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INVESTIGATION OF THE INFLUENCES OF AFP PROCESS PARAMETERS ON THE CRYSTALLINITY AND MECHANICAL PROPERTIES OF LM-PAEK COMPOSITES

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Abstract: *AFP (Automated Fiber Placement) is a composite material manufacturing process that allows parts to be manufactured in an automated way. It consists of depositing tows or tapes of composite material with an AFP head (mounted on a robotic arm or on a gantry or column type structure) that applies pressure and temperature, and that moves at a specific speed. Until now, AFP technology has been widely used in the aeronautical industry to manufacture large parts, such as wings or fuselages, mainly using thermosetting matrix composite materials. In recent years, the aeronautical industry has begun to investigate the use of thermoplastic matrix composite materials, since their properties allow for shorter manufacturing processes, taking advantage of their reprocessing properties. Currently, one of the main research goals on the processing of thermoplastic materials with AFP is the In-Situ Consolidation (ISC), which allows to manufacture the piece and consolidate the material at the same moment of its deposition, resulting in the part being finalized at the end of its lay-up, avoiding the need to perform an autoclave cycle.*

In this work, ISC manufacturing with AFP of LM-PAEK tapes is studied, trying to optimize the quality of the composite specimens by understanding the relationship between the manufacturing parameters, crystallinity, and the obtained mechanical properties.

Keywords: Automated Fiber Placement (AFP); In-Situ Consolidation (ISC); Thermoplastic composites; Low melting Poly aryl ether ketone (LM-PAEK); Mechanical properties; Crystallinity

1. Introduction

The use of composites in the aeronautical industry has been present for decades. Automated manufacturing technologies, such as AFP (Automated Fibre Placement) have allowed the manufacturing of large structures, such as wings or fuselages, reducing production times, and increasing the repeatability and final quality of the parts. Thermoset based materials represent most composites used in aerospace, but in recent years, thermoplastic based composites are gaining relevance. The main advantage of thermoplastics over thermosets is that prior to curing, they have an unlimited shelf life at room temperature and do not require refrigeration. Also, recycling is possible, they can be remelted and reformed post cured, offering flexibility and sustainability advantages [1].

The processing of thermoplastics in AFP is one of the main technologies on which the development of thermoplastic materials is based. In this technology, the material is deposited automatically, heating it with a laser system and applying pressure, to achieve consolidation between the material that is applied and the one that is already part of the substrate deposited in previous layers.

Several works [2],[3], indicate that the processing temperature of the Nip Point (point of contact between the incoming tape and the substrate) should be high enough to decrease the matrix viscosity and allow an intimate contact between the tapes when the pressure is applied. This contact is essential for allowing the polymeric chain reptation and diffusion between the tapes, and thus, for a good bonding. Also, other publications[4],[5], make reference to the importance of crystallinity in the bonding of thermoplastic tapes, characterizing interlaminar fracture toughness as a combined effect of matrix ductility (inversely proportional to crystallinity) and fibre-matrix interface bond (proportional to crystallinity).

In the present work, the structural performance of the union between AFP deposited tapes isbe studied and correlated with the processing temperature and the crystallinity in the union area. The processing temperature in the nip point and the crystallinity is studied also in relation to the layup tool temperature (and thus, the cooling rates in the processed tape).

2. Materials and Methods

2.1 Materials

The material studied in this work is a unidirectionally reinforced composite tape, composed by a semi-crystalline low melt PAEK thermoplastic matrix (LM-PAEK TC1225) and a T800G carbon fibre reinforcement. The material was supplied by TORAY and is commercially identified as Cetex® TC1225 LMPAEK / T800G UD tape.

The material properties of the neat resin used as matrix is listed in *Table 1*, and the format of the composite tape is specified in *Table 2*.

Table 1: Material properties of matrix.

Material	Tm (°C)	Tg (°C)	Recommended processing temperature by supplier (°C)
LM-PAEK TC1225	305	147	340-385

Table 2: Composite tape format.

