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EFFECTS OF PROCESSING PARAMETERS ON THE PROPERTIES OF CF/PPS LAMINATES MANUFACTURED BY LASER-ASSISTED IN-SITU CONSOLIDATION

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ABSTRACT

In-situ consolidation of thermoplastic composites can be realized through laser-assisted automated fiber placement (AFP) technology, and the overall quality of the composites was significantly affected by the processing parameters. Understanding the mechanisms which affect the properties of the composites was imperative to optimization of the processing parameters. In this work, the effects of processing parameters on the properties of continuous carbon fiber reinforced polyphenylene sulfide (CF/PPS) laminates manufactured by laser-assisted AFP were investigated. Properties including micro morphology, void content, crystallinity of the laminates were investigated and the mechanisms were discussed. The interlaminar shear strength (ILSS) was evaluated and analyzed considering void content, crystallinity, and interlaminar bonding. The ILSS of the laminates was strongly dependent on interlaminar bonding and void content. Conversely, there was no apparent corresponding relationship with crystallinity and ILSS.

1 INTRODUCTION

High performance fiber reinforced thermoplastic composites are widely used in the aerospace industry due to their excellent heat resistance, high toughness, excellent creep resistance, damage tolerance, and impact resistance[1]. The consolidation of thermoplastic composites mainly depends on the diffusion bonding of molecular chains under temperature and pressure[2]. Combined with automated fiber placement (AFP), the one-step manufacturing process called “in-situ consolidation (ISC)” could be achieved. However, due to the high viscosity of high-performance thermoplastics and short dwell time of the consolidation temperature and pressure, as well as the lack of tackiness for thermoplastic composites, the properties of the composites manufactured by ISC are hardly reach the levels achieved by autoclave.

A schematic illustration of the laser-assisted AFP ISC process of thermoplastic composites is shown in Figure 1. There are two main challenges with ISC: firstly is the high melting viscosity of high-performance thermoplastic composite and short consolidation time results in insufficient interlayer contact and fusing bonding; secondly for semi-crystalline thermoplastics, the extremely fast cooling process leads to incomplete matrix crystallization. The corresponding processing parameters including laser temperature, tool temperature, placement speed, and compaction pressure, etc. And the overall properties of the laminates were significantly affected by these parameters. A deep understanding of the various manufacturing mechanisms at play was essential to the optimization of the processing parameters and improvement of product quality and manufacturing efficiency.

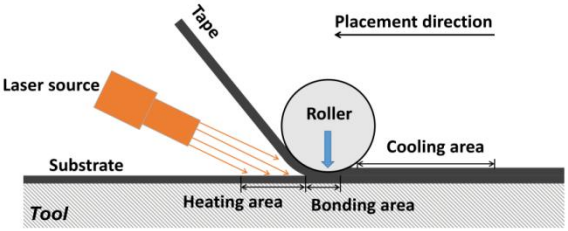

Figure 1. Schematic illustration of laser-assisted AFP in-situ consolidation process of thermoplastic composites.

As one of the high-performance thermoplastic composites, carbon fiber reinforced polyphenylene sulfide (CF/PPS) composites have great potential in aviation applications. In this study, the effects of AFP processing parameters on the micro morphology, crystallinity, void content, and interlaminar properties and their mechanisms of the laminates were investigated and discussed.

2 MATERIAL AND EXPERIMENT

2.1 Material

CF/PPS unidirectional tape (TU100) was provided by Barrday Co. Ltd, with a resin content of 34% (by weight). The width and thickness of the tape were 6.35 mm and 0.14 mm, respectively. The density, areal density and glass transition temperature (Tg) were 1.62 g/cm^3, 227 g/cm^2 and 95 ℃, respectively.

2.2 Manufacturing of CF/PPS laminates by AFP

A robot-style four-tows lay-up machine developed by M.Torres (Pamplona, Spain) was used. A diode laser of 4 kW with a rectangular spot was used to heat the thermoplastic composites. The laser power was adjusted in real-time utilizing internal software to keep the tape surface temperature constant during laying. The maximum compaction force was 2000 N. Two types of unidirectional laminates including four-plies ($[0]_4$) and twenty-plies ($[0]_{20}$) were manufactured. The $[0]_4$ laminates were used to investigate the micro structure and properties, as the $[0]_{20}$ ones were used to analyze the mechanical properties. The corresponding manufacturing parameters are listed in Table 1 and Table 2. The $[0]_{20}$ laminates manufactured at laser temperatures of 250, 300, 350, 380 and 400 ℃ were named L250, L300, L350, L380 and L400, respectively.

