 25mm where the beam distribution can be biased towards the incoming tape or the substrate. A silicone consolidation pressure roller was pressed onto the substrate by a pneumatic cylinder and cooled externally using compressed air to prevent the tape adhering to the roller during processing.

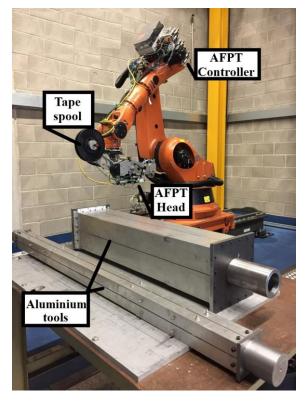


Fig. 1. LATP setup and aluminium tools.

The quality of the final laminate depends on the optimised selection of LATP processing parameters including: (a) laser power, (b) laser angle, (c) roller pressure, (d) tool temperature, (e) lay-down speed and (f) roller temperature which are shown in Table 1, for the different stacking sequences. These parameters are based on previous manufacturing trials and suggestions made by the LATP head manufacturer [6, 7]. Three different laminates were manufactured: (a) a unidirectional laminate of $[0^{\circ}]_{16}$ layup, (b) a quasi-isotropic laminate [- $45^{\circ}/0^{\circ}/45^{\circ}/90^{\circ}]_{2s}$ and (c) an angle ply laminate $[\pm 45^{\circ}]_{4s}$. The laser beam was focused at the nip point (50/50: incoming tape/substrate) for all lay-ups. The tape was laid down by a continuous winding process onto unheated square profile aluminium tools of dimensions 200 mm and 300 mm, producing four panels for each lay-up configuration, with a laydown speed of 3 m/min.

LATP process parameters	ILSS	ОНС	IPS
Lay-up	[0°] ₁₆ , [-45°/0°/45°/90°] _{2s}	$[-45^{\circ}/0^{\circ}/45^{\circ}/90^{\circ}]_{2s}$	[±45°] _{4s}
Lay-down speed	3 m/min	3 m/min	3 m/min
Target temperature	380 °C	380 °C	380 °C
Tool temperature	Unheated	Unheated	Unheated
Roller material	Silicone	Silicone	Silicone
Roller pressure	2.5 bar	2.5 bar	2.5 bar

Table 1. Laser assisted tape placement process parameters.

2.2. Repass treatment

Shadmeri et al [12], describe the term repass as the application of heat and pressure though the ATP head, without using the spool, onto the uppermost surface of the already laid tapes. Laser repass assists thermal stress relief and cold crystallization phenomena. The details of repass treatment for $[0^{\circ}]_{16}$ laminates, quasi-isotropic $[-45^{\circ}/0^{\circ}/45^{\circ}/90^{\circ}]_{2s}$ and angle ply $[\pm 45^{\circ}]_{4s}$ laminates are shown in Fig. 2. The following steps were performed for the repass treatment of $[0^{\circ}]_{16}$ laminates (Fig. 2b): (a) completion of a single layer laid on the four faces of the square tool, (b) a single repass (R1) was performed in parallel to the fibre direction on one side of the square tool (profile of 200 mm), (c) the opposite side was laser repassed twice (R2) in parallel to the fibre direction (double repass) and (d) the third side was laser repassed once perpendicular to the fibre direction (PR). The steps for the repass treatment of the quasi-isotropic and angle ply laminates were as follows (Fig. 2c): (a) completion of the material lay-up on the four faces of the square tool (profile of 300 mm), (b) a single repass (R1) was performed in parallel with the 0° direction on one side of the square tool, (c) the opposite side was laser repassed twice (R2) in parallel with the 0° direction (double repass) and (d) the third side was performed in parallel with the 0° direction on one side of the square tool, (c) the opposite side was laser repassed twice (R2) in parallel with the 0° direction (double repass) and (d) the third side was laser repassed twice (R2) in parallel with the 0° direction (double repass) and (d) the third side was laser repassed twice (R2) in parallel with the 0° direction (double repass) and (d) the third side was laser repassed twice (R2) in parallel with the 0° direction (double repass) and (d) the third side was laser repassed once; parallel to

0° direction (tool-side repass, R3). The notation for the repass treatment described in the text is also depicted in Fig. 2.