Material designation	Fiber Areal Weight, FAW (g/m²)	Resin content (%)	Ply Thickness (mm)	Width (mm)
TC1225 LMPAEK / T800G UD	145	36	0.145	25.4

2.2 AFP manufacturing

The equipment used in this work is an AFP (Automated Fibre Placement) head system, a PrePro3D model from Conbility manufacturer. The AFP system is mounted in a FANUC R-2000iC/165F Robotic Arm. The AFP system controls (among other parameters not listed for clarity) the placement speed, the applied compaction force (that defines the applied pressure, together with the application area resulting from elastomeric roller deformation), and the applied laser power, that sets the processing temperature in a closed control loop, receiving temperature feedback from two pyrometers pointing at the Nip point (one focused on the incoming tape and the other focused on the substrate). Temperature measurements at the nip point are taken from both pyrometers, and in this work are presented as an average of both of them for determining the nip point temperature. Also, measures from all the coupons of every case of study are averaged. The temperatures are selected from the third ply of each coupon, since this is the ply where the interlaminar properties are studied in the mechanical experiments, as described in 2.4.

The main parameters used in this work are listed in *Table **3***.

The laser used for heating up the nip point (incoming tape and substrate) is a Laserline diode laser source, model LDF6000-40, of 6300W and operating wavelengths ranging from 940 to 1060 nm.

The material is deposited on a heated layup tool that consists of an aluminum plate heated with resistances.

Table 3: AFP parameters used in this work.

AFP Parameter	Value
Layup speed [m/s]	0.25
Pressure [bar]	5.5

2.3 Annealing

Heat treatments of the samples were carried out in a furnace at equal annealing cycles for all the coupons subjected to annealing. The temperature was increased at the rate of 10°C/min until 220°C, and this temperature was maintained for 90 min. After this, the samples were cooled at 5°C/min until they reach ambient temperature.

2.4 Mechanical testing

Single Lap Shear Strength properties were evaluated according to the experimental procedure and specimen preparation proposed in [6]: four tapes where laid-up with the AFP process, on top of each other and in the same direction. In the middle of the stacking, two Kapton film strips of 20 mm width were placed with a separation of 5 mm, so that the top two plies and the bottom two plies are overlapped 5 mm. The separation in the bottom and top plies was made cutting the tapes on top of the Kapton film strips.

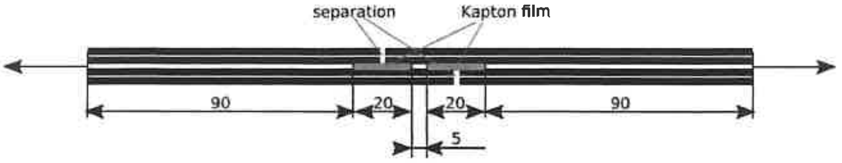

Figure 1. Simplified Single Lap Shear test specimen proposed in [6].

Single Lap Shear Strength tests were performed to characterize the samples using a 250 kN capacity standard tensile/compression machine (MTS®). Shear strength was measured at a constant speed of 1 mm min^{-1}. At least five repeated specimens were tested, and the data was averaged.

2.5 Differential Scanning Calorimetry (DSC)

The thermal properties of the materials were analyzed by Differential Scanning Spectroscopy (DSC). The experiments were carried out in a DSC Q2000 (TA Instruments). The DSC tests were performed with N_2 as the purge gas. The samples were sections cut from the fabricated specimens, in the area of study of the mechanical tests. Samples with a mass of 20-30 mg (7-10 mg of neat resin) were put on an alumina crucible and subjected to a heating cycle from r.t to 360 °C at a rate of 10 °C/min. After the first heating cycle, the temperature is held at 360 °C for 15 min, that is about 50 °C above the melting temperature to erase the thermal history, then cooling to r.t at 10 °C/min, and finally, a second heating from r.t to 360 °C at 10 °C/min. Crystallinity was determined by the equation 1.

$$\chi = \frac{\Delta H_m}{130} \times 100\% \qquad 1$$

Where ΔH_{endo}, ΔH_{exo} are the integral area of the endothermic peak and exothermic peak, respectively, and 130 J/g is the melting enthalpy of a 100% crystalline LMPAEK sample.

