THE EFFECT OF PLY ORIENTATION ON BOND STRENGTH IN FIBER-PLACED COMPOSITES

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ABSTRACT

The Laser-Assisted Fiber Placement (LAFP) process is an additive manufacturing technique with the potential for out-of-autoclave manufacturing. A thermoplastic composite pre-impregnated tape is heated above a laser above melting temperature and subsequently pressure is applied via a roller to bond the tape to previously laid down plies. LAFP offers the opportunity to combine the stacking and the consolidation, thereby reducing cost and process time compared to conventional processes, such as autoclave and press-consolidation. However, the consolidation quality achieved with LAFP is not yet at the same level as the conventional processes. One of the measures of consolidation quality is the interlaminar bond strength. The mandrel peel test is used as a method to quantify the effects of process parameters on the interlaminar bond strength. The aim is to optimize the interlaminar bond strength achieved and to increase the understanding of governing bonding mechanisms.

A LAFP robot was used to place carbon/PEEK tapes onto pre-consolidated substrates with various laser power settings and two different top ply fiber orientations. The fracture toughness of the bond was measured using the mandrel peel test. The results showed that the fracture toughness strongly depends on laser power. Underheating and overheating resulted in a decline in fracture toughness and the highest fracture toughness of about 1.30 kJ/m² was obtained for a nippoint temperature of around 500 °C. A similar maximum fracture toughness at equal laser power settings was measured for both 0° and 90° top ply orientations. However, the process window of the 90° top ply orientation seems to be smaller.

1 INTRODUCTION

The Laser-Assisted Fiber Placement (LAFP) process has the potential for out-of-autoclave manufacturing of thermoplastic composites [1]. Out-of-autoclave manufacturing can reduce production costs substantially. During the LAFP process, a thermoplastic composite pre-impregnated tape is heated above the melting temperature by a laser and subsequently bonded to previously laid down plies by applying pressure via a roller. Figure 1 shows this process schematically. In this way, a component is built up and consolidated ply-by-ply. Heat and pressure are applied to consolidate the material during the stacking process, in contrast to conventional consolidation processes, such as autoclave and press-consolidation, where stacking and consolidation are two consecutive steps. The combination of simultaneously stacking and consolidating is termed in-situ consolidation. A post-consolidation step can be omitted in case the in-situ consolidation quality achieved is sufficiently high.

Consequently, in-situ consolidation yields a reduction in process time and cost. Furthermore, LAFP offers several other advantages. The fully automated stacking improves the reproducibility of components. Defects in the laminate lay-up originating from errors of stacking by hand can be prevented, resulting in higher quality components. Components can also be manufactured in near-net shape, resulting in a reduction of material waste after trimming. Finally, component performance can be improved by tailoring the laminate, in terms of local fiber orientation and part thickness. First, the lay-up can be optimized with different orientations.
for every single ply, without an increase in costs. Second, LAFP can place the thermoplastic tape material in curved paths, so as to manufacture variable stiffness laminates. In variable stiffness laminates not only the orientation of individual plies in the lay-up is tailored, but the local ply orientation is optimized to load paths. Substantial weight reductions are feasible with variable stiffness laminates [2]. Third, LAFP can also be used to tailor laminates by local reinforcement with extra plies. The final performance of a LAFP-produced component is based on the consolidation quality achieved in terms of void content, crystallinity, residual stresses and interlaminar bond strength. This research focuses on the interlaminar bond strength, since it is essential for transferring the load between plies.

Lasers are an ideal heat source for the fiber placement process, offering a high energy density and short response times [3]. The laser light of the equipment used is absorbed by the carbon fibers in the composite and the fibers in turn heat the surrounding thermoplastic matrix. Consequently, the fiber orientation of the substrate, besides the laser incidence angle and surface roughness of the thermoplastic composite tape, affects the heat absorption and therefore the achieved interlaminar bond strength.

Process parameters, such as laser power and placement velocity, will affect the achieved interlaminar bond strength [4]. The effects of process parameters and substrate orientation needs to be quantified to be able to optimize the interlaminar bond strength, to enhance component performance.