Table 1. Manufacturing parameters of $[0]_4$ CF/PPS laminates

Samples	Laser temperature(℃)	Tool temperature (℃)	Placement speed (mm/s)	Compaction force (N)
1#	380	40	60	500
2#	280	40	60	500
3#	280	120	60	500
4#	280	120	100	500
5#	280	120	200	500
6#	380	120	100	500
7#	380	120	100	1000
8#	380	120	100	1500
9#	380	120	100	2000

Table 2. Manufacturing parameters of $[0]_{20}$ CF/PPS laminates.

Factors	Parameters
Laser temperature (℃)	250, 300, 350, 380, 400
Tool temperature (℃)	120
Placement speed (mm/s)	60, 100, 200
Compaction force (N)	500

2.3 Characterization

For the laminates made by AFP, the crystallinity was determined by differential scanning calorimetry (DSC). The samples were heated from room temperature to 320 ℃ at 10 ℃/min. The crystallinity of the sample could be calculated as:

$$X_C = \frac{\Delta H_m - \Delta H_c}{(1 - w_f) \cdot \Delta H_f} \quad (1)$$

Where X_c is the crystallinity, ΔH_m and ΔH_c are the melting and crystallization enthalpy of the samples, respectively, w_f is the fiber weight content of the composites (66%) and ΔH_f is the melting enthalpy of fully crystalline PPS (80 J/g)[4].

The surface and cross-sectional morphology of the laminates were observed by SU8010 scanning electron microscope (SEM, Hitachi Limited, Tokyo, Japan), which was operated at an accelerating voltage of 5 kV.

2D microscopy was utilized to estimate the void content in the CF/PPS laminates. All samples were polished to obtain a smooth cross-section. The morphology of the cross-section of the specimen was observed using Olympus BX41M-LED microscope, and the void content was calculated by Image J software.

Interlaminar shear strength (ILSS) of CF/PPS laminates was determined by short beam shear test according to ASTM D2344 on ETM 204C electronic universal testing machine (WANCE, Shenzhen, China). The sample size was 18 mm×6 mm×3 mm and the cross-head displacement rate was 1 mm/min. Ten samples were tested for each laminate. ILSS of the laminate was determined as,

$$ILSS = \frac{3P_m}{4bh} \quad (2)$$

Where P_m was the maximum load； b and h were sample width and thickness, respectively.

3 RESULTS AND DISCUSSION

3.1 Micro morphology of the laminates

The surface and cross-section morphology of the laminates are shown in Figure 2. At high laser temperature (1#) and tool temperature (3#), the surface of the laminates appear smooth. With the increase of placement speed (3~5#) and compaction force (6~9#), the resin texture on the surface of the composites disappeared. Generally, the resin viscosity decreased with the increase in temperature, which was conducive to the interlaminar bonding between layers, and the resulting smooth surface is beneficial to the entire quality of the laminates. Also, it can be seen from the cross-section morphology that all samples showed good compactness.

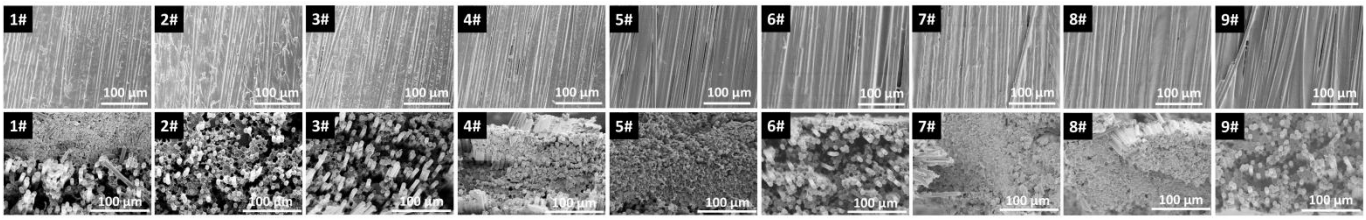

Figure 2. Surface and cross-section morphology of the laminates.