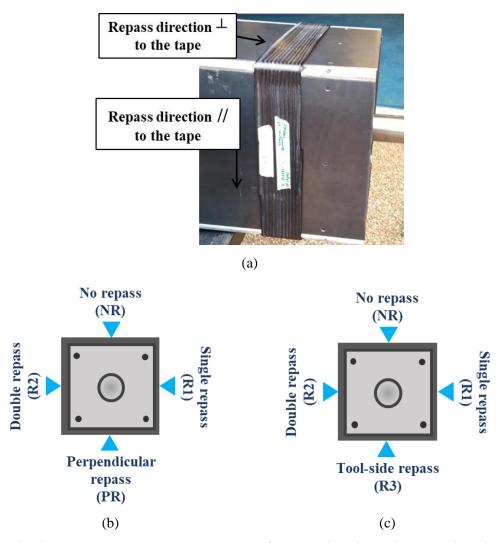


Fig. 2. Laser repass steps. (a) Examples of repass directions with material laid on the aluminum tool, (b) laser repass steps for the [0°]₁₆ laminates and (c) laser repass steps for the quasi-isotropic [-45°/0°/45°/90°]_{2s} and angle ply [±45°]_{4s} laminates.

Laser repass annealing was carried out with a laser power of 500W, producing a surface temperature of approximately 343 °C (T_m of PEEK), and passing over the panel at the laydown roller pressure. Four different types of laser repass annealing were performed for the current study, i.e. single repass (R1), double repass (R2) and single repass onto the tool-side (R3) on the uppermost layer of the laminate and a perpendicular repass (PR). These treatments were carried out on two sides of the lay-up, respectively, while they were done in-situ on the tool. In addition, composite panels of similar stacking sequences were laid-down by LATP and then consolidated using standard autoclave manufacturing procedures, according to the supplier recommended processing parameters, for comparison with the laser annealed respective panels. The

consolidation pressure and cure temperature were monitored throughout the cure cycle and the cooling rate was controlled at 2 °C/min.

2.3. Physical properties of the composites

After manufacture, physical properties including thickness, density, fibre volume fraction and void content were measured following the appropriate ASTM standards. The thickness of each specimen was measured at six different locations. The density was measured as per ASTM D792 [15], fibre volume fraction (FVF) and void content were measured according to ASTM D3171 [16]. Density values were obtained from the water displacement method and FVF was measured by performing burn-off tests. The nominal dimensions of both the density and FVF samples were 10 mm x 10 mm. For the measurement of density, samples were weighed dry and then immersed into distilled water, for which the temperature measured 16.3 °C, using the appropriate holder from the density kit of an analytical Sartorius balance. For FVF measurements, specimens were placed in crucibles that were kept for 2 hrs at a temperature of 585 °C. Specimens were weighed before and after burn-off. Then, FVF values, based on the weight fraction values were calculated using empirical relations provided in [16]. After estimating density and FVF values, void content was also measured [16]. At least ten specimens were used to measure these physical properties.

3. EXPERIMENTATION

3.1. Differential scanning calorimetry analysis

DSC analysis on the CF/PEEK composites was performed using a Netzsch Polyma 214 system. The objective was to evaluate the thermal properties of LATP manufactured laminates. Specimens of approximately 15 mg were analysed. Thermal history varied from 20 °C to 400 °C at a heating rate of 10 °C/min. The amount of heat absorbed by the materials during melting was also estimated for each case by calculating the area under the melting peak. The results were obtained as input energy versus temperature. Samples were tested from each differently processed laminate and also from the unprocessed prepreg CF/PEEK tape.

3.2. Surface roughness

Surface roughness measurements assessed the smoothness of the laminate from different repass treatments, using a HOMMEL T500 surface roughness tester. A minimum of ten samples for each repass case were tested and the surface roughness values were estimated at six locations. Surface roughness calculations identified mean deviation values of the assessed profile (R_a) which were based on the five highest peaks and lowest troughs over the entire sampling length.

3.3. Mechanical characterisation

ILSS, IPS and OHC tests were performed following the ASTM D2344 [17], D3518 [18] and D6484 [19] standards, respectively. ILSS characterisation was carried out for the treated and untreated laminates and a minimum of ten samples were tested.

The nominal dimensions of the ILSS samples were: 20 mm x 10 mm x t mm where t corresponds to the thickness of different laminates and a span of length-to-specimen thickness, l/t ratio of 4.0 was maintained. A Tinius Olsen H25KS test machine with a 5 kN load cell was used to carry out the ILSS tests. Tests were performed using a three-point bending test rig, the diameters of the support cylinders and the loading nose were 3 mm and 6 mm, respectively. The crosshead displacement rate was set to 1 mm/min and the peak load for each sample was recorded to estimate ILSS strength. The schematic representation and typical test setup to perform the ILSS tests are shown in Fig. 3(a-b). In addition, failure modes in ILSS samples were studied through digital images and scanning electron microscopy (SEM).

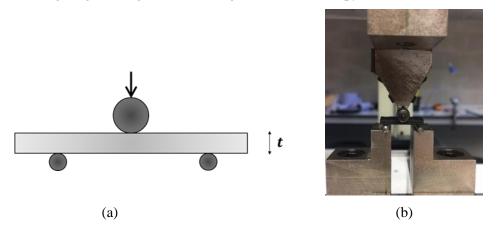


Fig. 3. ILSS test details: (a) Schematic representation and (b) ILSS test setup.