3. Results and Discussion

3.1 Thermal analysis

In this work, several coupons were manufactured with different layup tool temperature conditions, to have different scenarios regarding cool down rates. The heated layup tool, that acts as a substrate to the material, was heated up to 3 different temperatures: 25, 160 and 220°C. For each layup tool temperature, two groups of 5 coupons were manufactured. One of this groups was submitted to the annealing cycle described in Section 2.3, and then tested following the mechanical test procedure described in Section 2.4. The other group was mechanically tested (as described in Section 2.4) without a prior annealing cycle.

Crystallinity of the samples was measured using DSC technique (as described in section 2.5), resulting in the values listed in *Table **4***. Our results show that, the coupons manufactured with low layup tool temperatures (25°C) have crystallinity levels around 5%. The layup tool temperature of 160°C, despite being higher than the Tg of the material (147°C), renders also crystallinity values similar to the previous. However, the coupons manufactured at the highest layup tool temperatures (220°C) give markedly higher crystallinity levels (around 25%), similar to the post-processed samples.

Table 4: Average transitions temperatures, enthalpies, and estimated crystallinities correspondent to the first heating cycle.

Layup tool temperature [°C]	T_g (°C)	T_{cc}(°C)	ΔH_{cc} (J/g)	T_m(°C)	ΔH_m (J/g)	X (%)
25	149.36	192.20	9.37	305.58	12.08	5.8
160	149.89	191.33	9.53	307.4	12.79	7.0
220	152.28	235.00	-0.81	304.43	10.15	26.5
Annealed coupons*	159.74	206.60	-1.06	307.12	11.24	26.3

*(independently of layup tool temperature)

3.2 Mechanical properties

In a first round of experiments, coupons were manufactured to study the influence of the tool temperature on the processing nip point temperature, and in the interlaminar shear strength properties. The layup tool temperature was increased from 25°C to 220°C in five steps, and five coupons were manufactured in each step. The laser power and incidence angle on the nip point was maintained, and consequently the temperature in the substrate material was different in all the iterations. The nip point temperature at the substrate and shear strength values are listed in *Table 5* and represented in *Figure **2***.

Table 5: Nip point temperatures and Shear Strength values (variable nip point temperature).

Layup tool temperature [°C]	Nip point temperature (substrate)*1 [°C]	Shear Strength [MPa] Without Annealing
25	265	26.70±5.09
160	335	36.00±5.10
220	385	42.58±5.45

*1 Temperature measured at the substrate in the nip point area. The incoming tape was kept at a stable temperature of 350±15°C.

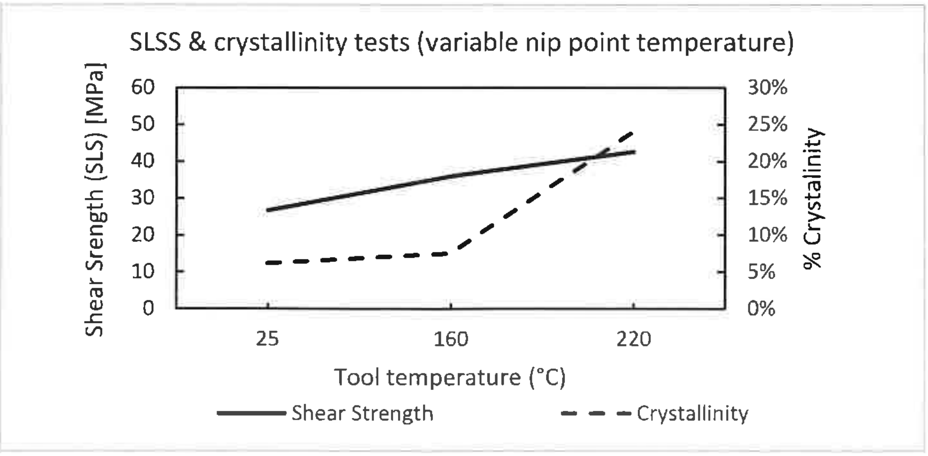

Figure 2. Interlaminar Shear Strength values with variable nip point temperature.

