IN-SITU MONITORING OF CONSOLIDATION PROCESS FOR HIGH-PERFORMANCE THERMOPLASTIC COMPOSITES BY FIBRE BRAGG GRATING

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ABSTRACT

In the present paper, a process monitoring by an embedded FBG sensor for the in-situ consolidation of a high-performance carbon/thermoplastic composite (LM-PAEK) was investigated. First, the distribution of thermoplastic matrix and carbon fibre in the prepreg was observed using an optical microscope and a profilometer. The main thermophysical properties of the thermoplastic matrix such as its glass transition, crystallisation and melting point temperatures were obtained by DSC measurements. Second, a temperature calibration of the FBG sensor close to 400 °C was carried out. A specific protocol for the integration of the FBG in the laminate and for the in-situ process monitoring up to 400 °C was then designed and set up. The analysis of the FBG response during and after manufacturing of the laminate provided information on the different phenomena occurring during the process such as matrix crystallisation as well as the Coefficient of Linear Thermal Expansion of the carbon/thermoplastic composite. The consolidation phenomena of the thermoplastic matrix were detected and information on melting and glass transition temperatures was provided and found similar to data collected during DSC testing. These preliminary results tend to validate the use of optical fibres for the manufacturing process monitoring of a high-performance carbon/thermoplastic composite.

1 INTRODUCTION

The field of aviation undergoes nowadays multiple environmental challenges, linked on the one hand to the reduction of transport costs and on the other hand to an increasingly strict legislation on greenhouse gas emissions. Therefore, the development of non-corrosive carbon/thermoplastic composite materials with outstanding mechanical properties and resistant to high temperature conditions represents a major challenge for the aeronautical industry. In that case, the use of thermoplastic matrix such as PolyArylEtherKetone (PAEK) offers advantages in terms of production by allowing, in particular, assembly by welding. However, during composite manufacturing, a poor consolidation process could induce certain initial manufacturing defects (porosities) in the thermoplastic [1]. Potential factors that can lead to a poor consolidation process are linked to material features (prepreg thickness and consolidation phenomena such as crystallisation and melting) as well as process parameters (cooling rate and coefficient of thermal expansion of the thermoplastic matrix) [2]. However, the use of FBG (Fibre Bragg Grating) for an in-depth understanding of consolidation phenomena, manufacturing defects as well as the impact of the process parameters on the final properties of a high-performance carbon/thermoplastic composite is yet to be investigated. In the purpose of better understanding these aspects, SHM (Structural Health Monitoring) could be a promising solution to monitor both the manufacturing process of the thermoplastic as well as its thermomechanical behavior over time. The present study provides an overview on the development of an experimental protocol for the in-situ monitoring of the thermoplastic consolidation by an embedded FBG.

2 PREPREG THERMOPLASTIC CHARACTERISATION

2.1 Prepreg presentation

The considered carbon/thermoplastic composite was a CFRP (carbon-fibre-reinforced polymer) from Toray advanced composites with a semi-crystalline low-PAEK melt (LM-PAEK) resin. The prepreg is 0.14 mm thick, with a fibre volume fraction of 66%.

The first step in the material characterisation was to observe the fibre-matrix distribution (fibres repartition) and distribution of the porosities. In that way, we would be ensured for any resin-rich regions at the surface or any initial porosities in the prepreg ply, and the appropriate impregnation of fibre strands could be also checked. The optical microscope (Axiocam 305 color imager from Zeiss) was used to monitor perpendicularly to carbon fibres, a carbon/thermoplastic specimen of 30 mm length. The monitoring of a small area in the thermoplastic prepreg (see Fig. 1 with the carbon fibre in white, matrix in dark grey and fibre tearing during surface polishing in black) highlighted a feature of the prepreg; while the fibre-matrix distribution is rather uniform with a low-void content and practically no initial porosity, the ply thickness is irregular.