IPS tests were carried out using a Tinius Olsen H25KS test machine with a 25 kN load cell. The nominal dimensions of the IPS samples were: 250 mm x 25 mm x 3.3 mm and tests were carried out at a crosshead displacement of 2 mm/min. Both axial and transverse displacements were recorded using a biaxial extensometer (Epsilon, model 3560) with a gauge length of 25 mm, as shown in Fig. 4. The peak load was retrieved for each sample and the shear strength and shear modulus were calculated.

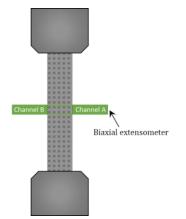


Fig. 4. Test set up for the IPS experiments.

The OHC tests were carried out using a Dartec test machine with a 100 kN load cell as shown in Fig. 5b. The nominal dimensions for the OHC samples were: 300 mm x 36 mm x 3.3 mm with a 6 mm diameter hole. The tests were carried out at a crosshead displacement rate of 2 mm/min. The test set up for OHC characterizations is shown in Fig. 5a. The OHC specimens were supported using a standard support fixture; displacements were recorded using an axial extensometer (Epsilon, model 3562) with a gauge length of 25 mm (Fig. 5a). The peak load was recorded for the estimation of ultimate strength values. Details from the failure region were captured using an optical microscope (VMS-005-LCD-Portable Microscope). Areas of interest for the assessment of failure focused on the hole and sides.



Fig. 5. OHC test set up: (a) Schematic representation of the specimen fitted with the extensometer and (b) Dartec 100kN test machine used for the experiments.

4. RESULTS AND DISCUSSION

4.1. Thickness, density, fibre volume fraction and void content

The physical properties of LATP manufactured laminates are shown in Table 2. Repassing along the fibre direction had insignificant effect on the thickness of the laminate. However, repassing in the transverse direction affected the thickness by reducing by 1.23%. The density of the reference laminate (no repass treatment) was found to be 1.44 g/cm³, while for the treated

laminates an increase was found within a range of 2.04-2.70% after the single, double and perpendicular repasses. The increased density values correspond to reduced void content from the untreated to the treated laminates. Void content significantly reduced progressing from no repass to single, double and perpendicular repasses by amounts of 17.81%, 17.57% and 19.00%, respectively. The autoclave consolidated laminates provided the lowest measured thickness and void content values (void content reduction of 31.35% compared to the reference LATP laminates) and the highest density and fibre volume fraction values.

Repass type	Thickness (mm)	Density (g/cm ³)	FVF (%)	Void (%)	
NR	3.28±0.05	1.44±0.07	61.36±0.85	4.21	
R1	3.29±0.04	1.47±0.01	62.00±0.57	3.46	
R2	3.28±0.04	1.47±0.01	62.05±0.50	3.47	
PR	3.24±0.03	1.48±0.01	62.36±0.46	3.41	
Autoclave	2.88±0.03	1.57±0.01	65.97±0.23	2.89	

Table 2. Density, thickness, volume fraction and void content values of CF/PEEK [0°]₁₆ laminates.

4.2. Surface roughness

Surface roughness was measured to identify the influence of repass on surface quality (Table 3). Fig. 6 shows the surface roughness values of the LATP treated samples and their comparison with laminates manufactured with gas-heated ATP treated samples [12]. From the surface roughness values obtained from LATP samples $[0^{\circ}]_{16}$, it is clear that these values are better for LATP than gas-based ATP. This improvement in surface roughness was in the range of 18.46-74.71%, considering all repass trials. The R_a values for the angle ply samples $[\pm 45^{\circ}]_{4s}$ were also measured after using a sampling length parallel to the direction of the repass treatment. The results indicated that the R1 laser repass treatment improved surface roughness significantly, R2 treatment marginally improved the surface roughness, while R3 treatment achieved similar results to R2.

Repass type	<i>R_a</i> (µm)			
	Present study [0°] ₁₆	Present study [±45°] _{4s}	Shadmeri et al. [12], [0°] ₂₄	
NR	4.88	3.17	19.30	
R1	2.36	2.73	3.80	
R2	2.12	2.59	2.60	
PR	2.02	NA	6.20	
R3	NA	2.60	NA	
Autoclave	0.48	0.30	0.50	

Table 3: Surface roughness results for the treated and untreated CF/PEEK laminates.

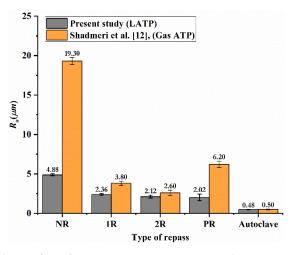


Fig. 6. Comparison of surface roughness between LATP and gas based ATP.