Several tests can be used to characterize the bond strength between plies. The mandrel peel test is one of these [5]. It characterizes the fracture toughness of the bond by peeling a ply from a substrate. The complete layup of the laminate can be peeled ply-by-ply with this measurement technique, to assess each interface. Grouve et al. showed that the mandrel peel test is able to quantify the effects of process conditions on the interlaminar bond quality of UD-tapes on woven substrates [5]. This makes the mandrel peel test an ideal testing method to optimize the bond quality obtained with the LAFP process. This research concentrates on the effects laser power and ply orientation on the interlaminar bond quality for UD carbon/PEEK on a UD substrate using the mandrel peel test. The effects of various laser power settings and two different ply orientations on the fracture toughness were investigated.

2 INTERLAMINAR BONDING

2.1 Bond strength development

The bonding process during the LAFP process can be divided into two steps, as shown in Figure 2. The first step is the **heating** step, in which the tape and substrate are heated by a laser. Sufficient heat should be supplied to melt the tape and the surface of the substrate in order to be able to bond them in the second step. Pressure is applied via a roller during the **consolidation** step to consolidate the tape and substrate. Underneath the roller the bond between the tape and substrate is obtained. No more heat is added during this consolidation step and the material will cool down during this step. Once the pressure is released, the substrate with the bonded tape should be cooled down sufficiently to prevent deconsolidation.

The heated tape and the substrate are brought into contact in the consolidation step to allow diffusion of polymer chains across the interface. Interdiffusion can only take place where the two surfaces are in intimate contact. Intimate contact development comprises flattening of the tape and substrate surface roughness. Increasing the applied consolidation pressure will accelerate the deformation of surface asperities, thereby
facilitating the intimate contact development. The deformability of the polymer matrix material increases with temperature, since the viscosity reduces. Therefore, a higher process temperature and consolidation pressure will facilitate the development of intimate contact [6, 7].

The actual bonding of the two surfaces in contact, is based on polymer movement described by the reptation theory of chain mobility in which the thermoplastic polymer matrix is considered as an entanglement of polymer chains [8]. The mobility of chains is restricted by neighboring chains to movement in axial direction. The chain ends are free to move, resulting in a crawling movement of chain through the entanglements. Polymer chains can therefore migrate over the interface of the two surfaces in contact, merging the two interfaces. The fusion bonding mechanism is usually referred to as healing. A sufficient amount of heat should be supplied to increase the polymer chain mobility, thereby reducing the time needed for healing. The heat is also needed to melt all the crystals in a semi-crystalline polymer, such as the PEEK used in this research. The crystals impede the chain movement and therefore increase the time needed for healing or even inhibit healing of the tape and substrate [9, 10]. However, if too much heat is applied, the polymer degrades which may result in a cross-linking polymer in the case of PEEK [11]. The thermal degradation not only reduces the polymer properties, but the cross-linking also reduces chain mobility, thereby slowing down the healing process [4]. The optimal process temperature is therefore a compromise. Enough heat should be supplied to reduce the viscosity and to melt all crystals, while preventing thermal degradation of the matrix. Under and overheating would both result in a reduction in bond strength.

2.2 Bond strength assessment

Traditionally, the Short Beam Strength (SBS) test has been used to test the interlaminar bond strength of composites laminates. This method is also used for LAFP-produced laminates to help optimizing the process parameters [3, 12]. The interface in which the failure occurs during testing, however, can not be predicted. Therefore, it is difficult to assess the bond strength of individual interfaces or the interface between differently oriented plies with the SBS test. As a consequence, the SBS test is not the ideal test for optimizing process parameters for individual plies.

Peel tests can be used instead to assess the bond quality of a predefined interface. The wedge peel test uses a wedge to fracture two bonded tapes, starting from a predefined crack initiator to test the selected interface [3, 12]. The fracture toughness can be calculated from the peel force required to propagate the crack over the interface. It is assumed that the fracture toughness, like interlaminar shear, is a measure for interlaminar bond quality. However, specimens for this test usually consist of two plies, therefore it is difficult to test the fracture toughness between differently orientated plies. The mandrel peel test, however, does offer this opportunity. Figure 3 shows the mandrel peel test schematically. In this test a tape is peeled from a substrate, which can have various orientations or can even be manufactured from another type of material [13]. The interface between different plies of the layup can be characterized which allows optimization of process settings for each individual ply. Essentially this test is a modified 90° peel test, with the addition of a mandrel to be able to peel a stiff composite tape without introducing permanent deformation or tape
breakage. The specimen, a laminate substrate with a bonded tape on top, is connected to a sliding table and the tape is bent around the mandrel. A force is applied to peel the tape from the substrate, while a constant alignment force is required to ensure conformity of the tape to the mandrel. The mandrel ensures a certain radius of the tape, to prevent breakage of the tape. The fracture toughness is calculated from equation 1 as derived by Grouve [5]:

$$G = \frac{1}{w}(F_p(1 - \mu) - F_a)$$  \(1\)

where \(F_p\) is the peel force, \(F_a\) is the alignment force, \(\mu\) is the friction coefficient of the setup and \(w\) is the width of the peeled tape. Two force cells are used to measure the peel and the alignment force respectively. The friction coefficient is obtained by performing the test on a tape which is not bonded.