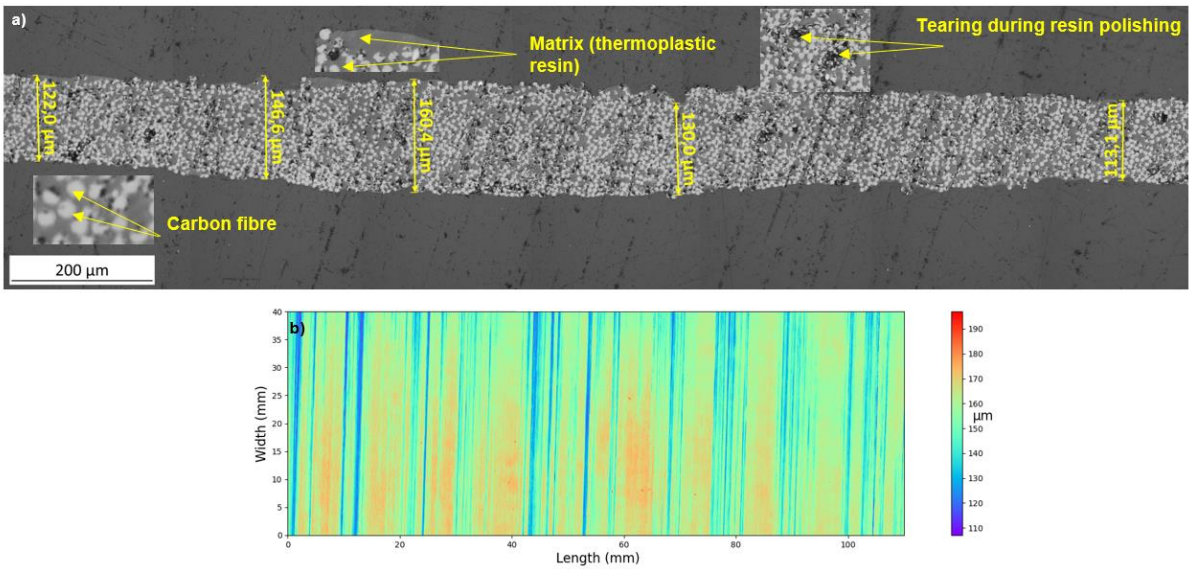

Figure 1: a) Microscopic observation of thickness variation in prepreg, b) prepreg thickness measured using a profilometer.

This difference of thickness throughout the prepreg was also confirmed by a second investigation on another prepreg sample from the same carbon/thermoplastic batch (40 x110 mm^2). The prepreg sample was characterized using an optical profilometer with a measuring velocity of 0.10 mm/s and a measurement interval of 0.5 µm, in order to inspect its thickness (see Fig. 1b).

Through these first two steps of prepreg characterisation, it was confirmed that the thickness of the prepreg is not uniform, a characteristic that has already been observed in literature on prepreg C/PEEK by Cytec [3]. Thus, a thickness variation of about +/-50 µm was established during both microscopic and profilometer observations. Consequently, during thermoplastic consolidation process, a thickness variation creates voids at the ply interface, which may prevent the thermoplastic matrix from quickly fulfilling these voids. One possible solution would be to increase the necessary time for the establishment of perfect intimate contact between adjacent plies during thermal annealing [4].

2.2 DSC measurement for prepreg characterisation

The last step in the prepreg characterisation process consisted in DSC (Differential Scanning Calorimetry) measurements in order to study the thermal properties of the polymer matrix. The model device TA DSC25 was used for this experiment and the procedure had to be executed under neutral gas (argon) to homogenize the temperature in the enclosure while preventing the realization of an oxidation reaction. From this experiment, a curve of total heat flow (W/g) versus temperature was extracted (see Fig. 2). Fig. 2 shows one test result for a sample of approximately 6 mg of LM-PAEK prepreg. The thermal cycle applied here was a heating ramp at 5 °C/min from 25 °C to 380 °C, an isothermal stage of 15 minutes applied at 380 °C and a cooling ramp at 5 °C/min down to 25 °C.
Several conclusions can be deduced:

- During the first heating, the thermoplastic matrix is heated above its glass transition temperature (T_g) which is identified around 150+/-2 °C. Cold crystallisation (T_{cc}) seems to occur approximately between 180 °C and 200 °C (appearance of a first exothermic peak). The endothermic peak spotted between 260 °C and 323 °C can be associated with melting point of the thermoplastic matrix (T_m).
- During the first cooling, the polymer is solidified again by cooling below its melting temperature. The existence of an exothermic peak between approximately 270 °C and 225 °C is a result of hot crystallisation (T_{hc}). These first results are found similar to DSC thermal characterisations of LM–PAEK indicated in literature [5].
- During the second heating, the glass transition temperature of the matrix is no longer visible while the absence of the cold crystallisation indicates that the polymer was completely crystallised during the first thermal cycle. The endothermic peak spotted between 254 °C and 323 °C is due to resin melting (T_m). Finally, a second exothermic peak is obtained during second cooling between approximately 268 °C and 227 °C, which represents the second hot crystallisation. As a result, it can be assumed that the phenomena of glass transition and cold crystallisation only occurred during the first thermal cycle.

