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School of Aeronautical and Space Engineering

Characterization and real-time process
monitoring of thermoplastic composites
manufacturing processes

Doctoral Thesis

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MSc. Physics

2020

Department of Aerospace Materials and Production

School of Aeronautical and Space Engineering

Polytechnic University of Madrid

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2020

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Abstract

There is a renewed interest on thermoplastic composites by the aerospace industry. This is mainly due to the main advantages that thermoplastic composites may offer, along with the recent development of promising manufacturing techniques, which are very close to reach the maturity and the production rates that the industry demands. The main advantages that thermoplastic may offer mainly consist of high strength and impact resistance, unlimited shelf life, low water absorption, short processing times and the opportunity of alternative out-of-autoclave (ooA) processing technologies. Some examples of the mentioned promising technologies that are nowadays being improved are compression moulding, induction welding, over-moulding or automated fibre placement (AFP) with in-situ consolidation system (AFP-ISC). AFP-ISC process consists on the automated lamination of thermoplastic tapes which are melted by a heat source and consolidated by a compaction unit over a previously consolidated substrate. The part is quickly cooled down and solidified. In this way, melting and consolidation of the part can be performed in a single step

This study aimed to characterize and compare different OoA manufacturing methods with a particular emphasis on laser-assisted AFP-ISC. In addition to that, the present investigation also aimed to develop a real-time process monitoring methodology by means of optical fibre Bragg grating sensors (FBG).

The characterization of thermoplastic manufacturing processes was focused on two main issues. The first issue was to investigate the existing gap of mechanical properties observed in the laminates produced by AFP-ISC technology in comparison with other techniques. With this aim, an approach based on the porosity was adopted, since it is considered the main defect responsible of a drop of properties in composites. Thus, void distribution, size and content within laminates manufactured by vacuum bag-only in oven (VBO), hot-press and AFP-ISC were characterized. The characterization campaign included other tests which evaluated other key aspects, including ultrasonic non-destructive testing (NDT), degree of crystallinity by means of differential scanning calorimetry (DSC) and mechanical testing.

The second issue was to investigate the impact of void content on the final mechanical behaviour of the composites; in order to establish interrelations between ultrasonic NDT, void content and mechanical properties. For this purpose, several methodologies for manufacturing defect-induced laminates were investigated. One of them was selected for manufacturing a set of laminates in a wide range of porosity. The laminates were characterized by means of ultrasonic response, void content and mechanical testing; and the interrelation was performed.

Regarding the process monitoring with FBG sensors, it was detected that thermal history is the parameter governing the key phenomena taking place in laser-assisted AFP-ISC. Thus, only infrared (IR) thermographic cameras are typically employed for temperature control during lamination, which are very limited to surface measurements. Thus, different strategies for FBG

sensors embedment were investigated. After the optimization of the strategy, different FBG sensors were embedded with real time process-monitoring purposes. The analysis of the monitored data allowed identifying the main stages of the process, such as heating-up, consolidation and cooling down; and several key findings were made. Also, a set of FBG sensors was encapsulated, which acted as a temperature probe allowing the thermal history monitoring.

The results provided by the performed characterization showed that some properties of AFP-ISC laminates can be up to a 30% lower than VBO or hot-press laminates. The analysis of voids size and distribution revealed that bigger voids can be found in AFP-ISC laminates, which, moreover, are accumulated in interlaminar regions. These findings were related to the drop of mechanical properties. Interrelation of porosity, ultrasonic attenuation and mechanical properties was performed after characterizing the set of laminates manufactured within a wide range of porosity. These correlations allowed obtaining critical void content values, where the mechanical properties of the laminates start decreasing. The results suggested that each processing technology should have its own quality control criteria. The performed characterization is useful for engineering applications of thermoplastic composites and the creation of NDT acceptance criteria.

On the other hand, the application of FBG sensors for process monitoring of laser-assisted AFP-ISC was validated. The main phenomena occurring during the process were identified after analysing the monitored data. Also, thermal history was successfully monitored applying the sensor encapsulation approach. First steps were taken toward strain monitoring during the process, but further work is required. Post-consolidation residual strains remained in the part were assessed by the embedded sensors after demoulding.

Resumen

Durante los últimos años, hemos presenciado un interés renovado en los materiales compuestos de matriz termoplástica por parte del sector aeronáutico. Esto se debe principalmente a las numerosas ventajas que dichos materiales pueden ofrecer, pero también a los recientes avances en el desarrollo de prometedores procesos de fabricación, que están más cerca que nunca de alcanzar el nivel de madurez y las tasas de productividad que demanda la industria. Entre las principales ventajas que ofrecen los materiales termoplásticos se encuentran una alta resistencia, la no necesidad de almacenamiento en frío, baja absorción de humedad, cortos tiempos de procesado y la oportunidad de usar métodos alternativos de fabricación fuera de autoclave (OoA, *out of autoclave*). Entre esas tecnologías que se encuentran en desarrollo a día de hoy, se pueden encontrar el moldeo por compresión, soldadura por inducción, *over-moulding* o el encintado automático de fibra (AFP) con consolidación in-situ (AFP-ISC). Este último proceso de fabricación consiste en encintar y consolidar material de manera automática sobre un sustrato que ha sido previamente consolidado. Para ello, un cabezal realiza el aporte de material mientras que una fuente de calor calienta la resina para que posteriormente una unidad compactadora realice la consolidación con el sustrato y se prosiga con el enfriamiento y la solidificación. Este proceso permite la fabricación de una pieza de material compuesto en un solo paso, sin la necesidad de ciclos de consolidación posteriores en estufas u autoclaves.

El presente proyecto de investigación ha pretendido caracterizar y comparar diferentes métodos de fabricación OoA, con especial énfasis en el proceso AFP-ISC con un sistema láser como fuente de calor. Además de la caracterización, el otro objetivo principal del proyecto se basó en desarrollar un sistema de monitorización a tiempo real del proceso con sensores de fibra óptica de redes de Bragg (FBG), que son principalmente sensibles a variaciones de temperatura y deformación.

La caracterización de los procesos de fabricación estuvo centrada principalmente en dos factores. El primero fue investigar acerca de la brecha existente entre las prestaciones mecánicas de laminados de material compuesto termoplástico fabricados por AFP-ISC en comparación con otros métodos. Esta problemática se abordó desde la perspectiva de que la porosidad y los huecos son los principales defectos que pueden causar una caída de propiedades mecánicas en el material compuesto. De este modo, se realizó un estudio de la distribución tamaño y la forma de la porosidad en laminados fabricados bajo tres métodos OoA, incluyendo bolsa de vacío en estufa (VBO), prensa de platos calientes y AFP-ISC. La campaña de caracterización también incluyó otro tipo de ensayos, como ensayos-no destructivos (END) por ultrasonidos, el grado de cristalinidad por calorimetría de barrido y una serie de ensayos mecánicos.

El segundo de los factores que se investigó fue el efecto del volumen de huecos en el comportamiento mecánico final de laminados termoplásticos, para poder establecer correlaciones entre la atenuación de los END por ultrasónicos, la porosidad y las propiedades mecánicas. Con este objetivo se investigaron diferentes estrategias de fabricación que permitieran obtener laminados con diferentes valores de porosidad. Los laminados fabricados se caracterizaron por medio de END por ultrasonidos, contenido en huecos y propiedades mecánicas. Dicha caracterización permitió realizar la correlación previamente mencionada.

La monitorización del proceso con sensores FBG vino motivada porque se detectó que la historia térmica del proceso es probablemente el parámetro que gobierna todos los fenómenos que ocurren durante el proceso AFP-ISC; y, que a día de hoy solo se monitoriza la temperatura mediante el uso de cámaras termográficas infrarrojas (IR). El uso de dichas cámaras termográficas está muy limitado a medidas superficiales, que no permiten conocer el comportamiento térmico dentro del laminado. En este proyecto de tesis se investigaron diferentes estrategias de embebido de sensores FBG en laminados fabricados por AFP-ISC, lo que supuso un reto debido al agresivo entorno de fabricación por las altas temperaturas y presiones alcanzadas. Una vez que se encontró una estrategia favorable, se investigó el uso de diferentes sensores FBG para monitorizar el proceso a tiempo real. El análisis de los datos monitorizados permitió identificar las principales fases del proceso a las que se ve sometido el material, tal como calentamiento, fundición, consolidación y enfriamiento. Los resultados obtenidos permitieron ampliar el conocimiento acerca del proceso de fabricación y de las diferentes etapas. Para poder monitorizar la historia térmica del proceso, un conjunto de sensores FBG fueron adecuadamente encapsulados para que actuaran como sensores únicamente de temperatura.

Los resultados de la campaña de caracterización revelaron que algunas propiedades de laminados fabricados por AFP-ISC pueden caer hasta un 30 % en comparación con los otros métodos OoA bajo estudio. El análisis de la porosidad sugirió que en los laminados fabricados por AFP-ISC hay huecos de un mayor tamaño; y que además, se concentran en localizaciones específicas, como en las regiones interlaminares, lo cual está relacionado con la caída de propiedades mecánicas.

La fabricación y caracterización de los laminados fabricados en un amplio rango de valores de porosidad permitió realizar la correlación entre valores de porosidad, atenuación de la señal ultrasónica y las propiedades finales. Dicha correlación permitió establecer valores críticos de volumen de huecos, a partir de los cuales las propiedades mecánicas empiezan a decrecer. Debido a la diferente morfología, los resultados sugirieron que los laminados fabricados por métodos distintos deberían de tener criterios de aceptación diferentes. Los resultados pueden ser de gran utilidad para aplicaciones en ingeniería y certificación de materiales compuestos de matriz termoplástica, así como para elaborar criterios de aceptación.

Por otro lado, la aplicación de sensores FBG para monitorizar a tiempo real el proceso AFP-ISC con fuente láser ha sido validada durante la presente tesis doctoral. Los sensores se mostraron capaces de identificar los principales fenómenos del proceso (calentamiento, consolidación, enfriamiento). Del mismo modo, la historia térmica del proceso fue monitorizada con éxito a partir de la estrategia previamente descrita del encapsulado de sensores FBG. Por último, se dieron los primeros pasos acerca de monitorizar la evolución de las deformaciones durante el proceso de fabricación, aunque se trató de un estudio muy preliminar que requiere de futuras investigaciones. Por últimos, se demostró la viabilidad de medir las tensiones residuales acumuladas en el material tras la fabricación mediante los sensores FBG integrados en la pieza..

Agradecimientos

En primer lugar, me gustaría agradecer a mi director de tesis, Alfredo Güemes, por darme la oportunidad de realizar la presente tesis doctoral. Es un honor haber realizado el doctorado bajo la supervisión de una eminencia en el mundo de los composites. De igual manera, me gustaría agradecer a Fernando Rodríguez Lence, ya que sin su colaboración, este proyecto no hubiera sido posible.

De igual manera quería agradecer a todo el personal y doctorandos del departamento de Materiales y Producción Aeroespacial de la ETSIA, Antonio, Ángel, Patricia, Bea y Renato.

No hay espacio suficiente en esta tesis para mostrar todo mi agradecimiento a todas las personas de FIDAMC que han apoyado de una manera u otra en la realización de esta tesis. El departamento de termoplásticos, quienes me guiaron en mis primeros pasos: Edu, Salva, Mar, Katia, con mención especial a Isa, mi compañera de fatigas en esta aventura. Al departamento de laboratorio, por su gran apoyo y colaboración: Silvia, María G, Vanessa, María Z, Margarita, Ingrid, Fran, Carlita, Carolo, José, Rodri y Mikel. Gracias a todos, sé que os alegráis tanto como yo de haber llegado al final de este camino y que no haya más paneles “puzzle” que cortar y ensayar. Un agradecimiento especial a Rafa, puesto que todo lo que sé de inspección por ultrasonidos lo sé gracias a él. No hay suficientes niveles según la EN 4179 que reflejen todos sus conocimientos. También quería agradecer especialmente al personal de la ISC, Rubén, Miriam, Juan Pablo, Javi C, Javi A, Miguel y Pilar, que realizan un trabajo insustituible. A mis compañeros Fernandito, Dani, Gloria, Carlos, Vero, Sofía, Gemma, Lucas, María M, Juanma y Sergio; por todo su apoyo durante estos años.

Gracias a toda las personas que forman la familia FIDAMC.

Quería agradecer a las personas que desinteresadamente han ofrecido soporte técnico a esta tesis, como Ben de Pauw de la universidad VUB. También quería agradecer a las personas del CACM de Auckland, que hicieron posible mi estancia en el extranjero. Gracias a Mark Battley, Tom Allen y, en especial, a Abhi Ramesh por su colaboración en esta tesis doctoral.

Por último quería agradecer a familia y amigos, quienes muestran su apoyo y confianza incondicional en nuestro día a día; no sólo en los buenos momentos, sino también en los no tan buenos. Gracias a mis padres, Mari Carmen e Ignacio. A mi hermano y corrector Nacho. A mis tíos, en especial a Susi y Rafa, por ser ellos. Y a mis abuelos, Pepe y Trini, y Lola y Luis.

Gracias a todos.

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Chapter 1

Introduction

1.1 Introduction

A historic milestone was reached by the aerospace industry in the last generation of aircrafts, since main aircrafts' manufacturers Airbus and Boeing launched Airbus A350 and Boeing 787 with more than 50% of their structure made out of composite material. Airbus A350 counts with a 53 % of its structure made out in composites while Boeing 787 has a 50 % [1,2]. The percentage of materials used for the manufacturing of the Airbus A350 is shown in Figure 1.1. Carbon Fibre Reinforced Plastics (CFRP) market has been growing during the last years without interruption not only in the aerospace sector, but also in other sectors such as automotive, wind energy, construction and sporting goods. As reported by Sauer et al. [3], the global CFRP market is growing nowadays at a yearly rate of 10-12 %; and it is expected to continue growing the upcoming years, due to their application in several sectors, as mentioned.

Figure 1.1 Materials used in the Airbus A350 [1].

The main reason the application of composite materials has been increasing is their excellent mechanical properties combined with low weight and fatigue behaviour [4]. CFRP structures are made of a polymeric matrix material reinforced with fibre which results in an anisotropic behaviour. Thus, the constituents confer to the composite physical properties not present in the constitutive materials taken alone [5]. The aerospace parts are typically manufactured by stacking composite plies and combining the fibre orientation, resulting in multi-directional, lighter and more efficient structures which allow the reduction of fuel consumption.

The current picture of the CFRP in the aerospace industry is clearly dominated by the use of thermoset prepreg cured in autoclave. This is mainly due to the evolution and the high production rates achieved by the innovative automated lamination techniques, such as automatic tape laying (ATL) and automatic fibre /tape placement (AFP or ATP). The high technology readiness level (TRL) of those technologies, along with the properties of the final structure allowed the main application of CFRP in opposition more traditional materials, such as aluminium or titanium. Other composites materials and processes are used for the production of aerospace parts, such as resin transfer molding (RTM) of honeycomb structures. However, the current situation is completely dominated by thermoset composites, as mentioned before.

Thermoplastic composite materials are employed for aerospace application, but their use is limited to secondary structures, such as clips, leading edge or ribs [6]. Regarding thermoplastic polymers, high-performance semi-crystalline polymers are typically employed when it comes to aerospace structures. Those polymers mainly consist of the PAEK (poly-aryl ether-ketone) polymers family, including PEKK (poly-ether-ketone-ketone) and PEEK (poly-ether-ether-ketone). Other high-performance polymers such as PA (polyamide), PPS (poly-phenylene-sulphide) or PEI (polyetherimide) are also employed. The polymers pyramid is shown in Figure 1.2, where polymers are divided by categories depending on their crystalline structure and performance [7]. All these polymers are characterized by having high melting and glass transition temperatures, in addition to a high viscosity. This is why high processing temperature and pressure are typically required in thermoplastic composites processing.

Figure 1.2 Pyramid of thermoplastic polymers, categorized by crystalline structure and performance [7].

Thermoplastic composites offer a number of advantages over conventional thermosets. These advantages have been widely reported in the literature, and can be listed as follows, according to Vodicka [8]:

- High chemical stability, which allows the use of thermoplastic composites in a wide range of operational temperature. Also, thermoplastic resins have high chemical resistance and exhibit excellent resistance to jet fuel and hydraulic fluids.
- High strength and impact resistance which allow the use of thinner structures, leading to a lightweight part and reduction of fuel consumption.
- Unlimited shelf life. Due to the fact that there is no curing reaction in thermoplastic polymers (they are only melted), there is no need of cold storage in industrial fridges. This avoids problems with material age and storage.
- Thermoplastics may be re-melted and re-formed after consolidation. This opens new opportunities for repair and recycling. Thus, a structure may be repaired by re-melting and re-forming to the optimal shape. Regarding recycling, thermoplastic structures may have a second useful life after the life cycle of an aircraft.
- Low water absorption. The moisture absorption of thermoplastic polymers is around one order of magnitude less than thermoset resins. This avoids a drop of mechanical properties caused by in-service moisture absorption. Also, thermoplastic composites exhibit excellent mechanical performance in hot/wet environments.
- Short processing time. Since there is no curing reaction in thermoplastic polymers, the consolidation dwell time may be reduced in thermoplastic processing. Manufacturing processes are mainly focused on melting the resin and compacting the part. The part must then be cooled at a set rate which allows correct level of crystallinity.
- Alternative manufacturing processes. Due to some of the previous mentioned advantages, thermoplastic composites can be processed in a different way than thermosets. Alternative methods such as welding, press forming or over moulding can be employed. AFP can be also used for thermoplastic, in addition with the so-called in-situ consolidation method

(AFP-ISC). This manufacturing method is based on the automated lamination of thermoplastic tapes which are melted by a heat source and placed on the top of a previously consolidated substrate. Consolidation is performed by a compacting unit and the system is quickly cooled down. In this way, melting and consolidation of the part can be performed in a single step. AFP-ISC process is extensively described in next chapter. In this aspect, the greatest advantage of the mentioned alternative manufacturing processes is that an autoclave is not required. This reduces final part costs substantially, offsetting the high raw material costs.

On the other hand, thermoplastic composites also have disadvantages, which can be summarized as:

- High processing temperature is required. As mentioned before, thermoplastics are characterized by having high melting points (T_m) in comparison with thermoset curing temperatures. Typically, service temperatures of composites do not exceed their glass transition temperature (T_g) in wet conditions (also called wet T_g). Thermosets are cured at their dry T_g , which is approximately wet T_g plus 80 °C. On the other hand, T_g of thermoplastic is approximately their T_m divided by a factor of two. In this way, if thermoplastic composites are to be incorporated in aerospace structures under same service temperatures, it is needed to use thermoplastic polymers with T_m in the ranges of 300-350 °C which will result in similar T_g values to thermoset composites. This is the case of the previously mentioned polymers such as PEKK, PEKK or PPS. For instance, PEEK-based composites are commonly processed at 380-400 °C since its T_m is 342 °C. The T_g of PEEK is 143 °C. This fact leads to the use of different ancillary material (bagging films, sealant tapes, etc.). Furthermore, conventional autoclaves may not reach these temperatures and should be customized.
- High processing pressure is required. Thermoplastic are highly viscous materials even when melted, which causes the need of high pressure in most of the manufacturing processes, especially in injection methods.
- High raw material cost. The cost of thermoplastic raw material, whether prepreg-format or resin pellets is higher than the cost of thermosets. These costs are expected to decrease over the next years. However, final part costs can be offset if the processing costs are decreased.
- Processing techniques not mature. Some of the most promising thermoplastic processing technologies have not yet reached proper TRL in order to be incorporated in the production lines of the aerospace industry. Thus, some research is still required for improving those technologies.

Currently, there is a regained interest in thermoplastic polymers for aerospace application in primary structures, which is mainly caused by the evolution of promising processing techniques. These techniques such as AFP with in-situ consolidation, ultrasonic welding or stamping may achieve, in the coming years, the corresponding TRLs and production rates demanded by the industry. There are several indicators that Europe is promoting the research on thermoplastic composites for aerospace applications. This is mainly due to the established goals regarding the reduction of noise, CO₂ and NO_x levels. There are several thermoplastic research projects across Europe framed within programmes funded by the European Commission. Some of those

European programmes which define the EU roadmap for future airframes are TAPAS (Thermoplastic Affordable Primary Aircraft Structure), Clean Sky and CORAC (French Strategic Advisory Board for Civil Aviation Research) [9–11]. The mentioned programmes aim to develop and manufacture aerospace demonstrators made out of thermoplastic composites by the previously mentioned manufacturing technologies, in order to improve their corresponding TRL. TAPAS 1 and TAPAS 2 projects were carried out by a Dutch consortium mainly led by Fokker, the Netherlands Aerospace Centre (NLR) and TU Delft. The main goals of the projects are to develop primary stiffened thermoplastic structures. The CF/PEKK centre torsion box demonstrator of TAPAS project is shown in Figure 1.3 (i). The project called OUTCOME (Out of Autoclave Composite Wing) – framed within CleanSky programme – aims to develop and manufacture thermoplastic wing demonstrators fabricated by laser-assisted AFP-ISC. The project is carried out by a consortium mainly formed by Airbus Defence and Space (ADS), MTorres and FIDAMC (Foundation for the Application and Development of Composite Materials). A CF/PEEK thermoplastic stiffened curved panel manufactured via AFP-ISC is shown in Figure 1.3 (ii). A CF/PEEK thermoplastic stiffened curved panel manufactured via AFP-ISC is shown in Figure 1.3 (ii).

Figure 1.3 Thermoplastic composites aerospace demonstrators manufactured during European projects: (i) CF/PEEK torsion box produced during TAPAS project; (ii) CF/PEEK curved stiffened panel produced by means of AFP-ISC during OUTCOME project; and (iii) CF/PEKK fuselage demonstrator manufactured during CORAC project [9].

In a similar way, CORAC project also aimed to develop a full-scale thermoplastic fuselage demonstrator to allow an evaluation of the use of high performance thermoplastics within a next generation single aisle aircraft [12]. Within CORAC project (led by Stelia Aerospace), AFP of thermoplastics was also investigated; with further out-of-autoclave (OoA) consolidation step. The

CF/PEKK thermoplastic fuselage demonstrator –exhibited at JEC exhibition– manufactured during the CORAC project is shown in Figure 1.3 (iii).

Those industrial projects typically aim to increase the TRL of a certain technology and end up with a final large-scale demonstrator in order to proof the investigated concept. However, some gaps of knowledge may be detected during the evolution of industrial projects, which are more focused on other aspects, such as production rates.

This brief introduction presented the current situation of thermoplastic composites in the aerospace industry, along with the main challenges that still need to be addressed.

1.2 Motivation

The present investigation is the result of the collaboration between FIDAMC and the Polytechnic University of Madrid (UPM). FIDAMC have been researching the thermoplastic composites during the last decade due to its participation in R&D project associated to the aerospace industry. In particular, thanks to its participation in CleanSky-funded OUTCOME project, FIDAMC had the opportunity to develop thermoplastic OoA manufacturing techniques, such as oven consolidation via vacuum bag-only (VBO), compression moulding or laser-assisted AFP-ISC.

During the development of the research projects, some gaps of knowledge were detected in the field of thermoplastic composites processes that the industry was not covering. In this way, the aforementioned collaboration between FIDAMC and UPM was initiated, resulting in several PhD projects.

The present study focuses on the characterization of thermoplastic laminates manufactured by different OoA methods, including VBO, hot-press and AFP-ISC. AFP-ISC is one of the promising technologies that is being developed not only by FIDAMC, but also by other international research institutions. However, the technology is not yet ready for industrial application in the manufacturing of primary aerospace structures.

Among the different challenges to be addressed, it was detected that there is a gap of mechanical properties between thermoplastic parts manufactured by means of autoclave and AFP-ISC. The present study aims to investigate this issue from the point of view of porosity, since voids are one of the main defects responsible of a drop of properties in composite structures. Regarding porosity, average void content is typically only checked when a composite part is manufactured in an industrial environment. In this study, the investigation of other key aspects of porosity, such as void size or distribution is also performed by cutting-edge methods, such as 3D X-Ray computed tomography. Other aspects related to the final mechanical performance of thermoplastic composites are also investigated, such as the degree of crystallinity.

It was also detected that all the previous investigations regarding the interrelationship of ultrasonic non-destructive testing (NDT), porosity and mechanical properties were totally focused in thermoset-based composites. Only few or even no studies were found regarding ultrasonic testing of thermoplastic composites and its relation to void content and mechanical properties. Some of the required activities before an aerospace structure can be manufactured by means of a specific material and process (M&P) are certification activities. Among all the tasks performed

during this step, the acceptance criteria for the quality control of the manufactured parts should be defined. For this purpose, the relation of void content with the mechanical properties is required, along with the behaviour of a specific NDT technique (typically ultrasounds). This study aims to perform the required investigations regarding the mentioned interrelations.

During the development of thermoplastic AFP-ISC manufacturing process, other issues were identified that are investigated in the present study. Hence, the thermal history of the process governs the main physical phenomena occurring during process (essentially melting, bonding and consolidation, which are in-depth analysed in next chapter). However, the current state of the technology is lacking of a robust monitored system, since temperatures are only recorded by means of infrared (IR) thermographic cameras. This monitoring system is limited to surface measurements which prevent a comprehensive understanding of the thermal behavior of the material on the inside. This study proposes the development of an on-line process monitoring system based on the use of optical sensors. The monitoring system is based on fibre Bragg grating (FBG) sensors, which aims to identify and analysed all the key phenomena of the process.

The sensors are known for being sensitive to both strain and temperature. Therefore, a monitoring strategy is proposed for monitoring not only the thermal history of the process, but also to provide information about the strain accumulation on the manufactured part which may eventually cause residual deformation in the part. Summarizing, the proposed monitoring approach aims to enhance the understanding of thermoplastic AFP-ISC manufacturing process in order to address the issues that are not yet solved.

1.3 Objectives

According to the technological challenges and issues to overcome described in the previous section, general objectives of the project were established. Those objectives are based on the characterization of the behaviour of CF/PEEK thermoplastic laminates manufactured under different OoA methods. The OoA methods under investigation in this work are VBO, hot-press and innovative laser-assisted AFP-ISC, with special emphasis on the latter. In addition to that, the development of an on-line process monitoring system based on FBG optical sensors was established as a main objective. The monitoring system may allow the monitoring of the thermal history of the process.

In order to achieve the aforementioned main objectives, they were broken down into specific objectives, as specified hereafter:

- To investigate void content, distribution and size in thermoplastic laminates manufactured under OoA technologies, including laser-assisted AFP-ISC.
- To characterize and compare the mechanical response of thermoplastic laminates manufactured under different OoA methods. In addition to that, the gap of mechanical properties of AFP-ISC laminates should be analysed.
- To evaluate the effect of voids in thermoplastic composites. With this aim, the development of a manufacturing methodology which allows obtaining a set of laminates

in a wide range of porosity is required. Once the defect-induced laminates are manufactured, it is essential to characterize the laminates in terms of physical and mechanical properties.

- To establish a relation between void content, ultrasonic testing and mechanical properties of thermoplastic composites. The previously mentioned manufactured set of laminates will allow to correlate the results provided by NDT, void content and mechanical tests.
- To develop a methodology for embedding optical fibre FBG sensors into laminates manufactured under laser-assisted AFP-ISC process.
- To validate the application of FBG sensors for on-line process monitoring of laser-assisted AFP-ISC process.
- To identify the main key phenomena occurring in laser-assisted AFP-ISC process by means of monitoring the process with FBG sensors.
- To monitor the thermal profile of laser-assisted AFP-ISC process with FBG sensors and to gain some knowledge about the post-consolidation strains in a structure.

1.4 Thesis outline

The present thesis manuscript is organized in different chapters, which are listed and detailed hereafter:

Chapter 2 consists of review of literature relevant for the investigation. The chapter was divided into three sections. First section describes different manufacturing methods of thermoplastic composites, including autoclave and OoA technologies, such as VBO, compression moulding and AFP-ISC. The second section is related to the defects in composites, with particular focus on voids and porosity. The section addresses key aspects such as void formation mechanisms, void characterization methods and the most relevant investigations to date regarding the impact of porosity in the mechanical response of composite materials. In the third and final section, a literature review regarding the FBG sensors and their application in composites process-monitoring is presented.

In Chapter 3, the characterization of CF/PEEK laminates manufactured under the three OoA methods under investigation in this study is presented. Thus, the laminates were subjected to ultrasonic NDT, void analysis, differential scanning calorimetry (DSC), 3D X-Ray tomography and a set of mechanical tests. Special attention is given to the void distribution and size within the manufactured laminates. A thorough discussion is carried out regarding the different void formation and evacuation mechanisms in the different OoA methods. Also, the mechanical response exhibited by the different laminates as a function of the porosity distribution is discussed.

Chapter 4 focuses on the effect of void content on the thermoplastic laminates. In this chapter, different methodologies for defect-induce laminates manufacturing are presented and discussed. One manufacturing methodology was selected and the manufacturing of a set of laminates in a

wide range of porosity is presented. This chapter also presents the correlations performed by linking ultrasonic NDT, void content and mechanical properties of thermoplastic properties, which is useful for certification activities.

In Chapter 5, a methodology for successfully embed FBG sensors into laser-assisted AFP-ISC process is investigated and presented. Furthermore, on-line process monitoring with different FBG sensors is presented. The results provided by the monitoring experiments are presented and discussed, which allowed identifying key phenomena and stages of the process, such as heating-up, compaction and cool-down.

Chapter 6 investigates a methodology for using FBG sensors as temperature probes. The applied approach was based on the encapsulation of the FBG sensor inside a metallic capillary for isolating the sensor from external strains. The chapter presents the experiments carried out by different optical sensors and configurations and reports the monitored thermal histories. In addition to that, first steps were taken towards on-line strain monitoring during the process, even though further work is required. The use of embedded FBG sensors for evaluating the remaining final residual strains within the thermoplastic samples after demoulding is presented.

Chapter 7 compiles the key findings and conclusions achieved during the development of this investigation.

In Chapter 8, the potential future investigations which may continue and complete the present study are presented.

1.5 List of publications and milestones

During the development of the present doctoral thesis, some of the results were disseminated by means of journal articles or conference papers. The detailed information about the publications is given below, along with the description of the conducted research stay.

Journal articles

Saenz-Castillo, D., Martín, M.I., Calvo, S., Rodriguez-Lence, F., Güemes, A., Effect of processing parameters and void content on mechanical properties and NDI of thermoplastic composites, *Composites:Part A* (2019), doi: <https://doi.org/10.1016/j.compositesa.2019.03.035>

Saenz-Castillo, D., Martín, M.I., Ramesh, A., Battley, M., Güemes, A., A comparison of mechanical properties and X-ray tomography analysis of different out-of-autoclave manufactured thermoplastic composites, *Journal of Reinforced Plastics and Composites* (2020).

Status: Accepted April 2020.

Saenz-Castillo, D., Martín, Güemes, A., Real-time monitoring of thermal and strain evolution during thermoplastic automatic lamination with FBG sensors and process modelling validation, *Smart Materials and Structures* (2020).

Status: Under peer review process in Smart Materials and Structures (submitted on 21st April 2020)

Conference papers

Saenz-Castillo, D., Martín, M.I., Rodriguez-Lence, F., Güemes, A., On line monitoring of a laser assisted fiber placement process with CFR thermoplastic matrix by using Fiber Bragg Gratings, *8th European Workshop on Structural Health Monitoring (EWSHM 2016)*, July 5-8, 2016 in Bilbao, Spain (EWSHM 2016).

Proceeding available at: <https://www.ndt.net/search/docs.php3?id=20174>

Saenz-Castillo, D. , Rodriguez-Lence, F., Güemes, A., Implementation of FBG optical sensors within a laser-assisted fiber placement process for on-line monitoring of high temperature thermoplastic materials consolidation, *VIII ECCOMAS Thematic Conference on Smart Structures and Materials SMART 2017*. June 5-8, 2017, Madrid, Spain.

Saenz-Castillo, D. , Rodriguez-Lence, F., Calvo, S.; Güemes, A, Impact of processing parameters on void content and mechanical properties of PEEK/CF thermoplastic composites, *ECCM18 - 18th European Conference on Composite Materials*. Athens, Greece, 24-28th June 2018.

Proceeding available at: <https://az659834.vo.msecnd.net/eventsairwesteuprod/production-pcoconvin-public/1c12cd5927a548778332d885b6be1507>

Saenz-Castillo, D. , García-Martínez, V., Calvo, S., Rodriguez-Lence, F.. Güemes, A., Critical void content of CF/PEEK thermoplastic composites: impact on NDT and mechanical properties. 22nd International conference on composite materials (ICCM22). Melbourne, Australia, 11-16th August 2019.

Proceeding available at Engineers Australia, 2019: 3826-3834:

<https://search.informit.com.au/documentSummary;dn=897462765002128;res=IELENG>

Research stay

During the development of the present PhD, the author had the opportunity to conduct a international research stay at the Centre for Advanced Composite Materials (CACM) of the University of Auckland in Auckland, New Zealand, under the supervision of deputy director Mark Battley.

Ultrasonic Testing (UT) – NDT Level II Certification

Due to the collaboration with FIDAMC and the work carried out regarding ultrasonic testing of composites, the author had the opportunity to be certified as UT-Level II inspector under standard EN 4179.

Chapter 2

Literature review

2.1 Manufacturing technologies for thermoplastic composites

2.1.1 Autoclave processing

In Fernández et al. [13], the processing of thermoplastic composites by autoclave is thoroughly analysed, also defined as autoclave forming. The main difference between autoclave processing of thermoset and thermoplastic composites resides in the fact that thermoset processing involves chemical reactions; while thermoplastic processing involves physical changes in the matrix, which needs to be melted and consolidated. In contrast with thermosets, thermoplastic processing requires the use of autoclaves adapted to work at very high temperatures, since the employed temperatures for thermoplastic consolidation are typically above 330 °C. In Figure 2.1, a consolidation adapted for processing thermoplastic processing is shown.

Figure 2.1 High-temperature autoclave, prepared for working at very high temperatures up to 650 °C [13].

Processing of thermoplastics in autoclave allows manufacturing large and complex composite parts. One of the main advantages is the co-consolidation of different elements, such as a skin with reinforcements (for instance, T or Ω stringers) in a single forming cycle [14]. The UPM participated in the past in several programmes related to the thermoplastic advanced composite structures, in collaboration with other institutions such as the INTA or EADS [15].

The phases involved in thermoplastic processing in autoclave are quite similar to those carried out for thermoset processing. However, some differences can be identified, especially those related to the lack of tacking in thermoplastic prepreg. Due to this, the lay-up is commonly performed manually and the layers are joined by employing welding guns and locally melting the resin (automated lamination of thermoplastic composites by means of AFP is discussed in further subsections). After lamination, a vacuum bag is then prepared for consolidation in oven. Due to the employed high temperatures, the ancillary materials used for the vacuum bag are different from those applied with thermosets. Thus, ancillary materials may consist of high-temperature tacky tape, polyimide vacuum bag and high-temperature breather (glass fibre fabric can be used for this purpose). An example of the preparation of a high-temperature vacuum bag is shown in Figure 2.2.

Figure 2.2 Preparation of a high temperature vacuum bag for thermoplastic composites processing [16].