3 EXPERIMENTAL METHOD

The effectiveness of the mandrel peel test to quantify the effects of process conditions on the bond strength of Carbon PPS tapes on woven substrates was shown by Grouve [5]. In this research the mandrel peel test was used to characterize the bonding of carbon/PEEK tapes on substrates from carbon/PEEK UD tape. The fracture toughness was measured for various laser power settings to investigate the effects of the amount of heat supplied. Furthermore, the orientation of the substrate was varied, to investigate the effects of substrate orientation on interlaminar bonding. Quantification of the effects of substrate orientation is essential to the development of LAFP in-situ consolidation for tailored laminates. The results were compared to show the potential of the mandrel peel test to be able to assess the effects of process settings on interlaminar bond strength and to show effects of substrate orientation on interlaminar bond strength.

Pre-consolidated laminates (300 by 300 mm, lay-up: [0\2]/90\s), manufactured by press consolidation from Ten Cate AS4 carbon/PEEK tape, were used as substrate to ensure uniform surface properties for all specimens. The tapes were placed on the substrate using a LAFP robot from Coriolis Composites at a placement speed of 100 mm/s. The laser power was varied from 475 W, which was just enough to bond the tape, to 750 W where smoke was a clear sign of thermal degradation. Tapes were placed at a certain distance from each other, to make sure the heat supplied to place a tape did not affect the bonding of an adjacent tape. Two placement directions were considered. First, the placement direction was parallel to the fiber direction of the top ply of the substrate was designated as 0° substrate orientation. The second placement direction was perpendicular to the fiber direction of the top ply of the substrate and this orientation was designated as the 90° substrate orientation. The temperature during the process was measured using a thermal camera mounted on the head of the LAFP robot. The accuracy of the thermal measurements is highly depended on the used emissivity settings of the camera. Beforehand, the emissivity values were defined by comparing thermal camera measurements with thermocouple measurements in a steady state oven setup. These validation measurements showed that the camera was able to measure the temperature accurately to within 5°C in the temperature range of interest.

The specimens were cut from the laminate to test each tape. After testing, the fractured specimen was
tested again to measure the friction in the mandrel peel test setup, so as to correct the measured peel force. The width of the tape was measured at two locations with a micrometer afterwards, to be able to calculate the fracture toughness. Also the thickness of the peeled tape was measured at the begin and end of the crack to analyze the change in thickness. A large change in thickness indicated a fracture plane which was not on the interface and the measured value was therefore the intralaminar fracture toughness of the tape or substrate. The results of specimens with a large change in tape thickness were discarded or the fracture toughness was only calculated for part of the measurement. Several specimens, under various process conditions, were investigated with an optical microscope to determine the failure mode of the specimen.

4 RESULTS

4.1 Effect of laser power on fracture toughness

Figure 4 illustrates a typical result of the peel experiments. The graph shows the effective peel force and friction force as a function of the peel distance for a specimen manufactured at 100 mm/s, 600 W and with a 0\(^\circ\) substrate orientation. The graph shows the start-up effect of the measurement between 0 and 20 mm. During this start-up the pressure on the pressure cylinder for the alignment force is applied and the movement of the head is started. The mean of the effective peel force, shown with a black dotted line, is corrected with the mean of the friction force to calculate the fracture toughness.

![Figure 4: Measured peel and friction force during peel experiment of a specimen on a 0° substrate orientation, manufacture with 600 W of laser power at 100 mm/s.](image-url)
Figure 5 shows the fracture toughness for different laser power settings. The markers show the calculated mean value of each specimen and the error bars show the standard deviation of the average value of each power setting. These results are from 3 sets with 2 to 4 specimen per sample. A large decline can be observed for the lowest and highest laser powers. A plateau is visible for laser powers of 500 to 700 W. A slight peak is shown for a laser power of 575 W with a maximum average fracture toughness of 1.28 kJ/m².