For the thermoplastic, the second heating scan is identical to the first scan while the temperature is below T_g. A slight deviation should be noted for the melting between the two cycles as well as the crystallisation during cooling. Both peaks (see Fig. 2: in green for heating and in orange for cooling) are more intense during first thermal cycle indicating a remodelling of the crystallite's morphology [5].

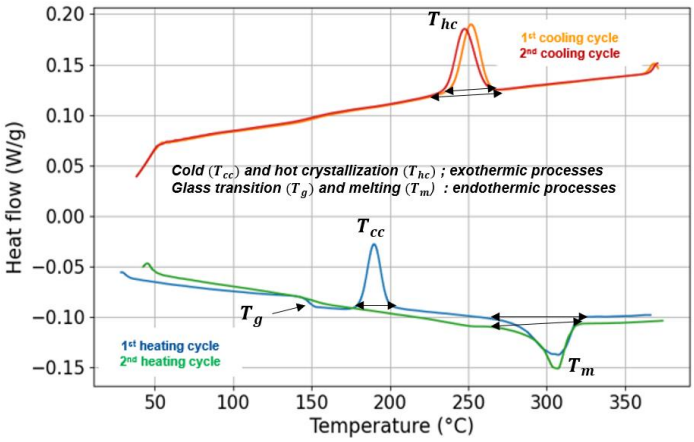

Figure 2: Evolution of exothermic (cold and hot crystallisation) and endothermic (glass transition and melting) heat flow versus temperature in DSC.

3 THERMAL CHARACTERISATION OF THE FBG

3.1 Optical sensor

The goal of our experiment was to monitor the temperature and deformation in the composite

material during its manufacturing process. In that purpose, FBG sensors that could withstand temperatures up to 400°C during thermoplastic consolidation process were used. The FBG is a temperature and strain sensor inscribed on an optical fibre. In the present study, the considered FBG was a single mode fibre (Ø125µm without recoat, Ø250µm with recoat) with one Bragg grating (5mm length). The Bragg grating corresponds to a periodic modulation of the refraction index in the core of the optical fibre [6]. It reflects a portion of an incident light signal in the form of a spectrum composed of a wavelength, called the Bragg Wavelength (λ_B), defined by the reflection peak as follows:

$$\lambda_B = 2n_{eff}\Lambda \tag{1}$$

Where n_{eff} is the effective refractive index and Λ the grating period [6].

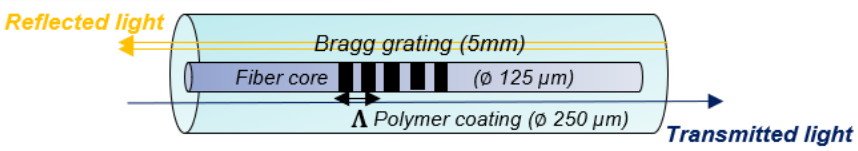

Figure 3: Complete structure of the FBG sensor and its principal function.

When a stress ε and a temperature variation ΔT are applied to the FBG, the Bragg length shifts as follows:

$$\Delta\lambda_B = K_T\Delta T + K_\varepsilon\varepsilon \tag{2}$$

Where K_T and K_ε are respectively defined as the coefficients of thermal and strain sensitivity of the FBG.

3.2 FBG Thermal calibration

In order to characterize the FBG's thermal sensitivity, a calibration test was carried out. The experiment took place as follows: a thermal cycle composed of a heating ramp at 3°C/min up to 350°C was applied to the system followed by a free cooling. The goal of this experiment was the estimation of thermal sensitivity of the FBG around temperatures close to 400°C. Hence, in equation (2), the thermal coefficient was assessed by a second-degree polynomial fitting in order to get sufficient agreement (see Fig. 4) as follows:

$K_T = 0.00913T^2 + 0.0087\,T + 1565$ nm/°C while the strain coefficient $K_\varepsilon = 1.2\,x\,10^{-3}$ nm/µdef was based on literature [7].