Once the vacuum bag is prepared, the samples are typically consolidated by using temperatures above the melting point of the polymer and pressure between 6 and 10 bars. Due to the lack of

curing reaction, dwell times are significantly shorter than those applied with thermosets. In this case, the dwell time should only guarantee proper void evacuation and consolidation between plies. However, cooling rate should be taken into account, since it has an impact on the final degree of crystallinity of the structure, which will play a key role in its final structural behaviour. In Figure 2.3, some examples of thermoplastic parts manufactured in autoclave by Fernández et al. [13] are shown. Figure 2.3 (i) shows a double-hat stiffened panel demonstrator after co-consolidation in autoclave; meanwhile Figure 2.3 (ii) shows a PPS/carbon fibre demonstrator for the A340-600 elevator leading edge skin.

Figure 2.3 Thermoplastic structures manufactured in autoclave: (i) Double-hat stiffened panel demonstrator obtained by autoclave co-consolidation techniques and (ii) PPS/carbon fibre demonstrators for the A340-600 elevator leading edge skin [13].

2.1.2 OoA processing

In this section, the literature review regarding the OoA processing methods under research in this study is presented, along with additional information about other trending OoA processing methods.

2.1.2.1 Vacuum bag-only in Oven

The fabrication of thermoplastic structures by means of vacuum bag only or vacuum bag in oven (commonly shorted as VBO) follows similar procedures to those carried out during autoclave procedures. The main difference in this case resides in the absence of applied pressure during the consolidation cycle. In this way, a correct extraction of the voids during the cycle is the main concern in VBO processing.

Also, the forming of complex structures is often more complicated due to the lack of pressure. Thus, the consolidation of specific parts or regions, such as corners, stringers or curved zones results in a higher complexity. Usually, longer dwell times are required for correct void extraction and proper consolidation. Figure 2.4 shows the preparation of complex thermoplastic geometries prior being consolidated by VBO. Figure 2.4 (i) shows a CF/PEEK L-shape stringer ready for consolidation and Figure 2.4 (ii) shows same stringers inside the oven at FIDAMC facilities.

Figure 2.4 VBO manufacturing of a thermoplastic L-shape stringer. (i) Vacuum bag prepared ready for consolidation and (ii) stringer inside the oven at FIDAMC facilities.

One of the main challenges of VBO processing is to produce parts with a final void content under 1% or 2%, which may fulfil typical aerospace requirements [17]. Several authors have investigated the optimization of VBO processing and the void dynamics in the process. For instance, Zhang et al. published a series of articles where they studied the consolidation of thermoplastic composites by VBO, the material permeability and voids dynamic [18–20]. Those studies reported the feasibility of manufacturing thin and thick laminates with final low void content. They also predicted a minimum dwell time for voids evacuation corresponding to 11 minutes.

2.1.2.2 Compression moulding

Forming of thermoplastic composites has been investigated since the end of the last century [21] until today [22–25], due to the extensive possibilities that press forming may offer, apart from cost and time-savings. Compression moulding is a fast, repeatable and cost-effective manufacturing process. The process consists of locating a thermoplastic lay-up in a tool which is located in a hot press which heats the preform above the melting point T_m of the employed thermoplastic polymer. Pressure is then applied to perform the forming of the part and the final consolidation. In contrast with the previously described manufacturing methods, there is no vacuum vent during compression moulding; therefore the void extraction mechanisms are mainly driven by the applied pressure. However, low void content parts can be achieved by increasing the dwell time when the pressure is applied [23].

Compression moulding methods may vary depending on the type of heating system and the moment when the preform is located inside the press. For instance, there is a variation of the technology, which is denominated stamping, where the preform is typically heated by fast methods such as infrared lamps and when the preform reaches a temperature above the forming temperature T_{form} it is transferred to a mould in a press which can form the part to the final shape in few seconds. Time and pressure are applied while the system cools down. Pressure is release when the temperature drops below the crystallization point. Figure 2.5 shows forming process of a thermoplastic laminate into an omega-shape stringer. Figure 2.5 (i) shows the laminate holding by springs and Figure 2.5 (ii) shows the laminate located into the press ready for forming.

Figure 2.5 Press forming of a thermoplastic laminate into a omega stringer. (i) Laminate held by springs and (ii) laminate located into the mould inside the press for consolidation [23].

For characterization and research activities, hot platen press are commonly used. The process is essentially similar with some slight differences. The main difference is that flat laminates for characterization are commonly manufactured in a hot platen press, rather than complex structures. The prepreg laminate is located on the top of the bottom plate. The press is closed and temperature and pressure is applied. In this method, the heat is commonly applied through conduction; since the press plates are in direct contact with the laminate under consolidation (or in direct contact with caul plates or demoulding sheets). A metallic frame is also commonly used; acting as resin retainer and ensuring the final thickness of the laminate. A hydraulic hot-press for composites manufacturing, along with the manufacturing set-up scheme is shown in Figure 2.6.

Figure 2.6 Hot-press manufacturing of composites plates. (i) Schematic representation of hot-press manufacturing method [26]; and (ii) industrial hot-press for composites manufacturing [27].

The importance of the processing parameters in the final quality of composites has been investigated and reported by different authors [28,29]. Thus, consolidation temperature and pressure were identified as key factors for the final quality (and void content) of the final structure, along with the applied dwell time.

2.1.2.3 Automatic Fiber Placement with in-situ consolidation (AFP-ISC)

As described in the previous chapter, AFP or ATL of composite materials and subsequently autoclave curing are the current preferred option for composite processing in the aerospace sector, due to the high production rates that can be achieved. However, ATL method is limited to quasi-flat manufacturing; meanwhile AFP processing combines the advantages of ATL and filament winding, providing the possibility of laminating complex structures at high production rates.

AFP of thermoplastic composites has been under research since the end of the last century [30–38], but a regained interest on thermoplastic materials along with promising new generation equipment caused a new intense wave of research during the last decade [39–63], mainly motivated for the possibility of avoiding any further consolidation steps (especially autoclave due to high energy costs), if the automated lamination is performed accompanied by the in-situ consolidation method. A typical thermoplastic ISC process is shown in Figure 2.7. The process is based on an incoming unidirectional thermoplastic prepreg tape which is fed and guided by a spools system to the surface of a previous consolidated substrate. A heat source is used for heating up both the incoming tape and the substrate in order to elevate their temperatures above the melting point of the polymer, followed by the application of a consolidation force by a compaction roller, resulting in the bonding of the surfaces.

ISC consists on a high complex process where many physical-chemical phenomena take place, which are governed by optimal processing parameters. In this way, the heating source should heat both the incoming tape and the substrate in order to facilitate the bonding, also conducted with the help of the compaction pressure. When the two surfaces are in contact (intimate contact), the polymer chains start reptating and cross-linking is then performed, which will determine the degree of bonding (D_b). In this way, the parallelism between surfaces is highly recommended so a good intimate contact can take place. Also, temperature at nip point should guarantee a proper viscosity of the matrix which facilitates the bonding, but also the voids compression (further discussed in next sections). After the roller passes, the polymer starts cooling down and therefore crystallization begins. Crystallization is mainly driven by the cooling rate, which is especially high during ISC process.

Since those phenomena take place at very short-times, processing parameters should be optimized for a proper bonding and crystallization, which ensure a high quality final part. However, due to the short processing times, the process window is very narrow.

Figure 2.7 Schematic set-up of a typical thermoplastic AFP system with in-situ consolidation [64].

Different systems can be employed for providing the main processing parameters. Depending on the specifications of an AFP-ISC machine, several configurations are possible, as described in next sub-section.

2.1.2.3.1 Thermoplastic AFP-ISC machines systems.

AFP-ISC thermoplastic head machine may differ in the different hardware aspects. Among the main hardware systems, the main ones may consist of the head machine, heating source, compaction roller, number of feeding tapes and temperature control systems. The different possibilities are described in this sub-section.

▪ Head machine

First of all, a thermoplastic AFP-ISC machine needs to be installed allowing the head movement. Typically, main lamination machine suppliers such as MTorres, Automated Dynamics, Electroimpact or Coriolis provide multi-axial gantry-style machines; however, at a research stage, replaceable and modular heads mounted on industrial multi-axial robots can be a good choice for performing investigation at laboratory scale, while the process is still being optimised.

Figure 2.8 shows three examples of ISC machine heads mounted in multi-axial robots. Figure 2.8 (i) shows Evo I developed at the Institut für Verbundwerkstoffe, Figure 2.8 (ii) shows a customized ISC head mounted in an arm and Figure 2.8 (iii) shows an Automated Dynamics ISC machine mounted on a robotic arm.

Figure 2.8 Thermoplastic AFP-ISC machines mounted on multi-axial robotic arms. (i) Evo I developed at the Institut für Verbundwerkstoffe [65], (ii) ATP-ISC mounted in a robotic arm [44]; and (iii) Automated Dynamics thermoplastic ISC head mounted on an industrial robot [66].

On the other hand, Figure 2.9 shows thermoplastic gantry-style ISC machine heads. Figure 2.9(i) shows MTorres gantry-style AFP-ISC machine at FIDAMC facilities [67] and Figure 2.9 (ii) shows Accudyne Systems thermoplastic gantry-style AFP-ISC head [68].

Figure 2.9 Thermoplastic AFP-ISC machines mounted on multi-axial gantry. (i) MTorres ISC machine at FIDAMC facilities [67]; and (ii) Accudyne Systems thermoplastic gantry-style ISC head [68].

▪ Heating source

Heating systems is one of the key elements of the ISC process which will eventually impact on the thermal history of the process. Different methods on heating up the material substrate and the incoming tow have been investigated during the last decades. Earliest studies were performed by choosing hot gas torch, meanwhile most recent studies have been performed by using diode lasers. Other investigations also reported the use of ultrasonic welding or the innovative VCSEL (Vertical cavity surface-emitting laser) system.

Hot-gas torch heating typically consists of the use of nitrogen, which is heated by the torch and then directed to the composite through a nozzle. Apart from the heat source itself, there are different approaches for the heating strategy. Thus, several investigations were carried out with two heating sources, instead of the typical single nozzle. Examples of hot-gas torch heating are given in Figure 2.10. A two hot gas nitrogen torches system is presented in Figure 2.10 (i), where the first nozzle pre-heats the substrate and the incoming tape; meanwhile the main nozzle

provides through thickness heat in order to perform consolidation and bonding [69]. On the other hand, Figure 2.10 (ii) shows a single hot gas torch system with a customize nozzle concept which ensured heat flux [44]. In any case, the flow gas rate (L/min), lamination speed and final temperature at nip point have been demonstrated to have a huge impact on the final quality of the manufactured laminates.

Figure 2.10 Hot-gas torch heating system for thermoplastic AFP-ISC. (i) Double nozzle system [69]; and (ii) single nozzle system [44].

Regarding diode-laser heating, first investigations employed CO₂ lasers, followed by the use of Nd-YAG lasers and finally diode-laser. It is mandatory to know the interaction of the laser radiation with the composite material when processing with a laser source, especially the absorption and emission coefficients, as described by Martín [70], Figure 2.11 shows two examples of laser-assisted AFP-ISC systems. Figure 2.12 (i) shows a Coriolis AFP-ISC machine located at the NCC (National Composites Centre, UK) with a diode-laser supplied by Laserline GmbH [71]. Figure 2.12 (ii) shows the MTorres single-tow AFP-ISC machine at FIDAMC facilities with also a diode laser supplied by Laserline GmbH [67]. Similarly to the case of hot-gas torches, different approaches focused on the use of a second laser can be followed. In addition to that, key parameters as laser power or radiation angle define the process thermal history, which is decisive in the part quality; impacting on the degree of bonding, intimate contact, healing and therefore final mechanical performance.

Figure 2.11 Thermoplastic laser-assisted AFP-ISC. (i) Coriolis AFP-ISC with a diode laser at NCC facilities [71]; and (ii) MTorres single-tow AFP-ISC machine with a diode laser at FIDAMC facilities [67].

Other alternative heating systems have been investigated, such as ultrasonic consolidation or VCSEL. Chu et al. [72] reported activities involving innovative ultrasonic vibration automated fibre placement (UAFP). The mechanism consisted of the molecule being frictionally heated by the low-amplitude, high-frequency ultrasonic vibration. The range of ultrasonic frequency is 20–120 kHz, and the mechanical wave produced by the ultrasonic vibration heats the interface. Processing of glass fibre/polypropylene was reported.

VCSEL heating consists on several arrays of micro laser diodes and each line arrays can be controlled independently. This way, heating profiles can be arranged during a process, and the use of VCSEL may result in some advantages, such as different local heating zones [73]. Figure 2.12 (i) shows a VCSEL unit [73] and Figure 2.12 (ii) shows a thermoplastic VCSEL-assisted AFP-ISC at Fraunhofer IPT facilities [74].

Figure 2.12 VCSEL heating system. (i) VCSEL unit [73]; and (ii) thermoplastic VCSEL-assisted AFP-ISC at Fraunhofer IPT facilities [74].

Even though hot-gas torch systems consisted of a cost-effective option, the response time is considerably lower than those obtained by diode laser, which is currently the preferred option. Schledjewski et al. [65] reported in 2009 a graph after conducting a cost-efficiency analysis, where they gave scores to different heating sources for thermoplastic AFP-ISC according to different factors, including (i) costs (acquisition, energy, consumables, personnel qualification, maintenance), (ii) feasibility (complexity, know-how, potential) and (iii) power output (material degradation, maximum power, depth of penetration, adaptability to tape material, accuracy, reaction time and degree of automation).

Figure 2.13 Efficiency of heating systems for thermoplastic AFP-ISC [65].

▪ Compaction unit

The main advantage of thermoplastic AFP-ISC technology is to offer the possibility of consolidation in one single step, without needing any extra autoclave or oven consolidation, as Schledjewski highlighted in [65]. Due to the nature of the process, consolidation may take place in very short times of few seconds and, due to size limitations, the area where the consolidation force can be applied is also very limited. This limitation makes the compaction unit selection of a high importance. In [65], two different approaches were discussed: (i) area pressure systems like slides shoes which offer pressure during longer times but have significant dimensions which limited the adaptability to curvatures and (ii) pressure rollers, which offer higher flexibility but less covered surface.

According to the literature, compaction rollers are the preferred option as compaction unit and can be found in most of the research publications. Within the compaction roller approach there are different options, regarding the material and heating system. Thus, Henne et al.[75] investigated different roller concepts which are shown in Figure 2.14. As observed, the compaction roller may consist on a rigid system (metallic roller) or flexible roller (for instance, silicone rollers) regarding the material and may be heated or refrigerate. Non cooled rollers may tend to overheat and therefore their service life is limited. Rigid rollers offer less compaction area and can't be property adapted to curvatures. Henne et al. concluded that the most promising option consisted of flexible silicone rollers, in accordance with Schledjewski, who reported higher peel force values for adaptive rollers in comparison with non-adaptive rollers [65].

	Concept 1: Flexible silicone roller (air cooled outside)	Concept 2: Flexible silicone roller (conduction cooled)	Concept 3: Rigid metal roller (water cooled inside)	Concept 4: Flexible silicone roller with water cooled core
Sketch				
Result				

Figure 2.14 Different compaction roller concepts investigated by Henne et al. [75].

▪ Number of feeding tapes

Regarding the productivity of the process, several studies claimed that there exists a maximum threshold of the lamination speed; and therefore a good degree of bonding cannot be achieved above this threshold [76]. Other aspect which may contribute to improve the productivity is the number of tapes or tows than are laid in each consolidation pass and therefore improve the productivity in terms of Kg/hour. Thus, most of the investigations performed at a research stage consisted of single-tow machine heads; but in the recent years, multi-tows head machines are emerging as a promising option for increasing productivity. Figure 2.15 shows multi-tow AFP-ISC systems. Figure 2.15 (i) shows MTorres multi-tow AFP-ISC machine head at FIDAMC facilities [67] and Figure 2.15 (ii) shows a Coriolis 8-tow feeding system [77].

Figure 2.15 Multi-tow AFP-ISC systems. (i) MTorres multi-tow AFP-ISC machine head at FIDAMC facilities [67]; and (ii) Coriolis 8-tow feeding system [77].

It is important to note that multi-tow concepts required larger heating areas and therefore a more complex laser heating profile (or other heating source) should be applied which ensures homogeneous and proper heating of incoming tapes and substrate for a good quality bonding. Other studies claim that higher productivity may be achieved by other approach, consisting of laminating at the maximum possible tape speed (even with poor consolidation) and a subsequent oven consolidation step.

▪ Temperature control systems

During AFP-ISC process, it is of high importance to monitor the processing temperature, since the thermal history has a great impact on the quality of the manufactured part, and it is the parameter which governs the main phenomena like melting, bonding and crystallization. The temperature monitoring allows modifying the applied power of each heating source (hot gas, laser, etc.) in order to obtain the desired processing temperature at the nip point.

In the current state-of-art of the technology, the most extended temperature control system is the infra-red (IR) thermographic cameras. In this way, an IR camera is attached to the head machine in a way which allows temperature monitoring, as depicted in Figure 2.16, where a FLIR IR camera is attached to the MTorres single-tow AFP-ISC machine at FIDAMC facilities [67], as shown in Figure 2.16 (i). Thermal profile monitored by Stokes-Griffin et al. [47] is shown in Figure 2.16 (ii). Several disadvantages are found in the use of IR cameras. Thus, the emission and absorption coefficients of the elements of the manufacturing set-up should be known, in order to obtain reliable results; resulting in time and cost consuming activities. Thermal models of the process have been deeply investigated in order to predict the thermal history of the process and to compare the experimental measurements. Other drawbacks of IR camera monitoring are directly related to some of the key objectives of the present investigation. Those drawbacks consist of the lack of temperature measurements in the cooling stage, the contaminating effect of the reflections into the reliability of the IR measurements and the limitation to non-contact surface measurements. As previously commented during the introduction, an embedded monitoring system based on FBG sensors is proposed in this investigation.

Figure 2.16 IR thermal cameras for temperature control. (i) FLIR IR camera in the MTorres single-tow AFP-ISC machine at FIDAMC facilities [67]; and (ii) monitored thermal profile by an IR camera [47].

Other research studies investigated the embedding of thermocouples into the substrate for the thermal history monitoring [33,46,78,48,50,79]. Most of the investigations reported the use of thermocouples in order to validate thermal models and to correlate the results. Other studies were interested in knowing the experimental thermal history of the process for a better understanding of the process itself and the main phenomena which take place. Whatever the reason, most of the authors used K-Type thin thermocouples, which consisted of diameters below 100 μm (thinner than a typical single composite ply), in order to not cause detrimental effects on the structure. Regarding the reliability of the measurements, some of the authors rejected the validity of the

monitored data during the first layers due to direct irradiation over the thermocouple and possible air accumulation in the vicinity of the thermocouple [57]. Besides, overestimations in the temperature measured due to the irradiation on a metallic wire may take place during thermocouple monitoring.

Alternative solutions are proposed in this study for ISC thermal monitoring. Thus, the use of FBG optical sensors is proposed for temperature monitoring. To the best of the author's knowledge, the present study (and the already disseminated activities [80]), along with the research developed by Oromiehie et al. [49,81–83]; consist of the only research investigations carried out regarding the use of FBG sensors for thermoplastic AFP-ISC process monitoring.

2.1.2.3.2 Issues to overcome

According to the revised literature and the FIDAMC experience, the development of thermoplastic AFP-ISC has overcome many challenges thanks to the research performed in the last decades. Despite those research efforts, there are still several issues or challenges that need to be addressed and overcome. The main identified issues are compiled and discussed in this subsection.

- **Gap of mechanical properties of thermoplastic AFP-ISC structures**

There are many studies which investigated different mechanical properties of thermoplastic AFP-ISC laminates [39–41,47,48,50,54,68,69,84–87]. The main investigated properties consisted of fast screening techniques such as SBS (short beam shear) and interlaminar shear strength (ILSS). Also, it was found that tests such as fracture toughness (G_{IC}), single-lap shear (SLS), wedge peel or in-plane shear strength (IPSS) were directly related to the quality of the bonding and were also investigated in other research studies. Overall, it was generally accepted that there is a gap of mechanical performance between ISC-manufactured laminates and autoclave processed laminates; or even other OoA methods as hot-press. Depending on the mechanical test performed, some authors revealed a difference of 10-15 % of mechanical properties between ISC and autoclave, as the case of Lamontia et al. [88], who reported the results of a collaboration between Accudyne systems, the University of Delaware and NASA, revealing values of IPSS (154 MPa for ISC, 174 MPa for autoclave), open hole compression (294 MPa for ISC, 330 MPa for autoclave) and axial compression (481 MPa for ISC, 495 MPa for autoclave). Other cases reported a more abrupt gap, as the results obtained by Comer et al. [48], who reported better response of ISC laminates in comparison with autoclave in terms of interlaminar toughness (134 %), but they found poorer mechanical behaviour in flexure strength (68 %), ILSS (70 %) and open-hole compression (78 %).

Many other similar results can be found in the literature, but the important aspect to highlight is that regardless of the mechanical property, it is accepted the lack of mechanical properties which ISC structures can achieve nowadays. There is no a single reason behind the gap of mechanical properties in ISC laminates, but multiple reasons. It is important to take a global perspective to understand the issue. After evaluating the existing literature, it can be said that the primary reasons leading to a drop of properties may consist of a poor bonding quality (degree of bonding

or bond strength), elevated void content and residual stresses; which may be largely solved by the combination of processing parameters optimization, correct lamination strategy and AFP-grade thermoplastic tapes.

Regarding the bonding quality, several studies can be found which investigated the related phenomena such as the intimate contact and the autohesion (healing) [39,65,68,69]. As previously mentioned, the short times (typically few seconds) during which temperature and compaction pressure can be applied make very difficult to achieve same degree of bonding obtained by autoclave processing. This is why many authors suggested that processing parameters must be optimized, since many phenomena (intimate contact, healing, void compression) are mainly driven by processing parameters such as laser power, laser orientation, compaction loads and tape speed. This is also why several authors defended that there is a maximum tape speed for allowing proper bonding and healing of the polymer chains; and if that tape speed is exceeded, bad consolidation is expected. For instance, Sonmez et al. [76] defended that there is a maximum tape speed of 8 cm/s; equivalent to 13.3 m/min. Along with the parameters optimization, there is a general demand of specific AFP-grade raw tapes. It is accepted that raw thermoplastic tapes having flat surface, rich matrix at the surface, constant thickness, low intralaminar voids and good fibre/matrix distribution can be of huge help for good intimate contact, low final void content, good consolidation and therefore final improvement of the mechanical response [41,87,89,90]. The studies of Rodriguez-Lence et al. [89] and Gruber et al. [41] reported significant improvements when using experimental AFP-grade APC2-AS4 material supplied by Cytec, as depicted in Figure 2.17. Figure 2.17 (i) shows preliminary results obtained in FIDAMC, highlighting the difference between the microstructure of laminates manufactured with commercial APC2-AS4 and experimental flat APC2-AS4 tapes supplied by Cytec, resulting in a higher quality laminate with less intra and interlaminar voids [89]. Figure 2.17 (ii) shows similar results reported by Gruber et al., who additionally reported an increase of 12 % in SBSS by using experimental flat material. The investigation performed by Lamontia et al. [68,90] concluded by demanding placement grade tape specification consisting of: FAW = 145 g/m², resin weight fraction 35 % ± 1 % with uniform fibre/resin distribution, surface resin content thickness equal to one filament diameter, thickness variation less than 6 % across entire tape width, with variation +0.00 mm, - 0.10 mm, and intralaminar void content < 1 %.

Figure 2.17 Use of experimental AFP-grade raw tapes. (i) Figure 2.17 (i) shows the difference between the microstructure of laminates manufactured with commercial APC2-AS4 and experimental flat APC2-AS4 tapes supplied by Cytec, resulting in a higher quality laminate with less intra and interlaminar voids [89]; and (ii) shows similar results with fewer voids and better mechanical properties [41].

▪ Placement of the first composite ply

Due to the lack of tacking of thermoplastic composites and the bad adhesion to the metallic tooling, the placement of the first ply of a laminate or structure under fabrication has been a concern for the research community.

The most extended procedure in the current state of art consists on manually placing a first ply of the composite over a modified tooling. The tool generally consists of a drilled tool connected to a vacuum port. In that way the first composite ply is fixed to the surface and the automated lamination is performed on the composite; instead on the metallic tool, as depicted by Hoang [91] in Figure 2.18, where a first ply located manually over a vacuum-assisted tooling can be observed.

Figure 2.18 Use of a vacuum-assisted tool for manual first ply positioning [91].

Different strategies can be found in the literature, which are less time-consuming than the manual location of the first ply, but are still at a research stage. Among those, there is a promising strategy based on the use of polymer films to coat metallic tools. In [52], the authors reported a good adhesion of a CF/PA6 composite on a metallic tool by coating the tool with films of the same polymer (PA6 in this case). A schematic representation of that investigation is presented in Figure 2.19

Figure 2.19 Schematic representation of the lamination of a composite tape on a metallic tool by locating a PA6 film coating on the tooling [52].

In order to improve the productivity of the process, a fast and cost-effective strategy for the placement of the first ply should be achieved.

▪ Residual stresses

Other drawback of the thermoplastic processing via AFP-ISC is that in most cases the final manufactured structure presents undesired deformations owing to residual stresses. In Ghayoor and Hoa [92] it is said that due to the fact that the cooling down process is non-isothermal and the material is anisotropic along with other parameters, the tape is subjected to thermal and mechanical stresses. The final state of the tape regarding its deformation and strains is hard to predict. This factor results in residual stresses and strains in the part that often leads to major unwanted deformations.

A possible solution was proposed by Hoa et al. [54], based on the assumption that the reason for the distortion of the plate is due to the interaction either between layers at different temperatures

or between the layers and the mandrel. They stated that the cause for the interaction was the difference in thermal expansion between the different materials. Thus, the proposed method was based on laying the composites on a heated mandrel, where the temperature of the mandrel is above the T_g of the materials. Thus, all layers should be deposited while the mandrel is above the T_g of the material. After the lamination of the structure, they proposed cooling down the entire system at the same time. In this way, there would be no significant difference in stiffness among the layers during cooling to give rise to distortion. The difference among a distorted CF/PEEK laminate and a quasi-flat laminate is shown in Figure 2.20, where a distorted CF/PEEK laminate is shown in Figure 2.20 (i) and a CF/PEEK laminated on a steel tooling heated up to 200°C is shown at Figure 2.20(ii). They also indicated that the tooling temperature should be reduced when laminating structures with more layers, since the substrate acts as a heat retainer making the substrate soft and therefore complicating the lamination of subsequent layers.

Figure 2.20 Difference of residual stresses of CF/PEEK laminates. (i) Distorted laminate and (ii) flat laminate manufactured on a steel tool heated up to 200°C [54].

According to FIDAMC experience, the residual stresses may be relieved with the use of self-heating tools up to 200 °C; but, although the problem seems to be solved for characterization laminates (around 300 × 300 mm or 500 × 500 mm), it is not completely sorted out when it comes to larger stiffened demonstrator. A considerable improvement may be achieved, but this issue still needs to be addressed.

Apart from addressing the manufacturing issue, it is also important to be able to measure and quantify the existing residual stresses. Parlevliet et al. [93] studied available experimental techniques developed to measure residual stresses or strains in thermoplastic composites for aerospace industry. In their study, the residual stresses were categorized as laminate stresses, which take place in the thick laminates and large parts. They discussed about the available techniques for measuring the residual stresses, since typical strain gauges cannot be used due to the temperature range employed in thermoplastic AFP-ISC. Methods for residual strain measurements were proposed, such as Micro Raman Spectroscopy, metallic particles embedding and subsequent X-ray diffraction or Digital Image Correlation (DIC). Also, and more linked to the present study, the use of Fibre Optic Sensors (FOS) was proposed, especially FBG sensors. In this study, not only AFP-ISC process monitoring with FBG sensors is proposed, but also the measurements of residual strains after manufacturing (experimental results in Chapter 6).

▪ **Reliable quality control system**

In the thermoset composites production in aerospace industry, ultrasonic NDT is the preferred option for ensuring the quality of the manufacturing process and therefore the quality of the manufactured part. In many cases, control coupons (travellers) are introduced in the same autoclave curing cycle which are later physical-chemically and mechanically tested for ensuring the curing cycle quality (typically determination of T_g and ILSS). As mentioned in the previous point regarding the gap of mechanical properties of ISC-manufactured part, one of the main issues affecting the ISC-manufactured structures is that in most of the cases there is a low degree of bonding Db , owing to low interdiffusion of the polymer chains and resulting in a weak physical-chemical bonding between layers. In those cases, ultrasonic NDT is not able to detect a weak bonding of layers, since there is continuity between the layers and therefore a good transmission of the ultrasonic wave. In that way, ultrasonic NDT is useful mainly for detecting porosity (interlaminar voids or intralaminar voids) or other discontinuities derived from the ISC process, but it is not enough for validating the quality of the structure.

Within this context, it is necessary to develop a reliable quality control procedure for determining the quality of the final part. It is highly recommended to define tools which allow monitoring the processing parameters and also to check the quality of the final thermoplastic parts. Due to the different nature of the materials and the processes, those procedures employed for thermosets should not be extrapolated for thermoplastic composites.

▪ **Productivity**

In order to introduce thermoplastic ISC-AFP technology in the production lines of the main aircraft manufacturers, the productivity (in terms of Kg/hour) must be increased, apart from overcoming all the other issues discussed in this sub-section.

With this purpose, there are mainly three different approaches which were also mentioned before. The two approaches consist of (a) increasing the lamination speed while performing ISC, (b) increasing the number of laid tows in each pass and (c) increasing the lamination speed over the maximum threshold and using a second consolidation step in oven, therefore abandoning the one-step consolidation.

Different research centres are concentrating their efforts on different approaches. Thus, Stokes-Griffin et al. reported studies with tape speeds of 100 mm/s (6m/min) or 200 mm/s (12 m/min) which are challenging speeds. Following a different strategy, MTorres developed a 8-tow AFP-ISC machine which is located at FIDAMC facilities in the frame of the European project OUTCOME. In recent research, FIDAMC has also investigated deposition speeds above 10 m/min by using AFP-grade raw tapes, consisting on experimental PAEK tapes supplied by Vycortex. The performed trials generated promising results, observing good final quality and suggesting that the quality of the raw tapes is crucial for optimising the process.

Whatever the followed approach, it is mandatory to increase to productivity rate of the thermoplastic AFP-ISC process in order to make the process competitive in comparison with the typical thermoset processes.

2.2 Defects in composites

One of the main objectives of this study is to understand the impact of the defects on the mechanical performance of thermoplastic composites, and perform interrelations with the existing non-destructive testing systems; since many investigations can be found about thermoset composites but a gap of knowledge was found regarding thermoplastics.

In this section, the defects which may appear in composites structures are described. Also, a review of the more relevant studies regarding the detrimental effects of the defects in the mechanical response of the composites is presented.

2.2.1 Types of defects

According to Smith [94], defects in composites can be categorized in: (i) manufacturing-induced defects which appear during the manufacturing stage and (ii) in-service defects which take place during the normal service life of the component. In this study, only manufacturing-induced defects are described.

Smith reported that the manufacturing processes have the potential to cause a wide range of defects into the final structure. Depending on the commercial sector, the manufacturing technique selected depends on the size and quality required, apart from the final cost of the product. For instance, a surfboard is typically manufactured by wet lay-up, consisting on hand lay-up of the fabric and consequently application of the resin with the help of a brush. The board is then cured at ambient temperature. On the other hand (and completely related to the present study), aerospace industry demands high quality composite parts and therefore more complex manufacturing processes are used, such as automated lamination and autoclave curing, which minimizes the defects.

Regarding the defects which may appear in aerospace production, they are intimately related to the main processing parameters employed (such as processing pressures, vacuum level or processing temperature), along with some unintended events such as a vacuum bag puncture. Some other effects are related with the automated lamination of composites, and are discussed later.

The manufacturing defects related to the manufacturing key parameters were identified by Smith [94]. Some of the most relevant are listed and described hereafter:

- **Voids (Porosity)**

The appearance of air in the final composite structure is usually denominated porosity or voids, depending on the size of the pores and the distribution. According to Birt and Smith [95], voids can be classified in discrete voids, porosity and delaminations. In this way, discrete voids consist of those voids which are big enough to be of structural significance and can also be detected individually. Porosity is used for describing a large number of microvoids which are distributed along the matrix. Delaminations can be also understood as large planar voids. Due to their relevance in the composite materials, an individual point is assigned to delaminations (next point). An example of different types of voids in composites recovered from Yang et al. [96] is given in Figure 2.21. Figure 2.21 (i-iii) shows discrete voids with different shapes existing in the plies of

laminates and Figure 2.21 (iv) shows large and planar voids which led to the formation of an interply delamination.

Also due to the relevancy of the effects of voids in composite materials and the relationship with the present study, voids and porosity are presented in more detail in next section 2.2.2.

Figure 2.21 Different types of voids: (i-iii) Voids with different shapes existing in the plies of laminates and (iv) large and planar voids parallel to the ply which led to coalescence of neighbouring voids, contributing to the formation of interply delamination [96].

▪ Delaminations

Delaminations are planar defects usually at ply boundaries and can be caused either during the manufacturing stage or during in-service. As described before, delaminations may appear from air entrapped during manufacturing, which led to an accumulation of air between layers, causing the lack of bonding between plies, and the consequent decrease of strength. Also, delaminations may appear as a result of any incident during in-service, such as an impact.

▪ Bonding defects (debonding)

During manufacture, components may be bonded together and it is possible for defects to occur in the bondline due to incorrect cure conditions for the adhesive or contamination of the surfaces to be bonded. Debonding can be typically observed in honeycomb structures.

▪ Foreign object debris

During any of the manufacturing steps, any foreign object can be left inside the composite lay-up, coming from a possible human error. Typically, release films or sections of vacuum bags may be forgotten within the laminate, which can eventually act as a material delamination.

- **Incorrect fibre or matrix volume due to insufficient resin**

Depending on the selected manufacturing technology, matrix leaking may take place, causing local variations of the contents and deviations from the material specifications. Several procedures are applied in composites manufacturing to avoid matrix leakage, such as the use of cork dams in autoclave of VBO processing or metallic frames in hot-press manufacturing.

Regarding the automated lamination of composites performed via ATL, AFP or AFP-ISC; other manufactured-induced defects which cause a structural detrimental effect in the final part may be found. Harik et al. [97] identified the most relevant AFP defects. Some of them are presented hereafter:

- **Gaps/Overlaps**

A gap appears in the lamination when there is excessive space between two adjacent tapes or tows and there is a gap between them. An overlap appears when the two adjacent tapes or tows are overlapped on the top of each other. Gaps and overlaps are mainly caused due to steering during layup or when the lamination programme has not been correctly designed. Also, gaps and overlaps can naturally occur when laminating on a complex 3D tool. An example of gaps and overlaps is given in Figure 2.22 (i).

- **Wrinkles and tow waviness**

A wrinkle is typically observed as a wavy pattern along the edge of a tow when it is steered over double-curved surfaces or following a steered path on a flat surface. Tow waviness can be also observed when an incoming tape loses the tape tension provided by the spools systems, leading to a lack of alignment in the path. Wrinkles in a steered lamination can be observed in Figure 2.22 (ii).

- **Bridging**

Bridging defects occur when a tow is not fully bonded to the substrate or to the concave surface of the tool, leaving a gap between the substrate and the new tape. Bridging is mainly caused by too much tension on the tape or lack in the consolidation temperature, which will force the tow to lift up. Bridging effects are shown in Figure 2.22 (iii).

- **Twisted tow**

A twisted tow (or tape) can be observed after the incoming tow is rolled onto itself and then compacted by the roller. This effect can be caused by errors in the tapes guiding system. Twisted tows are shown in Figure 2.22 (iv).