4.3. DSC analysis

DSC analysis was conducted on UD $[0^{\circ}]_{16}$ and angle ply $[\pm 45^{\circ}]_{4s}$ composite laminates, CF/PEEK dry prepreg tape and autoclave consolidated samples. Thermal transitions of $[0^{\circ}]_{16}$ and $[\pm 45^{\circ}]_{4s}$ laminates, which were both treated and untreated are shown in Fig. 7(a-b), respectively, as retrieved from the Netzsch Polyma 214 system. The area under the exothermic crystallization peak was used for the calculation of the crystallinity content and the enthalpy of fully crystalline PEEK was set at 130 J/g for the analysis.

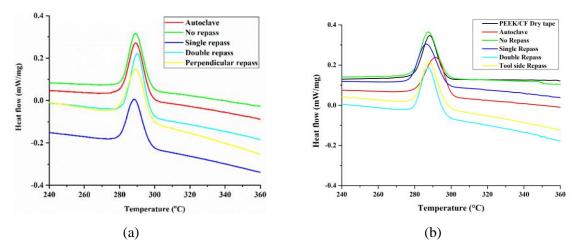


Fig. 7. Heat flow (exo↑) to temperature curves (Thermal transitions): (a) Thermal transitions for UD [0°]₁₆ laminates and (b) Thermal transitions for angle ply [±45°]_{4s} laminates.

The heat flow to temperature curves for all case studies presented in Fig. 7(a-b), have their peak values varying within the range of 250°C-320°C. The CF/PEEK prepreg tape used for the manufacture of the laminates, was found to have 20.98% crystallinity. Moreover, the angle ply LATP manufactured and laser annealed samples along with the autoclave samples were of similar crystallinity values (Table 2). UD [0°]₁₆ LATP manufactured and laser annealed samples

showed a similar trend with respect to crystallinity values 25.03% and 25.68%. The angle ply laminate's crystallinity content for the laser treated samples slightly increases for the single laser repass but remains within a similar range as the untreated samples. The dense arrangement of carbon fibres within the composite material does not allow for the full growth of crystals, as mentioned by Gao et al. [20] and affects crystallinity. The effect of the latter parameter can be observed from the results of the double laser repass treatment on UD $[0^{\circ}]_{16}$ laminates. The double laser repass direction was parallel to the fibre direction which enhances crystallinity levels and is opposite in effect to the case of angle ply laminates where the laser direction does not coincide with the fibre direction. The high cooling rates incurred by LATP limit the full crystallisation of PEEK and promote trans-crystalline growth, a phenomenon which affects the interphase regions between the layers as mentioned by Ray et al [1]. Ostberg et al [21] studied the annealing effect on crystal growth for PEEK and its composites and mentioned that the crystalline content of non-optimally (i.e. not full densification) shaped crystals increases at a given temperature, during annealing. The crystallinity estimation of the autoclave consolidated samples was expected to be higher due to the favourable conditions for equi-axed crystal growth. Two main factors can hinder the quality of these results, namely the applied thermal history for the DSC experiment does not necessarily correspond to the same history followed for the autoclave consolidation, and sampling from the uppermost layers of laminates may not be representative of the overall laminate. Mazumdar et al. [22], found that the crystallinity levels of uppermost layers were found to be smaller than the crystallinity levels estimated for the first few layers. In this study, the present results show insignificant effects to the crystalline content by annealing; however, it is possible to affect the amorphous region of PEEK which macroscopically evidences itself as matrix-dominated mechanical properties.

The estimated values of crystalline content for all samples with different laser repasses are shown in Table 4. The areas under the exothermic crystallization peaks are also included in Table 4.

Layup	Repass type	PEEK Crystallinity (%)	Area (J/g)
CF/PEEK tape	-	20.98	13.64
	NR	25.03	16.27
	R1	21.65	14.08
[0°]16	R2	26.28	17.08
	PR	25.52	16.56
	Autoclave	25.68	16.69
[±45°] _{4s}	NR	25.64	16.66
	R1	25.78	16.76
	R2	25.49	16.57
	R3	25.47	16.55
	Autoclave	24.24	15.75

 Table 1: Crystallinity percentages for CF/PEEK composite laminates.

4.4. Interlaminar shear strength

The effect of laser repass treatment on the ILSS response of LATP manufactured CF/PEEK unidirectional laminates is shown in Fig. 8(a-c). As shown in Fig. 8a, the single laser repass treatment on each layer has an insignificant effect on ILSS values. However, the double laser repass treatment increases ILSS values compared to the reference (NR) and single laser repass ILSS values by 3.80% and 2.79%, respectively. Similarly, laser repass perpendicular to the direction of fibres also increases ILSS values compared to the reference (NR) and single laser repass repass ILSS values by 2.23% and 1.91%, respectively. Nevertheless, both double and perpendicular laser repass treatments achieve similar ILSS values. The autoclave consolidated samples exhibit the largest ILSS value \approx 105 MPa, which differs by a factor of 2 or more compared to the ILSS values of the laser treated laminates. The ILSS value achieved by hot-gas ATP, 94 MPa [23], is closer to that achieved by autoclave.