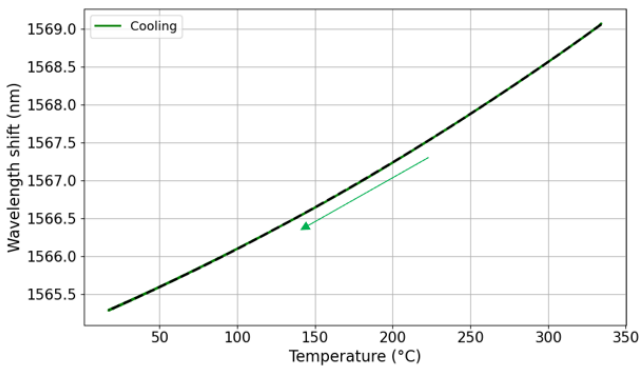

Figure 4: Second degree polynomial fitting for the estimation of the thermal sensitivity of the FBG.

4 PROCESS MONITORING DURING THERMOPLASTIC CONSOLIDATION

4.1 Instrumentation & manufacturing method

An embedded FBG sensor was used to follow the evolution of the deformation in the carbon/thermoplastic composite during the manufacturing process. Here, eight-ply laminates ($[0]_{8S}$) were investigated. Each eight-ply laminate was instrumented with an embedded FBG and a K-type thermocouple, both in the middle of the ply stack (as shown in Fig. 5), specifically between the 4th and 5th plies. Another thermocouple was placed on the surface of the last ply (8th) in order to follow any possible thermal gradient through the prepreg thickness. The integrated thermocouple was positioned as close as possible to the FBG in order to obtain the best possible thermal compensation. The FBG was introduced perpendicularly to carbon fibres in order to be more sensitive to the impact of the physicochemical phenomena of the matrix during consolidation. The entire FBG spectrum over the [1500,1600] nm wavelength range and temperatures measured by both thermocouples were recorded during the experiment every 5 seconds.

Concerning the manufacturing process, the Vacuum Bag Only (VBO) process was chosen (see Fig. 5). The process consists in sealing the system in a vacuum bag in which the maximum pressure applied is 1 bar. During the heating phase, the target temperature was set to 380 °C with a heating rate of 5°C/min followed by a temperature stage of 15 minutes. This was followed by a free cooling phase down to room temperature (around 25°C) and a pressure relief.

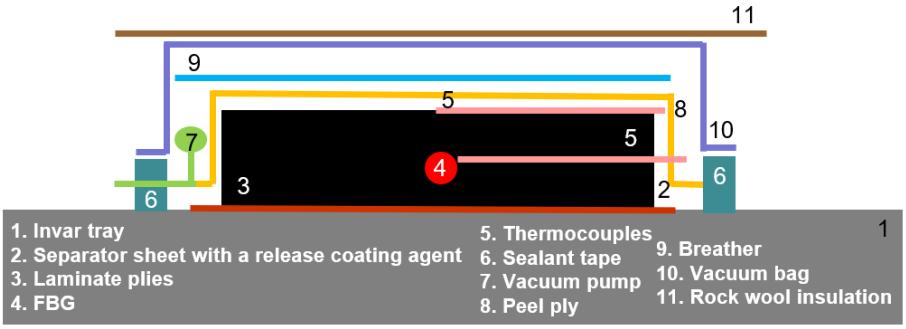

Figure 5: Schematic representation of the experimental setup for vacuum bagging process.

4.2 Process monitoring during consolidation

During this experiment, the evolution of Bragg wavelength during thermoplastic consolidation was investigated. Data collected during thermal calibration of the optical sensor (thermal coefficient of the FBG) was used to estimate the strain of the thermoplastic during its consolidation. The temperature was measured by the embedded thermocouple. At the end, a curve of strain (µdef) of the thermoplastic matrix versus temperature could be extracted (see Fig. 6) thanks to the following equation:

$$\varepsilon = \frac{1}{K_\varepsilon}(\Delta\lambda_B - K_T\Delta T) \tag{3}$$

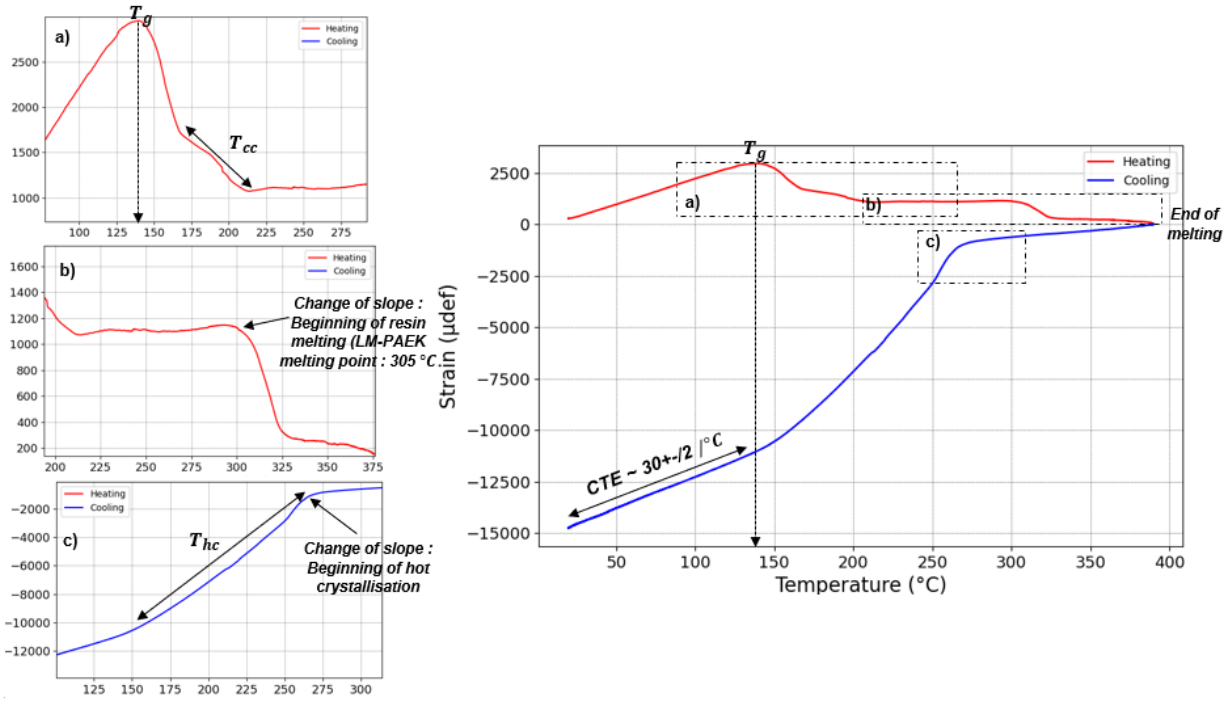

Figure 6: Thermoplastic strain measured by FBG during consolidation process:
a) glass transition, b) melting point c) hot crystallisation.

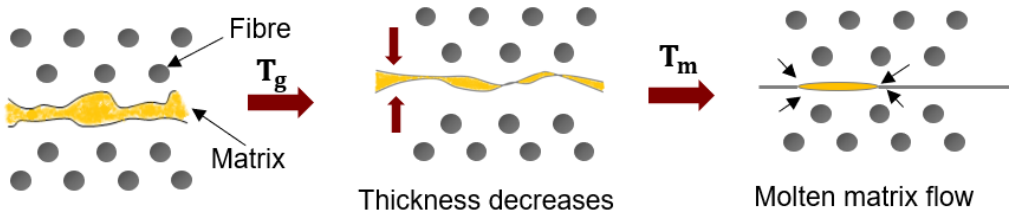

Figure 7: Phenomenon of intimate contact between prepreg plies.

Several conclusions can be deduced:

- During heating, the glass-transition temperature, detected with the DSC measurements, is observed around 150°C (see Fig. 6a). The FBG seems to be already submitted to the thermal expansion of the thermoplastic matrix due to heating. A strain diminution appearing after glass transition temperature is hypothetically assigned to the establishment on intimate contact between thermoplastic prepreg plies (see Fig. 7); In fact, movements of portions of chains in the amorphous phase of the semi-crystalline polymer are responsible for this phenomenon [8].
- In the temperature range of 168 °C and 212 °C, based also on data obtained in DSC, cold crystallisation seems to take place. Another assumption was the release of residual stresses induced by thermal effects [9] (strong thermal gradients induced by prepreg manufacturing process) and variations in the degree of crystallinity. However further research will be conducted in order to examine both hypotheses.
- Similarly to DSC results, a sudden change of slope marks the melting region, which takes place at around 305 °C.
- Cooling curve shows a characteristic step in the temperature range between 278 °C and 150 °C, marking the post crystallisation region (hot crystallisation), detected also in DSC.