Figure 2.22 Typical manufactured-induced defects during AFP lamination: (i) gap and overlap, (ii) wrinkles; (iii) bridging; and (iv) twisted tow [97].

Some other AFP manufacturing defects such as pucker, boundary coverage, angle deviation, fold, wandering tow or missing tow can be consulted in the investigation performed by Harik et al.

2.2.2 Voids and porosity

As introduced by Costa et al. [98], voids and porosity are among the most common manufacturing induced defects in composites. This, along with their implication in the present study, is why a single section is dedicated for voids and porosity.

In this section, the most relevant investigations regarding voids formation mechanisms, characterization techniques and the impact on composites are compiled and described.

2.2.2.1 Void formation mechanisms

It is known that voids may appear in the composite due to several reasons such as air entrapped during lamination, volatile resin components, moisture acquired by the prepreg during the storage or air not properly evacuated during the processing cycle (deviations from the process window such as inadequate values or curing temperature or pressure) [95,99,100]. However, the formation and evolution of voids during composites processing are not the same for all manufacturing technologies, since there are different thermodynamic and rheological phenomena taking place among processes, as suggested by Mehdikhani in its very comprehensive review [101]. It is important to highlight that most of the available research papers in the literature investigated the void formation and dynamic mechanism of thermoset based composites. In next points, the relevant investigations regarding void formation mechanisms in OoA processes such as VBO, hot-press and AFP are presented.

2.2.2.1.1 Voids mechanisms in VBO

As introduced by Mehdikhani [101], the investigations regarding prepreg processing technologies, in contrast with resin infusion processes; focused on the lamination and curing/consolidation stages. This is mainly due to the fact that the material impregnation stage is typically performed by material suppliers and the investigations. However, several investigations concluded in the need of better quality prepreg material for a quality final product, as indicated in previous sections.

The exact mechanisms of void formation and growth during curing are not yet fully understood [102], but there is a general agreement on the main sources of voids, including air entrapped in the raw prepreg (intralaminar voids) or during layers deposition, volatiles emerging in the resin during curing (only thermosets) and moisture dissolved in the resin [103–105].

Most of the investigations carried out regarding the void formation in OoA methods were based on previous investigations about autoclave processing. Autoclave mainly differs from other methods in the high pressures employed during the cycle which facilitates voids removal. At the end of the last century, several void growth, void dynamics and curing models were created by Loos and Springer, and Kardos et al. [105,106], where they mainly suggested that after a void is formed, it can grow due to thermal expansion, diffusion effects, or changes in the void pressure because of pressure changes. They set out the basis for future investigations. More discussion about void modelling can be found in recent years. For instance, in 2016 some research groups were still investigating the appearance of voids in autoclave processing, such as Parscau et al., who performed some improvements in the models by considering water diffusion due to polymerization in epoxy resin and surface tension. However, they argued that the highly complex physics behind the void formation and growth is not yet completely clear. Regarding experimental studies, Shim et al. [107] found that interlaminar voids appeared in the laminates without the assistance of vacuum in the autoclave, but the highest void content was found with vacuum but without autoclave pressure. Same results were achieved by Liu et al. [108], who concluded that autoclave pressure is the key parameter for obtaining low void volume laminates. Olivier et al. [109] suggested that voids size, shape, and distribution change depending on the cycle parameters. Mehdikhani et al. [101] concluded their review on void formation in autoclave by saying that pressure may be the crucial parameter but the entire set of processing parameters (pressure, temperature and vacuum) should be optimized as a whole, rather than separately.

The interest on investigating VBO process resided mainly in reducing costs related to expensive and high energy consuming autoclave cycles. As described in previous section, the main challenge of VBO processes is being able to produce composite structures with similar quality to those obtained through autoclave processing; and this fact is directly related to low porosity levels. In contrast with autoclave, where voids are reduced by means of high pressure, VBO only counts with the assistance of combined temperature and vacuum for this task. Mehdikhani et al. [101] made a classification between different kinds of voids in VBO composites: “flow-induced” voids resulting from insufficient resin impregnation of fibres; and gas-induced voids, caused by the presence of entrapped air, moisture, or resin volatiles. They also differentiated between different voids co-existing in an unconsolidated prepreg preform, consisting on micro-voids, within dry tow cores, meso-voids within the resin-rich regions around tows, and inter-laminar voids between the plies. The mechanism of void evacuation during VBO processing corresponds to first evacuation of air through the dry material when the vacuum is connected so the

interlaminar voids may reduce. Once the system is heated up and the viscosity of the resin is reduced, the resin starts flowing to dry areas; reducing the dry regions. In a series of research publications, Centea et al. [102,110–113] performed a series of investigations focused on specific OoA-grade prepreg material, consisting of a partial impregnated material, which assisted void evacuation. In 2015, they focused on void formation and void reduction mechanisms in the before mentioned thermoset OoA-grade prepreg. They found that many voids remained in the laminate surface due to low out-of-plane permeability and that the main sources of voids were those entrapped in the interfaces of the lay-up (especially in the tool-laminate interface) and those caused by the moisture acquisition during the prepreg storage. In 2017, they concluded that the quality of the received prepreg material (special attention to low intralaminar voids was given) is of high relevancy for the quality of the manufactured part.

As mentioned, most of the investigations have been carried out with thermoset materials. However, the conclusions achieved by thermoset investigations may not be completely extrapolated to thermoplastic composite, mainly due to the following facts: (i) there is no chemical reaction taking place in thermoplastic consolidation, so there is no chemical volatiles inside the system, (ii) low moisture absorption by thermoplastic and (iii) the use of higher processing temperatures, which cause a higher void expansion due to larger thermal expansion. In this way, the main void formation and mitigation in thermoplastics are more related to the entrapped air between layers in the lay-up (interlaminar voids) and the occluded air within the raw tapes (intralaminar voids). Depending on the manufacturing method, the air needs to be evacuated (as in VBO processing) or compressed (as in hot-press or AFP-ISC). The investigations conducted by Zhang et al. [18–20,114] are of particular interest for the present work since they investigated void mitigation mechanism in VBO processing for CF/PEEK composites. In their first investigation in 2012 [114], they highlighted the large amount of intralaminar voids in the raw material (Figure 2.23). They investigated the transport and reduction of voids by applying different dwell times (2, 4 and 5 hours). They applied long vacuum times due to the high viscosity of the resin and the lack of autoclave pressure. They concluded that void compression coupled with gas diffusion are key factors for achieving low void content thermoplastic laminated by VBO processing. In 2013, they used a resin infusion void filling for determining optimal dwell time for achieving low content CF/PEEK laminates, which was established as 11 minutes. They highlighted the importance of the initial state of the voids (size, distribution and internal pressure) as governing factors in the void dynamics. In 2014 [19], two mechanisms of void extraction were investigated. First, they sealed the perimeters of the unconsolidated lay-up and air removal was limited to diffusion through the entire laminate thickness, leading to limitations. Secondly, they left the edge perimeters open and therefore air removal occurred through single layer diffusion. In-plane dimensions of the part in conjunction with the interlayer permeability were determined as crucial for the time needed for air removal. The results were validated experimentally and they concluded that single layer diffusion followed by interlayer air evacuation were key mechanism for void mitigation in thermoplastic VBO processing. This also pointed out the importance of rough surfaces in thermoplastic prepreg tapes for optimal VBO processing.

Figure 2.23 Intralaminar voids in AS4/APC2 raw tape as received [114].

2.2.2.1.2 Voids mechanisms in hot-press

In contrast with VBO, hot-press methods are characterized for the application of high pressures and the lack of vacuum assistance. Most of the systems apply the pressure through a hydraulic system acting on two hot plates. Fewer references were found regarding void mechanisms in hot-press compared to VBO.

Gu *et al.* [115,116] investigated the effect of applied pressure and absorbed moisture in thermoset laminates. They found that wet storage of the material led to higher initial void content in the prepreg. However, a decreasing exponential trend was reported for the void content versus the applied pressure, even for those prepreg which were subjected to moisture. Thus, an optimal applied pressure was declared to be enough to mitigate the voids coming from raw material with initial voids. They also reported the importance of gel temperature in thermosets. An increase in gel temperature caused higher void content values. This was explained by attributing less internal pressure on the voids at lower processing temperatures. In this way, voids growth is limited at lower temperatures and therefore voids are easier to eliminate. In a similar work, Anderson and Altan [117] investigated the combined effect of pressure and humidity on the void formation and growth. They used a model based on the Kardos model for predicting the void evolution during the process, based on the internal pressure of the voids. The model is based on when the pressure of the resin surrounding a void is higher than the internal void pressure; then the void collapses. They achieved similar results to those of Gu *et al.*, reporting a negative exponential trend of void content with the applied pressure.

Hernández *et al.* [118,119] investigated the final porosity distribution in thermoset laminates manufactured under different pressure and temperature cycles in hot-press. They observed that the resin flow is prone to occur in the fibre direction since a channel-type structure is created which enables void transport and coalescence along the fibres. This causes an inhomogeneous pressure distribution which impacts in the void volume, size and distribution. They also observed by tomographic analysis that voids cannot be treated as spheres since most of voids have rod-like shapes with a significant aspect ratio. They concluded that viscosity of the resin along the cycle is decisive to the final void content.

Regarding thermoplastic composites, Brzeski and Mitchang [120] investigated the deconsolidation effect on polypropylene reinforced composites. They summarized the main causes affecting deconsolidation in thermoplastics as: (1) decompaction of the fibre reinforcement network, (2) void expansion of the interlaminar and intralaminar voids due to thermal gas

expansion and internal void pressure, (3) void shrinkage and coalescence of smaller voids into larger voids due to surface tension and (4) thermal expansion and viscoelastic behaviour of the matrix. They developed a deconsolidation model and made a comparison with experimental results which consisted of CF/PP laminates deconsolidated in hot-press under different pressure conditions and subsequently analysed by 2D microscopy. An example of a deconsolidated sample is shown in Figure 2.24. The study concluded by saying that internal void pressure, volume energy of the void, and surface tension can reduce deconsolidation, while the fibre reinforcement network and thermal gas expansion of the voids enhances deconsolidation.

Figure 2.24 Comparison of a well consolidated CF/PP sample (left) with a deconsolidated sample by applying heat without sufficient pressure [120].

2.2.2.1.3 Voids in ISC

Due to the interest in AFP-ISC of thermoplastic composites caused by the before mentioned possibility of processing thermoplastic composites in a single step without requiring further consolidation steps, different investigations can be found regarding the void formation mechanisms during thermoplastic AFP-ISC process.

Pitchumani et al. [121] described the dominant AFP-ISC process mechanism as heat transfer (influencing the rest of phenomena), intimate contact between the incoming tow and the substrate layer (where interlaminar voids may be removed), healing as the interdiffusion of polymer molecules at the interfaces, consolidation/squeeze flow by the application of compaction force by a compaction device (where intra-laminar voids within the tows may be reduced), void growth due to thermal expansion resulting from the high processing temperatures and polymer degradation. Thus, the quality of the final part is mainly driven by the final interfacial healing (diffusion of the chains) and the final void content. They developed a void growth model taking into account all the mentioned phenomena. They concluded that an increase of the compaction roller force is related with an increase of the interfacial bonding and void content reduction. On the other hand, an increase of the transferred heat may promote the void growth and increase the final void content. They suggested that a fast cooling performed immediately after consolidation may induce a stop of the void growth and reduce the final void content.

Tierney and Gillespie [122] developed a heat transfer model coupled with a microscopic void dynamics model, determining the void distribution along the thickness. They reported a gradient of voids through-the-thickness, showing more voids in the bottom and upper part of the specimen and less voids in the centre region. This was attributed to deconsolidation taking place in some of the layers. In this way, they reported that middle layers were consolidated a higher number of times compared to the top layers and therefore those multiple passes may facilitate the reduction

of voids. In the case of the bottom layers, they suggested the accumulation of heat by the substrate (or by the tool acting as a heat sink) may cause deconsolidation and void growth. Lamontia et al. [88,90] reported that the key manufacturing parameters such as temperature, compaction force and lamination speed govern the void control in the process. As commented before, they placed emphasis in the quality of the raw material since there is a very narrow window time for mitigating the voids. They claimed that flat tapes with low intralaminar void content for facilitating low void content in the manufactured part.

Simacek et al. [123] developed a non-local void filling model which was able to predict the final porosity. They reported that the final void content is highly influenced by the initial void distribution in the raw tape. Additionally, they suggested that volatiles may get entrapped during processing between two layers due to the surface roughness. They concluded that the applied force is expected to collapse and consolidate the voids. In an experimental investigation, Comer et al. [48] compared the porosity of CF/PEEK samples produced through autoclave and AFP-ISC. They suggested that interlaminar voids remain entrapped between the layers during lamination and that void mitigation can be achieved by reducing the tapes roughness and the processing parameters. They also discussed the possibility of an effect called void rebound, which may consist of the thermal expansion of voids after the pass of the compaction roller, if the resin is still above the melting point. This effect may take place when the substrate material has a significant thickness, since it may act as a heat retainer which slows down the cooling stage.

2.2.2.2 Voids characterization methods

Due to the detrimental effect of the voids over the mechanical behaviour of composite laminates, the evaluation of void content in composite parts is usually required by aerospace production procedures, especially for structural components in order to guarantee mechanical performance and safety standards, as introduced by Di Landro et al. [124]. Depending on the application, void content as low as 1 %, 2 % or even up to 5 % are usually considered acceptable for structural components [125]. While typical aerospace procedures focus on evaluating average void content, it is more and more frequent to find research investigations who suggest that the void size and distribution are factors as important as the void volume fraction. In this sub-section, different techniques that can be employed for voids characterization are presented, along with a discussion about their capabilities and limitations.

2.2.2.2.1 Archimedes method (Density method)

The determination of the void volume fraction is possible by using the Archimedes method (also called densities method). As described in different scientific research articles [101,126], the procedure is based on calculating the density of a given composite coupon by measuring their weight in the air and within the water applying Archimedes' principle. The coupon volume is then calculated following equation (4.2), where V_c is the volume of the coupon, W_c and W_c^{water} is the weight of the composite in the air and submerged; respectively, and ρ_{water} is the density of the distilled water.

$$V_c = \frac{W_c - W_c^{water}}{\rho_{water}} \quad (2.1)$$

The density of the coupon can be then calculated by knowing the volume of the coupon and following equation (2.2), where ρ_c is the density of the composite coupon.

$$\rho_c = \frac{W_c}{V_c} \quad (2.2)$$

The void content can be then estimated by following equation (2.3), where V_v is the void volume fraction, W_r and W_f are the matrix and fibre mass fraction; respectively, and ρ_m and ρ_f are the density of the matrix and the fibres, respectively. It is important to note that material properties as the constituents (matrix and fibre) fractions and densities should be known for being able of applying Archimedes methods.

$$V_v = 100 - \rho_c \left(\frac{W_r}{\rho_r} + \frac{W_f}{\rho_f} \right) \quad (2.3)$$

Archimedes method is a destructive test since coupons should be extracted, Some of the advantages reside in the fact that it is a low cost and fast screening method which allows the density and the void volume of a composite coupon within relative short times. The main disadvantage resides in the low accuracy of the method. Thus, Judd and Wright [127] reported a maximum accuracy of $\pm 0.5 \%$. Special attention should be given in the weight measurements performed in the water, since the undesired presence of bubbles may take place in the coupon surface. It has been declared that the method may provide negative results, which are physically impossible. The method can only provide information about the average value of the void content. Therefore for further information about void distribution and size, other methods should be applied.

2.2.2.2.2 Acid digestion

Acid digestion is the most extended method in the aerospace industry for void content assessment and it can be found in most of the research articles that investigate about voids and porosity [98,100,124,126,128–130]. For the void content assessment, the weight and density of the coupon is first determined. After that, the resin of the coupon is digested in sulphuric acid and the product is neutralized by means of hydrogen peroxide. The fibres are then recovered, washed and weighted. After this procedure, void, matrix and fibre contents can be calculated by following next equations (2.4) to (2.8), where m_o and m_f are the initial and final mass of the sample, V_r , V_f V_v are the final volume fraction of the resin and the fibre, and ρ_m , ρ_f ρ_c , W_r , W_f and V_v were previously described.

$$W_f = 100 \left(\frac{m_f}{m_o} \right) \quad (2.4)$$

$$V_f = W_f \left(\frac{\rho_c}{\rho_f} \right) \quad (2.5)$$

$$W_r = 100 - W_f \quad (2.6)$$

$$V_r = (100 - W_f) \frac{\rho_c}{\rho_r} \quad (2.7)$$

$$V_v = 100 - V_f - V_r \quad (2.8)$$

Due to the good accuracy of the method, some studies that compare different characterization methods, establish the values provided by acid digestion as the reference values [124]. Time and cost consumptions are considered the main drawbacks of this technique, along with the limitation of only providing average void content information.

2.2.2.2.3 2D optical microscopy

Void characterization is possible by means of 2D optical microscopy. With this method, not only average void content values can be assessed, but also void size, shape and distribution. This is why optical microscopy was selected as the void characterization technique in many research articles that can be found in the literature [100,120,124,126,128,131–133]. For void characterization via optical microscopy, a coupon should be extracted from a composite part and subjected to a surface preparation procedure, typically consisting on polishing one cross-section. The quality of the polishing will determine the definition of the captured micrographs. Once the coupon is prepared, a set of micrographs is taken by means of an optical microscope to the desired magnification. The acquired images are then processed with image analysis software. Typically, micrographs are taken in a grey scale and a colour threshold is applied for highlighting the voids. As described by Daniel et al. [134], optical micrographs typically contain three major intensities: bright (fiber), grayish (matrix), and dark (porosity). The images may be thresholded into a bi-level image for porosity analysis. Thresholded pixels for porosity are counted over the entire image, and then the void volume ratio is determined as the ratio of the number of thresholded pixels and the size of the image. However, they also reported that sometimes it was difficult to set the threshold from the histogram, especially when the image was not clearly trimodal. In that case, they suggested that the value may be determined by comparing the threshold image and the original micrograph, since it is easy to distinguish between porosity and matrix. Since this technique was somewhat subjective, they recommended the use of skilled personnel. An example of the grey-scale and its different phases reported by Santulli et al. [131] is given in Figure 2.24 (i). In that case, it was reported that typical grey-scales have four different phases corresponding to fibre, matrix and porosity, but also to fibre ends. In Figure 2.24 (ii), an example of image thresholding for porosity determination is shown.

Figure 2.25 2D optical microscopy analysis. (i) Different phases of the grey-scale of a micrograph and (ii) image thresholding for porosity determination [131].

As a clear advantage of the microscopy technique in contrast with Archimedes and acid digestion methods, is that void size, shape and distribution can be analysed. This is the case of different studies, such as Liu et al. [128], who investigated the void distribution and shape of CF/Epoxy laminates manufactured under different autoclave pressures. Bodaghi et al. [132] investigated void size, shape and distribution of composites processed by high pressure RTM by using 2D optical microscope analysis. On the other hand, there is an obvious disadvantage, consisting on the limitation to a single cross-section of the coupon. As concluded by Mehdikhani [101], the results provided by microscopy are dramatically section-biased, which means that characterization of voids in different cross-sections does not necessarily give the same results. To correct this problem, Bodaghi et al. [132] proposed the analysis of at least 20-25 images for obtaining an acceptable accuracy. Other authors suggested that optical microscopy may be used for analysing local regions, while other technique (such as acid digestion) should be used for determining average void fractions.

2.2.2.2.4 Ultrasonic testing

Ultrasonic testing (UT) is one of the most extended non-destructive tests used for the quality control of the composite manufactured parts in the aerospace industry. It is commonly employed after a structure is manufactured for evaluating not only the existence of porosity, but also other manufacturing defects, as those described in previous section 2.2.1.

Regarding the use of UT for porosity evaluation, the attenuation of the ultrasonic wave is dependent on the void fraction and the fibre volume fraction, as introduced by Birt and Smith [95]. From the end of the last century, many investigations have studied the possibility of evaluating the porosity within composite materials by means of UT. However, up to the date, only relations of void content with the ultrasonic attenuation can be found in the literature regarding thermoset composites.

Stone and Clarke in 1975 [99] were the first authors that reported the relationship curves of the ultrasonic absorption coefficient (at different inspection frequencies) as a function of the void

content. A bi-linear behaviour was reported for UD laminates with different values of void content in the range of 0 to 4 %; which is shown in Figure 2.26 (i). They described that voids were mainly volatile induces up to a value of porosity of 1.5%, but entrapped air forming interlaminar voids predominated beyond 1.5%, causing higher ultrasonic attenuation. A non-linear relation was reported by Daniel et al. in 1992 [134], who fabricated graphite/Epoxy laminates with a void range from 1 to 11 % by controlling the vacuum level during the curing process. The non-linear relation is shown in Figure 2.26 (ii). In 1994, Almeida et al. [135] fabricated a set of CF/Epoxy laminates in a range of porosity between 1 and 6% by limiting the autoclave pressure with metallic frames. They assessed the void content following matrix digestion and investigated the impact of the porosity on UT and ILSS. In 2006, Liu et al. [108] investigated the ultrasonic response of a set of CF/Epoxy laminates manufactured under different autoclave pressures, resulting in a porosity range of 0 to 3.5 %. They reported a linear behaviour of the ultrasonic attenuation coefficient with the void content, which is shown in Figure 2.26 (iii).

The investigations above mentioned are some of the most relevant studies regarding void content assessment by UT. Other investigations reported theoretical expressions which related the ultrasonic attenuation to the void content, including the radius of the pores in the equation [136]. However, those expressions are not accurate since the assumption of spherical voids is not valid [101]. Other studies reported that the variability of the ultrasonic attenuation with the porosity is not only related to the average void content, but also to the morphology of the voids [137].

Overall, it is accepted that the porosity within composite structures has an effect on the ultrasonic attenuation coefficient; therefore more porosity is related to more ultrasonic attenuation. However, there is no consensus regarding the relationship of ultrasonic attenuation coefficient and void content. This is mainly due to the facts that every material has a different behaviour and that voids morphology is also a key factor impacting on that relationship. However, there is no doubt about the capacity of the UT to detect existing porosity in composite structures; and the important role of the UT for establishing acceptance criteria in aerospace production; as will be described in next section 2.2.2.3.

Figure 2.26 Ultrasonic attenuation coefficient as a function of void content. (i) bi-linear relation thermoset UD laminates inspected at different frequencies, reported by Stone and Clarke [99]; (ii) non-linear relation for CF/Epoxy laminates reported by Daniel et al. [134]; and (iii) linear relation reported by Liu et al. [108].

2.2.2.2.5 X-Ray CT

In the last decade, 3D X-ray computed tomography or microtomography (CT or micro-CT) has emerged as an attractive technique for void characterization and therefore many research articles can be found regarding the investigation of porosity by means of micro-CT [118,119,124,126,138–151]. As described by de Souza et al. [149], the method consists on scanning cross sections of specimens by mapping with an x-ray beam. The x-ray source irradiates an object in thin slices and the transmitted intensities are detected using a detector device. The transmitted intensity depends on the energy attenuation caused by the specimen, driven by the density and atomic number, as depicted in equations (2.9) and (2.10), where I is the intensity of the transmitted radiation, I_0 is the initial intensity, t is the specimen thickness, μ is the attenuation coefficient dependent on the density (ρ) and atomic number (Z) of the scanned material; and E is the energy, $\alpha(E)$ and $\beta(E)$ are functions of x-ray energy. The set of images produced during the scanning allows reconstructing a 3D volume which is basically a density mapping obtained by means of different coefficients of absorption of the component materials of the sample (matrix, fibres and voids).

$$I = I_0 e^{-\mu t} \quad (2.9)$$

$$\mu(E) = \rho Z^{3.8} \alpha(E) + \rho \beta(E) \quad (2.10)$$

A schematic representation of the micro-CT scanning and the 3D volume reconstruction is shown in Figure 2.27 (i).

Figure 2.27 3D X-Ray microtomography. (i) schematic representation of the micro-CT scanning and 3D volume reconstruction [149]; and (ii) micro-CT scanning set-up [150].

As described by Nikishkov et al. [139], after the volume reconstruction, the scanned specimen is represented as a set of voxels (3D pixels) where each voxel has the intensity (grey value) related to the density. Different grey scales may be found if the employed software was 8-bit (255 grey tones) or 16-bit (65535 grey tones). The distinction of the porosity in a sample is typically performed by applying thresholds in the grey scale, in a similar procedure to the one applied in microscopy. Tretiak and Smith [151] investigated the differences among results given by different applied thresholds. Figure 2.28 shows a typical grey-scale representation of a scanned specimen, with different zones corresponding to composite, resin and air. The analysis of the reconstructed volume allows performing an extensive void characterisation. Most of the investigations showed that void size, shape, aspect ratio and distribution, apart from average void fraction can be analysed by means of micro-CT.

Figure 2.28 Grey-level histogram showing the ‘air’, ‘resin’, and ‘composite’ of a scanned specimen [151].

Regarding the experimental investigations, Kastner et al. [138] reported in 2010 the micro-CT void characterization of several CF/Epoxy samples in a porosity range of 0-10 %, highlighting the importance of proper threshold selection. They reported a good match between porosity values assessed by micro-CT and acid digestion and ultrasonic testing. In 2011, Hernández et al. [118,119] initiated a series of investigations where they studied the existing porosity in different CF/Epoxy samples (0 to 3 % of porosity). They reported void distribution along the thickness, voids size and shape. Figure 2.29 (i) shows the void spatial distribution of the laminates manufactured in Hernández et al. [118]. Regarding the shape, they found that voids were rod-like, as depicted in Figure 2.29 (ii). In 2012, Little et al. [126] reported a comparison between void characterization techniques including micro-CT, Archimedes and optical microscopy. They suggested that, having no section-bias and less location-bias error, micro-CT is the most accurate and reliable technique for void content assessment, which also offers the possibility of analysing void shape, size and distribution. In 2013, micro-CT scanning of a CF/PEEK raw tape was performed by Zhang et al. [18]. They found that 2D optical microscopy gave higher void content than micro-CT. This was attributed to the higher resolution of microscopy. With the same reasoning, the number of detected small voids increased by improving the resolution of micro-CT [101]. Madra et al. [140] conducted in 2014 a micro-CT analysis of voids in three scales: composite bulk, a textile layer, and a single yarn; and the development of a set of image processing algorithms and procedures. In 2018, Ramesh et al. [148] performed the characterization of the manufacturing-induced defects in the curved part mast of a sailing boat, evaluating porosity and wrinkles. Regarding different applications, Sket et al. [142] employed tomographic scanning for the investigation of matrix cracking growth. Same authors investigated the IPSS damage evolution within a composite by means of micro-CT. Stamopoulos et al. [150] proposed the use of micro-CT during the composite manufacturing stage, in order to develop smart manufacturing processes that are able to predict the mechanical properties of a manufactured part by using an Artificial Neuronal Network.

Figure 2.29 3D micro-CT of CF/Epoxy laminates reported by Hernández et al. [118]: (i) void spatial distribution of the manufactured laminates and (ii) Rod-like shape of voids.

To conclude, there is a general agreement that micro-CT is a powerful technique for void characterization in composite materials. Its main advantages consist of being able to characterize most of the voids characteristics (content, size, shape and distribution) and a high accuracy. There are more and more studies suggesting that not only void content has a final impact on the composites performance, but also the location and size of individual defects at critical locations. This way, micro-CT appears as an optimal alternative. However, the main drawback of the technique is the sample size. The spatial resolution is inversely proportional to the sample size, meaning that high-resolution analysis (several micrometres/pixel) requires very small samples (several millimetres), as concluded by Medihkahni [101]. Other drawbacks should be noted, as the high equipment and maintenance costs, along with the need of high skilled personnel.

2.2.2.3 Effect of voids on composite materials

Voids and porosity are the most investigated defects in composite materials due to their detrimental effect on the mechanical properties and final structural behaviour. As depicted by Mehdikhani et al. [101] in their thorough review, there is a large number of investigations regarding the effect of voids on the mechanical properties of composite materials. Again in this case, thermoset composites are the most investigated; and only few studies were found regarding thermoplastic composites. Overall, there is a general agreement that voids and porosity cause a drop of the mechanical properties of composites. However, voids do not affect all properties to the same extent; and it has been observed that matrix-dominated properties in particular (such as ILSS or IPSS) are strongly dependent on the presence of porosity in comparison to those properties which are driven by the fibre (such as tensile). In this study, a brief but comprehensive

review of the effect of voids on composites is presented, due to the large number of available investigations.

The earliest investigations date from the 70's, when Stone and Clarke [99] investigated the effect of voids in UD thermoset laminates, showing a linear decrease of ILSS with the void content. In 1977, Hancox et al. [152] investigated the longitudinal shear modulus of cylindrical UD CF/Epoxy specimens by means of torsion test, finding a decrease of 10 % of the property each 1 % of void content above 1.5 %; meaning a drop of 30 % of the property at 5 % of void content. In 1987, Tang et al. presented a study based on the investigation of void content, compression and short-beam shear (SBS) in Fiberite T300/976 and T300/934 UD prepreg laminates manufactured under different applied pressures. They reported that matrix dominated properties such as compression and SBS are especially affected by the cure pressure and therefore by the void content. It was observed that those properties were not significantly affected until 3-4 % of void content; but a fast decrease was observed after those values. They suggested that it is not imperative to eliminate all voids from the laminates since it is sufficient to keep the void content below a certain limit. In 1992, Bowels and Frimpong [153] fabricated 38 graphite/Epoxy laminates by using different autoclave pressures and performed an evaluation of the void content and mechanical properties (ILSS and flexure). They found a decrease trend of ILSS. The data was fitted by using a power function. A predictive analytical equation for ILSS values as a function of the void content was given. They also concluded that porosity distribution should be considered as a significant factor. De Almeida et al. [135] proposed in 1994 a methodology for assessing critical values at which the mechanical properties begin falling. In this way, they manufactured several CF/Epoxy laminates in a range of porosity from 1 % to 6 % and performed ultrasonic testing, void content analysis and ILSS tests. They correlated all the results and represented ILSS values as a function of the ultrasonic attenuation coefficient following the Mar-Lin criterion. They observed that ILSS values remained constant at low attenuation values (related to low void content), while ILSS started falling down at a certain level; considered the critical value. They concluded that their results may be of a great value for engineering applications and development of acceptance criteria. In a similar study, Jeong et al. [100] produced in 1996 a series of laminates with different materials and lay-ups, consisting on CF/Epoxy and graphite/Polyimide. The reported void content was in the range 0.2 – 11.2 %; and they also investigated the ultrasonic response and ILSS. Void content vs. ILSS correlation was performed and they found similar results to those reported by De Almeida et al., since it was observed that ILSS values remained constant up to a certain void content value, where the property decrease. They concluded by suggesting that similar studies can be applied for quality control of composites regarding the determination of allowable ultrasonic attenuation. In 1996, Wisnom et al. [154] performed an study on failure mode of Glass/Epoxy laminates. In that case, artificial defects consisting on PTFE monofilaments were inserted into the laminate to simulate the voids. They reported a reduction in ILSS of between 8 and 31% for discrete voids from 0.28 to 3.0 mm long. Costa et al. [98] reported in 2001 an investigation regarding the effect of porosity in CF/Epoxy and CF/Bismaleimide laminates produced by different applied pressures in a range from 1.1 % to 5.6 %. They analysed ultrasonic NDT, void content and ILSS, reporting critical void content values around 1 % where the mechanical properties started decreasing. They also proposed analytical equations for predicting fracture stress as a function of the ultrasonic absorption coefficient (related to void content). In a series of two scientific publications, Liu et al. [128] investigated the effect of the curing cycle of CF/Epoxy laminates in final porosity and mechanical properties.

They reported linear decreasing trends of tensile and ILSS properties. They found a decreasing trend of 2% tensile strength, 6% in tensile strength and 9 % in ILSS for each 1% increase in void content. Thus, they concluded that matrix-dominated properties are much more sensitive to porosity than fibre-dominated properties. Olivier et al. [155] reported in 2007 a methodology similar to Costa et al. for elaborating acceptance criteria for composites. Thus, they manufactured a set of CF/Epoxy laminates with different porosities and performed ultrasonic NDT, 2D microscopy and a set of mechanical tests, including ILSS, tensile, transverse tensile and IPSS. They performed an inter-relation of all tests and reported values of critical void content. It was observed that different materials may have different behaviour and therefore different void content critical values. In more recent investigations, Hernández et al. [118,119] manufactured a set of CF/Epoxy laminates by means of OoA and related the void content assessed by micro-CT with ILSS and compression after impact (CAI). They represented the mechanical values as a function of the void volume fraction, but they also pointed out the importance of void distribution, shape and size. Di Landro et al. also evaluated the drop of ILSS with void content in CF/Epoxy laminates manufactured under different pressure conditions. They evaluated the void content by means of different characterization methods, discussing the advantages and disadvantages of each method.

As may be observed, most of the available literature is limited to the investigation of thermoset composites. Regarding thermoplastic composites, fewer investigations can be found. In that way, Bowles and Frimpong [153] reported the influence of voids on ILSS in CF/polyimide laminates. Hou et al. [156] reported in 1997 the decreasing evolution of flexural strength and modulus with porosity in CF/PEI laminates. Fujihara et al. [28] investigated in 2004 the degradation effects of CF/PEEK specimens subjected to long consolidation times up to 60 minutes, where degradation effects were observed. A drop of flexural strength was observed in laminates processed at a high temperature of 440 °C during 60 minutes which caused degradation of the matrix in form of voids. Hagstrand et al. evaluated in 2005 the influence of void content on the flexural performance on UD GF/Polypropylene composites. They reported a drop of tensile and flexural strength and modulus with the void content. The negative reported impact consisted of a decrease of 1.5 % of flexural strength and modulus for each 1 % of voids up to a level of 14 % of porosity. However, a positive effect on the beam stiffness was reported with increasing void content. This consisted of one of the rare cases where positive effects of the porosity were declared. Comer et al. [48] compared in 2015 the mechanical behaviour of CF/PEEK laminates manufactured via laser-assisted AFP-ISC and autoclave. They found ISC laminates to have more voids and therefore lower mechanical properties. In 2016, Patou et al. [130] investigated the influence of void content on the mechanical behaviour of CF/PPS laminates. They reported the relation of flexure, tensile, transverse tensile and ILSS with void content, revealing that flexure strength and ILSS are more affected by the presence of voids than tensile and transverse tensile stresses. It was reported a drop of properties at 5 % of void content of around 35 % for flexure strength, 18 % for ILSS and less than 10 % for both tensile strengths. All of the investigated modulus experienced a drop of less than 10 % for 5 % of void content.

As described by Patou et al. [130], most of the available studies regarding process parameters, consolidation cycles and voids having an effect on the mechanical performance of composites are focused on thermoset composites. In this way, more research is required in thermoplastic composites, especially when innovative and promising manufacturing techniques are appearing, which are very attractive for industrial applications.

2.3 Process monitoring with FBG sensors

In the previous sub-section, the manufacturing-induced defects and their relation to the manufacturing parameters was described. In the recent years, more and more publications can be found regarding the so-called industry 4.0 which is aiming to incorporate smart manufacturing processes and therefore being a paradigm shift in the industry sector, including the aerospace industry. In this way, a smart manufacturing process should be understood as a process where the key manufacturing parameters are real-time monitored by a sensing network. The analysis of all the monitored data sets by means of big data and machine learning could lead to ultimate prevention of defect formation if all the manufacturing parameters are interrelated, allowing detecting and fixing a hypothetical deviation from optimal parameters. In an ideal case, optimized in-situ process monitoring could lead to reduction of NDT activities, involving a considerable reduction of operational time and costs.