In Fig. 8b, the ILSS values of the $[\pm 45^{\circ}]_{4s}$ layup are shown for the reference LATP manufactured samples (NR) compared to laser treated (R1) and autoclave consolidated (A) samples. Regarding angle-ply laminates, the laser repass does not exhibit a significant influence on ILSS. Fig. 8c shows a sample load-extension response for the no repass and treated laminates. The curves are of a similar trend indicating that the laser repass treatment has an insignificant effect on the interlaminar shear response of the laminates. The viscoelastic behaviour of the thermoplastic matrix is prominent within the non-linear response of the samples as shown in Fig. 8c.

It is interesting to note that the 0° ILSS samples exhibit a significantly higher strength than the $\pm 45^{\circ}$ ILSS samples, for both the autoclave and LATP processed laminates. This difference in strength can be attributed to a combination of factors. Generally for fibre-reinforced polymer composites, the shear modulus G_{13} is approximately 2.5 greater than G_{23} , indicating that 0° fibres have a greater influence on through thickness shear properties than any other fibre orientation. As such, the $\pm 45^{\circ}$ ILSS sample is significantly more compliant in transverse shear than the 0° ILSS sample. In addition, the $\pm 45^{\circ}$ ILSS sample is more susceptible to interlaminar damage due to the 90 degree difference in ply angle. Huang et al. [24] that suggests that laminates with relatively homogeneous microstructures with less resin-rich regions between the adjacent plies and within the plies have a higher interlaminar shear strength. Unidirectional laminates, like the autoclaved 0° ILSS sample will have almost indistinguishable interlaminar regions, whereas those in the $\pm 45^{\circ}$ sample will be more pronounced.

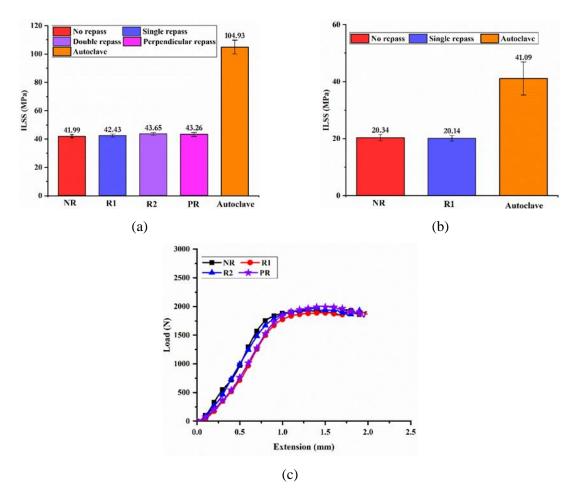


Fig. 8. Influence of repass on the ILSS: a. UD [0°]₁₆, (b) Angle-ply [±45°]_{4s} and (c) Typical load-extension response.

In Fig. 9, the failure modes of the LATP manufactured UD laminates are shown as digital images captured by a high definition camera. Specifically, the reference NR samples (Fig. 9a) exhibit delamination followed by the deep indentation of the loading nose, and subsequent fibre failure and deformation. All laser repass treated specimens, (Fig. 9b to 9d) exhibit failure with minor cracks and negligible delamination. Moreover, the above mentioned samples exhibit delamination failures that mostly occur in the bottom layers while small cracks appeared at the edges. The images presented in Fig. 9 suggest that the repass annealing treatment has embrittled the laminates relative to the no repass laminate. The NR laminate exhibits significant plastic deformation prior to failure not seen in the repass specimens.

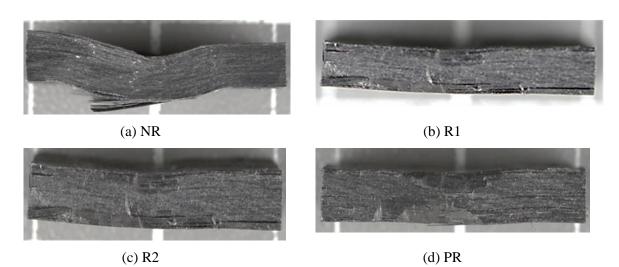


Fig. 9. Influence of laser repass on the interlaminar shear failure of UD [0°]₁₆ laminates.