The difference in characteristic temperatures (glass transition, crystallisation and matrix melting) obtained by FBG monitoring and the DSC method are due to measurement uncertainty. At last, from

150 °C until the ambient temperature, the FBG was uniquely submitted to the thermal expansion of the thermoplastic matrix. The Coefficient of Thermal Expansion (CTE /°C) of the carbon/thermoplastic composite was estimated from a linear fit to the data over the entire test-temperature range (130 °C – 45 °C) according to the following equation:

$$\frac{\Delta\varepsilon}{\varepsilon} = \alpha_L * \Delta T \tag{4}$$

Where α_L was defined as the Coefficient of Linear Thermal Expansion along the transverse direction of the carbon fibre with a value of approximately 30+/-2 /°C. This value is close to with data obtained in literature from an ATG (Analytical Thermal Gravimetry) on a prepreg CF/PEKK for temperatures inferior to glass transition temperature of LM-PAEK [8]. However, the accuracy and reliability of this measurement technique will be further verified.

4.3 Process monitoring after consolidation

The DSC calorimetric tests enable to establish that the thermoplastic matrix completely crystallised during first thermal cycle. Consequently, the second heating was characterized by an absence of cold crystallisation whereas glass transition temperature was unobservable. In order to investigate whether or not the embedded FBG sensor could still be used as a mean of matrix characterisation, a second thermal cycle was applied to the thermoplastic laminate after its consolidation. Data collected by the FBG during the second thermal cycle may be used to discriminate the evolutions of measured deformation induced by the consolidation of the material from the ones due the change of state of the matrix. In that purpose, the same thermal profile used during consolidation was applied, which ultimately led to the evolution of the matrix strain (µdef) with the temperature (see Fig. 8).

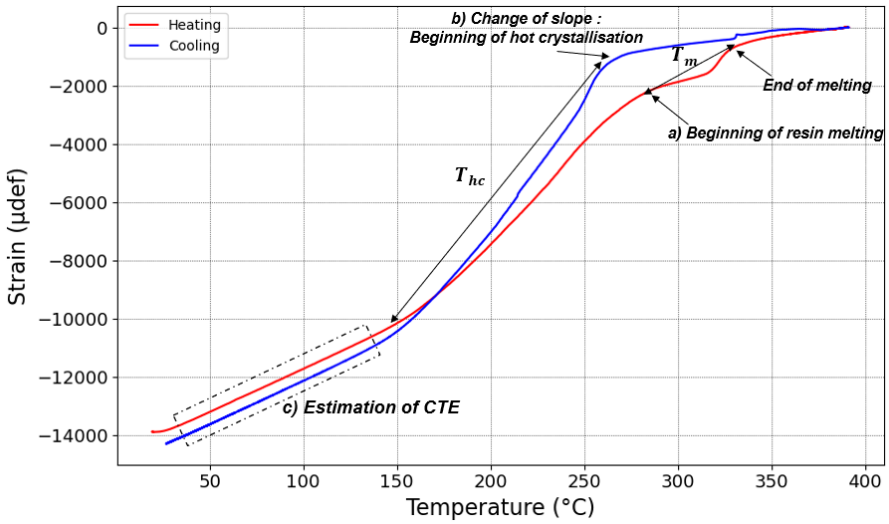

Figure 8: Thermoplastic strain measured by FBG after consolidation process

It is then confirmed that the optical sensor remains sensitive to thermoplastic deformation even after manufacturing of the laminate. The difference between figures 6 and 8 lies in the fact that during thermoplastic consolidation (Fig. 6), consolidation phenomena linked to establishment of intimate contact and glass transition are visible. On the contrary, once the laminate is consolidated and the sensor is well incorporated in the laminate structure, it can be considered that the optical sensor is only submitted to the thermal expansion of the thermoplastic matrix. In particular, during heating and for the temperature range of around 284 °C and 330 °C, melting of the polymer matrix takes place, observed also through DSC experiment. A sudden change of slope in cooling curve appearing at around 273 ° C

could be assigned to hot crystallisation. This assumption has not been yet validated.