Even though the sensing and processing technology is not yet ready for performing such huge data monitoring and processing, many investigations have been carried out during the last decades relative to process monitoring. Those investigations were not always motivated by defect prevention and process optimization by means of controlling manufacturing parameters, but by reducing processing costs and times. Fernando et al. [157] described the need for robust process monitoring techniques in composites production. They highlighted the main reasons for developing process monitoring in composites, which mainly consisted of resin curing kinetics, residual stresses which may appear in the composites after consolidation and thermal history of the process, which governs the main phenomena.

In the literature, many process monitoring investigations can be found which integrated different sensors in different manufacturing processes. Earlier investigations date from the 90s, when Lee et al. [158] incorporated dielectric sensors in a RTM process in order to reduce the process cost and time without losing end-product quality. Dielectric sensors (also called DEA, from Dielectric analysis) have been used since then in many studies which were mainly focused in the resin curing monitoring (degree of curing in thermoset composites) and the detection of resin flow front in RTM and vacuum assisted resin transfer moulding (VARTM) processes [159–162]. Other authors employed direct current sensors with the same goal consisting of resin flow front monitoring [163–165]. In the recent years, process monitoring has also been performed by means of IR cameras, in order to detect manufacturing defects [166] or to monitor the thermal history of open-mould processes [167].

However, among the employed sensors in process monitoring research, optical sensors have been one of the most investigated. This has been mainly due to the advantages that optical fibres may offer, which are listed below according to the reviewed literature [157,168–170].

- Optical fibres are immune to electromagnetic interference and therefore can be used in areas where electrical-based devices cannot operate without expensive shielding and protection.
- Integration or embedding of optical fibre sensors is relatively simple in composites because of their relative small dimensions and uniform cross-section. In the same way, it is also relatively straightforward to integrate optical fibres in the preforms and manufacturing processes.

- Coating or encapsulation techniques can be applied for providing protection to the sensors in order to be used in extreme and hostile environments.
- The mechanical properties of the composites are not dramatically affected when embedding optical sensors.
- Optical sensors can be multiplexed, meaning that similar or different sensors can be located along the same optical fibre
- Optical sensors offer a wide range of applications in structural health monitoring (SHM) and process monitoring (optical fibre sensors can be used for monitoring strain, temperature, vibration, pressure, torque, moisture ingress and acoustic emission)
- Optical sensors can be separated at a significant distance from the interrogation equipment due to the low transmission losses, allowing remote monitoring.

There are also some disadvantages which should be considered when choosing the sensor system for process-monitoring. The disadvantages or concerning issues are listed below:

- Brittleness of optical fibres. Optical fibres are not robust and therefore the survivability of the embedded sensors needs to be studied for each case. In most of the investigations, special care is given to the egress points, where the optical fibre may suffer shear loads and it is prone to be broken. Other studies have investigated coating, jacketing or encapsulation solutions.
- The efficiency of the measured magnitude transfer to the sensor. A good adhesion or bonding of the sensor to the host structure must be guaranteed in order to have a good transfer of the magnitude to the sensor and therefore a reliable measurement.
- The interpretation of the monitored data is not simple. The output provided by the sensor can be impacted by other unexpected magnitudes such as strains, temperatures, humidity, vibrations, etc.
- Long-term stability and reliability of the sensor and interrogation unit.

Within the family of optical fibre sensors, Fibre Bragg Grating (FBG) are the most investigated; and this is why an individual section is dedicated to them, apart from their application in the present study. However, the application of other optical sensors has also been investigated, such as optical fibre interferometer (Fabry-Pérot, Mach-Zehnder, Michelson, Sagnac), or optical reflectometry, where distributed sensors or optical time-domain reflectometer (OTDR) technology is grouped.

The motivation for the research of optical sensors has been mainly focused on SHM, where a sensing system evaluates the structural health of an in-service structure in order to predict damage. The Aerospace Materials and Production Department of the School of Aeronautical and Space Engineering of the UPM has been investigating the application of optical sensors in SHM of aerospace structures for more than two decades, in a series of doctoral theses, research articles and industrial projects [171–185]. In this way, key achievements have been addressed, such as the use of FBGs as strain sensors [171], the application of FBG at cryogenic temperatures [172], SHM of large aerostructures with optical and PZT sensors [176], including distributed OTDR sensing [177]. Figure 2.30 and Figure 2.31 show examples of the research of optical fibres for SHM applications performed in the Aerospace Materials and Production Department of the UPM.

Figure 2.30 (i) shows a mechanical test of a wind blade which is monitored by optical sensors. Figure 2.30 (ii) shows the mechanical test of a demonstrator stiffened panel which is monitored by distributed optical sensors [173,177].

Figure 2.30 Application of optical fibre sensors in SHM: (i) Mechanical test of a monitored wind blade and (ii) mechanical test of an aerospace stiffened panel monitored with distributed optical sensors

Figure 2.31 Distributed strain measurements with optical sensors (OTDR) for SHM of aerospace composite structures [176].

As concluded by Güemes et al. [184,186], distributed optical sensing (OTDR) has a huge potential due to its advantages, due to the capability of obtaining strain or temperature measurements all along the fibres. The distributed sensing technique is very useful for large structural tests; instead of bonding a large number of sensors, this can be substituted by bonding one or several plain fibres which will cover the whole length. However, there are also several drawbacks to address, such as the low frequency data acquisition and long times for data processing. In this study, real-time monitoring of thermoplastic AFP-ISC process is proposed, where the phenomena of interest take place in few seconds (or even milliseconds). For this reason, along with the other capabilities offers by the FBG sensors, resulted in the selection of FBG sensors for the investigations performed in this study.

2.3.1 FBG sensors

As aforementioned, FBG sensors have been also widely investigated for both SHM and process monitoring applications. This section is completely dedicated to the physical principles and applications of FBG optical sensors.

2.3.1.1 General features and physical principle

The definition of FBG sensors and the physical principles are well known since it has been widely depicted in the literature [5,93,184,187–193]. FBGs sensors are obtained by creating periodic variations in the refractive index of the core of an optical fibre. This is typically performed by transversally exposing the core of a photosensitive optical fibre to an intense interference pattern of ultraviolet light. Two methods are commonly followed, consisting of the holographic method [194] or a phase mask method [195]. Figure 2.32 shows an example of a FBG written by the phase mask method depicted by Allil et al.[188]. Even though there are multiple grating types (uniform, chirped, long period, apodized, etc.), uniform FBG sensors are the only sensors covered in this literature review.

Figure 2.32 FBG sensor being inscribed by a laser beam through a phase mask [188].

When the light passes through the grating, the grating itself acts as a dichroic mirror, causing the reflection of a particular wavelength; called the Bragg wavelength, which is amplified. The Bragg wavelength is driven by equation (2.11), where λ_B is the Bragg wavelength, n_{eff} is the effective refractive index of the fibre and Λ is the period of the grating.

$$\lambda_B = 2n_{eff}\Lambda \quad (2.11)$$

Figure 2.33 shows the FBG's principle of operation, depicted by Di Sante et al. [196], where the spectrum of the incident light, reflected light and transmitted light can be observed.

Figure 2.33 Fibre Bragg Grating's principle of operation [196].

A relative change in the Bragg wavelength may be caused by shifts in several magnitudes, such as strain, temperature, vibrations, pressure, etc. However, most investigations (including the present study) are mainly focused on strain and temperature monitoring. In this way, when strain is induced into an FBG, the relative change in the Bragg wavelength is related to the strain shift by equation (2.12), where ε is the longitudinal strain on the FBG and ρ_e is the effective photo-elastic constant of the fibre core material. ρ_e is in turn defined by expression (2.13), where p_{ij} are the silica photo-elastic tensor components and ν is the Poisson's ratio [190].

$$\frac{\Delta\lambda_B}{\lambda_B} = (1 - \rho_e)\varepsilon \quad (2.12)$$

$$\rho_e = \frac{n_{eff}^2}{2} [p_{12} - \nu(p_{11} + p_{12})] \quad (2.13)$$

In the case of thermal perturbations, the wavelength shift will be governed by expression (2.14), where α is the thermal expansion of the fibre, ξ is the thermos-optical coefficient of the fibre and ΔT is the external temperature change.

$$\frac{\Delta\lambda_B}{\lambda_B} = (\alpha + \xi)\Delta T \quad (2.14)$$

If a FBG sensor is simultaneously affected by external changes in the longitudinal strain and the thermal field, the Bragg wavelength will be then shifted by the combined effect of strain and temperature, in according to (2.15), which is typically shortened in the literature as depicted in (2.16), where κ_ε is commonly defined as the strain sensitivity coefficient and κ_T is the temperature sensitivity coefficient. Typical values found in the literature regarding κ_ε and κ_T of FBG sensors centred in a wavelength of 1550 nm are in the range of 1.0-1.4 pm/ $\mu\varepsilon$ for the strain sensitivity and 9-13 pm/ $^\circ\text{C}$ for the temperature sensitivity. However, those values may vary depending on the hosting material nature and the quality of the strain and temperature transfer from the host material to the sensor.

$$\frac{\Delta\lambda_B}{\lambda_B} = (1 - \rho_e)\varepsilon + (\alpha + \xi)\Delta T \quad (2.15)$$

$$\frac{\Delta\lambda_B}{\lambda_B} = \kappa_\varepsilon \cdot \varepsilon + \kappa_T \cdot \Delta T \quad (2.16)$$

2.3.1.2 Discrimination of strain and temperature

In many occasions, FBG sensors (both embedded in the composite or surface-mounted) are affected by the simultaneous effect of temperature and strain shift. This has been a major subject of research when it comes to FBG sensing techniques. As seen in the previous sub-section, the Bragg wavelength can be affected by both magnitudes, but is a complex problem to discriminate the contribution of each magnitude. Diverse techniques can be found in the literature, mainly based on monitoring the temperature with a different measurement device (or even another FBG sensor after applying some modifications) and perform the so-called temperature compensation or using FBG sensors with significantly different sensitivity coefficients. The two most widely applied techniques are listed and explained below.

- **Use of FBG sensors with different sensitivity coefficients**

One of the techniques which can be easily found in the literature is based on the use of sensors (typically two) with different strain and/or sensitivity coefficient, which will eventually result when affected by temperature and strain changes in an equation system of two equations with two unknowns. The equation system may be found in different ways in the literature; but in general it is expressed as detailed in expression (2.17), where $\Delta\lambda_{Bi}$ correspond to the wavelength shift of each i sensor, $\kappa_{i\varepsilon}$ and κ_{iT} are the strain and temperature sensitivity coefficients of each i sensor; and $\Delta\varepsilon$ and ΔT are the strain and temperature variations.

$$\begin{pmatrix} \Delta\lambda_{B1} \\ \Delta\lambda_{B2} \end{pmatrix} = \begin{pmatrix} \kappa_{1\varepsilon} & \kappa_{1T} \\ \kappa_{2\varepsilon} & \kappa_{2T} \end{pmatrix} \cdot \begin{pmatrix} \Delta\varepsilon \\ \Delta T \end{pmatrix} \quad (2.17)$$

Several sensor configurations may be found in the literature for the application of this technique. For instance, Patrick et al. [197] reported in an early investigation the combined use of an uniform FBG sensor and a long period grating (LPG) inscribed in the same optical fibre. In this way, they created a hybrid FBG/LPG sensor characterised by having different sensitivity coefficients and therefore it made possible to solve previous expression (2.17) and discriminate strain and temperature shift. Same procedure based on the use of FBG/LPG hybrid sensor was followed by other authors such as Yang et al. [198]. A schematic view of the hybrid sensor construction proposed by Patrick et al. is shown in Figure 2.34 (i).

Other way of achieving different sensitivity coefficient resided in the use of two FBG sensors inscribed in different types of optical fibres which are spliced together. The different nature and chemical content of the different optical fibres cause different sensitivity coefficients. This methodology can be found in investigations performed by Guan et al. [199], Cavaleiro et al. [200] and Yoon et al. [201]. The use of spliced sensors inscribed in optical fibres with different material and later spliced is shown in Figure 2.34 (ii).

Figure 2.34 Sensor heads with different strain and temperature sensitivity coefficients: (i) Hybrid FBG and LPG sensor [197]; and (ii) FBG sensors inscribed in optical fibres of different material which were spliced [201].

Also, strain and temperature can be discriminated by employing two different FBG sensors inscribed at different wavelengths. Even though 1550 nm is the most common choice for investigation, FBGs can also be written in other wavelengths such as 830 nm and 1300 nm. Different wavelengths lead to different sensitivity coefficients for both strain and temperature, as shown by Rao [202].

Other investigations proposed the use of two FBGs inscribed in individual optical fibres and laid down forming a small angle between the fibres. Thus, external strain and temperature perturbations will cause different reflections of the Bragg wavelength and may allow the solution of expression (2.17). This approach was followed by Pereira et al. [203], who embedded two optical fibres (with a single FBG sensor each) within two layers of a composite coupon, forming an angle of 37.5° between them, as depicted in Figure 2.35 (i). The sensors exhibited different reactions to tensile loads. Same approach was followed by Oromiehie et al. [82], who embedded two FBG sensors inscribed in different optical fibres between the layers of a thermoplastic sample being consolidated by hot-gas torch AFP-ISC method. The optical fibres were laid down forming an angle of 15° . This was performed for discrimination of temperature and strain variations which take place during the consolidation process. The set-up proposed by Oromiehie et al. is shown in Figure 2.35 (ii). In a recent study, this methodology was also employed by Armitage et al. [204], who embedded horizontal and angled FBGs into a host material.

Figure 2.35 Single-FBG optical fibres embedded in composite material forming an angled between fibres. (i) Two angled FBGs embedded in a tensile testing coupon [203]; and (ii) two angled FBGs embedded into a thermoplastic sample for process monitoring of hot-gas torch TP-ISC [82].

Other methodologies for strain and temperature discrimination based on the use of sensors with different sensitivity coefficients were found in the literature, such as the use of combined chirped FBG and Fabry-Perot sensors [205], tilted FBGs [206] or FBGs with different diameter [207].

$$\begin{pmatrix} \Delta\lambda_{B1} \\ \Delta\lambda_{B2} \end{pmatrix} = \begin{pmatrix} \kappa_{1\varepsilon} & \kappa_{1T} \\ 0 & \kappa_{2T} \end{pmatrix} \cdot \begin{pmatrix} \Delta\varepsilon \\ \Delta T \end{pmatrix} \quad (2.18)$$

▪ Encapsulation of FBG sensors

The methodology approach consisting of encapsulating a FBG sensor is the other alternative which can be found in most of the reviewed research investigations. By encapsulating an FBG, the sensor remains isolated from external mechanical strain perturbations, and thereby it acts as a temperature probe. The simplest way of discriminate temperature and strain while monitoring a perturbed system consists on using an FBG array construction consisting on a single optical fibre with two inscribed FBG sensors, where one of them is isolated of the external strain variations by being encapsulated in a capillary. The other FBG sensor remains bare and sensitivity to both temperature and strain shifts. Thus, expression (2.17) can be reformulated as (2.18), since the encapsulated sensor only monitors the thermal history of the system, and the so-called temperature compensation can be applied. The use of conventional thermocouples at the edges of the preform during processing can also be applied for subsequently temperature compensation. However, it is generally not advisable to insert thermocouples deep into the component and this is why the use of optical sensors is under research for online temperature metrology as a potential solution [157].

The application of encapsulation systems for temperature monitoring has been investigated for more than two decades. In 1997, Song et al.[208] designed a sensing head consisting of two FBGs where one of them was attached to a glass capillary which prevented the sensor from being affected by external strains. The system was validated by applying tensile loads and temperature. Figure 2.36 (i) shows a schematic view of the sensor construction whereas Figure 2.36 (ii) shows that the applied strain did not cause any wavelength shift of the encapsulated FBG sensor.

Figure 2.36 Encapsulation of FBG sensor performed by Song et al. [208]: (i) schematic view of the sensor construction and (ii) wavelength shift of the FBG sensors while applying tensile loads (note that the encapsulated sensor is not perturbed by the applied strains).

In 2000, Frazao et al. [209] designed a similar sensing head consisting of an optical fibre with both bare and encapsulated FBG sensors. The isolated sensor was encapsulated inside a steel capillary tube with inner diameter of 0.5 mm, outer diameter of 0.8 mm and 40 mm length. The system was evaluated by performing strain and temperature variations, revealing that the κ_ϵ coefficient of the encapsulated sensor was 0.0000 pm/microstrain. In 2003, Kang et al. [205] encapsulated a FBG sensor inside a glass capillary which acted as a temperature probe assuming a strain-free condition $\epsilon = 0$. The designed sensing head (depicted in Figure 2.37) also included a Fabry-Perot sensor. They used the sensing system for process monitoring of an autoclave cycle.

Figure 2.37 Hybrid FBG/EFPI sensor designed by Kang et al., which included an encapsulated FBG acting as a temperature probe [205].

Fernández et al.[210] measured in 2007 the coefficient of thermal expansion (CTE) of a composite plate with a sensing array consisting on a bare sensor and a sensor encapsulated in a glass capillary. They analysed the results provided by embedding the sensors compared to mounting the sensors on the laminate surface. An schematic view of the sensor array constructed by Fernández et al. is depicted in Figure 2.38.

Figure 2.38 Schematic view of the sensing head created by Fernández et al., consisting of a bare FBG and an encapsulated FBG [210].

In 2010, Parlevliet et al. [211] employed in 2010 a sensing device with an encapsulated FBG for discriminating thermal and strain contributions. The encapsulation was performed by surrounding the FBG sensor by a stainless steel capillary with an inner diameter of 0.6 mm and an outer diameter of 1.0 mm, which was sealed at the edges with a high temperature resistant epoxy glue. They reported that a metallic capillary was chosen due to its good thermal conductivity and there is no affinity (static attraction) between the optic fibre and inner metal surface of capillary, unlike what may happen with a glass ferrule. They used the sensing system for investigating the residual stresses into thermoplastic composite parts. Chen et al. [212] used in 2015 an encapsulated FBG sensor acting as a thermal probe in medical laser ablation of tumours for temperature monitoring. Encapsulated FBG sensors can be observed in Figure 2.39 (i) and (ii), where they are compared to commercial thermocouples. The results monitored by the sensor were compared with those provided by an IR thermographic camera, as depicted in Figure 2.39 (iii) and (iv), where Figure 2.39 (iii) shows a view of the IR camera and Figure 2.39 (iv) shows the thermal history monitored by the encapsulated FBG.

Figure 2.39 Encapsulation of FBG sensors: (i) thermocouple (a) compared to FBG sensors (b) bare, (c-d) embedded into the capillary; (ii) detail of encapsulated FBG sensor; (iii) infrared picture provided by IR camera; and (iv) thermal history provided by the encapsulated FBG sensor [212].

In 2016, Mulle et al. [213] encapsulated a FBG sensor into a hermetic tube for temperature monitoring and subsequently temperature compensation of a bare sensor. The sensors were embedded between layers of a GF/PP laminate. A schematic view of the encapsulation system is shown in Figure 2.40 (i), while the sensors embedded into the GF/PP laminate are shown in

Figure 2.40 (ii). The sensing device was used for process monitoring purposes, which are detailed in next section. The thermal history of the process was also monitored with standard thermocouples. The comparison of the results exhibited a good agreement between the encapsulated FBG and the thermocouple. Wang et al. [214] also employed an encapsulated FBG sensor for temperature monitoring in 2017. The FBG sensor was encapsulated in a steel capillary tube.

Figure 2.40 (i) Schematic view of the encapsulation system followed by Mulle et al.; and (ii) sensors embedded into a GF/PP laminate [213].

Kinet et al. [5] elaborated a review of the applications of FBG sensors into composites for SHM activities. In their review, they evaluated the performance of different methods for the discrimination of temperature and strain contributions. Their results suggested that the encapsulation approach was the most competitive solution. However, other parameters should be taken into account when selecting a methodology approach, such as ease of use, the integration into the composite or multiplexing capability. They suggested that each case should be analysed in an individual manner for selecting the best solution.

Even though it has been demonstrated that transverse or out-of-plane strains are attenuated by the use of coatings on the optical fibres; the use of modified FBG sensors is emerging for the discrimination of transverse and axial loads. In this way, microstructured optical fibre Bragg gratings (MOFBG) are a promising candidate for the discrimination of transverse and axial strains. Those sensors are characterised by having a high bi-refrindex, which causes the existence of two Bragg wavelength peaks. Those peaks may allow the discrimination of longitudinal and transverse loads, as investigated by Berghmans et al. and Geernaert et al. [215,216].

2.3.2 Application of FBG sensors on process monitoring

In this last section of the literature review, the most relevant investigations concerning the application of FBG sensors for on-line/real time process monitoring of composites manufacturing processes are presented.

Earliest investigations date from 1998, when O'Dwyer et al.[217] performed an investigation based on monitoring the curing process of a CF/Epoxy specimen. For that investigation, the authors employed a sensing system consisting of a thermocouple, a FBG sensor and a dielectric sensor. The sensors were embedded between two layers of the laminate, parallel to the fibres direction. The curing cycle inside an oven was monitored and the acquired results consisted of the

conductivity (provided by the dielectric sensor), the thermal history (monitored by the thermocouple) and the strain history (provided by the FBG sensor after performing temperature compensation with the thermal history provided by the thermocouple). The process monitoring set-up is depicted in Figure 2.41 (i), whereas the monitored results are shown in Figure 2.41 (ii). Curing monitoring was also investigated in subsequent years by Murukeshan et al. [218], who applied a different approach. Murukeshan et al. embedded bare FBG sensors into a composite and not only monitored the curing cycle, but they also demonstrated the survival of the sensor after the cycle, and therefore the manufacturing of a smart composite, which can be used for SHM applications. In this case, the raw monitored data (wavelength shift) was analysed to identify the main physical phenomena of the curing process, such as vitrification of the resin. After that, the laminates were subjected to 3 and 4-point bending and the provoked strains were monitored.

Figure 2.41 Monitoring of the curing process of a CF/Epoxy laminate by a sensing system (Dielectric sensor, FBG and thermocouple): (i) Monitoring set-up and (ii) monitoring results consisting of conductivity, thermal history and strain history [217].

In 2003, Kang et al. [205] investigated the simultaneous measurement of temperature and strain by means of a hybrid sensing device consisting of a FBG and an EFPI, which has been detailed in previous Figure 2.37. The FBG sensor was encapsulated inside a silica capillary to be isolated from external strain and the whole sensing system was embedded into an uncured thermoset prepreg stacking. The thermal and strain histories which take place during the curing of the laminates in an autoclave were monitored and reported. The monitoring set-up and the monitored thermal and strain histories are shown in Figure 2.42 (i) and (ii). A very good correlation of the thermal history monitored by the encapsulated sensor and the thermocouple was also reported. The research article was concluded by confirming that optical fibre sensors can definitely be used to monitor several parameters such as the fabrication strain and temperature during the cure of composites. The authors advised the use of sensing devices for efficient smart processing of composites and therefore gaining insight into the thermal behaviour of composite under fabrication.

Figure 2.42 Real-time monitoring of the curing cycle of a thermoset laminate by means of a hybrid FBG/EFPI sensing device: (i) Monitoring set-up and (ii) thermal and strain monitored histories [205].

Kuang et al. [219] monitored the manufacturing process of a sandwich structure with GF/PP skin. They embedded FBG sensors between the layers of the skin for detecting singular point of the resin behaviour during manufacturing. The selected manufacturing process was compression moulding and the sensors monitored the induced strains and were able to identify key phenomena such as melting, solidification and recrystallization. Wavelength peak split was observed which lead to the use of the full width at half maximum (FWHM) instead. After the stress profile was decoupled from thermal strains, it was compared with post-manufacturing mechanical tests that correlated well with FBG strains. A good agreement was reported between the theoretical strains and DSC results.

Antonucci et al. [220] employed in 2006 a hybrid optical fibre system composed of a FBG and a refractometer (Fresnel), along with a thermocouple for monitoring the curing process of an epoxy resin. The authors reported that the sensing head was able to monitor real-time the main phenomena occurring during the non-isothermal polymerization of a thermoset resin: the cure reaction and the gelification onset.

Sorensen et al. [221] published a research paper where the residual stresses appearing on thermoplastic composites during manufacturing were investigated. In their investigations, they embedded FBG sensors into CF/PPS laminates during manufacturing in order to monitor the strain history during the consolidation process. The egress points of the optical fibre were protected by using PTFE tubes. Temperature compensation was performed by means of a K thermocouple. They were able to identify key moments of the consolidation process such as application of pressure and material changes. Also, they evaluated the remaining residual strains within the material and compared it with modelling results.

De Oliveira et al. [192] investigated in 2008 the internal stresses that may appear in CF/Epoxy autoclave manufacturing and the effect of different manufacturing tools. With that aim, they embedded FBG sensors in UD and cross-ply laminates and monitored the curing cycle. The strain history for each case was reported and the results suggested that the FBG sensors may suffer peak splitting when embedded in cross-ply laminates, due to the effect of transverse loads. However, they were able to conclude that FBG sensors are under tension when curing on a metallic tool, suggesting that internal stresses are transferred to the laminate during curing.

Gupta and Sundaram [222] reported in 2009 the possibility of monitoring the resin flow front in VARTM processes by means of FBG sensors, highlighting the fragility of the optical fibres, which are susceptible to failure during embedment. Mulle et al. [223,224] published a series of investigations focused on the study of curing residual stresses through-the-thickness in CF/Epoxy laminates using FBG sensors. For their investigation, several FBG sensors were embedded into the samples at different layers (bottom and top) during stacking operations. Arrays consisting of two FBG sensors where one on them was encapsulated for strain isolation were employed. The samples were cured into an autoclave and thermal and strain histories were monitored by the sensing devices. The authors reported that the various phases of the curing process were well characterized; in particular that of the cooling, at which time one clearly noted interactions between the metal mould and the composite part. Residual strains were estimated at the end of the curing. Transverse strain effects due to pressure or contraction did not have significant influence on the longitudinal strain information. This guaranteed a good degree of confidence in the strain measurements. Sensor installation and monitoring set-up is presented in Figure 2.43 (i) and the monitored results provided by the FBG sensors are shown in Figure 2.43 (ii).

In 2010, Parlevliet et al. [211] incorporated FBG sensors (some of them encapsulated acting as temperature probes) and performed the curing monitoring of several resins. The post-curing strain development was investigated, and they successfully measured the polymer cure shrinkage responsible for the residual strains in the composites. They highlighted the need of encapsulated FBG sensors for obtaining reliable results after performing temperature compensation. In similar investigations, Nielsen et al. [193] investigated the residual stresses appearing on thick GF/Epoxy during vacuum infusion of wind turbine blades.

In more recent studies, Mulle et al. [213] employed FBG sensors in 2016 for process monitoring of CF/PP samples consolidation in hot-press. They embedded FBG sensors in parallel and transverse direction of the fibres into 8-ply unidirectional laminates. Sensor arrays composed of a bare and an encapsulated FBG were embedded between 1st and 2nd plies and between 4th and 5th plies of the samples. Monitoring set-up is shown in Figure 2.43 (iii) and the monitored strain and temperature profiles are shown in Figure 2.43 (iv). Each phase of the process was identified by the measurements provided by the FBGs. Thus, the authors were able to identify melt and solidification transition phases, and they reported identical results to those provided by DSC. Residual strains after manufacturing and releasing from the mould were estimated and reported.

Figure 2.43 Process monitoring of composites fabrication with FBG sensors. Investigations performed by Mulle et al. (i) Instrumentation of CF/Epoxy laminates processed by autoclave and (ii) corresponding monitored results. (iii) Set-up for GF/PP consolidation monitoring; and (iv) corresponding monitoring results [213,223].

In 2016, Oromiehie et al. [81] started a series of investigations based on the on-line monitoring of hot-gas torch AFP-ISC of thermoplastic composites. In their investigation, two bare FBG sensors were integrated in GF/HDPE material substrate followed by automated lamination of the rest of the layers. The location of the FBG sensors can be observed in Figure 2.44 (i). The lamination and consolidation of the rest of the layers was successfully monitored. The authors reported raw monitored data (wavelength shift, Figure 2.44 (ii)) and highlighted interesting findings. Thus, in each consolidation pass, the changes in the wavelength were caused by the combined effect of consolidation loads and heat. An inverse correlation was reported between the wavelength shift and the thickness of the laminate. In addition to that, it was reported that after the lamination of a new layer, the FBG sensor did not recover the initial base level, as shown in Figure 2.44 (ii). This was attributed to residual strain effects and the authors opened the possibility of measuring residual stresses of thermoplastic ISC laminates by means of FBG sensors.

Figure 2.44 On-line monitoring of thermoplastic AFP-ISC technology by means of FBG sensors: (i) Placement of the FBG sensors on a GF/HDPE sample; and (ii) Wavelength shift monitored results [81].

In 2017, same authors employed FBG sensors for investigating the detection of manufacturing defects [66]. In that way, the sensors were again embedded into the substrate but artificial laminating defects were induced, consisting of gaps and overlaps. The analysis of the monitored wavelength shift suggested that depending on the type of defects, significant changes in the wavelength profiles were observed. These changes included the shape of wavelength profiles, the maximum wavelength shifts in corresponding layers, and the slope of the wavelength profiles during the cooling down. In other research article published in 2017, the authors employed a different strategy for discriminating the contribution of strain and temperature during hot-gas torch AFP-ISC process [81]. Thus, they embedded two FBG sensors within the composite substrate, but this time, the sensors were located forming an angle of 15° . This strategy has been previously detailed in this chapter as a possible methodology for discriminating strain and temperature contributions. Thus, the sensors exhibited different sensitivity coefficients for solving previous equation (2.17). The monitored raw data (wavelength shift) was reported, but regarding thermal and strain histories, only maximum values of strain and temperature monitored during the lamination of each new layer were reported. The maximum temperature was reported to decrease with the number of layers; however, increasing compressive strains with the number of layers was reported. The authors highlighted that further experimental is required for validating the monitored temperature and strain.

Figure 2.45 On-line monitoring of thermoplastic AFP-ISC process with two angled-FBGs: (i) Monitoring set-up; (ii) wavelength shift monitored during lamination; and (iii) temperature values [81].

In 2017, Takeda et al. [225] investigated the use of FBG sensors for monitoring the strain variations when consolidating a CF/PPS L-shape in a hot-press. One FBG sensor was located at the centre part and other FBG sensor was located at 25 mm, as indicated in Figure 2.46 (i). After sensor installation, the laminate was moulded in a hot-press as shown in Figure 2.46 (ii). The FBG sensors were monitoring during the whole process and temperature compensation was performed by using the thermal history monitored by a thermocouple. The monitored data was acquired with a frequency of 1 minute. The authors concluded that high-speed monitoring and temperature compensation should be applied to investigate the press and cooling process in detail.

Figure 2.46 On-line process monitoring of a CF/PPS thermoplastic L-shape by means of FBG sensors: (i) Placement of FBG sensors at different locations; and (ii) forming process in hot-press [225].

In a very recent study, Lehman et al. [226] presented the industrial application of FBG sensors for resin flow front detection during the infusion of a large scale composite part like a booster casting for space applications. The investigation was framed within an industrial research project called Infusion 4.0, carried out by BMW, Fraunhofer LBF and MT Aerospace ranked 3rd in the 2017 Innospace Masters Award for the DLR Challenge. The investigation consisted of embedding 63 FBG sensors between the dry layers of the cylinder; and subsequently monitoring the resin infusion with process control purposes. The dry preform with the installed sensors can be observed in Figure 2.47 (i). The monitoring control system is depicted in Figure 2.47 (ii). The authors concluded by saying that the experiments showed very promising results which can raise the infusion technology to a new level, by reducing infusion time. In addition to that, the sensors remained in the structure and the authors suggested their use for SHM activities. In relation to the present study, the authors highly recommended the use of FBG sensors for real-time monitoring of thermoplastic AFP-ISC process, since key parameters as thermal history and compaction during lay-up should be well known during manufacturing step.

Figure 2.47 Flow front detection during the infusion of an industrial booster casting by means of FBg sensors: (i) Dry preform with 63 FBG sensors; and (ii) control station outside the oven [226].

Chapter 3

Characterization of thermoplastic composites manufacturing processes

3.1 Introduction

This chapter presents a detailed characterisation and comparison of thermoplastic laminates manufactured under the three different OoA manufacturing methods under investigation in this study. Experiments including non-destructive testing (ultrasonic, X-Ray micro-CT), physical-chemical (matrix digestion, DSC) and mechanical characterisation including fibre and matrix dominated properties (tensile, flexure, IPSS and ILSS) are presented.

The increasing interest of the aerospace industry in thermoplastic composites along with the opportunity of using fast processing methods have caused the development of innovative manufacturing processes for thermoplastics, such as ISC. However, a lack of some mechanical properties has been found in the structures manufactured under ISC: In this chapter, a comparison of the performance of VBO, hot-press and ISC laminates in order to enlighten the possible causes which may induce a drop in some properties is carried out.

According to the performed tests, several points of view were investigated, including the ultrasonic propagation, porosity, voids distribution and degree of crystallinity. However, among the results, those obtained by the micro-tomography may have revealed the most interesting conclusions based on the voids distribution of ISC laminates, since the manufacturing process may be causing a characteristic void distribution pattern which can eventually cause a drop in the mechanical properties.

In this chapter, all the performed tests have been deeply analysed and discussed. This chapter consists on an extended version of the published research article entitled “A comparison of mechanical properties and X-ray tomography analysis of different out-of-autoclave manufactured thermoplastic composites” [227]

3.2 Materials

The material selected for this chapter and the rest of the present doctoral dissertation was the thermoplastic composite material APC2/AS4. The material is an unidirectional (UD) CF/PEEK tape supplied by Cytec-Solvay Group. Two different width formats were purchased for the manufacturing via different processing methods. UD tape 300-mm width was purchased for oven and press manufacturing, meanwhile same material but 6.35 mm wide slit was acquired for the automatic lamination by in-situ consolidation. The 300 mm width roll of APC2-AS4 is shown in Figure 3.1 (i), while the spool of 6.35 mm width APC2-AS4 tape is shown in Figure 3.1 (ii). The nominal prepreg fibre areal weight (FAW) was 145 g/m^2 and the initial prepreg matrix weight fraction was 34%. Nominal thickness per ply was 0.135 mm. The main properties of the raw material can be also found in the supplier data sheet [228].

Figure 3.1 APC2-AS4 material employed in this study: (i) 300 mm width roll for VBO and hot-press manufacturing; and (ii) spool of 6.35 mm width tapes for AFP-ISC manufacturing.

3.3 Manufacturing of laminates

For the study proposed in this chapter, a set of six CF/PEEK laminates were manufactured by using three different out-of-autoclave manufacturing methods, including vacuum bag-only in oven, hot-press and laser assisted AFP-ISC. Figure 3.2 shows the manufacturing set-up for the manufacturing processes: (i) a laminate inside the vacuum bag ready for the cycle, (ii) a laminate

being consolidated by means of hot-press; and (iii) laser assisted AFP-ISC equipment along with the manufacturing tooling ready for lamination. The manufactured specimens consisted of laminates with two different stacking sequences: $[0]_{16}$; and $[45,-45]_{4S}$, and final dimensions of $300 \times 300 \times 2.16$ mm (length $\times$ width $\times$ thickness). One laminate of each lay-up sequence was manufactured by each of the manufacturing technologies.