4.5. Open hole compression

The influence of different repasses on the OHC strength of LATP manufactured laminates is shown in Fig. 10. The single laser repass treatment increased OHC strength by only 1.2% while the double laser repass treatment had a decreasing effect of 4%. These low strength values may be attributed to poorer consolidation and larger void content as observed in [25]. The autoclave consolidated specimens achieved 72.6% higher OHC strength as compared to the LATP consolidated specimens. The laser repass treatment on the tool-side showed similar results to the single laser repass specimens.

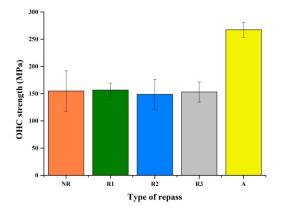
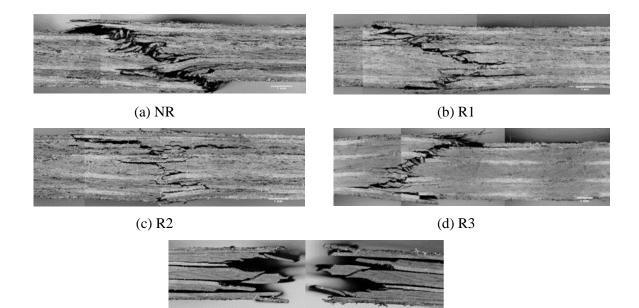


Fig. 10. Variations of compressive strength values for all cases of consolidation treatments.

Fig. 11 shows fractographic images from the lateral sides of OHC specimens captured using an optical microscope. Observation of the undamaged areas of the untreated sample (Fig. 11a), and the laser treated samples (Fig. 11(b-d)), indicate that they appear to be more porous than the autoclave consolidated sample (Fig. 11e). The failure modes observed were similar for all processing procedures, indicating either lateral or angled failure within the gauge area.

In Fig. 12(a-e), the top views of the laser treated open-hole specimens are shown. While delamination, fibre and matrix cracking occurred in the LATP specimens (Fig. 11(a-e)), they did not exhibit the same clean fracture as observed in the autoclave consolidated specimens (Figs. 11(e), 12(e) and 12(f)). The images presented suggest that the laser annealing treatment has an insignificant effect on the LATP specimen failure modes, and as mentioned in Ref. [12] the effect of the treatment is more evident on the surface finish, the void content and the crystallinity content. The test results clearly indicate the LATP in-situ consolidated specimens do not achieve the same level of consolidation as exhibited by the autoclave processed laminates.



(e) A

Fig. 11. Failure modes of OHC samples (side view) delamination and buckling of composite layers.

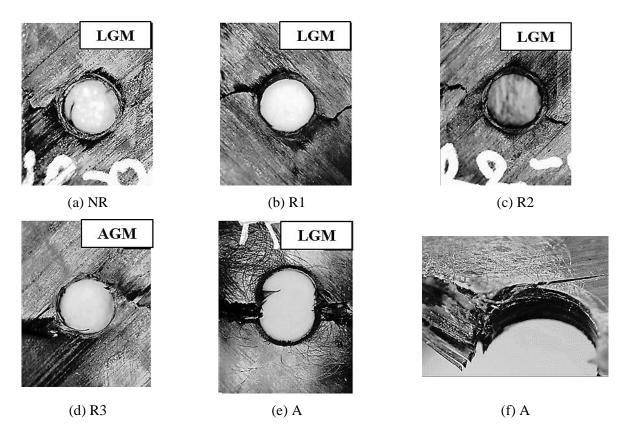


Fig. 12. Details of failure modes from the top view of the hole areas for the OHC specimens, both lateral (LGM) and angled (AGM) failure types: (a) no repass, (b) single repass (c) double repass, (d) tool-side repass, (e) autoclave-1 and (f) autoclave side view.

4.6. In-plane shear

The in-plane shear strength (shear yield strength) values from the laser annealing treatment samples are shown in Fig. 13. The in-plane shear test results for the LATP samples was within the same range as reported in the literature for similar thermoplastic composite systems [26]. It was found that the in-plane shear strength of single laser repass samples increased by 3.57%, but for the case of double laser repass a reduction of 7.75% in yield stress was observed. The laser repass treatment on the tool-side had a negligible effect on in-plane shear strength and the values were comparable to the untreated specimens. However, the autoclave consolidated specimens achieved an increase of 35.8% in in-plane shear strength compared to laser treated laminates. This is likely due to the fact that the LATP processed laminates appear to have higher levels of porosity than those processed by autoclaving, reducing the overall shear strength. In-plane shear performance is closely linked to fibre-matrix interface performance. The lower values achieved for the R2 and R3 repasses may be attributed to the annealing repass alleviating the residual stresses in the matrix that help to anchor the fibres in the matrix. The results presented here suggest that one annealing repass can improve the laminate properties, but subsequent repasses have either minimal effect or can reduce the mechanical properties.