Finally, results of strain monitoring after consolidation provided an estimation of the CTE of the thermoplastic composite for two curve regions (see Fig. 8, part c) in which the FBG was uniquely submitted to the thermal expansion of the thermoplastic matrix. The CTE (/°C) of the thermoplastic composite was estimated from a linear fit to the data over the entire test-temperature ranges (25 °C - 140°C and 140 °C - 45 °C respectively). The value of this coefficient was approximately 30+/-2 /°C in the beginning of heating and in the end of cooling during this second thermal cycle. This value corresponds to data published in literature using a linear regression between ambient temperature and 200 °C for a similar high-temperature thermoplastic composite as mentioned in part 4.2.

4.4 Observations before and after laminate manufacturing

In order to compare FBG's response reproducibility during and after manufacturing of the laminate, two more trials were carried out following the same experimental setup. The strain – temperature curves (see Fig. 9) demonstrated a similar however not identical response of the FBG during and after laminate manufacturing. This was due to the establishment of the experimental setup which can sometimes differ from one trial to another (FBG slipping relatively to thermoplastic ply in the beginning of heating).

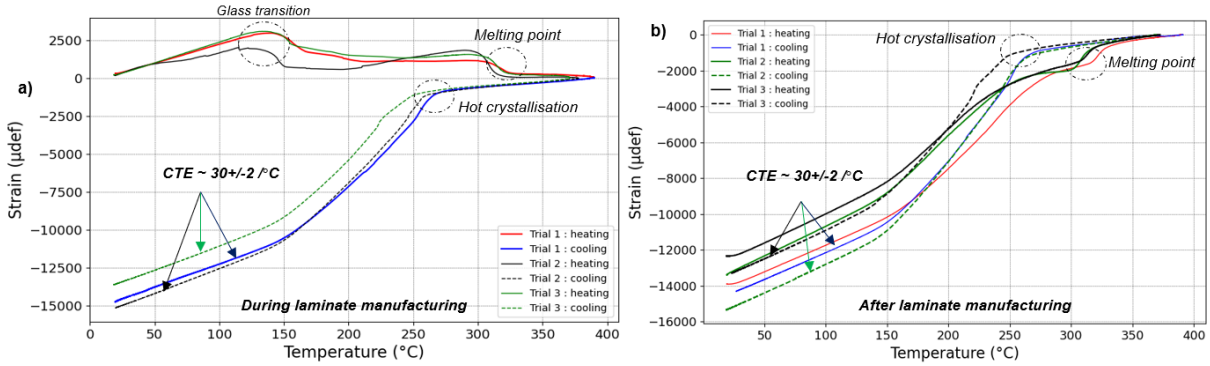

Figure 9: Study of FBG's reproducibility response for trials 1-3: a) before thermoplastic manufacturing b) after thermoplastic manufacturing.

The CTE (/°C) of the thermoplastic composite was estimated from a linear fit to the data over the entire test-temperature range (130 °C – 40 °C) and found similar for the three trials with a value of approximately 30+/-2/°C, demonstrating a similar material behaviour during cooling between the consolidation and post-consolidation cycles. Finally, the hot crystallisation could also be observed during cooling.

On the other hand, different profiles were observed during the heating phases before and after laminate manufacturing. In the first case, the FBG sensor delivers information on phenomena linked to the material consolidation (intimate contact, glass transition). Therefore, once the thermoplastic material is consolidated and maximally crystallised, the above phenomena are unseen.

5 CONCLUSION

Through this study, it was possible to experimentally monitor the consolidation of a high-performance carbon/thermoplastic composite using an embedded FBG. The experimental setup which was designed, demonstrated that an FBG sensor seems to be an appropriate solution to characterize the thermoplastic laminate during as well as after its manufacturing. Thus, it was confirmed that the optical fibre could detect the phenomena occurring during thermoplastic consolidation and deliver relevant information on thermal properties of the thermoplastic matrix such as its glass transition, matrix

crystallisation and melting point temperatures after comparing to data collected by DSC experiments on the thermoplastic prepreg. In perspective, further studies will be carried out in order to use the embedded FBG for detecting initial defects in the laminate during thermoplastic consolidation. An optimal solution could be the establishment of an efficient SHM monitoring system used for both the manufacturing process of the thermoplastic as well as its thermomechanical behaviour.

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