Figure 3.2 OoA manufacturing technologies used in this work: (i) vacuum bag-only in oven (VBO), (ii) compression molding in hot-press; and (ii) laser assisted AFP-ISC.

A detailed summary of the manufactured laminates including key manufacturing parameters is given in Table 3.1. A specific nomenclature has been used for the identification of each laminate. Thus, the codes referred to the manufacturing method: VBO= vacuum bag-only, HP = hot-press and ISC= laser-assisted AFP-ISC; and also referred to the lay-up: 0= $[0]_{16}$ and 45= $[45/-45]_{4S}$.

Table 3.1 Summary of manufactured laminates and manufacturing parameters

Manufacturing technology	Laminate code	Stacking sequence	Manufacturing process parameters			
			Consolidation temperature (°C)	Dwell time (min)	Vacuum level (mbar)	
Vacuum bag-only in oven	VBO_0	[0] ₁₆	400	15	100	
	VBO_45	[45/-45] _{4S}				
Hot press			Consolidation temperature (°C)	Dwell time (min)	Pressure on laminate (MPa)	
	HP_0	[0] ₁₆	400	15	0.7	
	HP_45	[45/-45] _{4S}				
TP-AFP with in situ-consolidation (ISC)			Average laser power (W)	Tooling temp (°C)	Consolidation force (N)	Lamination speed (m/min)
	ISC_0	[0] ₁₆	180	200	500	2
	ISC_45	[+45/-45] _{4S}				

3.3.1 Vacuum bag in oven

Thermoplastic laminates manufactured by VBO in this study were processed following aerospace standard procedures. Two 16-ply laminates corresponding to stacking [0]₁₆ and [45/-45]_{4S} were consolidated in an oven. A flat steel tool with an incorporated grid as air evacuation path was used. The thermoplastic preform was located between two polyimide sheets (kapton) with demoulding agents. A high temperature breather was located on the top of it and the vacuum bag was sealed against the tooling by using high temperature sealant tape. Manufacturing parameters applied during consolidation have been already listed in Table 3.1. During the consolidation cycle, the temperature was maintained at 400 ± 10 °C for a dwell time of 15 min. The temperature and the vacuum were monitored during the cycle with embedded thermocouples and vacuum sensor. Heating and cooling ramps were performed at 2 °C/min, in order to obtain an adequate degree of crystallinity. The VBO manufacturing set-up is shown in Figure 3.3 (i), whereas a schematic illustration of the process is shown in Figure 3.3 (ii).

Figure 3.3 Vacuum bag only manufacturing: (i) manufacturing set-up with a high temperature vacuum bag inside the oven ready for consolidation; and (ii) schematic illustration of the process.

3.3.2 Hot press

Figure 3.4 (i) shows the hot-press manufacturing set-up, while Figure 3.4 (ii) shows a schematic illustration of the process. The prepreg material was first laid-up manually applying some welding points with the help of a manual welder. The laminate was then located inside a metallic frame which acted both as material retainer (dam) and thickness constraint. Two kapton (polyimide) sheets with release agent were located on the bottom and the top of the laminate. Two metallic caul plates were used for obtaining flat surfaces. The detailed manufacturing parameters have been shown in Table 3.1. The temperature was increased up to 400 °C and maintained for 15 min before cooling down. Contact pressure was applied until the cycle reached consolidation temperature, after which the pressure was increased and maintained to 0.7 MPa for the rest of the cycle. The temperature was monitored by an embedded thermocouple and the hot-press control device. Heating and cooling ramps were performed at 2 °C/min.

Figure 3.4. Compression moulding manufacturing in hot-press: (i) Manufacturing set-up with a laminate located inside the hot-press ready for consolidation; and (ii) schematic illustration of the process.

3.3.3 In-situ consolidation TP-AFP

The laminates manufactured by in-situ consolidation were consolidated using a gantry-style single-tow deposition machine developed by MTorres (Pamplona, Spain) and located in FIDAMC facilities (Getafe, Spain). A diode laser was used as heating source which created a profile with a heating area of 140 mm in the lamination direction and 7.5 mm in transverse direction. A spool

system orientated an incoming tow which was also heated by the laser. A self-heated tooling was used for minimizing the effect of residual stress and also preventing the tool from acting as a heat sink. Manufacturing parameters are compiled in Table 3.1. Thus, tooling temperature was kept at 200 °C, deposition rate was fixed at 1 m/min, laser worked at an average power of 180 W and the silicone compaction roller applied a force of 500 N. The processing temperature was recorded real-time with an infrared thermal camera supplied by FLIR. Set-up of ISC process is detailed in Figure 3.5 (i), where the main elements are indicated: IR camera (a), diode laser (b), spools system for material feeding (c), compaction roller (d), manufacturing tool with self-heating system (e) and substrate material (f). A schematic illustration of the process is shown in Figure 3.5 (ii).

Figure 3.5 Thermoplastic laser-assisted AFP-ISC: (i) lamination set-up; and (ii) schematic illustration of the process.

3.3.4 Manufacturing of CF/PEEK NDT reference standard (calibration block)

All the NDT activities performed during the present study were performed according to industrial standards and procedures. Within this context, a thermoplastic CF/PEEK reference standard (also known as calibration block) was manufactured following AITM6-0012 [229]. All the laminates manufactured during this study were around 2 mm thickness. According to the standard, a composite part must be ultrasonically inspected after calibrating the system in a reference standard with different thicknesses. The thinnest step should be 1 ± 0.1 mm and the thicker step should be thicker than the composite part without exceeding a 120% of the major thickness. Thus, a 240 x 300 mm standard reference with 3 different steps was designed. All the details about the lay-up of the three included steps are given in Table 3.2.

Table 3.2 Manufacturing details of CF/PEEK standard reference

Step number	Lay-up	Number of layers	Nominal thickness (mm)
1	[0/90] _{2S}	8	1,08
2	[0/90] _{4S}	16	2,16
3	[0/90] _{6S}	24	3,24

Typically, reference standards (composite or metallic made) must include defects or indication in order to validate that the inspection equipment and personnel are able to detect and characterize the defects. In this case, several defects (material inclusions) were embedded into the reference block at different depths and positions. Polyimide (Kapton) was selected for the material inclusion. Figure 3.6 shows the schematic views of the reference standard including the location of the artificial defects. The materials inclusions should be located at different depths of the thickness, including, at least, at the thickness corresponding to two plies from the top surface, in the middle plane and also two plies to the bottom surface, as indicated in the figure. The two columns of defects that can be observed correspond to the two used different defect sizes. Those sizes were $4 \times 4 \text{ mm}^2$ and $6 \times 6 \text{ mm}^2$. The defect sizes were defined according to the minimum detectable size given in the AITM6-4005 [230] industrial standard.

Figure 3.6 Schematic view of CF/PEEK NDT standard block with the location of artificial defects (kapton inclusions): (i) top view; and (ii) profile view.

After design, the CF/PEEK calibration block was manufactured. Firstly, the plies and the kapton inclusions were cut by hand, as indicated in Figure 3.7.

Figure 3.7 Preparation for the reference standard manufacturing: (i) CF/PEEK plies cutting; and (ii) kapton artificial inclusions.

After cutting the plies, the kapton artificial defects were embedded in the defined positions, as shown in Figure 3.8 (i). The plies were then manually laid-up with the help of a manual welder as shown in Figure 3.8 (ii). The use of a welder was required due to the lack of tacking, characteristic of the thermoplastic prepreg material.

Figure 3.8 Reference standard manufacturing: (i) location of kapton inclusions in the designed locations; and (ii) hand lay-up with a manual welder.

After the manual lamination of the calibration block, the vacuum bag set-up was carried out, following the same procedures as depicted in previous section 3.3.1. However, the use of caul plates was included in order to obtain an optimal quality surface finish which ensures the proper entry of the ultrasonic wave into the composite plate, reducing the attenuation of the wave due to superficial irregularities. The preparation of the vacuum bag is shown in Figure 3.9. The placement of the metallic caul plates is shown in Figure 3.9 (i), while the bag closure is shown in Figure 3.9 (ii). Once the vacuum bag was prepared, the system was introduced into the oven and submitted to a typical consolidation cycle consisted of maintaining a consolidation temperature of 390 °C during a dwell time of 15 minutes. Heat-up and cooling-down rates were maintained as close as possible to 1-2 °C/min following material supplier suggestions, as shown in Figure 3.10.

Figure 3.9 Vacuum bag preparation for the consolidation of the CF/PEEK NDT reference standard: (i) placement of caul plates for finish surface; and (ii) vacuum bag closure.

Figure 3.10 APC2-AS4 consolidation cycle recommended by supplier.

3.4 Experimental methods

The main purpose of this chapter is to study the different properties and response of the manufactured CF/PEEK laminates under the three different OoA manufacturing methods. A set of coupons were extracted from all the laminates in order to analyse different physical-chemical properties, mechanical performance and ultrasonic response. A detailed description of all the experimental methods and equipment is given in the next sub-sections.

3.4.1 Ultrasonic NDT of laminates

All the manufactured laminates manufactures not only in this chapter, but also in the rest of this study were submitted to ultrasonic non-destructive testing. Double through-transmission following AITM6-4011 was the ultrasonic method selected for this work. The method is also known as reflector plate. In this methodology, the composite laminate is located inside a water tank (full immersion) on a reflector plate manufactured with a reflective material (e.g. methacrylate) leaning on supports. In this technique, a single transducer is required working as emitter-receiver (same configuration as the well-known pulse-echo). The ultrasonic wave is generated by the transducer and travels through the laminate until it is reflected by the reflector plate and travels all the way back until it is received by the probe working as receiver. In the double through transmission technique, the energy drop of the wave during its travel is analysed. If the wave is especially attenuated, an indication may be further studied and determined as defect or healthy area.

The automated equipment that was used was an industrial Triton 8000TT+ supplied by Tecnitest (Madrid, Spain). The equipment consisted of a 5-axis machine operated by Tecnitest-made software. The NDT equipment used in this study is shown in Figure 3.11, where the immersion water tank and the probe are shown.

Figure 3.11 Ultrasonic equipment Triton 8000TT+ (Tecnitest).

All the specimens manufactured during the study were inspected following the same testing conditions. The selected inspection frequency was 5 MHz, which is generally considered optimal for the inspection of carbon fibre composite plates. The inspection probe automatically displaces tracing lines ensuring the inspection of the 100% of the area of the specimen. The pitch between scanning lines was set at 2 mm. The use of an automated inspection equipment involved the generation of C-scan registers, but also A-scan for each point of the register. The use of the reflector plate technique allowed generating c-scans in different modes. Those modes were attenuation and amplitude, which represent the attenuation of the wave during its travel through the specimen and the amplitude of the back-wall echo in each point, respectively.

3.4.2 DSC

Samples extracted from the thermoplastic laminates were subjected to Differential Scanning Calorimetry testing. DSC tests were performed in order to obtain thermograms and the degree of crystallinity of all the specimens. At least three different specimens of different regions of each laminate were tested, with a mass of at least 15 mg. DSC tests were performed following the standard ISO 11357-3 [231]. The used equipment consisted of the Q2000 from TA Instruments shown in Figure 3.12. The calibration of the equipment was carried out by using indium and cyclohexane. All the samples were enclosed inside metallic capsules which were hermetically sealed.

Figure 3.12 Q2000 (TA Instruments) DSC equipment.

Dynamic tests were performed with a heating rate of 20 °C/min up to 390 °C starting from room temperature; followed by a 5-min isotherm. Cooling was then performed at a rate of 10 °C/min. The degree of crystallinity X was calculated using equation (3.1).

$$X = \frac{\Delta H_m - \Delta H_c}{\Delta H_f(1 - \alpha)} \quad (3.1)$$

Where ΔH_m is the enthalpy of fusion at melting point, ΔH_c is the enthalpy of cold crystallization (which can be observed in some cases), ΔH_f is the enthalpy of the completely crystalline polymer (considered as 130 J/g) and α refers to the weight fraction of carbon fibre within the laminate. Experimental values of weight fibre fraction obtained after matrix digestion testing (detailed in next sub-section) were applied to determine the degree of crystallinity.

3.4.3 Void content

The void volume fraction values of all the manufactured CF/PEEK laminates were assessed by means of matrix digestion method, also known as acid digestion method. A minimum of three samples from different regions of the laminate were tested in order to obtain a reliable void volume value. All tests were performed at the physical-chemical FIDAMC laboratory facilities in a ventilated chamber, following internal procedures. The tests were performed in accordance to EN 2564 standard [232]. The method is based on the comparison of the coupon weight before and after the elimination of the composite matrix by degradation in heated sulphuric acid. For these testing, an Ovan MCH300E heating plate, 250 ml flasks, concentrated sulphuric acid, hydrogen peroxide, acetone and distilled water were used. The matrix was removed from the composite owing to the chemical reaction and; after that, the weight of the coupon is compared before and after the test in order to obtain the void volume fraction. Figure 3.13 shows the details of the void content assessment by matrix digestion method: (i) coupons inside volumetric flasks where the digestion reaction is ongoing by means of heated sulphuric acid, (ii) ventilated chamber, (iii) thermoplastic coupon after matrix is degraded (only carbon fibres remained); and (iv) microbalance for weight determination.

Figure 3.13 Void content assessment by matrix digestion method: (i) specimen matrix being removed in sulphuric acid, (ii) ventilated chamber, (iii) remaining carbon fibres after matrix digestion; and (iv) microbalance for weight determination.

For the void volume assessment, the following expressions (4.3) to (4.5) were used. In these expressions, m_o and m_f refer to the initial and final mass of the sample (in mg), W_r and W_f refer to the weight fraction of the resin and fibre into the sample (in %), ρ_r and ρ_f are the resin and fibre densities. V_r , V_f and V_v are the final volume fraction of resin, fibre and voids in the sample.

$$W_f = 100 \left(\frac{m_f}{m_o} \right) \quad (3.2)$$

$$V_f = W_f \left(\frac{\rho_c}{\rho_f} \right) \quad (3.3)$$

$$W_r = 100 - W_f \quad (3.4)$$

$$V_r = (100 - W_f) \frac{\rho_c}{\rho_r} \quad (3.5)$$

$$V_v = 100 - V_f - V_r \quad (3.6)$$

3.4.4 X-ray micro-computed tomography (micro-CT)

At least one specimen of each of the six laminates characterised in this chapter were submitted to x-ray micro-CT for void distribution and size analysis. Specimens with approximated dimensions 10×10 mm were extracted from each laminate and scanned using a Bruker SkyScan 1172 X-ray Micro-CT Scanner located at the University of Auckland Bioengineering Institute, which is shown in Figure 3.14. The samples were mounted on the top of the rotary tool and fixed to the bottom using adhesive tape, as depicted in Figure 3.15. The settings used for the scans are provided in Table 3.3.

Figure 3.14 Bruker SkyScan 1172 X-ray Micro-CT Scanner located at the University of Auckland Bioengineering Institute.

Figure 3.15 CF/PEEK sample mounted in the rotary tool for CT scanning.

The x-ray projections obtained from the scans were reconstructed using NRecon version 1.6.9.18 using the InstaRecon version 2.0.3.7 engine into 16-bit three-dimensional volumes split into equally spaced TIFF image sequences. The reconstructed data were re-oriented to align the x, y and z planes to the actual specimen orientation and their histograms were stretched to provide adequate contrast using ImageJ [233]. Uneven greyscale intensities between the specimen centres and edges were corrected using fast Fourier transform filter within ImageJ. This was required before converting the adjusted image sequences into 8-bit formats. The image sequences were then processed to quantify the void morphology and locations using the method proposed by Ramesh et al. [148]. The image processing involved stretching the histogram of the reconstructed

volume to improve contrast and binarising the volumes by applying an adaptive threshold to isolate the voids before removing any residual noise. Finally, the locations, shapes and sizes of voids were quantified for all six specimens. Image processing, as well as the employed algorithm were developed with the assistance of University of Auckland research personnel

Table 3.3 X-ray micro-CT scan settings

Parameter	Value
Voxel size	4.47 μm
Source voltage	85 kV
Source current	118 μA
Specimen to source distance	106.93 mm
Camera to source distance	215.234 mm
Exposure time	600 ms
Rotation step	0.20°
Total rotation	360°
Frames per projection	2
Shuttling / random movement	Yes

3.4.5 Mechanical characterization

For the mechanical characterisation of the thermoplastic laminates manufactured in this chapter, a set of mechanical properties was selected, including both matrix-dominated and fibre-dominated properties. The main purpose of the mechanical characterisation is to know and to compare the mechanical response of the laminates manufactured under different OoA methods and investigating the mechanical properties of the innovative laser-assisted AFP-ISC process in comparison to the other two process under research. All the mechanical tests were performed at the FIDAMC mechanical laboratory. The machines used for the mechanical tests consisted of a universal Z010 Zwick testing machine with a 10 kN load cell; shown in Figure 3.16 (i); and a universal MTS testing machine model 370.10 with a 100 kN load cell, shown in Figure 3.16 (ii).

Figure 3.16 Universal testing machines used for the mechanical characterisation of this study: (i) Z010 Zwick testing machine with a 10 kN load cell; and (ii) 370.10 MTS with a 100 kN load cell.

In the next sub-section, all the details about each mechanical testing performed for this chapter of the investigation are given.

3.4.5.1 ILSS (inter-laminar shear strength)

All the ILSS tests were performed following EN 2563 standard [234], using the Z010 Zwick testing machines with 10 kN load cell. Batches containing at least five coupons machined from each laminate were tested. The final nominal dimensions of ILSS coupons were $20 \times 10 \times 2.16$ mm with $[0]_{16}$ stacking sequence. The cross-head stroke was set to 1 mm/min. The test fixture used for ILSS testing is shown in next Figure 3.17, where the loading nose and the supports are shown. In order to induce a pure interlaminar shear failure, the distance between supports (l_v) was calculated as a function of the averaged coupon thickness following expression (3.7).

Figure 3.17 ILSS test fixture

$$l_v = (5h_m) \pm 0.1 \quad (3.7)$$

After the test, the apparent ILSS was calculated from the force-displacement curve, following expression (3.8), where τ is the apparent interlaminar shear strength (MPa), P_r is the maximum load at the moment of first failure (in N), b is the coupon width (in mm) and h is the coupon thickness (in mm).

$$\tau = \frac{3P_r}{4bh} \quad (3.8)$$

After the test, the failure mode of the coupons was evaluated by means of optical microscopy. The possible failure modes are compiled in next Figure 3.18. If a not valid failure is detected in some of the tested coupons, that coupon may be excluded for the final analysis and repeated (if possible).

Figure 3.18 Possible failure modes of ILSS coupons

3.4.5.2 IPSS (In-plane shear strength)

The IPSS tests were performed in accordance with EN 6031 standard [235], by using the 370.10 MTS machine with a 100 kN load cell. In addition to that, two extensometers supplied by Epsilon were used for the monitoring of strains. Batches containing a minimum of six coupons per laminate were tested. The coupons had nominal dimensions of $230 \times 25 \times 2.16$ mm coupons with $[+45/-45]_{4S}$ stacking sequence. Load was applied at a rate of 1 mm/min and the longitudinal and transversal strains were continuously recorded during each test. When a longitudinal strain of 2% was reached, the crosshead speed was increased to 10 mm/min until failure. Maximum load was recorded and finally, the ultimate IPSS τ_{I2} and G12 in-plane shear modulus were calculated using next expressions (3.9) and (3.10), where P_{max} is the maximum monitored load during test, b represents the averaged coupons width, h is the average coupon thickness, $\Delta\tau$ is the difference of shear strength between 2500 and 500 $\mu\epsilon$, $\Delta\epsilon_0$ is the longitudinal strain difference between 2500 and 500 $\mu\epsilon$ and $\Delta\epsilon_{90}$ is the longitudinal strain difference between 2500 and 500 $\mu\epsilon$.

$$\tau_{12} = \frac{P_{max}}{2bh} \quad (3.9)$$

$$G_{12} = \frac{\Delta P}{2 b h (\Delta \varepsilon_0 - \Delta \varepsilon_{90})} = \frac{\Delta \tau}{(\Delta \varepsilon_0 - \Delta \varepsilon_{90})} \quad (3.10)$$

3.4.5.3 Flexure

All the flexure tests were performed in accordance to EN 2562 (type B), using the Z010 Zwick testing machine with 10 kN load cell. Batches containing a minimum of six coupons were tested from each thermoplastic laminate. Flexure coupons had dimensions of $100 \times 10 \times 2.16$ mm and unidirectional $[0]_{16}$ lay-up. Cross-head speed was fixed at 5 mm/min during testing. The testing fixture used for flexure tests is shown in Figure 3.19. Separation between bottom supports was fixed at 80.0 mm.

Figure 3.19 Flexure test: (i) Z010 Zwick testing machine set up for flexure test, (ii) close detail of coupon supports; and (iii) distance between bottom supports with millimetre ruler (iii).

Deflection as a function of load and load at failure were recorded during the test. After the tests, flexural strength and modulus were calculated by using the following equations (3.11) and (3.12), where σ_b (MPa) is the flexure strength, P_R (N) is the peak load at the failure moment, b (mm) represents the averaged coupons width, h (mm) is the average coupon thickness, l_v (mm) is the distance between bottom supports and f_1 and f_2 are the deflection corresponding to $P_R/2$.

$$\sigma_b = \frac{3 P_R l_v}{2 b h^2} \quad (3.11)$$

$$E_b = \frac{P_R l_v^3}{10 b h^3 (f_2 - f_1)} \quad (3.12)$$

After each test, the failure mode of the coupons was evaluated by means of optical microscopy. The possible failure modes are compiled in next Figure 3.20. If a not valid failure is detected in some of the tested coupons, that coupon may be excluded for the final analysis and repeated (if possible).

Figure 3.20 Possible failure modes of flexure coupons

3.4.5.4 Tensile

Tensile testing was performed according to EN 2561 (type A) standard [236], 370.10 MTS universal testing machine with a 100 kN load cell. Batches containing a minimum of six coupons were tested from each thermoplastic laminate. Tensile coupons had final dimensions of $250 \times 10 \times 2.16$ mm and unidirectional $[0]_{16}$ stacking sequence. During the test, the load was applied at a constant rate of 2 mm/min, while recording strain in longitudinal direction with an extensometer. Load at failure was also recorded for the calculation of tensile strength, which was calculated after testing following expressions, where σ_T (MPa) is the tensile strength, P_R (N) is the peak load at the failure moment, b (mm) represents the averaged coupons width, h (mm) is the average coupon thickness, (εB) is the applied force in the fibre direction and (εA) is the force applied in the fibre direction corresponding to $P_R/10$.

$$\sigma_T = \frac{P_R}{b h} \quad (3.11)$$

$$E_T = \frac{0.4 P_R}{b h (\varepsilon B - \varepsilon A)} \quad (3.12)$$

3.5 Results and discussion

All the above described tests characterise the ultrasonic NDT, degree of crystallinity, void distribution, size, average void content and the mechanical response of all the CF/PEEK laminates manufactured by the three different OoA methods under investigation. In this section, all the results that come from the experimental methods are compiled and discussed below.

3.5.1 Ultrasonic testing

Figure 3.21 shows the amplitude c-scans of the six laminates manufactured for the study compiled in this chapter. The figure shows the c-scan in grey scale (showing attenuation in dB) of each of the CF/PEEK laminates, along with an equivalent attenuation scale bar. The black squares which can be observed in some of the c-scans were caused by methacrylate supporting tools.

Figure 3.21 Ultrasonic amplitude c-scans of the CF/PEEK laminates manufactured by VBO, hot-press and ISC.

If a typical 6 dB attenuation threshold used for the quality control of composite materials in the aerospace industry is applied, the new filtered c-scans can be observed in Figure 3.22. By using this filter, laminates VBO_0, VBO_45, HP_0 and HP_45 can be considered as defect-free since no indication can be found in the c-scan. On the other hand, laminates ISC_0 and ISC_45, which were manufactured by ISC process, showed some attenuated indications. In the case of ISC_0, the areas with more attenuation observed in the top of the c-scan correspond to the region where the incoming tow is cut and the pre-tensile is lost during lamination, which may cause a bad degree of consolidation in that area. However, those areas were avoided for the coupons trimming. In addition to that, other attenuated regions can be found in both ISC laminates located in the gap between adjacent tapes, which may be due to air entrapped between plies. However, due to the elongated size and small dimensions of the indications (especially the low width), those defects may not be considered as defects according to aerospace standards, where typical minimum reportable size may be around 10 x 10 mm, depending on the applied standard.

Figure 3.22 Ultrasonic amplitude c-scans (filtered at a corresponding attenuation of 6 dB) of the CF/PEEK laminates manufactured by VBO, hot-press and ISC.

3.5.2 DSC

The values of the degree of crystallinity of the CF/PEEK laminates calculated after performing the DSC tests are shown in Table 3.4. Figure 3.23 shows the representative DSC thermograms performed to samples manufactured with the different methods under study.

Table 3.4 Average value of the degree of crystallinity of the CF/PEEK laminates manufactured by VBO, hot-press and ISC

Manufacturing method	Average degree of crystallinity (%)	Relation to VBO reference (%)
VBO	40.1 ± 0.2	N/A
Hot-press	42.7 ± 6.3	107
ISC	36.9 ± 2.8	92

Figure 3.23 Representative DSC thermograms of the manufactured CF/PEEK laminates.

It may be observed in the table that ISC laminates presented a slightly lower degree of crystallinity (36.9%) than the laminates manufactured by the rest of OoA manufacturing methods. The other two methods (VBO and hot-press) showed degrees of crystallinity higher than 40 %. Final degree of crystallinity of a thermoplastic part is mainly related to the cooling rate experience during the manufacturing process [237]. Therefore, fast cooling rates are mainly associated to lower values of crystallinity, due to the fact that low times during the cycle in the crystallization temperature window may limit the correct formation of the crystals. Thus, ultra-fast thermal cycles (and therefore cooling rates) characteristic of ISC process are the main cause of lower degrees of crystallinity in the ISC manufactured laminates. During ISC manufacturing, typically values of cooling rates above 10000 °C/min may take place, which differs from typical cooling rates of 1-5 °C/min considered optimal for achieving full crystallization. Due to this fact, CF/PEEK thermoplastic laminates manufactured by ISC process could be expected to have low values of crystallinity, even below 30%.

However, previous experience of the author in FIDAMC using unheated tooling during lamination resulted in values of degree of crystallinity below 30%. For the manufacturing of laminates ISC_0 and ISC_45 in this study, a heated lamination tooling was used, heated up to a temperature of 200°C, which considerably assisted in obtaining crystallinity values above 30%. Although ISC_0 and ISC_45 panels presented lower crystallinity values when compared with VBO and hot-press panels, it is worth noting that none of these ISC panels experienced the cold crystallization event, so the lower values of crystallinity could also be associated to different crystallization developments in comparison with VBO and hot-press. Moreover, as it can be appreciated in Figure 3.23, VBO and hot-press laminates exhibited multiple melting peaks which can be related to two distinct crystal morphologies or to a melting and recrystallization phenomenon [238].

These results are on the same line as the investigations found in the literature. Comer et al. [48] reported crystallinity values of 17.6% corresponding to laser assisted AFP laminates manufactured using unheated tooling. Tierney and Gillespie [69,122] determined the degree of crystallinity of a consolidated tape to be in the range of 10-29.5% also using an unheated tooling. By performing this comparison, it can be confirmed that the use of heated tools can significantly assist in increasing the final crystallization of a ISC thermoplastic structure.

It has been widely reported in the literature that the degree of crystallinity is directly related to the final mechanical behaviour of thermoplastic composite structures [48,237]. In the research article of Gao et al. [237], the relation between crystallinity and mechanical performance was deeply studied. They concluded that there should not be a negative impact in the mechanical response of thermoplastic composites caused by a low crystallinity as long as crystallinity values are maintained above 35%. Thus, even though laminates ISC_0 and ISC_45 exhibited lower crystallinity than VBO and hot-press laminates, the drop of mechanical properties observed in ISC laminates which is shown and discussed later in sub-section 3.5.5, should not be attributed to a lower crystallinity, since the values were above the threshold of 35% reported by Gao *et al.* and considered for an optimal mechanical response.

3.5.3 Void content

The results of average void content of the laminates obtained after matrix digestion testing are compiled in Figure 3.24, where a red dotted line has been added representing typical 2% porosity threshold used in aerospace applications.

The results suggest that the laminates fabricated by ISC had a level of porosity around 3%, which is higher than the void content shown by the VBO and hot-press laminates, which were within a porosity range of 0.5-1%. As mentioned in Chapter 1, accepted void content threshold in composites can vary between 1% and 2% for primary structure aerospace applications. 2 % threshold is typically applied; and this is why it was established as reference in Figure 3.24. However, for some other applications, higher porosity levels may be accepted, up to 5%, especially in structures with less mechanical requirements [108]. Also, it has been previously reported that thermoplastic composites can be tougher than thermoset composites, even at higher porosity levels [239].

OoA manufacturing methods are usually associated to higher porosity when compared to traditional autoclave cycles. However, the values of void content reported in this study for VBO and hot-press manufactured laminates can be considered equivalent to the void content achieved during autoclave manufactured.

This low void content achieved by OoA processes is only possible when the manufacturing methods are optimized. Thus, a proper combination of the most relevant manufacturing parameters should be established, combining optimal levels of consolidation temperature, pressure and dwell time, allowing the correct void diffusion and compression during the process.

In their investigation, Zhang et al [18,19] developed a void filling model which determined that a dwell time of 11 min should be enough for obtaining a void-free laminate manufactured by VBO. During the manufacturing of VBO and hot-press laminates included in this chapter, the applied

dwelling times consisted in 15-20 min, which correlate well with the dwelling times reported by Zhang et al.

Due to the nature of the ISC process, it is not possible to reproduce similar dwelling times in such a fast process, where heating, consolidation and cooling down occur in few seconds. The fact that the laminates manufactured under this technology contained higher porosity is directly related to the manufacturing process itself and the void reduction mechanisms. The void reduction mechanisms are directly related to those taking place during consolidation in dwelling times.

As pointed out above, proper (longer) dwelling times may be required to be maintained during VBO and hot-press. During this time, process temperature is held above the polymer melting point in order to facilitate the evacuation of the trapped air at intralaminar regions of the composite (inside the prepreg raw tape) and at interlaminar region (air occluded between different layers). The resin is usually at its lowest viscosity level during this period, therefore void reduction mechanisms such as migration or diffusion may take place, mainly assisted by vacuum or hot-press pressure.

On the other side, AFP-ISC process works significantly different, since the main phenomena occur at very short times. Due to the nature of the process, it is almost impossible for mechanisms of void reduction as migration or evacuation are almost impossible to take place. By contrast, void compression is the mechanism most likely to take place during the consolidation of the tapes assisted by the compaction roller. Thus, the material is melted by the laser irradiation and consecutively compressed by the compaction roller force. Such force is responsible of compressing interlaminar voids occluded in the raw material tapes.

A lamination speed of 1m/min was set during ISC lamination in this study. A laser heating profile length of 130-140 mm was established, resulting in the application of heat during several milliseconds followed by a very-fast cooling down (up to 10000 °C/min). Finally, a compaction force of 50 N was also set for ISC fabrication. These parameters (lamination speed, laser power and compaction force) should be completely optimized for proper void removal during ISC, which represents a challenging investigation. The manufacturing of free-void ISC structures could be achieved by having a good degree of intimate contact and an optimal void evacuation, which are mainly driven by key parameters above mentioned.

In addition to that, the quality of the raw thermoplastic tapes is crucial. Low intralaminar voids, good parallelism between the faces of the tows are essential factors for an optimal final part, but also a good distribution of fibre/matrix. A micrograph of a APC2-AS4 thermoplastic raw tape used in this study is shown in Figure 3.25, in which poor fibre and matrix distribution, significant intralaminar voids and uneven surfaces can be seen. Those factors have been reported in the literature to have a great impact on the final quality of laminates manufactured by ISC. Thus, some studies reported similar porosity levels to the ones reported in this study. For instance, Tierney and Gillespie Jr. [122] reported up to 4% of void content in in-situ consolidated samples and Comer et al. [48] declared a void content of 2.83% in their samples, determined by 2D microscopy. Other manufacturing strategies such as re-pass or higher tooling temperature or other phenomena as void rebound have been reported to impact on the final void content of ISC parts, and it is discussed in the next sub-section, where micro-CT results are presented.

Figure 3.24 Void content of the CF/PEEK laminates manufactured by VBO, hot-press and ISC.

Figure 3.25 Micrograph of an APC2/AS4 raw tape used in this study for ISC lamination [227].

3.5.4 X ray micro-CT

The micro-CT analyses were performed to investigate the inner structure of the manufactured thermoplastic samples. These analyses consisted of the study of voids distribution and voids size, in addition to average void content in order to compare the final quality of the parts when are manufactured by different methods. The results obtained from X-Ray CT scans are presented and discussed in this section.

All the performed CT scans generated a set of images corresponding to projections of the scanned sample. Each set of projections was reconstructed by NRecon software in order to generate a final 3D volume of the sample. Figure 3.26 (i) shows a single X-ray projection of a sample, while Figure 3.26 (ii) shows the reconstruction of the 3D volume performed by NRecon. The software also allowed modifying the brightness and contrast of the reconstructed volume.

Figure 3.26 CT scanning: (i) X-ray projection of a CF/PEEK sample; and (ii) 3D volume reconstruction of the sets of images with NRecon software.

After reconstruction, all the sets of images were processed by ImageJ. The processing consisted of re-orienting, cropping and filtering all the images, in addition to stretching the histograms in order to achieve the best possible contrast and cleanliness.

After the reconstruction with NRecon and the preparation of the image sequences with ImageJ, all the sets of images were then processed for the quantitative analysis of void content, distribution and morphology by using Matlab. Figure 3.27 shows an example of the 3D reconstruction and the void extraction process (voids highlighted in red). Figure 3.27 (i) and (ii) shows the 3D reconstruction of samples VBO_45 and ISC_45, respectively. Void extraction of samples VBO_45 and ISC_45 is shown in Figure 3.27 (ii) and (iv), respectively. The image displays the homogeneous distribution of small voids in sample VBO_45 whereas larger voids may be observed accumulated in the upper layers of ISC_45.

Figure 3.27 3D reconstruction and void extraction process (voids highlighted in red) of two different samples. 3D reconstruction of (i) sample VBO_45; and (iii) sample ISC_45. Void extraction of (ii) sample VBO_45, and (iv) sample ISC_45.

The results of the void distribution analysis are shown in Figure 3.28, where the porosity of each sample is represented in relation to the thickness of the sample. Figure 3.28 (i) shows the void distribution of samples VBO_0, HP_0 and ISC_0; while Figure 3.28 (ii) shows the void distribution of samples VBO_45, HP_45 and ISC_45.

In general terms, a considerable higher void volume can be observed in the ISC samples compared to VBO and hot-press samples, which is in a good agreement with the previously discussed results of matrix digestion. In this case, the information about the porosity can be extended in terms of void location. The figure shows some peaks which correspond to the interphases between layers. This suggests that the porosity of ISC laminates is mainly found at interlaminar locations.

Figure 3.29 shows different views of sample ISC_45 during the image processing and void extraction which suggest the interlaminar location of the voids within the sample. In Figure 3.29 (i), a XZ slice (cross-section) is shown, where voids can be found between plies. Figure 3.29 (ii) shows a XY slice corresponding to the space between layers, where voids can be also found. Finally, Figure 3.29 (iii) shows again a XY slice, but during the binarizing phase, where voids are highlighted in white.