3.2 Crystallinity

DSC curves and related crystallinity of all laminates is shown in Figure 3. It was found that there was a typical cold crystallization peak around 138 ℃ for samples manufactured on a tool temperature of 40 ℃, and the crystallinity was extremely low (~18%) (samples of 1# and 2#). When the tool temperature increased to 120 ℃, the cold crystallization peak disappeared for most samples (except sample 5#), and the highest crystallinity reached 46%. As mentioned above, the glass transition temperature (Tg) of CF/PPS composites as received was 95 ℃. When the tool temperature was 40 ℃, the layer temperature rapidly dropped below Tg once in contact with the cold tool, and molecular segments were frozen; when the tool temperature was 120 ℃, the layer temperature was still above Tg after contacting with tool, which means segment movement still occurs. Therefore, high tool temperature was conducive to the improvement of the degree of crystallinity. Furthermore, for the samples manufactured at a tool temperature of 120 ℃ and laser temperature of 280 ℃, when the placement speed increased from 60 mm/s to 200 mm/s (samples of 3#~5#), the crystallinity decreased from 40% to 26%. This may be attributed to the difference in cooling rate at different placement speeds.

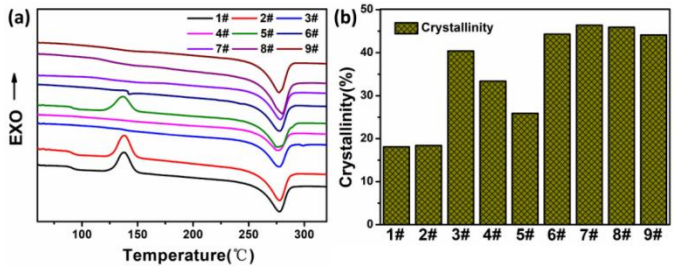

Figure 3. DSC curves (a) and related crystallinity (b) of all laminates.

It is worthy to note that when the placement speed was 200 mm/s, the cold crystallization peak appeared. When the laser temperature increased from 280 ℃ to 380 ℃, the crystallinity increased from 33% to 44%. The results showed that the crystallinity of the samples changed considerably with the variety of laser temperatures and placement speeds. This was because high laser temperature would slow down the cooling rate of the area around the polymer molten pool, which enhanced the movement ability of the molecular chains[5]. The results of samples 6-9# indicated that there was no apparent correlation between crystallinity and compaction force. The above results indicated that high laser temperature, tool temperature, and low placement speed were conducive to the improvement of the crystallinity of the composites, and the maximum crystallinity reached 46%. Ultimately, the tool temperature was the main influencing factor.

3.3 Void content

The void content of each laminate is presented in Table 3. When the laser temperature was increased from 280 ℃ to 380 ℃, the void content of the laminates decreased from 5.7% to 4.2% (1#, 2#). The void content of 4#

and 6# samples also showed the same trend. With the laser irradiating, the resin melted and extruded under pressure. However, due to the high melting viscosity of the thermoplastic resin, the void in the laminate would move with the resin. When a low laser temperature was applied, the resin encountered difficulty flowing because of the high viscosity, while the voids were confined to the resin and were difficult to discharge resulting in high void content. Conversely, high laser temperature enhanced the mobility of the resin, which was conducive to the discharge of the voids and the formation of good interlaminar bonding. When the placement speed increased from 60 mm/s to 200 mm/s, the void content of the samples increased from 5.6% to 6.6% (3#~5#). This was because with the increase of placement speed, the dwell time of temperature and pressure on the surface layer decreased. The voids in the layers did not have enough time to compress and discharge. Moreover, the voids formed by the influx of external air entering into the inter-layer during placement could not be effectively compressed, which led to the increase of the overall void content of the composites. As the consolidation force increased (6#~9#), the void content of the laminates decreased from 4.6% to 2.5%. This was because increasing the consolidation force better compressed the voids and made them smaller. Simultaneously, it allowed for easier movement of the voids in the thickness direction which led to more voids discharging. By comparing the samples 2# and 3#, it was found that the void content showed no discernible difference when the tool temperature was 40 ℃ versus 120 ℃.

Table 3. Laminates void content.

Sample	Void content (%)	Sample	Void content (%)	Sample	Void content (%)
1#	4.3±0.7	4#	6±1.1	7#	3.3±0.9
2#	5.7±0.9	5#	6.6±1.4	8#	3.0±0.8
3#	5.6±1.1	6#	4.6±0.7	9#	2.5±0.5

The cross-sectional morphology of laminate 1# is presented in Figure 4. It can be seen that the voids near the lower surface are fewer than those near the upper surface. This may be because the lower surface was subjected to rolling four times during the entire placement process. Under repeated high temperatures and pressures, the voids of the laminates near the lower surface were effectively compressed.