As it can be appreciated, lower tool and nip point temperatures result in lower SLSS values.

The increment of the nip point temperature decreases the polymer matrix viscosity, allowing a better flow and contact between the tapes, and enhancing the interdiffusion of the polymeric chains between tapes [2]. A higher processing temperature benefits the interlaminar union, as showed in the data from this first round of experiments, where the nip point temperature ranges from 265°C to 385°C, causing a variation on the SLSS values, that range from 27 MPa to 43 Mpa.

A second round of coupons was manufactured to study the influence of the tool temperature and the crystallinity in the SLSS values, without the influence of the nip point temperature. The laser power and orientation parameters were modified to obtain a uniform temperature distribution between the incoming tape and the substrate tape. The influence of the layup tool temperature in the processing temperature at the nip point was considerably reduced (but not eliminated) as it can be appreciated in the results listed in *Table 6*.

For each layup tool temperature two sets of 5 coupons were manufactured. Out of these 2 sets, one was subjected to an annealing cycle before the mechanical testing, and the other one was mechanically tested directly.

The mechanical results are listed on *Table 6*. In *Figure 3* are represented the mechanical test values of the samples manufactured, comparing the samples that were subjected to an annealing cycle and those that were directly tested after manufacturing. In *Figure* ***4*** are graphically represented the mechanical test results and crystallinity values of the coupons, without and with an annealing cycle, respectively. As it can be appreciated, the mechanical values of the SLSS tests of not annealed coupons are slightly higher than that of the annealed coupons. Accumulated tensions resulting from the annealing cycle, or the fragilization of the interlayer union caused by the higher crystallinity could be responsible of the slightly lower SLSS values in the annealed coupons, but further study should be do confirm this. Also, there is no significant difference on the SLSS values between the coupons manufactured with different tool temperatures, since there are no great differences on the nip point temperature.

Table 6 : Nip point temperatures and Shear Strength values (fixed nip point temperature).

Layup tool temperature [°C]	Nip point temperature*^{2} [°C]	Shear Strength [MPa]	
		Without Annealing	With Annealing
25	330	31.11±4.85	23.82±2.69
160	350	30.00±3.37	28.16±3.71
220	360	28.67±1.77	26.37±2.78

*^{2} Temperature averaged from the substrate and the incoming tape in the nip point area.

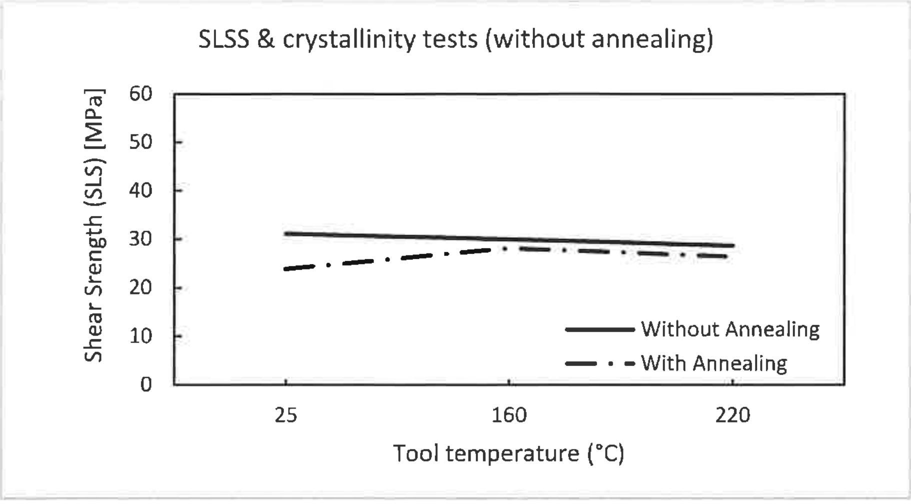

Figure 3. Interlaminar shear strength values comparison between annealed and not annealed coupons.