If void distribution results shown in Figure 3.28 of both ISC samples are compared, it can be observed that voids are equally distributed between layers across the thickness of ISC_0; while voids are accumulated in the outer five plies of sample ISC_45. Figure 3.30 shows a XY view of a slice of sample ISC_0, where interlaminar voids can be observed. Low void content was found for all the VBO samples laminates. Regarding HP samples, HP_0 resulted in a very low void content whereas HP_45 exhibited slightly higher void volume in the interply regions of some layers.

The phenomenon of void accumulation in the upper layers of ISC-manufactured structures has been previously discussed in the literature. This has been related to the fact that the external plies are consolidated a fewer number of times compared to the bottom layers [87,122]. Van Hoa et al. [54] proposed a methodology based in the “re-pass” which may assist in the reduction of voids accumulation in the external layers. Re-pass method is based on extra consolidation cycles

without material feeding, only by the application of heat and pressure. In addition to the number of consolidation passes, other factors can affect the accumulation of voids during ISC manufacturing. In this way, when the thickness of the substrate increases during the lamination process, it may act as a heat isolator and the heat may be retained by the composite substrate. This effect may be related with slow cooling down and facilitating a void rebound effect [48].

Figure 3.28 Void distribution (void content as a function of sample thickness) for coupons manufactured by VBO, hot-press and ISC with stacking sequences: (i) $[0]_{16}$; and (ii) $[45/-45]_{4S}$.

Figure 3.29 Different views of sample ISC_45: (i) XZ slice (cross-section), (ii) XY slice corresponding to space between layers; and (iii) XY slice during the binarizing phase, where voids are highlighted in white.

Figure 3.30 Different views of sample ISC_0: (i) XY slice corresponding to space between layers; and (ii) XY slice during the binarizing phase, where voids are highlighted in white.

The results of the performed analysis related to the void size of the samples are presented in Figure 3.31. The graph presents the number of voids (void count) in terms of the void size. Figure 3.31 (i) shows the results of samples VBO_45, HP_45 and ISC_45, while Figure 3.31 (ii) shows the results of samples VBO_0, HP_0 and ISC_0.

It can be deduced from the graph that regardless of the sample and manufacturing method, smaller voids are more common to occur than voids with higher dimensions. Similarities between all samples can be easily observed. Thus, all samples have similar number of smallest voids and there is a decreasing trend of the void count as the void size increases. Regarding the existing differences between samples divided by manufacturing methods, there is a clear contrast between ISC samples and all the other samples. In this way, the decreasing trend of void count with void size is softer in the case of both ISC laminates. This suggests that ISC coupons have voids with larger dimensions. The information of void distribution obtained from the CT scans, along with this information about void size will assist in the discussion of mechanical testing results in the next section. Thus, the fact that large voids have an impact on the mechanical response has been reported in the literature. Wisnom et al. [154] reported that voids may act as stress concentrations and can lead to an early failure of the structure, therefore impacting into the final properties of the part. In this way, the void distribution and void size analysed in this section may play a key role in the mechanical response of the manufactured laminates. Void distribution and size may be related to a decrease in the final mechanical properties of ISC laminates. The mechanical properties of all laminates is presented and discussed in the next section.

Figure 3.31 Void size analysis of the CF/PEEK manufactured samples. Number of voids (void count) as a function of void size of: (i) $[45/-45]_{4S}$ samples; and (ii) $[0]_{16}$.

Finally, a projection of the attenuation of the X-rays in ISC samples is shown in Figure 3.32. Figure 3.32 (i) and (ii) show the attenuation projection of sample ISC_0 and ISC_45, respectively. The data generated by the CT scans allowed plotting the energy attenuation through the thickness of the samples. In this case, the energy attenuation in the XY plane of ISC samples is presented, where a colour representation was chosen in order to highlight the void shapes, which correspond to less attenuated areas (lower density or air in contrast with the density of the composite). It can be observed that voids tend to be located along the fibre direction. In the case of ISC_0, voids can be found in the 0° direction, meanwhile rod-like voids are observed in the 45° and -45° direction of ISC_45. The location of voids in the fibre direction reported here is in line with the conclusions reported by Hernández et al. in [119].

Figure 3.32 Projection of the energy attenuation of the CT scans in the XY plane for samples: (i) ISC_; and (ii) ISC_45. A colour scale is included representing the attenuated energy.

3.5.5 Mechanical characterisation

The results of all the performed mechanical characterization are presented and discussed in this section. The mechanical properties such as ILSS, IPSS, flexure and tensile strength and modulus were evaluated. The main purpose of the mechanical characterisation performed during the work related to this chapter is to investigate the performance of ISC structures in comparison with the performance of the laminates manufactured by VBO and hot-press. In general terms, VBO and hot-press performed similarly with slight variations; meanwhile ISC laminates generally showed lower strength values. Each mechanical test is discussed in detail hereunder.

3.5.5.1 ILSS

All the force-displacement curves of the ILSS testing are compiled in Figure 3.33. In the figure it is compiled each test of every coupon (usually 6 coupons per laminate were tested) of the manufactured laminates (Figure 3.33 (i) – (iii)), along with comparison of a representative coupon of each manufacturing process (Figure 3.33 (iv)). The results of final ILSS strength (in MPa) of the laminates depending on the manufacturing process are shown in Figure 3.34, where a red dotted line has been included indicating the ILSS value corresponding to VBO, which was considered as baseline for this investigation. Also, a summary of the values obtained from the test is compiled in Table 3.5, where ILS strength, peak load and the deviation related to VBO reference are compiled.

It can be observed that VBO and hot-press laminates performed in a similar way. By contrast, the ILSS value of ISC laminate was 71 MPa, which represents a 33.8 % drop compared to the VBO reference (107 MPa). Also, less toughness was exhibited by ISC coupons, since the area behind the curve is smaller for ISC samples.

The ILSS values of VBO and hot-press laminates were at the same level as the ILSS values of 105 MPa reported by Lamontia et al. [90] with same material (APC2-AS4) but autoclave processing. This comparison aims to underline that ILSS autoclave levels can be achieved via out-of-autoclave techniques.

Figure 3.33 Force-displacement ILSS curves of CF/PEEK laminates manufactured by: (i) VBO, (ii) hot-press and (iii) ISC; and (iv) representative comparison between laminates.

Figure 3.34 ILSS values (in MPa) of CF/PEEK laminates manufactured by VBO, hot-press and ISC. Red line refers to the ILSS value of VBO laminate, set as baseline.

Table 3.5 Summary of results of ILSS testing.

Mechanical property		Manufacturing process		
		VBO	Hot-press	ISC
	Strength (MPa)	107 ± 3	109 ± 8	71 ± 2
ILSS	Peak Load (N)	3316 ± 74	3362 ± 227	2228 ± 59
	Comparison to VBO (%)	-	102	66.2

ILSS is a widely used screening test, mainly due to low material and time consumptions. During the interlaminar shear testing, the applied loads are expected to generate transverse shear stresses in the coupon, which will eventually cause a pure interlaminar failure. For a correct understanding of the ILSS drop observed in ISC laminates, it is important to note that ILSS is a matrix-dominated test which is strongly impacted by the presence of manufacturing defects especially voids and porosity, as reported by Wisnom et al. [154]. This study claims to find a relation between the mechanical response drop of ISC laminates and the results of the micro-CT and void content, which is thought to play a key role here. However, the drop of ILSS may not respond to a single reason. Thus, the crystallinity is another factor which has been proven to be related with the mechanical response of thermoplastics. A significant impact of the crystallinity in the mechanical properties has been previously discarded in this chapter (section 3.5.2) due to the achievement of values above 35% in the ISC laminates, which are considered to be within the optimal range.

Regarding the void volume fraction, it is very likely that the void fraction of ISC laminates (3%) in comparison to VBO (less than 1%) had a significant impact on the shear behavior of the laminate. However, micro-CT also allowed to study the distribution and size of the voids. Kastner et al. [138] concluded that voids distribution has a great impact on the mechanical response. As shown in the micro-CT results during the previous section, ISC laminates presented an accumulation of voids at interlaminar regions. The observed interlaminar voids may induce an early critical failure caused by poor adhesion between layers (or low degree of consolidation). This is in line with the results observed in Figure 3.33, where an earlier first failure is observed in the load-displacement curve of the ISC laminate compared with the other laminates.

In addition to that, it is important to note that all the samples experienced a first elastic behavior (linear zone) followed by plastic behavior. If compared with thermoset composites, the plastic region is greater and this suggests that the final failure mode is a combination of interlaminar shear and plastic deformation. This has been previously discussed in the literature; since plastic deformation is a non-valid failure mode in ILSS testing but it always occurs when testing high performance thermoplastic composites. The appearance of plastic deformation may lead to errors in the calculation of the final strength. Thus, a potential reduction of plastic deformation when thermoplastics are subjected to ILS testing was reported by Roselli and Santare [240]. They proposed an interlaminar shear device (ISD) consisting of a modification from the SBS testing causing pure shear loads and therefore avoiding plasticization. The fact that plastic deformation may take place during ILS testing of thermoplastic composites makes it necessary to resort to

another matrix-dominated testing such as in-plane shear stress. The failure modes of the samples tested during this investigation were analyzed and are presented in Figure 3.35. The analysis revealed that all the coupons presented interlaminar shear fracture as observed in the figure, despite possible plastic deformation.

Figure 3.35 ILSS failure mode analysis of CF/PEEK samples manufactured by: (i) VBO, (ii) hot-press (ii); and (iii) ISC.

Overall, the results suggest that ISC laminates which presented higher void content and interlaminar voids in previous sections are experiencing now a reduction in the ILSS. However, higher ILSS values may be achieved if an optimization of the process is investigated leading to a reduction of interlaminar voids. In addition to a process optimization, the use of a higher quality raw material (low intralaminar voids, constant surfaces and optimal fibre/matrix distribution) along with higher tool temperature may also assist in the achievement of higher ILSS. In that respect, Gruber et al. [41] concluded that specific ISC-grade raw tapes are needed for an optimal lamination. Schledjewski and Miaris [85] reported an ILSS of 89.2 MPa by laminating with a tool at 276°C (considerably higher than the temperature applied in this study).

3.5.5.2 IPSS

In this sub-section, the results of IPSS testing are presented. It is important to note that IPSS consisted of the other pure matrix dominated testing performed during this chapter. IPSS testing consists of a [+45/-45] coupon subjected to tensile loads in order to minimize the contribution of the fibre. Thus, a final failure in the matrix is expected.

IPSS force-displacement curves are compiled in Figure 3.36. The test of each coupon batch is presented (images i to iii), along with a comparison of a representative coupon of each batch (iv).

Figure 3.36 Force-displacement IPSS curves of CF/PEEK coupons manufactured by: (i) VBO, (ii) hot-press and (iii) ISC; and (iv) representative comparison between samples.

Average values of IPSS presented in Figure 3.37 (i), while G_{12} shear modulus results are presented in Figure 3.37 (ii). A summary of all the relevant IPSS values is presented in Table 3.6.

Figure 3.37 Experimental IPSS results of CF/PEEK samples manufactured by VBO, hot-press and ISC. Values of: (i) IPSS, and (ii) G_{12} modulus. Red line refers to the ILSS value of VBO laminate, set as baseline.

Table 3.6 Summary of results of IPSS testing.

Mechanical property	Manufacturing process		
	VBO	Hot-press	ISC
Strength (MPa)	222 ± 4	180 ± 12	155 ± 2
ILSS Comparison to VBO (%)	-	81	70
Modulus (GPa)	4.6± 0.1	4.3 ± 0.1	4.0 ± 0.0
Comparison to VBO (%)	-	94	87

The results obtained show that value of the hot-press manufactured laminate (180 MPa) was slightly different from the oven reference (222 MPa), resulting in a reduction of 19%.

The value achieved by the oven reference is especially high, even when compared to the autoclave level of 174 MPa reported by Gruber et al. [41], which matches with the results of the hot-pressed laminate. These results suggested an existent difference in the distribution of fibre-matrix between VBO and hot-press laminates. The fact of applying high pressure during hot-press manufacturing may induce to a more heterogeneous distribution of the fibres. In this way, internal waviness or resin rich areas can appear. Resin rich areas may act as stress concentrators and therefore reduce the final IPSS value. However, whatever the scope between the discrepancies of VBO and hot-press IPSS values, both laminates exhibited optimal IPS behaviour and may be associated to a defect-free behaviour in correlation with the ultrasonic c-scans. On the other hand, the ISC laminate presented a lower IPSS value of 155 MPa corresponding to a 30% drop in relation to the VBO but only a 14% regarding the hot-press laminate and 11% in relation to the autoclave level in [41]. Also, the IPSS value is in strong agreement with the ISC value reported also in [41], where 154 MPa was reported.

All the aspects discussed in the previous section for ILSS testing can be extrapolated here. One of the main reasons causing the IPSS drop of the ISC laminate is related to the void distribution and size observed in the micro-CT results discussed in section 3.5.4. It is widely accepted that void content decreases the mechanical response of composite material structures. Thus, the large elongated voids allocated in the fibre direction that were observed in the ISC laminates may weaken the fibre-matrix adhesion therefore facilitating matrix crack and leading to an early critical failure. A critical failure can be observed for the ISC coupons at a lower load level in Figure 3.36. The fact that the modulus is also lower suggests a more ductile behaviour, normally related with lower degree of crystallinity (more amorphous part). Again, the use of higher grade raw tapes may help to obtain better mechanical results. Also, other manufacturing issues may be faced, as the appearance of residual stresses or a imperfect interlaminar bonding between layers.

3.5.5.3 Flexure

Figure 3.38 shows the load-displacement curves of all the tested flexure coupons. Again in this case, the graph shows not only the curves for each one of the batches (VBO, hot-press and ISC), but also a representative comparison between batches. In all cases, a linear load can be observed up to a load level where the critical failure occurs. During flexure testing, one of the faces of the

coupon is subjected to tensile loads whereas the other side is subjected to compressive loads. The critical failure is expected to take place in the external plies and fibre breakage may be also observed. It can be deduced from Figure 3.38 that VBO and hot-press coupons behaved in a similar way, being loaded up to similar load levels. However, the critical failure of ISC coupons can be observed to occur at an early stage. Even though flexure is mainly considered a fibre-dominated property, there is an obvious effect of the matrix. Thus, a higher void content and a weaker degree of consolidation may impact in the final flexural properties of a composite material. Some of the ISC coupons exhibited a stepped failure during testing (Figure 3.38-c). This can be related with the fact that the failure started but the coupon was still able of being loaded. The failure mode of all coupons was analysed. As a representative example, the failure mode of baseline (VBO) and ISC samples are presented in Figure 3.39. Damage can be observed in the two faces of the samples corresponding to tensile and compressive effects. In addition to that, fibre breakage was also observed. The failure mode was considered valid according to the applied standard. It can be deduced from the failure mode that all specimens showed similar brittle behaviour, which is indicative of similar values of crystallinity. By contrast, Comer et al. [48] reported ductile failure during flexural testing of their ISC laminates. They concluded that it occurred due to a large amorphous part within matrix.

Figure 3.38 Force-displacement curves of CF/PEEK samples subjected to flexural testing. Coupons manufactured by: (i) VBO, (ii) hot-press and (iii) ISC; and (iv) representative comparison between samples.

Figure 3.39 Detail of failure mode of flexural coupons of (i) VBO and (ii) ISC coupons.

The average values of flexural strength and flexural modulus are shown in Figure 3.40 and compiled in Table 3.7. Flexural strength results are shown in Figure 3.40 (i), whereas flexural modulus results are shown in Figure 3.40 (ii).

It can be observed that VBO and hot-press laminates obtained practically same flexural properties with less than 3 % of deviation in both strength and modulus. On the other side, ISC laminate exhibited a drop of 16.9 % of flexural strength in relation to the VBO baseline. As it occurred in ILSS and IPSS, flexural strength of ISC laminates is also lower than VBO and hot-press properties. However, in this case the reduction in relation to the baseline ($\downarrow 16.9\%$) is less than in the cases of ILSS ($\downarrow 29\%$) and IPSS ($\downarrow 30\%$). The reduction of the flexural properties of ISC laminates is again related to the void content and the void distribution of the samples. Even though flexural properties are mainly fibre-dominated as previously mentioned, a sample with porosity is expected to perform worse than a defect-free sample. Thus, the void location analysed during the micro-CT analysis and the approximately 3% of averaged void content can be understood of being responsible for the flexural strength drop. Other studies, such as Fujihara et al. [28] reported reduction in compressive modulus of samples with porosity, which can eventually reduce the flexural properties. The results presented here are in line with the results of Liu et al. [108], who reported a 22% drop of flexural strength in laminates with 3.2 % of void content. The fact that the flexural strength is almost the same in all laminate may be related to a similar degree of crystallization. As earlier discussed, Comer et al. obtained a 17.6 % of crystallization, in contrast with the 36.9 % obtained in this study.

Figure 3.40 Experimental flexural testing results of CF/PEEK coupons manufactured by VBO, hot-press and ISC: (i) flexural strength; and (ii) flexural modulus. Red line refers to the value for the VBO laminate, set as baseline.

Table 3.7 Summary of results of flexural testing.

Mechanical property		Manufacturing process		
		VBO	Hot-press	ISC
Flexure	Strength (MPa)	1812 ± 100	1856 ± 45	1505 ± 104
	Comparison to VBO (%)	-	102.5	83.1
	Modulus (GPa)	125 ± 4	127 ± 5	120 ± 2
	Comparison to VBO (%)	-	101.7	96.5

3.5.5.4 Tensile

Figure 3.41 shows the load-displacement curves of all the performed tensile tests. The curves of each coupon batch is presented (figures i to iii), along with a comparison of a representative coupon of each batch (iv).

All the coupons behaved in a similar manner, in such a way that a lineal progression can be observed until a critical failure happening when the coupon starts to break. This is mainly due to the fact that tensile test is a pure fibre-dominated test. The loads are support by the fibres until explosive fibre breakage takes place. Thus, the effect of the matrix in tensile testing is minimal. However, it can be observed that VBO and hot-press held similar load levels above 50 kN, whereas ISC coupons did not reach such load levels, therefore an earlier critical failure could be observed within a 40-45 kN scenario.

Figure 3.41 Force-displacement curves of CF/PEEK coupons subjected to tensile testing. Coupons manufactured by: (i) VBO, (ii) hot-press and (iii) ISC; and (iv) a representative comparison between samples.

The earlier critical failure observed in the ISC coupons had a direct impact on the final tensile strength values, which are shown in Figure 3.42 and compiled in Table 3.8. Tensile strength and modulus results are shown in Figure 3.42 (i) and (ii), respectively.

Again in this case, VBO and hot-press laminates obtained approximately similar values with less than 5 % difference. On the other side, ISC laminates exhibited a drop of 13.2 % in the tensile strength but only a 2.2 % drop of tensile modulus, which was considered as not relevant. As mentioned before, tensile is a pure fibre-dominated testing but other authors have reported in the past a drop of tensile strength in composite samples with porosity. This is the case of Liu et al. [108], who reported a drop of 20% of tensile strength in laminates with 3 % of void content, which are very similar results to the ones reported in this study. Interlaminar voids as the ones observed during CT analysis may cause a bad adhesion between fibre and matrix which can directly affect to the tensile properties. However, the drop of tensile strength may be also affected by other issues which occur after ISC manufacturing. Those issues may include the residual stresses and the in-plane fibre waviness which generally appear in ISC-manufactured structures. Even though residual stresses are not the main topic of this study, its impact cannot be neglected, and their negative effect has been previously exposed in the literature. Thus, Van Hoa et al. [54] explained how mismatches in thermal expansion coefficients in different directions of the tape, temperature gradient, shrinkage and crystallization can lead to final residual stresses in the final structure. They proposed a fabrication methodology using tooling heated up to 250 °C when working with CF/PEEK. In this study, we used a tooling heated up to 200 °C during the lamination of ISC laminates but the effect of residual stresses could be still observed. Due to time and cost limitations of the projects, more laminates were not manufactured, but a future work with

higher tooling temperatures is proposed. The averaged values showed in Table 3.8 indicate that the tensile modulus of all laminates was practically identical. These values are mainly driven by the carbon fibre nature and are not changed by the manufacturing process.

Figure 3.42 Experimental tensile testing results of CF/PEEK laminates manufactured by VBO, hot-press and ISC: (i) tensile strength and (ii) tensile modulus. Red line refers to the value for the VBO laminate, set as baseline.

Table 3.8 Summary of results of tensile testing.

Mechanical property		Manufacturing process		
		VBO	Hot-press	ISC
Tensile	Strength (MPa)	2127 ± 44	2228 ± 72	1847 ± 81
	Comparison to VBO (%)	-	104.7	86.8
	Modulus (GPa)	139 ± 6	144 ± 3	136 ± 6
	Comparison to VBO (%)	-	103.6	97.8

3.6 Conclusions

In order to compare the quality and properties of thermoplastic structures manufactured under different manufacturing methods, a set of CF/PEEK laminates was manufactured by VBO, hot-press and ISC technologies and subjected to NDT, physical-chemical and mechanical tests. The test plan included ultrasonic NDI, void content (matrix digestion), DSC, X-ray micro-CT and a set of mechanical testing (ILSS, IPSS, tensile and flexure). In general, VBO laminates were set as reference (or baseline) for this study in order to compare. Table 3.9 compiles all the relevant results achieved after performing all the tests. Also, a relation of all the properties with the VBO laminate (set as baseline in this study) has been included in brackets. The property was considered equal ($\approx$) when a deviation less than 5 % was observed. In the case of quantitative properties. Symbol ↓ was used if the measured property presented a worse value than VBO and ↑ was used in the case that the property was improved.

The ultrasonic NDI revealed that all the VBO and hot-press laminates may be considered defect-free according to typical aerospace standards (mainly 6 dB threshold) but ISC laminates presented some indications suggesting accumulation of air and higher porosity. The void content analysis revealed that both VBO and hot-pressed laminates had a porosity level around 1 % while ISC laminates exhibited a higher porosity up to 3%. Additionally, the micro-CT analysis allowed studying not only the average values of void content, but also the position of the voids (distribution) and voids size. The analysis revealed that voids tend to accumulate at interlaminar locations in ISC samples. The mechanisms of void evacuation in composites have been discussed. Due to the nature of the ISC process, the voids should be compressed by the compaction loads applied by a roller, which must act in very short periods of time when the resin is melted. This is a very complex process which should be optimized in order to reduce the final void content in ISC-manufactured structures. Also, as other authors have pointed out in their studies, a specific raw ISC-grade material containing an optimal fibres/matrix distribution and good parallelism between tape faces would easily improve the final quality of the parts [45,65,68,68].

Regarding the DSC analysis, all the laminates showed final crystallinity levels above 35 %, which are considered optimal and may not affect the final mechanical response [237]. However, a slightly lower value (↓8 %) was achieved by the ISC laminates in comparison with the oven reference. This may lead to a slightly more ductile behaviour of the structure, therefore reducing the modulus of some matrix-dominated properties.

Regarding the mechanical analysis, the most relevant mechanical properties obtained in this chapter are also shown in the table. Overall, it can be said that VBO and hot-press laminates performed similarly except for the IPSS strength, which was especially higher for the VBO manufacturing. As before discussed, that value can be taken as particularly high even when compared to autoclave levels [41]. Regarding ISC mechanical response, the laminates experienced a reduction of approximately 30 % in the strength of matrix-dominated properties (ILSS and IPSS) and an approximated 15 % drop in the fibre-dominated properties (flexure and tensile). The higher drops found in matrix-dominated properties is related to the void content and distribution analysed during the CT testing. Thus, a higher void content at interlaminar and intralaminar locations and larger voids are the main reasons for the drop of matrix-dominated properties of ISC laminates. The drop of IPSS modulus suggests that the matrix of ISC matrix had a more ductile behaviour, mainly attributed to a large amorphous matrix proportion. Regarding the fibre-dominated properties, a smaller drop in the properties was expected compared to the properties driven by the matrix. Even at fibre-dominated properties, the drop was expected to be different to zero since porosity has been proven to also have a negative impact in such properties [108]. Also, other issues such as residual stresses or waviness may affect.

Table 3.9 Summary of mechanical properties of all laminates.

		Manufacturing process		
		VBO	Hot-press	ISC
Ultrasonic NDT				
(6 dB threshold)		✓	✓ (≈)	↓ (Indications above 6 dB)
DSC (Crystallinity)				
Degree of crystallinity		40.1	42.7 (↑ 7 %)	36.9 (↓ 8 %)
Void content				
Average values (%)		1	0.8 (↑)	3 (↓↓)
Micro-CT				
Void distribution		✓	✓ (≈)	↓ Interlaminar and intralaminar accumulation
Void size		✓	✓ (≈)	↓ More and larger voids
Mechanical testing				
ILSS	Strength (MPa)	107	109 (≈)	71 (↓↓ 33.8 %)
	Modulus (GPa)	4.6	4.3 (↓ 6 %)	4.0 (↓ 13 %)
IPSS	Strength (MPa)	222	180 (↓ 19 %)	150 (↓↓ 30 %)
	Modulus (GPa)	4.6	4.3 (↓ 6 %)	4.0 (↓ 13 %)
Flexure	Strength (MPa)	1812	1856 (≈)	1505 (↓ 16.9 %)
	Modulus (GPa)	125	127 (≈)	120 (≈)
Tensile	Strength (MPa)	2127	2228 (≈)	1847 (↓ 13.2 %)
	Modulus (GPa)	139	144 (≈)	136 (≈)

Values between brackets () refer to the relation with VBO properties, set as the baseline.
(≈) is used when the property remained approximately equal.

(↑) is used when the property improved.

(↓) is used when the property decreased.

One of the technical goals of the presented thesis is to support the application and research of thermoplastic composites in the aerospace industry. Thus, after analysing all the performed tests, IPSS mechanical testing appears to be an optimal candidate for evaluating the quality of a thermoplastic manufacturing process. It can be concluded that the quality of the thermoplastic manufacturing process will directly impact on the quality and strength of the bonding between plies, the amount of air entrapped in and within layers, as well as the final crystalline structure of the polymer. All these factors will in turn impact on the final mechanical behaviour of the manufactured part, so the choice of a good screening mechanical test is mandatory. Matrix-dominated tests are the ones that should be applied for this purpose. However, among the properties analysed in this study, IPSS is the one which is finally suggested since the results are more reliable when compared to ILSS testing, where a non-valid failure mode (plastic deformation) is more prone to occur. Furthermore, IPSS testing will not only provide data about

the in-plane shear strength (which eventually gives information about the goodness of the matrix quality) but also will add information regarding the degree of crystallinity if the shear modulus is analysed.

Chapter 4

Effect of void content on thermoplastic composites

4.1 Introduction

In this chapter, it is presented the impact of void content in CF/PEEK thermoplastic composites manufactured by the three different manufacturing methods under investigation in this PhD project, along with correlations of void content with both ultrasonic NDT testing and mechanical response. As previously mentioned, one of the main goals of this work is to support the application and research of the thermoplastic composites for the industry. Within this context, some of the main activities before incorporating a new material into the industry are engineering and certification activities. These activities may involve the study of the impact of the defects (such as voids) in the structural behaviour, the ultrasonic acceptance criterion or the limit loads that a material will eventually withstand, among others.

In order to make a proper contribution, this chapter proposes three different methodologies for manufacturing thermoplastic laminates with induced manufacturing defects (voids/porosity) which will finally have an impact on industrial quality control tests.

The investigated methodologies for manufacturing laminates with production induced-defects were based on the studied void creation and migration mechanisms and consisted of:

- (i) Sealing the perimeters (edges) of the stacked prepreg laminates before consolidation in order to avoid void migration.
- (ii) Submitting the preform to moisture absorption before being consolidated in order to create voids from evaporated water.
- (iii) Deviation of the optimal consolidation cycle: Modification of the key manufacturing parameters.

Those manufacturing methodologies were tested and a down-selection was performed. Only one manufacturing strategy was selected for the continuation of the research and the manufacturing of laminates with induced porosity.

The selected methodology was based on the deviation of the consolidation cycle parameters and a manufacturing plan was then created in order to have a population of laminates for the study. A test plan was defined, including similar tests to those presented in chapter 3 for the evaluation of the laminates quality. The performed tests consisted of typical quality control tests for composites, as did in the previous chapter. The test plan included ultrasonic NDT, void content analysis, DSC and matrix-dominated mechanical testing. In agreement with conclusions of Chapter 3, IPSS test was defined as a good candidate for evaluating the suitability of a thermoplastic manufacturing process and the final matrix quality and bonding between plies. For this reason, IPSS is the primary mechanical test performed during this chapter. However, ILSS testing was also performed (not meeting the standard due to stacking sequence) in order to have more valuable information.

In summary, a set of laminates was manufactured with a different range of porosity by following the selected strategy consisting of the modification of the key manufacturing parameters. The laminates were characterised; and intercorrelation was established between porosity, ultrasonic attenuation and mechanical performance.

This chapter and the results presented here consisted of an extended version of the scientific article published by the author, entitled “*Effect of processing parameters and void content on mechanical properties and NDI of thermoplastic composites*” [87].

4.2 Methodologies for manufacturing laminates with induced defects

Before presenting the experimental methods section, it is important to present first the investigated manufacturing methodologies for defect-induced laminates. Those methodologies consisted of: (i) sealing the perimeters of the raw prepreg laminates; (ii) moisture conditioning, and (iii) modification of key parameters of the consolidation cycle. Same material (APC2/AS4), manufacturing technologies and procedures to those described in chapter 3 were applied for the laminates manufacturing.

After this stage of the project, only one manufacturing strategy was selected for the continuation the research. The investigated methodologies, along with the final decision made, are presented in detail hereunder.

4.2.1 Laminates with edges perimeter sealed

It is accepted that phenomena as air/gas diffusion and permeability affect the void dynamics into the thermoplastic composites and impact the final void content in the fabricated part, especially when using out-of-autoclave processes where an homogenous external pressure is not applied (as VBO) [18,19,241,242]. A methodology based on sealing the perimeter edges of a preform has been proven to be a successful strategy for creating laminates with porosity [19]. Thus, the in-plane evacuation of the voids is limited during the consolidation cycle and the voids are forced to keep inside the laminates. With this purpose, different 300×300 mm samples with a different percentage of total sealed perimeters were manufactured by VBO to study the feasibility of the methodology. The selected stacking sequence was $[0/90]_{4S}$ to avoid unidirectional laminates. The manufacturing set-up is shown in Figure 4.1.

Figure 4.1 Manufacturing configuration of laminates with sealed edges.

The applied consolidation cycles consisted of a heated-up ramp of approximately $2\text{ }^{\circ}\text{C}/\text{min}$ up to a consolidation temperature of $400\text{ }^{\circ}\text{C}$ during a dwell time of 10 min. Table 4.1 compiles the manufacturing information of the samples with sealed edges. The acronym SE given to the specimens refers to Sealed Edges (SE).

Table 4.1 Manufacturing details of CF/PEEK laminates with sealed edges.

Specimen ID	Dimensions (mm)	Stacking sequence	Percentage of sealed edges (%)
SE-1 (baseline)			0
SE-2	300×300	$[0/90]_{4S}$	≈ 50
SE-3			≈ 70
SE-4			≈ 90

An sketch of the prepreg laminates (before consolidation) with sealed edges is shown in Figure 4.2 (left), while the real laminates before consolidation are presented in same figure (right). The consolidated manufactured laminates (after consolidation) are presented in Figure 4.3.

Figure 4.2 Sketch of laminates with different percentage of sealed edges (left). Laminates (before consolidation) with the tacky tape used for sealing the edges (right).

Figure 4.3 Laminates SE-1 to SE-4 with a different percentage of sealed edges, after consolidation.

The validity of this methodology was evaluated by means of ultrasonic NDT following similar procedures to the ones described in previous section 3.4.1. Ultrasonic testing – reflector plate technique using a frequency of 5 MHz in total immersion was applied. The attenuation c-scans generated by the inspection system are shown in Figure 4.4. If the c-scans are analysed, it can be easily observed that the laminates with sealed edges presented attenuated regions due very likely to the air occluded into the specimen. Laminate SE-4 presented a total attenuation of the signal near the 18 dB, suggesting a high level of porosity. SE-2 and SE-3 presented similar c-scans. It is important to note that the central region of those laminates presented low attenuation, while the attenuated regions were located closed to the edges. This suggests that air/volatiles were able to move from the central region during the cycle vent, but the tacky tape acted as a dam leading to porosity regions accumulated in the edges of the plates. It is very likely that strategies followed for laminates SE-2 and SE-3 may lead to void-free samples if longer dwell times were applied. As expected, laminate SE-1 presented a c-scan free of indications suggesting a low level of porosity.

The performed ultrasonic testing allow to confirm that the proposed manufacturing methodology is valid for manufacturing defect-induced thermoplastic laminates. However, the approach followed here based on sealing the edges of the preforms may not be extrapolated to the others investigated manufacturing process in this thesis (hot-press and ISC fabrication). The selection of a strategy for manufacturing defect-induced defects is given in section 4.2.4.

Figure 4.4 C-scan of samples SE-1 to SE-4.

4.2.2 Moisture absorption

The second approach for manufacturing defect-induced specimens investigated in this study was based on the moisture absorption by the raw prepreg laminates. It is well known that defects in composites can be derived from multiple factors, and moisture accumulation in the raw composite material is one of those aspects. In a real production environment, the moisture intake by a raw prepreg is mainly related with the cold storage in fridge or simply the long-time exposure of the material when it is located a taping machine or a cutting table. When compared to the moisture absorption by thermoset composites, thermoplastic composites are characterized by low moisture absorption, with typical saturation values around 0.1 %.

For the study of this approach, a set of 75×75 mm non-consolidated laminates was used with different number of plies, in order to study the effect of number of plies in the humidity intake. The used samples and configurations are compiled in Table 4.2. The “MA” naming given to the specimens refers to Moisture Absorption. Due to the lack of tacking of thermoplastic composites, the samples were stacked by using a manual welder, as depicted in Figure 4.5.

Table 4.2 Configuration of all the samples subjected to moisture absorption.

Specimen ID	Dimensions (mm)	Number of plies	Stacking sequence
MA-1	75 × 75	1	[0]
MA-2		2	[0/90]
MA-3		4	[0/90] _s
MA-4		8	[0/90] _{2s}
MA-5		16	[0/90] _{4s}

Figure 4.5 Preparation of samples for moisture absorption.

All samples were located into a climatic chamber located at FIDAMC facilities. The moisture absorption was performed following EN 2823 standard [243], where the procedure for H/W conditioning is indicated. Typical H/W conditions of $(85^{+5}_{-1})\%$ humidity and exposure temperature of 70 °C were selected. The condition of equilibrium is reached when the weight difference between three consecutives measurements comply equation (4.1), where M_j represents the value of the weighting at a time j .

$$\left[\frac{M_{j-2} - M_j}{M_j} \right] \leq 5 \times 10^{-4} \quad (4.1)$$

The specimens were weighted every 72 hours according to FIDAMC laboratory background. For each weight measurement, the coupons were taken out of the chamber and dried superficially with a paper towel to remove condensation. The time required until the specimen reached room temperature (RT) was waited. The evolution of moisture absorbed by the samples is given in

Figure 4.6. The evolution of the mass M_j (mg) of each coupon can be observed as a function of time in Figure 4.6 (i) to (v), while the evolution of mass increase (in %) of all samples is given in Figure 4.6 (vi).