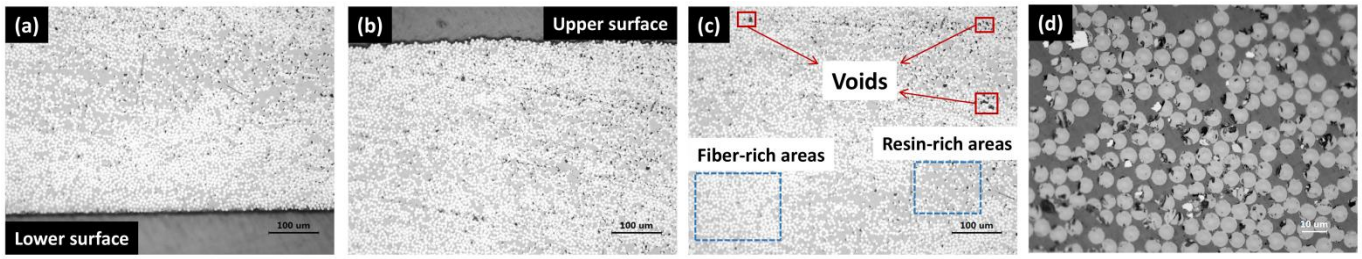

Figure 4. Cross-sectional morphology of laminate 1#. (a: lower surface; b: upper surface; c-d: different magnification)

3.4 Mechanical properties of CF/PPS laminates

The ILSS of $[0]_{20}$ CF/PPS laminates with different parameters are shown in Figure 5(a). The ILSS increased from 33.2 MPa to 61.3 MPa within a range of laser temperatures and placement speeds. When the laser temperature increased from 250 ℃ to 400 ℃ at a placement speed of 60 mm/s, the ILSS increased by 42.2% (from 43.1 MPa to 61.3 MPa). In the same temperature range, the ILSS increased by 53.6% and 67.8% at placement speeds of 100 mm/s and 200 mm/s respectively. With the laser temperature set at 250 ℃, and the placement speed increased from 60 mm/s to 200 mm/s, the ILSS decreased by 23.0% (from 44.1 MPa to 37.5 MPa). With the laser

temperatures set to 300, 350, 380 and 400 ℃, the ILSS decreased by 14.4%, 12.8%, 7.2%, and 9.1% respectively in the same range of placement speed.

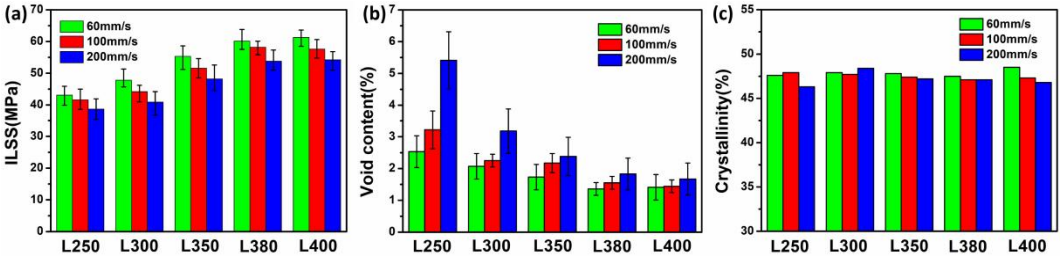

Figure 5. ILSS (a), void content (b) and crystallinity (c) of $[0]_{20}$ CF/PPS laminates with different parameters.

In addition, as laser temperature was increased, the reduction ratio of ILSS decreased when the placement speed increased. Simultaneously, ILSS at all placement speeds when the laser temperature was 400 ℃ was higher than that of the samples with the laser temperature of 250 ℃ and 300 ℃. Specifically, comparing the ILSS for samples at 250 ℃ & 60 mm/s (43.1 MPa) and 300 ℃ & 60 mm/s (47.8MPa), the ILSS of laminates with a laser temperature of 400 ℃ and placement speed of 200 mm/s increased by 29.2% and 16.5% respectively to 55.7 MPa. It can be concluded that the ILSS and placement efficiency could be simultaneously improved at higher laser temperatures.

Based on the results of crystallinity and void content of the laminates, the variation mechanism of ILSS was further analyzed. The crystallinity and void content of the laminates are shown in Figure 5(b,c). The results revealed that the crystallinity of all laminates was between 46% and 49%, without a significant difference. According to the results in Section 3.2 and the author's previous work[6], high tool temperature and the accumulation of heat in the substrate contributed to the improvement of crystallization. Therefore, the laser temperature and placement speed did not significantly impact the crystallinity for the $[0]_{20}$ laminates manufactured at a tool temperature of 120℃. In addition, it is worth noting that there was no apparent corresponding relationship between crystallinity and ILSS; as it turns out, interlaminar bonding and void content are more decisive for ILSS rather than crystallinity.

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