Figure 5: Fracture toughness as function laser power.

Figure 6 shows optical microscopy pictures of the typical fracture surfaces, perpendicular to the peeling direction, for different power settings. The substrate, tape, fracture interface and peel direction are indicated in Figure 6a. The fracture plane corresponds almost exactly with the interface between tape and substrate for the 500 W power setting, Figure 6a. This is typical for an interlaminar failure, suggesting incomplete bonding between the two plies. For the 600 W substrate, in Figure 6b, part of crack propagated is through the tape. This suggests a proper bonding between the tape and substrate. The measured fracture toughness, however, is partly the fracture toughness of the tape material. The fracture plane of the 750 W sample shown in Figure 6c lies on the interface between the tape and the substrate, again showing an interlaminar failure.

However, in Figure 6c failure of the second ply of the substrate is visible for the 750 W specimen. Large voids and are apparent in the second ply of the substrate for this power setting. These voids result in a delamination of the second ply, as shown in Figure 6c. A similar but less severe void growth is visible for the 600 W specimen and interlaminar voids are apparent between the first and second ply of the substrate. As a consequence, the 600 W specimen also shows an interlaminar failure between the first and second ply of the substrate.
4.2 Effect of substrate orientation on fracture toughness

Figure 7 shows the results of the fracture toughness measurements for the $0^\circ$ and $90^\circ$ orientation for different power settings. The measured fracture toughness of the $90^\circ$ orientation is lower for each power setting. This is particularly the case for the 500 W and 750 W, with reductions up to 30%. There is hardly any reduction for the 600 W power setting, it is within the standard deviation of the measurement. The scatter of the individual specimens is clearly larger for the 500 W and 750 W power settings.

The $90^\circ$ orientation results were also analyzed with an optical microscope and the images are shown in Figure 8. Similar failure mechanisms can be observed for both the $0^\circ$ and $90^\circ$ substrate orientation. The 500 W and 750 W show a clear interlaminar failure on the interface between the substrate and the tape. Only some fibers remain on the substrate for the specimen manufactured at 500 W. For the 600 W specimen a larger number of fibers remain on the substrate and part of the fracture plane propagated through the tape. This failure mode indicates a good bonding of the tape with the substrate. For the 600 W specimen less voids are visible in the substrate, compared to the $0^\circ$ substrate orientation. For the 750 W specimen large voids and delaminations are visible, similar to the specimen of the $0^\circ$ substrate orientation.
Figure 7: Comparison of the results of the $0^\circ$ and $90^\circ$ substrate orientation.

Figure 8: Microscopy images of fractured specimen with a substrate orientation of $90^\circ$. The white line indicates the fracture surface.
5 DISCUSSION

5.1 Effect of laser power on fracture toughness

The low fracture toughness values for the high and low laser power settings can be explained by the failure mode observed on the microscopy pictures. An interlaminar failure was observed for the 500 W and 750 W specimens, suggesting an incomplete bond. This supports the theory that no complete bonding can be achieved at low temperatures due to incomplete melting of the material. The incomplete bonding at 750 W, resulting in an interlaminar failure as determined by the microscopy pictures, seems to be caused by material degradation. This degradation was also observed during placement, as smoke development. The matrix material on the interface between the tape and the substrate needs to be analyzed further to be able to prove the thermal degradation. However, the high laser power showed a detrimental effect on the fracture toughness of the bond.

The LAFP process seems to have a devastating effect on the consolidation quality of the substrate. Severe interlaminar void growth between the first and second ply and intralaminar void growth in the second ply of the substrate was observed for the 750 W specimens. For the 600 W power setting some interlaminar voids can be observed between the first and second ply of the substrate. These interlaminar voids should be prevented, since they act as crack initiators. The consequence of these interlaminar voids are delaminations during the peel test. The interlaminar voids are only apparent for the 600 and 750 W samples and not for 500 W samples. Therefore, it seems that when more heat is added to the substrate, deconsolidation is facilitated. This effect might be less severe for the placed tape, since the roller can act as a heat sink. The effect of supplied heat on the substrate should be investigated further for various placement velocities to understand the observed interlaminar void growth.