Figure 4.6 Moisture absorption of CF/PEEK preforms: (i) to (v) Moisture absorption of samples MA-1 to MA-5; (vi) moisture absorption (in %) of all samples.

The results showed that all coupons absorbed a total amount of moisture less than 0.1 % at the saturation point. If compared with the values reported in the literature, it can be found that PEEK composites are typically characterized by low moisture absorption. Thus, Buchman and Isayev [244] reported low water absorption of 0.2% when APC2-AS4 reached saturation level after approximately 624 hours (26 days) of water immersion. Beland [245] reported a figure where moisture absorption of different polymer composites was shown (Figure 4.7). Those results are in a good agreement to the ones presented in this study, since APC2-AS4 is a semicrystalline thermoplastic and their water absorption is in the range of 0.2 %. Beland also came to the conclusion that PEEK based composites may be insensitive to moisture even when hot.

Figure 4.7 Moisture absorption of different polymer composites [245].

The results obtained during this sub-chapter suggested that the impact of moisture absorption in the thermoplastic raw material may not be big enough to cause laminates with significant void content. Due to this assumption, laminates were not consolidated after moisture absorption of the raw material. This manufacturing approach for defect-induced laminates was discarded.

4.2.3 Modification of processing parameters

Finally, an approach for manufacturing defect-induced laminates based on the deviation from the optimal key manufacturing parameters was proposed. This methodology is based on the fact that deviations from optimal parameters such as temperature may lead; for instance, to a higher viscosity of the resin avoiding a correct void migration and evacuation. Other inconvenient parameters such as low pressure may lead to a non-correct intimate contact between layers and deconsolidation. This manufacturing strategy is commonly used in composites research in order to investigate the optimal cure cycle or the impact of void content. For instance, Hernández et al. [118,119] investigated the relationship among the applied curing cycle with the later void content inside the laminate and the mechanical response. However, as highlighted several times in this study, many studies can be found in the literature regarding this topic for thermoset composites, while little information can be found regarding thermoplastic composites.

In order to validate this manufacturing approach for thermoplastic composites, three APC2-AS4 laminates were manufactured in oven by applying a modification in the consolidation temperature. The details of the manufacturing parameters of the three laminates are compiled in Table 4.3. According to material supplier, optimal consolidation temperature consists of 390 ± 10 °C under assisted vacuum. Vacuum should be maintained until temperature is below T_g during cooling down.

Table 4.3 Manufacturing details of laminates with deviations from optimal parameters.

Specimen ID	Dimensions (mm)	Stacking sequence	Consolidation temperature (°C)	Pressure	Dwell time
PAR-1 (baseline)	300 × 300	[+45/- 45] _{4S}	400	1 atm (vacuum bag)	15 min
PAR-2			370		
PAR-3			350		

PAR: Parameters modification

The nominal consolidation cycles carried out in this section are depicted in Figure 4.8. All laminates were consolidated by waiting the same dwell consolidation time of 15 minutes and applying the same pressure. In VBO manufacturing, the pressure is given by the vacuum pumps, which ideally produce 1 atm of pressure. Regarding the consolidation temperature, 400 °C was set as optimal temperature. Another two inconvenient (lower) consolidation temperatures were tested in this case. Those temperatures were 370 °C (medium temperature) and 350 °C (low temperature).

Figure 4.8 Different consolidation cycles used to consolidate laminates with induced porosity by modifications in the processing temperature.

As before mentioned, a lower processing temperature is expected to cause a higher viscosity of the resin and therefore less evacuation of voids. This polymer behaviour (and PEEK resin behaviour, in particular) has been widely investigated and reported by several authors. For instance, Yan et al. [246] investigated the rheological behaviour of PEEK under different isothermal conditions from 350 °C to 380 °C and reported clear differences in the viscosity at those temperatures, as can be observed in Figure 4.9. It was demonstrated that temperatures closed to the PEEK melting point (343 °C) are related to more viscosity of the resin. The closer the temperature is to the optimal processing temperature (390 ± 10 °C), the less viscosity is the resin. Besides, almost similar behaviour can be observed for the samples tested at 370 °C and 380 °C.

Figure 4.9 Zero-shear viscosity of PEEK under different isothermals [246].

Again in this case, the applied proceeding in order to validate this manufacturing approach was based in ultrasonic NDT inspection. Same inspection procedure based on water immersion using double through-trasmission with a 5MHZ probe and a reflector plate was performed. The c-scans of the laminates with the corresponding attenuation scale (in dB) are shown in Figure 4.10.

Figure 4.10 Ultrasonic c-scans of laminates manufactured with modified cycles: PAR-1 at 400 °C, PAR-2 at 370 °C and PAR-3 at 350 °C.

The c-scans show clear differences among the inner quality of the laminates. If the c-scans are evaluated by using the typical 6 dB acceptance criterion, it can be assumed that laminate PAR-1 consisted of a defect-free laminate and therefore the processing temperature of 400 °C can be considered optimal (as suggested by the material supplier) to process APC2-AS4 not only in autoclave, but also in VBO without the aid of external pressure. However, the other two laminates presented attenuated areas over 6 dB that can be considered defects. In the case of PAR-2, the results suggested that a temperature of 370 °C allowed the extraction of most of the air from the laminate, but there are also some regions which show porosity. Those regions can be observed forming a dark lattice in the c-scan. Finally, laminate PAR-3 presented a large dark region corresponding to a high attenuation of the ultrasonic wave. This suggests that a large amount of air could not be extracted during the consolidation process.

Based on this analysis, it can be concluded that the manufacturing of defect-induced laminates by making modifications in the consolidation cycle is a successful methodology.

4.2.4 Selection process of defect-induced manufacturing strategy

After the manufacturing tests performed for the investigation of manufacturing strategies of defect-induced laminates, a decision was made for the continuation of the project. Table 4.4 compiles the most relevant conclusions obtained from the results of the manufacturing trials. Thus, two of the manufacturing strategies were led to the point of laminates manufacturing (sealing edges and modification of parameters), meanwhile the other strategy (submitting the raw material to moisture absorption) was discarded. The reason why moisture absorption was discarded was based in the fact that the APC2/AS4 absorbed less than 0.1 % of moisture at saturation level and was considered not enough for impacting the final void content and the laminate mechanical response.

Regarding the other two manufacturing strategies, both shown promising results for manufacturing laminates with a different void content which compromise the final mechanical behaviour. However, the strategy based on sealing the perimeter edges of the preform can be

extrapolated to hot-press manufacturing but may not be applied for ISC manufacturing due to the automated lay-up process. Besides, the methodology based on modifying the manufacturing parameters may lead to an in-depth study since more correlations can be made, for instance the relation between manufacturing parameters and void content or with final mechanical response of the laminates.

For all the reasons discussed, modification of processing parameters was selected as the most convenient manufacturing methodology for the present study and was applied for the rest of the research of this chapter.

Table 4.4 Summary of conclusions during testing of induced-defects manufacturing strategies.

Methodology for induced-defects manufacturing	Manufacturing method tested	Defects induced?	Can be extrapolated to hot-press and ISC?
Perimeters sealing	VBO	Yes	No
Raw material moisture absorption	None	N/A	Yes
Deviation from consolidation cycle	VBO	Yes	Yes

4.3 Experimental method

After selecting a manufacturing strategy of defect-induced laminates, an experimental plan was designed in order to investigate the existing correlations between void content, ultrasonic attenuation coefficient and mechanical response. The next sub-sections compile all the information regarding the performed tests.

4.3.1 Laminates manufacturing

A set of laminates was manufactured using APC2-AS4 CF/PEEK; with same properties as described in section 3.2. All the laminates manufactured for the investigation compiled in this chapter had a configuration consisting of 300×300 mm, 16 plies and a lay-up of $[45/-45]_{4S}$. The $+45/-45$ lay-up was chosen based on the conclusions of chapter 3. Thus, the mechanical test selected as the most convenient for evaluating the quality of the thermoplastic laminates was IPSS.

The laminates were manufactured using the three different OoA methods under research in this thesis (vacuum bag in oven, hot-press and AFP with in-situ consolidation). The manufacturing procedures were described in previous section 3.3.1, section 3.3.2 and section 3.3.3. However, in this case, the key parameters were modified from the optimal cycle in order to have a set of laminates with different levels of porosity and therefore to investigate the relation between void content, ultrasonic NDT and mechanical properties. Processing temperature and pressure were selected as the main parameters governing the process and therefore they were modified (when possible) for obtaining porosity laminates. Hence, consolidation temperature was modified in the

laminates manufactured by VBO, meanwhile temperature and also pressure were modified in hot-press and ISC laminates. Effect of pressure was not studied on VBO laminates, since only vacuum vent was applied during manufacturing. Laminate ISC430 presents a different compaction force since a different compaction roller was used during manufacturing in order to avoid roller degradation during lamination due to high temperature. The roller consisted of a ceramic roller, based on previous experience of degradation in elastomeric rollers when applying high laser powers. Nevertheless, the final applied pressure was the same in all ISC laminates.

Table 4.5 compiles all the relevant information regarding the manufacturing of the laminates carried out in this section. A code was assigned to all the laminates indicating the manufacturing process, such as vacuum bag only (VBO), hot-press (HP) and AFP-ISC (ISC). Laminate VBO400 was considered as baseline in further discussion of results.

Nine laminates were manufactured by VBO under different temperature conditions, from low temperatures (close to the melting point) to high processing temperatures up to 460 °C. Other parameters were maintained for all the VBO laminates, as 15 min of dwell time and constant assisted vacuum. After consolidation, the oven was open and the laminates cooled down up to room temperature, at an approximate rate of 2 °C/min. The temperature was monitored with an embedded thermocouple and controlled in the oven control panel.

In the case of hot-press manufacturing, a total of eight laminates were consolidated by making deviations in the applied temperature or pressure. The range of covered temperatures was from 340 °C up to 500 °C. The upper limit temperature was higher than in VBO laminates since tacky tape is not required for hot-press manufacturing. The set of applied effective pressures was 0.2, 0.7 and 1.5 MPa. Dwell time was the same for all the laminates (15 min) and the laminates were cooled down at room temperature after consolidation with maintained pressure until T_g . The temperature was monitored with an embedded thermocouple.

Finally, six laminates were fabricated by ISC method. Temperature and pressure applied were modified from optimal processing conditions. Temperature was controlled manually during lamination. This was made by an experienced operator who adjusted the laser power according to the IR camera measurements. Regarding the pressure, it was established by the compaction roller force. Thus, averaged laser powers of 160, 180, 230 and 300 W were applied expecting to cause a temperature profile in the proximities of 350 °C, 370 °C, 400 °C and 430 °C, respectively. The different compaction loads applied by the silicone roller consisted of 200, 500 and 800 N. Those loads caused final pressures of 0.8, 2 and 3.2 MPa, respectively. Regarding the ceramic roller, it applied a load of 10 N, leading to a final pressure of 2 MPa. The control panel of AFP-ISC system for laser power and compaction load is shown in Figure 4.11.

Table 4.5 Manufacturing plan of defect-induced laminates.

Laminate code	Parameter modified	Consolidation temperature	Dwell time	Vacuum	Cooling rate	
		(°C)	(min)	(mbar)	(°C/min)	
VBO400	none (optimal)	400	15	100	2	
VBO340	Temperature	340	15	100	2	
VBO350		350				
VBO360		360				
VBO370		370				
VBO380		380				
VBO390		390				
VBO430		430				
VBO460		460				
Laminate code	Parameter modified	Consolidation temperature	Dwell time	Applied pressure	Cooling rate	
		(°C)	(min)	(MPa)	(°C/min)	
HP400	none (optimal)	400	15	0,7	2	
HP340	Temperature	340	15	0,7	2	
HP370		370				
HP430		430				
HP460		460				
HP500		500				
HP400_0.2	Pressure	400	15	0,2	2	
HP400_1.5		400		1,5		
Laminate code	Parameter modified	Temperature at nip point	Tooling temp	Load force	Cooling rate	Lay-up speed
		(°C)	(°C)	(N)	°C/min	m/min
ISC400	none (optimal)	380-400	200	500	10000	1
ISC350	Temperature	340-360	200	500	10000	1
ISC370		360-380		100		
ISC430		420-430		100		
ISC400_20	Pressure	380-400	200	200	10000	1
ISC400_80		380-400		800

Figure 4.11 Laser power and compaction force control panel of laser-assisted AFP-ISC equipment.

The number of laminates manufactured in this study was mainly limited by the total budget and time of the project, along with the quantity of available material and the accessibility to some of the manufacturing facilities.

4.3.2 NDT

All the laminates were subjected to ultrasonic NDT inspection after manufacturing. Automated double through transmission with 5 MHz and full immersion was used, as described in section 3.4.1. All the laminates were inspected following same inspection conditions.

In this chapter, discrete values of the sonic attenuation of the laminates were required in order to perform the correlation analysis between magnitudes. With this aim, ImageJ software was used for image analysis, since the Tecnitest software did not provide those average attenuation values. ImageJ can generate a colour histogram of a given figure in grey scale. Grey colour figures are composed of a combination of 255 grey colours from black to white. All the generated c-scans were processed with ImageJ in order to have an averaged attenuation value (in dB). An example of a created histogram of a c-scan is shown in Figure 4.12. According to the attenuation scale of a c-scan, the white end of the histogram scale corresponded to a less sound attenuated signal while the black end corresponded to more sound attenuation. Also, narrower histograms were related to more homogeneous c-scans, meanwhile wider histograms indicated more heterogeneous c-scans, caused by areas with porosity (or any defect).

Figure 4.12 Grey-scale histogram of an ultrasonic c-scan, generated by ImageJ processing.

4.3.3 Void content assessment

All the laminates were subjected to void content analysis to investigate the porosity of each laminate. In order to perform a comparison among characterisation techniques, different void content assessment tests were used, including liquid immersion method (also called densities or Archimedes), matrix digestion, and 2D microscopy were applied to estimate the void content within the CF/PEEK specimens.

A minimum of three coupons sized 20 x 10 x 2.16 mm were extracted from each plate for void content analysis. Regarding the location of the extracted sample, the ultrasonic c-scans were used to select different areas in order to have a population of coupons representative of the whole laminate. For those laminates with more heterogeneous c-scan which exhibited different attenuated areas, the number of analysed coupons was increased to obtain reliable and representative results.

The same coupons extracted from each laminate were tested under the three methods. Thus, the order of performing the tests was: first of all, immersion method was applied since consists of a non-destructive test. After immersion method, 2D microscopy was performed to one cross-section of the sample, which required only surface preparation. Lastly, matrix digestion analysis was performed since the coupon is destroyed during test.

Further details about each test are shown below.

Immersion method (Archimedes)

Density and void volume fraction of all the samples were measured. The procedure was carried out following ISO 1183-1 (Method A) [247]. For this purpose, a precision balance model XP205 Delta Range Analytical Balance supplied by Metler Toledo was used. A glass with distilled water was also required. The weight of each coupon was measured inside and outside the water, as depicted in Figure 4.13.

Figure 4.13 Weight measurements of CF/PEEK samples: (i) in air; (ii) inside water.

The density of each coupon was calculated according to expression (4.2), where $m_{c\ air}$ refers to the coupon weight in air, $m_{c\ water}$ refers to the coupon weight in water, ρ_c refers to the coupon density and ρ_o refers to the density of distilled water at test temperature. Distilled water density value at room temperature is 0.99756 g/cm^3 .

$$\rho_c = \frac{m_{c\ air} \cdot \rho_o}{m_{c\ air} - m_{c\ water}} \quad (4.2)$$

After density calculation, the fibre volume fraction can be calculated by following expressions (4.3) to (4.5), where V_f is the fibre volume fraction, V_c is the coupon volume which is given by the surface (S) multiplied by the thickness (t). FAW corresponds to the Fiber Areal Weight of the prepreg, m_f and ρ_f correspond to the fibre mass and density, n_{plies} is the total number of plies of the composite,

$$V_f (\%) = \left(\frac{V_f}{V_c} \right) \cdot 100 \quad (4.3)$$

$$V_f = \frac{m_f}{\rho_f} = \frac{FAW \cdot n_{plies} \cdot S}{\rho_f} \quad (4.4)$$

$$V_f (\%) = \frac{FAW \cdot n_{plies}}{\rho_f \cdot t} \cdot 100 = \frac{FAW \cdot n_{plies} \cdot S \cdot \rho_c}{m_{c\ air} \cdot \rho_f} \cdot 100 \quad (4.5)$$

Resin volume fraction can be also estimated by expressions (4.6) to (4.9), where previous fibre volume fraction is used, and V_r is the resin volume fraction, ρ_r is the theoretical resin density, and m_r is the resin mass.

$$\frac{\rho_c}{\rho_r} = \left(\frac{m_c/V_c}{m_r/V_r} \right) \rightarrow V_r = \frac{m_r}{m_c} \cdot V_c \cdot \frac{\rho_c}{\rho_r} \quad (4.6)$$

$$m_r = m_c - m_f \quad (4.7)$$

$$V_r = \frac{m_c - m_f}{m_c} \cdot V_c \cdot \frac{\rho_c}{\rho_r} = \left(1 - \frac{m_f}{m_c}\right) \cdot V_c \cdot \frac{\rho_c}{\rho_r} \quad (4.8)$$

$$V_r (\%) = \frac{V_r}{V_c} \cdot 100 = \left(1 - \frac{FAW \cdot n_{plies} \cdot S}{m_{c \text{ air}}}\right) \cdot \frac{\rho_c}{\rho_r} \cdot 100 \quad (4.9)$$

Finally, the final void volume fraction was assessed by knowing the fibre and resin volume fractions, in accordance with expression (4.10)

$$V_v (\%) = 100 - V_r (\%) - V_f (\%) \quad (4.10)$$

2D microscopy optical analysis

The samples were prepared for microscopy analysis after Archimedes method. As before mentioned, a minimum of three samples (more in the case of laminates with heterogeneous c-scan) were analysed by microscopy. The surface of one of the faces of each sample was polished to obtain higher resolution. Micrographs of the cross-section of each coupon were acquired using a Nikon Eclipse LV150. Micrographs were taken at different magnifications from 50X to 200X.

After the images acquisition, the images were post-processed with Leika Application Suite software v4.9.0. For the estimation of porosity by this method, a colour threshold was assigned to highlight and identify porous regions. Leika allowed counting the highlighted pixels, thus quantifying the final void content of the sample. An example of this procedure can be observed in Figure 4.14.

Figure 4.14 Void content assessment by 2D microscopy: (i) cross-section image of a CF/PEEK; (ii) same image after colorizing the porosity for void content quantification.

Matrix digestion

Void content was also finally determined by the matrix digestion method in accordance with EN 2564. The procedure was explained with detail in previous section 3.4.3. Same procedure was followed in this case.

4.3.4 DSC

DSC analysis was performed to several samples of each laminate, according to ISO 11357-3 [231]. Same procedure and equipment as detailed in previous section 3.4.2 was followed.

4.3.5 Mechanical testing

In the previous chapter, a set of mechanical testing combining matrix-dominated and fibre-dominated tests were performed in order to investigate the response of different laminates, manufactured under the three different OoA methods. It was concluded that IPSS was a good screening test for evaluating the quality of a thermoplastic manufacturing process, the final quality of the matrix and the interaction fibre/matrix. IPSS resulted to be very sensitive not only to voids within the laminate, but also to a bad interface of the matrix.

IPSS is the main test used in this study for the investigation of the porosity impact on the mechanical properties. For this reason, laminates were manufactured with a $[+45/-45]_{4S}$ lay-up (as indicated in previous section 4.3.1). At least 5 coupons of each laminate were subjected to IPSS testing in accordance to EN 6031 [235] standard. The applied testing procedure here was similar to the one described in section 3.4.5.2.

In order to take full advantage of the manufactured laminates, 20 x 10 mm coupons were also extracted from the laminates in order to also perform ILSS tests. The 20 x 10 coupons were trimmed forming an angle of 45° with the corner resulting in a final coupon lay-up of $[0/90]_{4S}$. A minimum of five coupons from each laminate were subjected to ILSS testing according to EN 2563 [234]. Due to the stacking sequence, ILSS testing did not fully meet the standard. Apart from that, same procedure as the exposed in section 3.4.5.1 was followed. Even though ILSS tests were performed without fully fulfilling the standard, the results were evaluated for intercomparison since the main goal of the research was to evaluate the drop of mechanical properties associated to void content.

4.4 Results and discussion

In this section, the results of the proposed characterisation program are compiled and discussed thoroughly.

4.4.1 Ultrasonic NDT

The ultrasonic c-scans of the manufactured thermoplastic laminates by different processing methods are shown in Figure 4.15 (VBO laminates), Figure 4.16 (hot-press laminates) and Figure 4.17 (ISC laminates). All the images are presented with a scale bar (grey-scale) of attenuation

values in dB. It is important to recall that during the ultrasonic inspection, the ultrasonic wave travels through the specimen and may be attenuated mainly owing to inner irregularities. Those irregularities may commonly consist of voids, distributed porosity, or bad consolidation between layers. Thus, laminates with more irregularities in their inner structure will eventually result in c-scans with more attenuated regions. According to the given scale, darker regions correspond to higher attenuation, probably meaning that certain amount of air is accumulated and prevented the ultrasonic wave propagation. In this study, inspection standard AITM6-4011 was followed, so a 6 dB attenuation threshold was applied for evaluating the c-scans, which is the most common threshold applied in aerospace industry [248,249].

According to Figure 4.15, laminates VBO400 (baseline), VBO380, VBO390 and VBO430 can be considered as defect-free specimens since no indication above 6 dB can be observed. Some circular black spots can be observed in VBO430, but they were caused by air bubbles during the inspection and were not considered. This suggests that processing temperatures in the range from 380 °C until 430 °C can be considered favourable conditions for a proper consolidation and air/voids evacuation. It is important to note that a high temperature of 430 °C applied during 15 minutes did not cause any detrimental effect to the laminate owing to matrix degradation. These results are in line with Fujihara et al. [28], who reported that detrimental effects on the properties of CF/PEEK laminates due to degradation only appeared when plates were processed at a temperature of 440 °C and holding time of 60 min.

The c-scans of VBO laminates consolidated at lower temperatures than 380 °C presented a progressive level of attenuation. Thus, VBO340 consolidated at a temperature very close to the melting point of the resin; presented a very high attenuation (over 18 dB) indicating the total deconsolidation of the panel and a large amount of occluded air. C-scan of VBO350 presented a large central area highly attenuated and two edges showing better consolidation. This suggests that a processing temperature of 350 °C was enough to allow movement of the voids. However, the resin was in such a viscous state that a dwell time of 15 minutes was not enough for remove all the inner air. VBO360 exhibited a similar behaviour with the difference that a larger area is well consolidated. Only a minor area of the central part was still considerably attenuated. In this way, the results suggested that air is evacuated through the edges of the laminates so the central part is the last area where the air is evacuated from. C-scan of VBO370 showed a central area free of air, but still some attenuated lines can be observed. Thus, processing temperature played a key role when the dwell consolidation time was fixed, especially when processing temperatures are below the optimal processing temperature causing a denser state of the resin. This matched with the results of Zhang et al. [19], who reported that proper balance between time and temperature is required for correct air evacuation. As before discussed, they also reported that air is evacuated through the perimeter of the laminate in VBO method.

Figure 4.15 Ultrasonic c-scans of laminates manufactured by VBO under different consolidation temperatures.

Figure 4.16 shows the c-scans of hot-press laminates. In this case, laminates processed at higher temperatures (up to 500°C) were manufactured, in addition to laminates whose applied pressure during manufacturing was also altered. According to the applied attenuation threshold, laminates HP400 (reference), HP370 and HP400_1.5 are considered defect-free. In hot press manufacturing, a consolidation temperature of 370 °C was enough to obtain an indication-free c-scan. In fact, c-scan of HP340 exhibited a large area with a good level of consolidation. Only an area in one of the edges presented discontinuities, suggesting that effective void evacuation or good consolidation did not take place in that region. HP400_0.2 presented similar c-scan to HP340, indicating that the applied low pressure did not manage to properly consolidate the full laminate. However, most of the laminate was well consolidated. These c-scans suggest that a low temperature of 340 °C which caused a unfavourable viscosity of the resin was sufficient to consolidate a large part of the plate with the aid of a higher pressure (0.7 MPa) when compared to VBO processing. On the contrary, an optimal temperature (400°C) may not be enough for obtaining full consolidation if a low pressure is applied (0.2 MPa). Low pressure has been reported in the literature to cause a detrimental effect in composites, leading to high porosity and mechanical properties drop. [108,115,129,250–252]. For instance, Koushyar et al. [129] reported similar c-scans to the reported in this study, where porosity was accumulated in the centre of the laminates.

Regarding high processing temperatures, c-scans of laminates HP430, HP460, and HP500 presented progressive attenuation levels. C-scan of HP430 showed indications mainly focused across the perimeter of the plate, suggesting a heat accumulation and resin degradation along the edges. HP460 presented a large attenuated region also in the edges, but attenuation areas can be also observed surrounding the centre of the plate. If c-scans of laminates VBO430 and HP430 are compared, it can be deduced that the attenuation along the perimeter of HP430 may be the consequence of oxidative degradation since, unlike VBO processing, hot-pressed laminates are processed in an oxygen atmosphere. Visual inspection of laminate HP460 indicated a different colour tone which can indicate matrix degradation. Finally, HP500 completely attenuated the ultrasonic signal above 18 dB. At the demoulding stage of the laminate, dark dust was covering the full laminate indicating advanced oxidative resin degradation, matching the results provided by Martín et al. [253].

Figure 4.16 Ultrasonic c-scans of laminates manufactured by hot-press under different temperature and pressure conditions.

In Figure 4.17, all the c-scans of the ISC laminates are presented. Laminate ISC400 was manufactured at optimal conditions but still present attenuated regions if compared with optimal laminates from the rest of the manufacturing methods. A pattern can be observed in the fibre

direction suggesting irregularities between tapes. All laminates presented attenuated regions regardless the manufacturing conditions.

ISC430 exhibited similar c-scan with the exception of two rectangular shaped black shapes which are caused by burnt tapes during manufacturing. Due to the use of an elevated laser power, two burnt tapes were observed during manufacturing which finally altered the ultrasonic register, mainly due to resin degradation. ISC350 also presented an individual defect associated to a twisted tow that was observed during manufacturing. That section was avoided for coupon trimming.

The rest of the laminates processed at different temperature and pressure conditions presented attenuated regions which may correspond to areas with higher distributed porosity. The distributed porosity is a consequence of the use of inconvenient parameters. As concluded in previous chapter 3, voids during ISC process are more complex of being eliminated since the whole process occurs at ultra-fast times. Deviations of temperature or pressure may lead to incorrect void compression and final higher porosity. However, as observed in c-scan of ISC400, and in agreement with the conclusions of Chapter 3, ISC process is still not able to produce optimal laminates, and higher porosity values may lead to low mechanical properties.

Figure 4.17 Ultrasonic c-scans of laminates manufactured by ISC under different temperature and compaction load conditions.

In Figure 4.18, the averaged values of attenuation (in dB) of each laminates are presented, along with their corresponding error bars. The attenuation values were obtained by analysing the histograms with ImageJ. The 6 dB threshold has been added to the figure, as a red dotted line. It is important to note that the averaged attenuation value can provide valuable information, but it is

also of high importance to analyse the deviation of each value, because it can be also extracted from that information if a laminate presented attenuated regions.

In next sub-sections, the attenuation values are used since the correlation between void content and absorption coefficient (dB/mm) is presented. The ultrasonic absorption coefficient α (dB/mm) is obtained from the divide of the ultrasonic attenuation A (dB) by the laminate thickness t (mm), as depicted in expression (4.11).

$$A = \alpha \cdot t \quad (4.11)$$

Figure 4.18 Ultrasonic attenuation values (in dB) of all manufactured laminates.

4.4.2 Void content

The void volume fraction values of each laminate are presented in Figure 4.19. In the graph, the values of void content assessed by the three proposed methods are included. The standard deviation was also added, along with a red-dotted line which indicates the typical aerospace porosity threshold of 2 % of void content. The selected manufacturing strategy of defects-induced laminates allowed to obtain laminates with porous in the range from almost 0 % up to 19 %. The results offered by different methods correlate well enough in many of the cases, but some discrepancies can be easily observed in other cases.

The highest discrepancies were observed in the case of ISC laminates, especially in laminates manufactured at lower temperature, such as ISC350 and ISC370. The higher deviation in ISC laminates was attributed to the fact that FAW theoretical values are required for void content assessment by Archimedes method, and it is believed that the real FAW value may be altered

during ISC processing. This may be caused by direct laser irradiation on the resin, followed by high compression applied by the roller. If the FAW of the material is modified during ISC processing, the values of void content found by Archimedes method may lead to not correct measurements and should be taken cautiously.

Regarding the values obtained by 2D microscopy, they should be considered reliable values since an experienced researcher or technician can look at the sample at the moment of taking the micrographs, but it is important to note that only one slice out of the full coupon can be analysed by this procedure. Even though the averaged void content values obtained by 2D microscope were obtained using at least three samples, only one cross-section of each sample was analysed. On the other hand, 2D microscopy may offer valuable information as porosity location and size. Much more information could be extracted from X-ray tomography as seen in previous chapter, but cost and time should be taken into account, in order to get to a compromise solution. Matrix digestion is a consolidated and reliable technique in composite material characterization, but longer times are required compared to the other two techniques.

A comparison among porosity assessment techniques can be found in literature. Little et al. [126] studied the accuracy and reliability of different techniques, highlighting the pros and cons of Archimedes and microscopy methods. In a good alignment with this study, they reported that Archimedes method may lead to inaccurate results if the material properties are not well known and also that it is a technique with large standard deviation. Microscopy results showed a good agreement with micro-CT. Kite et al. [254] reported a comparison between microscopy and matrix digestion methods, showing a good correlation and concluding that the results may vary from analysing one cross-section to another due to the voids morphology. Di Landro et al. [124] compared the results of resin digestion method with image analysis (micro-CT and SEM) reporting similar results between matrix digestion and micro-CT, which suggests that the results are reliable because the full specimen is analysed.

It is important to note that laminates which presented heterogeneous c-scans in previous subsection, led eventually to averaged porosity values with higher standard deviation. This suggests again that porosity is diversely distributed and some coupons from the same laminate may present very different levels of porosity. This is why a higher number coupons were analysed from laminates with heterogeneous c-scans in order to obtain reliable results, in a good agreement with Koushyar et al. [129], who extracted coupons from porous and non-porous regions of each laminate to obtain robust results.

Figure 4.19 Void content of all manufactured laminates assessed by three different methods.

In this study, the correlation between the void content and the processing parameters was performed. For the performed correlation, only values derived from matrix digestion were used, since it is the most reliable method according to the previous discussion, apart from being the most standardized method in the aerospace industry. Figure 4.20 shows the void content as a function of the processing temperature of the thermoplastic laminates which were manufactured by modifying the consolidation temperature, while Figure 4.21 shows the porosity as a function of the applied pressure. In the last case, the graph was divided showing the void content of the hot-press laminates as a function of the final applied pressure. Figure 4.21 (i), while Figure 4.21 (ii) shows the values of void content of ISC laminates as a function of the applied compaction roller force.

Some interesting conclusions can be drawn from these graphs. If the results are analysed by manufacturing method while the temperature was increased, a decreasing trend of the porosity is found while increasing the processing temperature in VBO laminates, whereas the opposite effect was observed for hot-press, since low void content is found at low temperatures, and it increases with temperature increase. Thus, low consolidation temperatures led to very high porosity in VBO laminates (14.5 % at 340°C and 3.8 % at 350°C) meanwhile the same temperature was enough for obtaining low porosity laminates with hot-press manufacturing (0.9 % for 340°C). Similar results were underlined while discussing the ultrasonic c-scans. This was related to the higher pressure applied during hot-press manufacturing. Therefore, a good diffusion of the voids is possible at low processing temperatures (when the resin is still in a viscous state) if a significant pressure is applied. VBO laminates processed at temperatures above 380 °C presented a porosity level below the 2 % threshold, while hot-press laminates manufactured in the range of 340 to 430 °C were

under this threshold. This suggests that hot-press manufacturing does not required such high temperatures for obtaining laminates under 2 % of porosity owing to the high pressure applied, meanwhile the appearance of voids in VBO method is very sensitive to the temperature of the cycle. It is also important to note that if VBO laminates would have been manufactured at higher temperatures (460 °C or 500 °C), same porosity increasing trend of the final porosity had been observed at those temperatures due to matrix degradation. However, the ancillary materials used in VBO manufacturing did not allow the manufacturing of laminates at such high temperatures. Hot-press laminates manufactured at very high temperatures reached the higher values of porosity. In a good agreement with the predicted by the c-scans, laminate HP500 showed a large porosity of 9%.

Regarding ISC laminates manufactured by modifying the processing temperature at nip point, a clear trend of the porosity was not observed. As investigated in chapter 3, the nature of the ISC process and the current state of maturity of the technology caused a porosity level within a range of around 2-3 %. In this case, only laminate ISC370 achieved a lower porosity than the 2 % threshold, in contrast to the investigation of other authors as Stokes et al. [47] who concluded that higher temperatures are required for process optimization, especially when working at higher deposition speeds.

Figure 4.20 Correlation of Void content as a function of consolidation temperatures of the laminates manufactured under different processing temperatures.

A close examination to Figure 4.21 revealed that an increase of the applied pressure in hot-press manufacturing led to a progressive decrease of the final void content in the laminate. Many authors studied the effect of applied pressure on the quality of composite laminates. For instance, similar behaviour was reported by Liu et al. [128] for composites processed at autoclave, where different autoclave pressures led to different porosities, concluding that low autoclave pressures were associated to higher void-content. Olivier et al. [155] reached the same conclusion, reporting a decrease of void-content values while increasing pressure.

However, a decrease of the void content was not observed while increasing the pressure in the ISC laminates manufactured by altering the compaction force. The different compaction forces applied did not cause a significant change in the final void content of the laminate. This suggests that a change of the manufacturing strategy is required in order to obtain very low void content results in ISC manufacturing. As discussed in previous chapter, solutions as consolidation re-passes, improvement of raw-material quality or the use of supplementary lasers may aim obtaining lower porosities.

Figure 4.21 Correlation of Void content with applied pressure: (i) void content as a function of consolidation pressure in hot-press manufacturing, (ii) void content as a function of compaction force in ISC manufacturing.

The optical micrographs acquired during the 2D microscopy analysis are shown in next Figure 4.22 to Figure 4.24. The results allowed performing a deeper study of the inner defectology in the laminates.

Regarding manufacturing in oven, it can be observed that VBO400, VBO430 were the only defect-free laminates manufactured by VBO method, in a good agreement with the predicted by the ultrasonic NDT. VBO380 and VBO390 only presented few intralaminar voids. The most inconvenient case was found to be VBO340, which showed large interlaminar and intralaminar porosity, confirming that the resin at 340 °C was in such a high viscosity state which prevented both void diffusion and adhesion between layers. The viscous state of the resin limiting a proper void evacuation led to intralaminar porosity since the inherent air occluded in the raw material finally stayed in the laminate. In the same way, the cycle was not able to favour the intimate contact and cross/linking of the resin among layers, leading to interlaminar porosity or even lack of consolidation. A progressive decrease of the porosity with the processing temperature was observed in laminates VBO350, VBO360 and VBO370. The results showed that the plies were bonded at those temperatures but some intralaminar porosity was observed inside the layers. This suggests again that processing temperatures of 350 to 370 were not enough for achieving proper void evacuation in vacuum bag manufacturing. The observed intralaminar porosity may drive to a drop in mechanical properties even though the layers were fully bonded. The fact that the plies are apparently bonded in those laminates does not mean that the chemical bonding consisted of a strong bond, since the cycle could prevent the optimal cross-linking between the polymer chains. However, there was no manner of evaluating the quality of the chemical bonding during optical

analysis. The goodness of the level of consolidation between layers is later study with the mechanical test results.