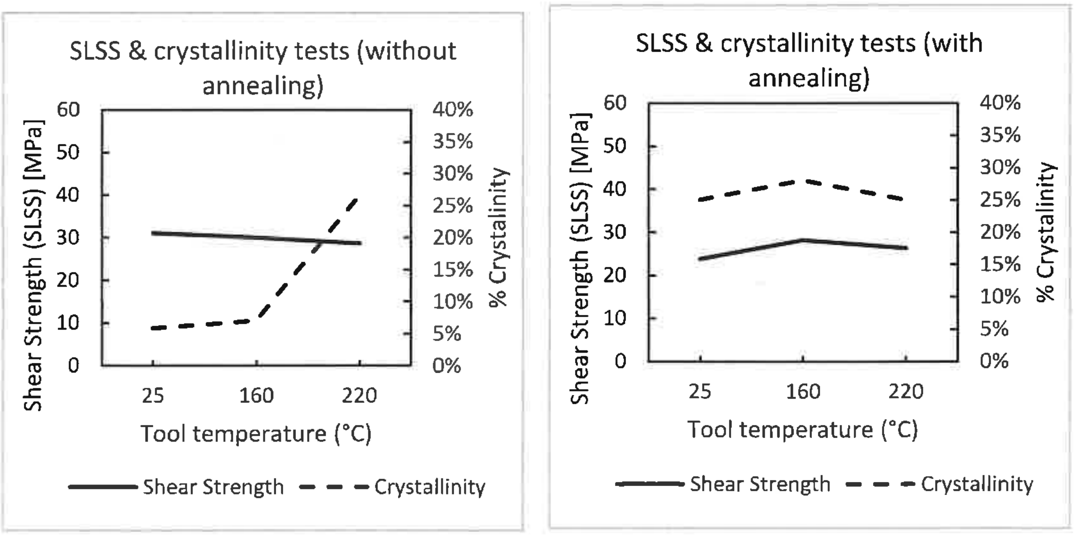

Figure 4. Graphical representation of crystallinity and SLSS load values of coupons tested without a previous annealing cycle (left) and with a previous annealing cycle (right).

4. Conclusions

This work has studied the thermoplastic AFP process, focusing on the influence of the layup tool temperature on the crystallinity and on the interlaminar mechanical properties (studied by SLSS method) and the influence of the crystallinity obtained with an annealing cycle in the mechanical properties.

The experiments performed show that achieving a sufficient nip point temperature has a great influence on the interlaminar shear strength values, showing a clear correspondence between the increment on the nip point temperature and obtained SLSS values.

Also, the results show that higher crystallinities do not improve the interlaminar shear strength properties of the coupons, when tested on a SLSS test. Low crystallinity specimens were compared with high crystallinity specimens obtained both by subjecting the specimen to an annealing cycle or by decreasing the cool down rates with the layup tool temperature, and the higher crystallinity specimens did not achieve higher shear strength values.

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References

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1. Veldman, S. L., Kortbeek, P., Wölcken, P. C., Herrmann, R., Kos, J., & Villegas, I. F. (2020, February). Development of a multifunctional fuselage demonstrator. In Aerospace Europe Conference 2020 (pp. 25-28).

2. Khan, M.A.; Mitschang, P.; Schledjewski, R. Identification of some optimal parameters to achieve higher laminate quality through tape placement process. Adv. Polym. Technol. 2010, 29, 98–111.

3. Levy A, Heider D, Tierney J, et al., Inter-layer thermal contact resistance evolution with the degree of intimate contact in the processing of TP composite laminates. J Compos Mater., 2014 48: 491–503.

4. Gao, Shang-Lin, and Jang-Kyo Kim. "Cooling rate influences in carbon fibre/PEEK composites. Part 1. Crystallinity and interface adhesion." Composites Part A: Applied science and manufacturing 31.6 (2000): 517-530.

5. Gao, Shang-Lin, and Jang-Kyo Kim. "Cooling rate influences in carbon fibre/PEEK composites. Part II: interlaminar fracture toughness." Composites Part A: Applied science and manufacturing 32.6 (2001): 763-774.

6. Dreher P, Chadwick AR, Nowotny S. Optimization of in-situ thermoplastic automated fiber placement process parameters through DoE. In: Proceedings of the 40th SAMPE Europe conference; 2019, p. 1–13.