The fracture toughness results show a plateau between 500 and 700 W, as can be seen in Figure 4. In this regime sufficient heat was added to bond the tape and the substrate properly, while preventing degradation. The plateau suggest a large process window for the laser power. This is beneficial for further optimization for other mechanical properties of LAFP-components. Figure 9 shows the fracture toughness of each specimen as function of the measured nippoint temperature. The carbon/PEEK processing temperature, 380°C, and the PEEK autoignition temperature, 595°C, are also shown [14]. The autoignition temperature is the temperature where the material starts to burn, however it is expected that degradation already occurs at lower temperatures [11]. Figure 9 shows that the measured nippoint temperature should be above the normal process temperature to obtain a good bond strength, but below the autoignition temperature to prevent degradation. The highest fracture toughness is measured for the specimen around 500°C.

![Figure 9: The fracture toughness as function of nippoint temperature](image)
The fracture surface should be studied in more detail to clarify the high standard deviation measured for specimens with a laser power of 600 W and higher. This can partly be explained by the microscopy results of the 600 W sample in Figure 6b. The high bond strength of the placed tape increases the probability of a deviation of the fracture plane. The fracture plane progressed into the tape for the 600 W specimen, as large parts of the tape remained on the substrate. The measured fracture toughness is affected by this behavior, since locally the fracture toughness of the bonded tape is measured. Furthermore, any interlaminar voids, as shown in Figure 6c, can act as crack initiators resulting in a second fracture plane in the substrate. The second fracture plane increases the area of the fracture plane, resulting in a higher peel force and as a result in an increased fracture toughness. Therefore, the failure mode of the specimen needs to be identified in order to discard specimens with incorrect failure modes.

The high bond strength at 600 W is also visible as a slight peak in fracture toughness around 550 to 650 W in Figure 4. This is the range of power settings which should be investigated further. It should also be noted that the crystallinity of the specimens is yet unknown and differences in crystallinity can affect the fracture toughness [15].

5.2 Effect of substrate orientation on fracture toughness

The 90° orientation specimens show a significant reduction in fracture toughness for the 500 W and 750 W power settings. This suggests a smaller processing window for the 90° substrate orientation. A larger number of power settings, similar to the settings used for the 0° substrate orientation, are needed to support this. The low fracture toughness measured and the failure mechanisms identified for the 500 W and 750 W show a low bond strength, due to underheating and overheating respectively. The fracture toughness of the 600 W power setting is similar for both the 0° and 90° substrate orientation. The microscopic images support the high fracture toughness measured, with a large number of fibers which remain on the substrate, suggesting a high bond strength.

The voids and delaminations apparent in the substrate in Figure 6 for the 0° substrate orientation, are also observed for the 90° substrate orientation in Figure 8. So for both the 0° and 90° substrate the added heat causes void growth and the voids in the substrate promote delaminations in the substrate.

6 CONCLUSIONS

The aim of this research was to quantify the effect of process parameters, the laser power and the substrate orientation, on the fracture toughness of tapes placed with LAFP. Tapes were placed on pre-consolidated laminates with various laser power settings and in two different placement directions. The fracture toughness of these specimens were measured with the mandrel peel test and the failure mechanisms were analyzed with an optical microscope.

It can be concluded that the mandrel peel test is suitable to quantify the effects of process conditions on interlaminar bonding. The measured fracture toughness is shown to be affected by laser power, particularly for insufficient heating and overheating. However, the fracture surface of the tested specimen need to be analyzed further to ensure that the fracture toughness measurements are not affected by deviating failure modes, such as both diversion of the fracture plane into the substrate and delaminations in the substrate. The results show only a slight variation in fracture toughness for a range of power settings. This implies a large process window in terms of laser power for interlaminar bonding, which is beneficial for further optimization of the process parameters for other consolidation quality measures, such as void content and crystallinity.

The effects of substrate orientation on the fracture toughness were also observable, although a larger number of power settings needs to be evaluated to confirm this. At the low and high power settings a decline of 30% in fracture toughness was measured for the 90° substrate orientation compared to the 0° substrate orientation. This suggest a narrower processing window for the 90° substrate orientation. The maximum bond strength achieve was similar for both substrate orientations, with a fracture toughness of about 1.30 kJ/m².

Finally a considerable effect of the LAFP process on the quality of the substrate was observed. The LAFP process facilitated void growth and deconsolidation of the substrate. This was particularly apparent
for the high laser power settings, but void growth was also visible for the power settings with the highest measured fracture toughness. This should be investigated further, to be able to prevent the deconsolidation of the substrate.

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