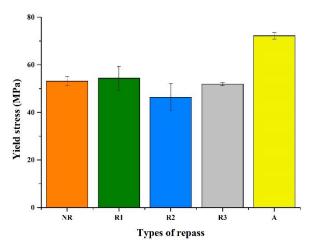


Fig. 13. Variation of the yield stress values for the annealed samples.

5. CONCLUSIONS

The purpose of this study was to assess the influence of laser repass annealing on the surface roughness, physical properties and mechanical properties of LATP in-situ consolidated CF/PEEK laminates. Laminates with layups $[0^{\circ}]_{16}$, $[-45^{\circ}/0^{\circ}/45^{\circ}/90^{\circ}]_{2s}$ and $[\pm 45^{\circ}]_{4s}$ were manufactured from IMS65/PEEK unidirectional tape (Teijin Carbon America, Inc.). Four laser repass strategies were examined; single repass, double repass, perpendicular repass and a tool-side repass. For comparison purposes, autoclave processed laminates were also included in the study.

Laser repass annealing was found to marginally increase density values and decrease void content for the $[0^{\circ}]_{16}$ laminates. The laser repass also improved the surface roughness of laminates; the surface roughness values determined were better than that reported in the literature for hot-gas torch repass treatment [12]. The crystallinity results indicated that the LATP laminates exhibited higher crystallinity then the prepreg tape used to manufacture the laminates. However, subsequent repass treatments did not increase the crystallinity level significantly. The autoclave laminates exhibited similar levels of crystallinity to the LATP specimens, this result is in contrast to the comparison between LATP and autoclave laminates reported in the literature [1] processed from CF/PEEK supplied from a different source.

The ILSS performance of the LATP laminates was significantly poorer than the autoclave processed laminates. Images of the failed LATP ILSS specimens indicated that the NR specimen exhibited more plastic deformation than the repass specimens. However, while the $[0^{\circ}]_{16}$ laser repass specimens exhibited a marginally higher ILSS value than the no repass specimens, the difference was within the standard deviation of the test results.

In the case of OHC and IPS performance, the specimens subjected to a single laser repass achieved the best IPS and OHC performance. The double laser repass specimens exhibited the poorest IPS and OHC performance, suggesting that double laser repass treatment may have a detrimental effect on mechanical properties, a 12.8% and 4% decrease for IPS and OHC test results respectively was observed. The lower properties achieved for subsequent laser repasses could be attributed to the annealing repass alleviating the residual stresses in the matrix that help to anchor the fibres in the matrix. The results presented here suggest that one annealing repass can improve laminate properties, but subsequent repasses have either minimal effect or can worsen them. Further work is required to define the exact mechanisms behind any improvement in mechanical performance.

Acknowledgements

The authors would like to thank Science Foundation Ireland (SFI) for funding Spatially and Temporally VARIable COMPosite Structures (VARICOMP) Grant No. (15/RP/2773) under its Research Professor programme. This project has received funding from Enterprise Ireland and from the European Union's Horizon 2020 research and innovation programme under the Marie-Sklodowska Curie grant agreement No 713654. The authors would also like to thank ICOMP for its help with the LATP.

REFERENCES

- Ray, D., Comer, A. J., Lyons, J., Obande, W., Jones, D., O'Higgins, R. M., McCarthy, M. A. "Fracture toughness of carbon fiber/polyether ether ketone composites manufactured by autoclave and laser-assisted automated tape placement." *Applied Polymer Science*, (2015), 132 (11).
- 2. Schledjewski, R. "Thermoplastic tape placement process in-situ consolidation is reachable.", *Plastics, Rubber and Composites*, (2009) 38:9-10, 379-386.
- Oliveri, V., Zucco, G., Peeters, D., Clancy, G., Telford, R., Rouhi, M., McHale, C., O'Higgins, R. M., Young, T. M., Weaver, P. M. "Design, Manufacture and Test of an In-Situ Consolidated Thermoplastic Variable-Stiffness Wingbox." *AIAA Journal*, (2019), 57 (4), 1671-1683.
- Bandaru, A. K., Clancy, J. G., Peeters, D., O'Higgins, R. M., Weaver, P. M. "Interface characterization of thermoplastic skin-stiffener composite manufactured using laserassisted tape placement" *Proceedings of the AIAA/ASCE/AHS/ASC Structures, Structural Dynamics, and Materials Conference*, AIAA SciTech Forum, (AIAA 2018-0481), 2018.
- Peeters, D., Clancy, J. G., Oliveri, V., O'Higgins, R., Jones, D., Weaver, P. M. "Concurrent design and manufacture of a thermoplastic composite stiffener." *Composite Structures* (2019), 212, 271-280.
- 6. Bandaru, A.K., Clancy, J. G., Peeters, D., O'Higgins, R. M., Weaver, P. M. "Properties of a thermoplastic composite skin-stiffener interface in a stiffened structure manufactured by

laser-assisted tape placement with in-situ consolidation." *Composite Structures* (2019), 214, 123-131.

- Clancy, J. G., Peeters, D., Oliveri, V., Jones, D., O'Higgins, R. M., Weaver, P. M. 2019, "A study of the influence of processing parameters on steering of carbon Fibre/PEEK tapes using laser-assisted tape placement." *Composites Part B: Engineering* (2019), 163, 243-251.
- 8. Denault, D. and Dumouchel, M. "Consolidation process of PEEK/Carbon composite for aerospace applications." *Advanced Performance Materials* 5 (1998), 83-96.
- Ray, D., Comer, A.J., Lyons, J., Obande, W., Jones, D., O'Higgins, R., O., McCarthy, M., A. "Fracture toughness of carbon fiber/polyether ether ketone composites manufactured by autoclave and laser-assisted automated tape placement." *Appl. Polym. Sci.* (2015).
- Papkov, V. S., Gerasimov, M. V., Dubovik, I. I. "Disordering of crystalline PEEK upon mechanical treatment." *Polymer Engineering and Science* (1997) Vol/37, No 8.
- 11. Zhang, Z. and Zeng, H. "Effects of thermal treatment on poly(etherether ketone)." *Polymer* (1993), Vol.34, 17, 3648-3652.
- 12. Shadmehri, F., Hoa, S. V., J. Fortin-Simpson, J., Ghayoor, H. "Effect of in situ treatment on the quality of flat thermoplastic composite plates made by automated fiber placement (AFP)." *Advanced Manufacturing: Polymer & Composites Science* (2018), 4:2, 42-7.
- Hoa, S., Shadmehri, F., Fortin-Simpson, J., Ghayoor, H. "Effect of in-situ treatment on the quality of flat thermoplastic composite tubes made by automated fiber placement (AFP)," *European Conference on Composite Materials -ECCM 18*, 25-28 June, 2018, Athens, Greece.
- Stokes-Griffin, C. M., and Compston, P. "An inverse model for optimisation of laser heat flux distributions in an automated laser tape placement process for carbon-fibre/PEEK," *Composites Part A: Applied Science and Manufacturing* Vol. 88, 2016, pp. 190-197.
- 15. ASTM D792., "Standard Test Methods for Density and Specific Gravity (Relative Density) of Plastics by Displacement," 2008.
- ASTM D3171., "Standard Test Methods for Constituent Content of Composite Materials," 2015.
- 17. ASTM D2344., "Standard Test Method for Short-Beam Strength of Polymer Matrix Composite Materials and Their Laminates," 2016.
- ASTM D3518., "In-Plane Shear Response of Polymer Matrix Composite Materials by Tensile Test of a 645° Laminate," 2005.
- 19. ASTM D6484., "Open-Hole Compressive Strength of Polymer Matrix Composite Laminates," 2005.
- Gao, S. L. and Kim, L. K. "Cooling rate influences in carbon fibre/PEEK composites. Part 1. Crystallinity and interface adhesion." *Composites: Part A* (2000), Vol. 31, 517-530.

- 21. Ostberg, G. M., K. and Seferis, J., C. "Annealing Effects on the Crystallinity of Polyetheretherketone (PEEK) and Its Carbon Fiber Composite." *Journal of Applied Polymer Science*, (1987), Vol. 33, 29-39.
- Mazumdar, S. K. and Hoa, S., V. "Determination of manufacturing conditions for hot-gasaided thermoplastic tape winding." *Journal of Thermoplastic Composite Materials* (1996), Vol. 9.
- 23. Khan MA, Mitschang P, Schledjewski R. Identification of some optimal parameters to achieve higher laminate quality through tape placement process. Adv Polym Technol 2010;29(2):98-111.
- Huang, C., M. He, Y. He, J. Xiao, J. Zhang, S. Ju, D. Jiang, "Exploration relation between interlaminar shear properties of thin-ply laminates under short-beam bending and meso-structures." Journal of Composite Materials (2017), Vol. 52(17), 2375-2386.
- Comer, A., J., Ray, D., Obande, W., O., Jones, D., Lyons, J., Rosca, I., O'Higgins, R., M., McCarthy, M., A. "Mechanical characterization of carbon fibre-PEEK manufactured by laser-assisted automated-tape-placement and autoclave." *Composites: Part A* (2015), Vol. 69, 10-20.
- 26. Lafarie-Freno, M., C., and Touchard, F. "Comparative in-plane shear behavior of long carbon fibre-composites with thermoset or thermoplastic matrix." *Composites Science and Technology* (1994), Vol. 52, 4417-425.