Figure 4.22 2D microscopy cross-sections of VBO thermoplastic samples manufactured following different consolidation cycles.

Regarding the optical analysis of hot-pressed samples, the results compiled in Figure 4.23 suggested that laminates HP400, HP370 and HP400_1.5 consisted of defects-free laminates according to the ultrasonic c-scans and also to the void content analysis which revealed porosities around 1% in those laminates. Samples of laminates HP430 and HP460 showed porosity in external plies suggesting that those layers were more affected by the high temperatures applied. This was attributed to the proximity of the external layers to the heat source (hot-plates of the press in this case), where the heat was accumulated. The c-scan of HP430 (Figure 4.16) showed a damaged area close to the edges of the laminate. Those results along with the cross-section, confirm that the heat is accumulated in some regions of the laminates if a high temperature is

reached. The contour of the laminates was in contact with air (no vacuum bag is used in hot-press method) which may act as a heat isolator. The most extreme case can be seen in HP500, where large voids can be observed, mainly due to resin oxidative degradation due to the very high temperature [28]. In the case of laminates manufactured by modifying the applied pressure, the low pressure (0.2 MPa) applied to laminate HP400_0.2 prevented the evacuation of intralaminar air, meanwhile high pressure (1.5 MPa) led to a defect-free laminate as before mentioned. However, an excess in the applied pressure may lead to other inconvenient effect, such as excessive loss of matrix, fibre misalignment, wrinkles of bad fibre/matrix distribution.

Figure 4.23 2D microscopy cross-sections of thermoplastic samples manufactured by hot-press method following different consolidation cycles.

The internal structure of the ISC-manufactured laminates can be observed in Figure 4.24. It is important to remark that the cross-sections of ISC samples are shown in a way that the upper face in the figure corresponds to the external side during manufacturing (also called laser face) meanwhile bottom section in the figure corresponds to the tool face of the sample. Similar trends in the results as the reported in the X-ray micro-CT results in chapter 3 can be observed here. Overall, interlaminar porosity can be found in almost all the samples, especially localized in external plies. This suggests that interlaminar porosity is inherent to the ISC process in the current state of the technology, and a change in the manufacturing strategy is required, as proposed by Van Hoa et al. [54]. However, the amount of interlaminar porosity present in the laminates manufactured in this study differed. Hence, ISC350 and ISC370 presented less interlaminar porosity, especially when compared with laminate ISC430, where several plies were affected. The different compaction forces applied did not cause a significant variation in the voids distribution. Some intralaminar porosity was also observed in a minor extent.

Similar conclusions were reached here based on the microscopy results to the ones derived from micro-CT analysis. In this way, the air is less prone to be evacuated during the lamination of external plies during ISC laminates. This was related to the fact that external plies are consolidated a reduced number of times compared to the bottom layers. Besides, it was previously discussed that the tool may act as a heat sink when a considerable amount of composite substrate is laminated on the top of it, possibly causing a void rebound effect [48]. The above mentioned change in the lamination strategy (re-pass, double diode-lase, etc.), along with better quality of raw material (for instance, better fibre-matrix distribution and flat surfaces) may aim to obtain better results. Future investigation focused on alternative lamination strategies which may improve the quality of the ISC-manufactured structures is proposed. However, the scope of this project could not make further progress due to cost and time limitations, along with limited access to the AFP-ISC facility.

Figure 4.24 2D microscopy cross-sections of thermoplastic samples manufactured by ISC method following different consolidation cycles.

After the assessment of averaged void content values and ultrasonic attenuation, the correlation of both magnitudes was performed and the results are shown in next Figure 4.25. As before mentioned in this manuscript, it is the first time to the best of the author's knowledge that this correlation has been performed for thermoplastic composites. It is commonly accepted that ultrasonic attenuation and the corresponding absorption coefficient increase with void content, therefore low void content laminates are associated to low ultrasonic attenuation [95,135]. Some authors reported a linear relation [99], which is in contrast with the results presented in this study. However, the range of porosity presented in those studies reporting a linear relation was reduced (usually around 5 %) compared to the porosity range presented in this study (beyond 14 %).

For fitting, bi-linear equation was selected as optimal function, since a dramatic increase in the void content was found with high attenuation values of the c-scan. Fitting was not carried out for the ISC results, since a clear trend was not observed. For ISC, even those laminates which presented a relatively low attenuation in the corresponding c-scan, values above 2% of void content were eventually found.

It can be concluded that the ultrasonic wave behaviour differs depending on the manufacturing method applied. This fact suggests that certification activities in industry should take this into account at the time of elaborating ultrasonic acceptance criteria for thermoplastic composites. From the graph, values of void content can be determined corresponding to typical aerospace acceptance criteria thresholds of 6, 12 or 18 dB (depending on the thickness of the structure).

Figure 4.25 Correlation between void content (assessed by matrix digestion) and ultrasonic absorption coefficient.

4.4.3 DSC

The crystallinity results provided by DSC testing are compiled in Table 4.6, along with the averaged value (given by manufacturing method) and the difference in relation to the established baseline (VBO400). Figure 4.26 (i), (ii) and (iii) show the crystallinity values of the VBO, hot-press and ISC laminates, respectively.

The region above 35 % of crystallinity was highlighted in the figure, since it is considered optimal for mechanical performance [48]. As previously discussed, the final degree of crystallinity of a given laminate is intimately related to the cooling rate that the polymer experiences during the cycle. As indicated in the table, the cooling rates of VBO and hot-press are 2 °C/min, while ultra-fast cooling rates can be observed in ISC manufacturing, up to 10.000 °C/min. In this study, and based on previous FIDAMC experience on ISC manufacturing, the self-heated tooling assisted to obtain considerable high values. In previous investigations, laminates manufactured using a cold tool at room temperature presented low crystallinity, even below 30%. Thus, Comer et al. [48]

reported a crystallinity of 17.6% in CF/PEEK samples manufactured by ISC methodology with an unheated tool.

Table 4.6 Crystallinity values of thermoplastic laminates manufactured under different consolidation cycles.

Laminate	Cooling rate	Degree of crystallinity			Averaged value
		α (%)	SD ($\pm$)	Deviation to reference (VBO400)	
VBO400	~2 °C/min	40.2	4.8	N/A	40.2 $\pm$ 3.9
VBO340		46.1	1.9	15%	
VBO350		34.1	2	-15%	
VBO360		39.4	4	-2%	
VBO370		38.8	0.6	-3%	
VBO380		43.4	1.2	8%	
VBO390		42.9	0.6	7%	
VBO430		36.6	0.8	-9%	
HP400	~2 °C/min	38.3	3.4	-5%	41.2 $\pm$ 2.6
HP340		39.7	3.4	-1%	
HP370		40	2.1	0%	
HP430		38.5	4.5	-4%	
HP460		43.3	4.3	8%	
HP500		-	-	-	
HP400_0.2		44.3	0.1	10%	
HP400_1.5		44.1	2.5	10%	
ISC400	~10.000 °C/min	38.0	2.1	-5%	35.9 $\pm$ 5.0
ISC350		34.2	0.7	-15%	
ISC370		45.0	3.0	12%	
ISC430		33.0	2.5	-18%	
ISC400_20		32.0	3.5	-20%	
ISC400_80		33.1	0.3	-18%	

Low polymer crystallinity has been proven to have a detrimental impact in the mechanical properties [237,255]. Gao et al. [237] reported the influence of cooling rate on final CF/PEEK crystallinity and the relation with the mechanical properties. In this study, only few laminates were in a crystallinity level below the mentioned 35 % threshold. The minimum achieved value of crystallinity was 32.0 % in the case of ISC400_20, which was close to the 35 % threshold. The detrimental effect of crystallinity in those laminates with 32-33 % of crystallinity was not validated, but was not considered, since the effect in the final mechanical properties may be minimal, especially since those laminates had a considerable high level of porosity (between 2.2 % and 3.8 %), which can definitely cause a more significant drop on the mechanical behaviour.

Figure 4.26 Crystallinity values of thermoplastic laminates manufactured under different processing conditions by: (i) VBO; (ii) hot-press; and (iii) ISC.

In this study, only crystallinity values were investigated by DSC technique since the project is mainly focused on the influence of voids. Different cooling rates may also lead not only to different crystallinity, but also to different crystallization morphology such as spherulites or lamellae, which could be related to the final performance of the thermoplastic structures. Different techniques may be used for a deeper characterisation of the crystallinity of the laminates, such as wide-angle X-Ray diffraction. Thus, further research is proposed for the investigation of crystallinity of AFP-ISC laminates.

4.4.4 Mechanical testing

IPSS and ILSS results are presented in this sub-section. Overall, the mechanical properties experienced a detrimental effect with the increase of void content, as previously reported by the investigations of other research groups [98,108,118,128,154,256].

4.4.4.1 IPSS

The IPSS values (strength and modulus) of all the manufactured thermoplastic laminates are presented in Table 4.7, along with the standard deviation and the difference of each laminate in relation to the selected baseline (VBO400). For better interpretation, the results were represented independently by manufacturing method and as a function of the modified processing parameter (temperature or pressure). The IPSS strength and shear modulus of all the laminates are shown in

next Figure 4.27 to Figure 4.29. Red-dotted lines were set for comparison in all the graphs indicating the values of the baseline laminate (VBO400).

Failure modes of all tests were analysed after testing and were considered valid according to the applied standard. All the coupons experienced elongation during loading up to matrix failure followed by ultimate fibre breakage.

Table 4.7 IPSS testing results: Values of the in-plane shear strength τ_{12} and modulus G_{12} of the thermoplastic laminates manufactured under different methods and consolidation cycles.

IPSS Testing				
Laminate	τ_{12} (MPa)		G_{12} (GPa)	
	Value	Difference to VBO400	Value	Difference to VBO400
VBO400	222.4 $\pm$ 4.0	0%	4.6 $\pm$ 0.1	0%
VBO340	32.5 $\pm$ 2.8	-85%	3.6 $\pm$ 0.1	-21%
VBO350	88.9 $\pm$ 32.0	-60%	4.1 $\pm$ 0.3	-9%
VBO360	150.1 $\pm$ 3.9	-33%	4.5 $\pm$ 0.1	-1%
VBO370	152.57 $\pm$ 2.6	-31%	4.7 $\pm$ 0.1	4%
VBO380	196.5 $\pm$ 1.5	-12%	5.2 $\pm$ 0.1	14%
VBO390	198.8 $\pm$ 4.4	-11%	4.4 $\pm$ 0.1	9%
VBO430	220.0 $\pm$ 6.2	-1%	4.3 $\pm$ 0.2	-6%
HP400	179.9 $\pm$ 12.0	-19%	4.3 $\pm$ 0.1	-5%
HP340	164.4 $\pm$ 34.0	-26%	4.5 $\pm$ 0.2	-1%
HP370	175.0 $\pm$ 8.4	-21%	4.7 $\pm$ 0.1	2%
HP430	203.6 $\pm$ 11.3	-8%	5.0 $\pm$ 0.1	9%
HP460	186.6 $\pm$ 5.6	-16%	4.7 $\pm$ 0.1	4%
HP500	51.33 $\pm$ 3.0	-77%	3.0 $\pm$ 0.3	-35%
HP400_0.2	182.8 $\pm$ 4.9	-18%	4.5 $\pm$ 0.2	-1%
HP400_1.5	181.4 $\pm$ 16.0	-18%	4.5 $\pm$ 0.1	-1%
ISO400	154.8 $\pm$ 2.0	-30%	4.0 $\pm$ 0.1	-12%
ISC350	170.8 $\pm$ 3.4	-23%	3.9 $\pm$ 0.1	-16%
ISC370	190.0 $\pm$ 3.5	-15%	3.4 $\pm$ 0.1	-25%
ISC430	148.1 $\pm$ 6.5	-33%	4.2 $\pm$ 0.1	-7%
ISC400_lowP	151.1 $\pm$ 1.7	-32%	4.1 $\pm$ 0.1	-11%
ISC400_highP	146.5 $\pm$ 1.8	-34%	4.3 $\pm$ 0.1	-7%

The results obtained from the testing of VBO laminates revealed that IPSS was very sensitive to the applied consolidation temperature. VBO400 was set as the baseline reference for comparison and furthermore, it presented the highest IPSS value (222.4 MPa). As detailed in chapter 3, that value was comparable to the reported of autoclave-manufactured laminates. An increasing trend of the IPSS was observed when increasing the processing temperature (Figure 4.27) matching the previous analysed results of NDT and void content. Highest IPSS values were found in laminates

VBO400 and VBO430. In this case, a slightly high temperature of 430°C did not cause any detrimental effect. On the other hand, from all the VBO laminates which were manufactured, it was found that laminates VBO340 and VBO350 had the lowest IPSS. The drop of IPSS at lower processing temperature was mainly attributed to the distributed porosity caused by inconvenient levels of resin viscosity at the manufacturing stage due to sub-optimal consolidation cycles. Regarding the shear modulus, a change was also observed with the consolidation temperature. In this case, a maximum value of shear modulus was observed in laminate VBO380 processed at 380°C. Slightly lower shear modulus were observed at lower and higher temperatures.

Figure 4.27 IPSS testing results of VBO laminates manufactured under different processing temperatures: (i) In-plane shear strength τ_{12} ; and (ii) shear modulus G_{12} .

The results of IPS testing of hot-press laminates as a function of processing temperature and pressure are represented in Figure 4.28. In this case, maximum value of IPSS was found in laminate HP430 (203.6 MPa) which even resulting the highest value for hot-press manufacturing, it was 9% below the oven reference. A slight and progressive increase of the IPSS values were observed with the temperature up to 460°C, where detrimental effects can be observed. As suggested by the ultrasonic c-scans, laminate HP340 presented a considerable higher IPSS (164.4 MPa) when compared to its oven counterpart VBO340. The lowest IPSS result was observed in laminate HP500, and was attributed to lack of resin due to degradation. Regarding the mechanical behaviour of the laminates manufactured under different applied pressures, it is important to note that no significant variations were either identified. IPSS coupons have a significant size (230 x 25 mm) and only a single coupon could be trimmed from the porosity affected region of laminate HP400_0.2 manufactured at low pressure.

Figure 4.28 IPSS testing results of hot-press laminates manufactured under different processing temperatures and pressures: (i) In-plane shear strength τ_{12} as a function of temperature and (iii) consolidation pressure. (ii) Shear modulus G_{12} as a function of temperature and (iv) pressure.

Finally, the IPSS results derived from the testing of ISC laminates are shown in Figure 4.29. Practically all ISC laminates behaved in a similar way, showing IPSS values around 150 MPa, meaning a drop of around 30 % in contrast with reference VBO400 and 17% in relation to HP400. However, higher IPSS values were found for laminates ISC350 and ISC370, manufactured by applying lower power laser than the used for ISC400. This suggests that applying lower consolidation temperature during ISC manufacturing may lead to higher IPSS values. However, the shear modulus of those two laminates were the lowest values obtained for the ISC panels, which means that the lower temperature applied might increase the IPSS but may also compromise the modulus of the laminate. Significant variation was not observed for laminates manufactured under different compaction loads.

A good correlation between the ultrasonic c-scans and the final mechanical behaviour was not found. It was found that after ISC manufacturing, the laminates may sometimes present impurities in the surface –such as resin small hairs or resin fluffs – which may alter the final c-scan register since it may interact with the ultrasonic wave. Polishing the laminate surface by using soft sandpaper seemed to work well for eliminating the impurities and then improving the quality of the c-scan. However, for the quality study of a thermoplastic laminate (especially if manufactured via ISC), it is highly recommended the application of several tests –not only ultrasonic NDT-. As studied during this project, the drop of mechanical properties in thermoplastic composites is a complex problem that has many causes and may involve good bonding between layers (related to

the crosslinking of the polymer chains), porosity (interlaminar or intralaminar porosity), resin degradation, optimal values of crystallization (recommended above 35%), among others. For all this reasons, it should be mandatory performing a set of tests rather than only NDT to ensure the structural integrity of the thermoplastic structure.

In any way, the mechanical response of ISC laminates is still slightly far from the mechanical values obtained with the other OoA methods, therefore a change in the lamination strategy should be done for optimization. As previously discussed, strategies as re-pass or double diode-laser, larger compaction rollers and of course, ISC-grade raw material may aim to obtaining better mechanical results.

Figure 4.29 IPSS testing results of ISC laminates manufactured under different processing temperatures and pressures: In-plane shear strength τ_{12} as a function of (i) temperature and (iii) compaction force. Shear modulus G_{12} as a function of (ii) temperature and (iv) compaction force.

After the evaluation of the IPSS results as a function of the applied consolidation temperature or pressure, a correlation between IPSS and porosity was performed for all laminates. Thus, the IPSS values as a function of void content are presented in Figure 4.30. A data fitting was added to the figure in order to establish values of critical void content for each manufacturing method. The critical void content is understood as the value of void content from where the mechanical properties may start to decrease. The strategy applied for the data fitting was based in the methodology raised by Olivier et al. and Costa et al. [155,257]. In their studies, the authors established a defect-free or optimal value of the investigated mechanical property, derived from the average value of all the laminates considered defect-free and traced a straight line until the

point where the mechanical property experienced a fall. That point was considered the critical void content. From that point forward, a best fit was performed to the remaining points. This methodology for critical void content assessment was also applied by Guo et al. [258] and Jeong et al. [100]. Table 4.8 compiles the averaged values of the laminates with low void content, along with the obtained critical void content for each manufacturing method.

If Figure 4.30 is deeply analysed, it can be observed that maximum IPSS values were obtained for VBO-manufactured laminates. Maximum IPSS value was given by the laminate VBO, set as baseline for this study. However, it is important to note that IPSS values of VBO laminates began to decline at a lower level of porosity than the other techniques. Thus, the critical void volume for VBO laminates was established at 1.2 %. From that point, a negative exponential fitting was applied.

All IPSS values of hot-press laminates remained in a range of 175-200 MPa (with the exception of laminate HP340 – 146.4 MPa) up to a porosity level of 2.7 %, which was established as the critical void content for hot-press laminates. At that point a linear fit was applied to connect with the lowest IPSS value, obtained for laminate HP500 (9% void content and 51.3 MPa), caused by matrix degradation. The linear fitting was applied due to the absence of experimental data at intermediate void content values. However, the existent experiment data allowed observing the clear drop-off rate. The established value of IPSS for void-free laminates was 182 MPa. For this value, laminate HP340 was not considered due to the largest standard deviation caused for a localized affected region by porosity. It is very likely that the value of void content for that laminate is underestimated due to the mentioned localized porosity. The averaged value for hot-press laminates (182 MPa) is in line with the optimal IPSS value (186 MPa) determined by the Cytac data sheet [259].

The experimental results of AFP-ISC laminates presented in this study have been extended in comparison with the research article published by the author [87]. A maximum IPSS value of 190 MPa was obtained for plate ISC370, which had a porosity level of 1.6 %, set as critical void content for ISC method. That IPSS level was not reached by any other of the ISC-manufactured laminates and a drop of properties was observed from that point onward, where a linear fitting was applied. The microscopy analysis suggested that the drop of mechanical properties is given by the interlaminar and/or intralaminar porosity observed in ISC samples.

The results derived from this study were in line with different scientific publications. Thus, Hancox [152] studied the evolution of shear strength in CFRP in a range of void content between 0% and 5%, reaching to similar results, with a drop of shear properties in samples with 5% void content. Similar results were reached by Olivier et al. [155], who investigated the fall of shear strength values in a porosity range up to 19%.

Figure 4.30 Correlation between IPSS and void content for all the thermoplastic laminates manufactured by different methods.

Table 4.8 Maximum-averaged values and critical void content for each manufacturing method.

Mechanical property		VBO	Hot-press	ISC
In-Plane Shear Strength (MPa)	Average value at low void content	209.4 ± 13.7	182.0 ± 11.9	190.0 ± 3.0
	Relation to VBO (%)	-	17.8% (↓)	14.2% (↓)
In-Plane Shear Modulus (GPa)	Average value at low void content	4.5 ± 0.2	4.6 ± 0.2	3.7 ± 0.6
	Relation to VBO (%)	-	3.5% (↑)	17.5% (↓)
Interlaminar Shear Peak Load (kN)	Average value at low void content	2834 ± 31	2573 ± 189	2142 ± 140
	Relation to VBO (%)	-	9.2% (↓)	24.4% (↓)
Degree of Crystallinity (%)	Average value at low void content	41.0 ± 4.9	42.9 ± 4.5	38.6 ± 6.0
	Relation to VBO (%)	-	4.6% (↑)	5.9% (↓)
Critical Void Content v_{crit} (%)		1.2	2.7	1.6

The shear modulus G_{12} of each laminate as a function of void content is presented in Figure 4.31. Overall, a decreasing trend is also observed in this case for VBO and hot-pressed laminates, meanwhile the trend is not sufficient clear for ISC laminates.

Same fitting strategy was performed for modulus behaviour interpretation. Thus, an average of the modulus data was calculated in the low porosity region, and the averaged data was plotted as a horizontal line. The average G_{12} values calculated in the low porosity region are compiled in Table 4.8.

Again, at a certain level of porosity, the modulus decreases. In the case of VBO laminates, G_{12} started decreasing around 2.4% of void content and a drop of 22% was experienced above 10% of porosity. For hot-press laminates, the drop started around 2.5% of void content and the modulus experienced a reduction of 33% at a corresponding porosity of 9%. Similar results were found and reported by Liu et al. [108].

In ISC laminates the same decreasing trend was not observed. All G_{12} values were relatively low compared to the other methods, and they were located in a 3.8-4.3 GPa range, with the exception of laminate ISC370 which presented an especially low shear modulus (3.44 GPa). Laminate ISC370 exhibited the lowest porosity in ISC manufacturing.

In general, irregularities found in ISC laminates such as concentration of voids between external plies or more irregular distribution of fibre and resin observed could led to a less stiff behaviour of the material, implying lower G_{12} values even at low void content levels. More ISC experimental data may be required to observe a clear drop-off rate of shear modulus with void content.

However, a clear trend may not be observed in ISC results with void content increase since the behaviour can be more related to the crystal morphology created during ultra-fast cooling, rather than the averaged degree of crystallization. Characterization techniques as wide-angle X-ray diffraction (WAXD) are recommended for extending this study.

Figure 4.31 Relationship between in-plane shear modulus G_{12} and void content for CF/PEEK laminates manufactured by different technologies.

4.4.4.2 ILSS

The results derived of ILSS tests are compiled in Table 4.9, along with the standard deviation and the difference of each laminate in relation to the baseline. Failure modes of all were analysed after testing. All were considered valid (with exception of laminate HP400_0.2 which experienced bending failure) according to the applied standard. All the coupons experienced elongation during loading up to matrix failure followed by ultimate fibre breakage. It is important to note that the main characterisation of mechanical properties performed in this chapter was IPSS and ILSS test was performed without fully compliance of the standard due to lay-up of coupons. Thus, ILSS testing was performed for intercomparison, in order to extend the study of the impact of the void content on the thermoplastic laminates.

Figure 4.32 shows the correlation between ILSS of each laminate with their according void content. Same fitting strategy as the previous performed for IPSS results was applied.

A decrease trend can be clearly observed for VBO laminates from a certain level of void content (approx. 1.1%) similar to the critical void content determined in last sub-section. ILSS values of hot-pressed laminates started to decrease at a higher level of 2.7% as observed with IPSS results. Laminate HP340 showed some inconsistencies since the ILSS value was particularly low for the given void content level. It may be assumed that the coupons for void content analysis were not completely representative of the final void content of the laminate, consequently the void content value of laminate HP340 may have suffered an underestimation, due to the specific location of the porosity (as seen in the c-scan). All values of ISC laminates were in the range of 57-78 MPa regardless the void content. The maximum ILSS value of ISC laminates (78.4 MPa) was 20% lower than the maximum for VBO manufacturing (VBO390 - 97.95 MPa) and 24% lower than the maximum for hot-press manufacturing (HP430 - 102.9 MPa). Those results are equivalent to the ones reported in Chapter 3 for optimal conditions manufacturing, along with the results reported by Comer et al. [48], who found ILSS values of ISC laminates to be 30% lower than those of autoclave levels.

Overall, the results suggest that ILSS is a property very dependent on the void content existing in the thermoplastic structure, caused by deviations in the consolidation cycles. However, the drop experience by ILSS might response to diverse reasons and not only to the averaged void content. The voids distribution, size and shape are crucial for the drop in mechanical performance, as concluded in Chapter 3. As previously discussed, this may not have been only the result of the void content of the samples, but also a result of the position and shape of the voids. The low peak load of the ISC samples may be attributed to the concentration of voids in the outer plies of the laminates due to the number of times they were consolidated by the roller and also to a more irregular distribution of fibre and matrix.

Table 4.9 ILSS testing results: Values of the ILSS (strength) and the peak load of the thermoplastic laminates manufactured under different methods and consolidation cycles.

ILSS Testing				
Laminate	ILSS (MPa)		Peak load (kN)	
	Value	Difference to VBO400	Value	Difference to VBO400
VBO400	88.0 ± 3.0	0%	2812 ± 181	0%
VBO340	5.0 ± 0.6	-94%	181 ± 22	-94%
VBO350	37.9 ± 12.3	-57%	1218 ± 377	-57%
VBO360	55.0 ± 1.9	-38%	1646 ± 63	-41%
VBO370	78.9 ± 2.85	-10%	2313 ± 77	-18%
VBO430	92.1 ± 3.6	5%	2856 ± 89	2%
VBO380	81,7 ± 2,3	-7%	2401 ± 73	-15%
VBO390	98.0 ± 1.9	11%	2973 ± 66	6%
HP400	100.4 ± 1.7	14%	2928 ± 51	4%
HP340	57.6 ± 8.9	-35%	1846 ± 279	-34%
HP370	86.0 ± 1.5	-2%	2478 ± 44	-12%
HP430	102.9 ± 3.9	17%	2807 ± 100	0%
HP460	91.1 ± 5.6	4%	2551 ± 146	-9%
HP500	18.3 ± 4.0	-79%	571 ± 124	-80%
HP400_0.2	74.4 ± 4.0	-15%	2301 ± 139	-18%
HP400_1.5	87.2 ± 5.0	-1%	2451 ± 100	-13%
ISC400	78.4 ± 2.7	-11%	2301 ± 78	-18%
ISC370	77.9 ± 2.7	-12%	2093 ± 70	-26%
ISC350	72.0 ± 3.0	-18%	1842 ± 70	-34%
ISC430	67.0 ± 2.6	-24%	2033 ± 67	-28%
ISC400_20	57.8 ± 1.5	-34%	1633 ± 53	-11%
ISC400_80	57.1 ± 3.1	-35%	1657 ± 79	-7%

Figure 4.32 Correlation between ILSS and void content for all the thermoplastic laminates manufactured by different methods.

4.5 Conclusions

In this chapter, the impact of the void content on the final behaviour and characteristics of CF/PEEK laminates manufactured by OoA methods (VBO, hot-press and ISC) were investigated. In order to obtain laminates in a wide range of porosity, different manufacturing technologies for obtaining induced-defects laminates were tested, including manufacturing laminates with sealed edges, submitting the raw material to moisture absorption and lastly, making modifications in the key parameters of the consolidation cycles. It was revealed that sealing the perimeters of the preforms and making variations in the consolidation cycle caused significant porosity in the manufactured laminates in a successful manner, meanwhile the strategy based on submitting the raw material to moisture absorption was aborted due to the low moisture absorption of APC2-AS4 of less than 0.1 % humidity. It was decided to continue the investigation with the manufacturing strategy based on modification the consolidation cycle parameters, since the results allowed the creation of valuable relations between key manufacturing parameters (such as consolidation temperature of oven or hot-press, applied pressure in hot-press, laser power or compaction roller consolidation force).

Following the fabrication methodology mentioned above, a manufacturing plan was designed and a total of 23 laminates with dimensions 300 x 300 mm and 16-ply with $[45/-45]_{4S}$ lay-up were manufactured by making modifications in the consolidation cycle, achieving a population of laminates with porosities in the range of 0 to 19 %. All the laminates were subjected to ultrasonic NDT, void content analysis, optical microscopy, DSC and mechanical testing (IPSS and ILSS).

The ultrasonic inspection presented a first scanning test which allowed to visualize the laminates or areas which were attenuated by porosity. The results suggested that laminates manufactured at the most adverse conditions presented large attenuated regions. Also, it was observed that same

processing temperatures may lead to completely different results depending on the manufacturing method (for instance, laminate VBO340 presented total attenuation, meanwhile HP340 presented a highly localized attenuated region). All ISC laminates presented an attenuated lattice and in general, more heterogeneous results. Image processing was performed with the aim of ImageJ and the histogram tool, which enable the extraction of averaged attenuation values (in dB) for each laminate.

After NDT, three different methods for void content assessment including Archimedes, matrix digestion and 2D microscopy were carried out, along with a comparison between results. The results correlated well with the predicted by ultrasonic testing, but the void content assessment methods presented some discrepancies among methods. As reported by other authors, each porosity assessment method has its pros and cons. Archimedes is a fast screening method but the reliability may be low and it is highly dependent on the knowledge of the material properties such as FAW, especially for ISC laminates. Matrix digestion is the most accepted method in the aerospace industry but it is a time and cost consumption method. Regarding 2D microscopy, it is an accurate method with provides highly valuable information but it is limited to a single cross-section per coupon.

With the experimental void content values obtained from matrix digestion method, a correlation was performed by representing the ultrasonic attenuation coefficient as a function of void content for each manufacturing method. A bi-linear fitting was performed for VBO and hot-press laminates, but the trend was not that clear for ISC laminates. This was attributed to the fact that ISC manufacturing may lead to resin impurities on surface caused by heating by laser, which may muddy the ultrasonic c-scans. The results presented in the correlation suggested that each manufacturing method should have its own NDT acceptance criterion.

The 2D microscopy analysis revealed the distribution of defects in the sample and showed those laminates having interlaminar and/or intralaminar porosities. The results indicated that laminates with low void content presented a good consolidation between plies with almost no pores, but a larger interlaminar and intralaminar porosity appeared when void content increased. Those laminates fabricated under the harshest conditions in terms of high temperatures presented degradation of the matrix. In general, ISC laminates exhibited large porosity and some of the laminates presented large voids accumulated between upper layers (as shown in chapter 3).

DSC results suggested that most of the laminates (with just few exceptions) were in the crystallization range considered optimal for mechanical properties and it was assumed that the crystallinity did not play a significant detrimental role in this study. However, it was suggested the future extension of the work including WAXD for a deeper study of the crystals morphology, which may affect to the structural behaviour, especially in ISC laminates, that are cooled down at very fast cooling rates.

For the mechanical characterisation of the laminates, IPSS test was selected as the main test for analysing the quality of the laminates, based on the results and conclusions of chapter 3. ILSS (not completely fulfilling the standard requirements due to lay-up) was also performed for intercomparison between laminates. From the experimental results, it was observed that IPSS is a property very sensitive to variations of the processing temperature, especially in VBO and ISC fabrication. The variations of IPSS and the shear modulus with temperature and pressure were analysed. Correlation of IPSS as a function of porosity was performed. An approach proposed by Olivier et al. [155] was followed for determining values of critical void content. Critical void

volume fraction was defined as the value of averaged porosity from where the mechanical properties of a given structure may start to decrease. Critical void volume values were obtained for VBO, hot-press and ISC thermoplastic laminates. Similar results were achieved with ILSS results.

From the combined analysis of all the results, it can be concluded that VBO processing may need temperatures closed to the recommended of 400°C for obtaining good quality laminates, since lower temperature may cause low matrix flow and incomplete void evacuation. On the other hand, hot-press method may lead to good quality laminates at lower temperatures due to the high pressure applied. In the case of ISC laminates, several discrepancies were observed. ISC manufacturing strategies still have to be optimized since higher porosity and lower mechanical strength values were found.

In a typical thermoset-aerospace production line, the quality control of a manufactured part is evaluated by means of NDT, ILSS and DSC (T_g) subjected in a traveller coupon. From the results analysed here, degree of crystallinity and void content evaluation is suggested, along with another matrix-dominated property (as IPSS), which may not suffer plastic deformation as easily as ILSS.

The correlations presented in this chapter (ultrasonic attenuation and IPSS as functions of void content) were the very first of its kind for thermoplastic composites. Similar investigations were have been conducted previously, but limited to thermoset composites [108,155]. Hence, this is one of the innovative points of this PhD project. Those correlations might be very useful for engineering application, especially for establishing an NDT acceptance criterion for thermoplastic aerospace structures depending on the level of porosity allowed in the structure.

Chapter 5

On-line monitoring of in-situ consolidation process with FBG sensors

5.1 Introduction

This chapter presents the first steps taken regarding the on-line process monitoring of thermoplastic AFP-ISC with FBG optical sensors.

Based on the results and conclusions of previous chapters, it can be claimed that the current state of laser-assisted AFP-ISC technology cannot nowadays provide laminates with similar performances to those manufactured under other OoA methods, such as oven or hot-press; but either to the laminates manufactured in autoclave. For this reason, ISC manufacturing should be optimized in order to reach those levels.

As concluded in the previous chapter, processing parameters play a key role in the final behaviour of the thermoplastic structures. Processing temperature appeared as the most crucial parameter among the manufacturing parameters. Consolidation temperature will eventually determine the

mechanical behaviour of a thermoplastic composite. Currently, only IR thermographic cameras are employed for monitoring the temperature during the process. One of the main handicaps of the IR camera is that the temperature measurements are limited to the surface. In addition to that, the emissive and absorption heat coefficients of all the elements surrounding the manufacturing set-up should be known in order to obtain reliable results of the thermographic monitored data. Together with the thermal history monitoring challenge, there are also other issues which should be addressed in order to improve the maturity of the ISC technology, such as the residual stresses which appear in the structures due to the fabrication process itself.

Within this context, a tool is required which may allow to gain knowledge about the critical issues of the process. Due to these reasons, optical FBG sensors emerged as a good candidate for ISC process monitoring. FBG sensors have been proved to be a strong tool not only for SHM purposes [260], but also for process monitoring activities [169], as described in Chapter 2. FBG sensors are sensitive to thermal and strain variations, so they can help to gain knowledge about the ISC process. The main goal of this study regarding ISC process monitoring is to obtain the thermal history of the process with FBG sensors, but this is mainly investigated in next chapter. This chapter focuses on the investigation regarding sensor embedding strategies and monitoring tests by using sensors from different suppliers.

First of all, it was required to investigate different embedding strategies, since optical fibres are very brittle and ISC process presented a harmful environment for sensor embedding due to several reasons including the high temperatures applied, the laser irradiation and the application of loads by the compaction roller. Thus, ISC process was an aggressive environment that compromised the survival of the FBG sensors. After selecting a successful embedding strategy; an experimental monitoring plan was defined, based on the process monitoring of the fabrication of different CF/PEEK samples. For this purpose, several FBG sensors were purchased from different suppliers in order to investigate different sensor features, such as the coating material. The results provided by the different sensors were compared and discussed.

The results exposed in this chapter helped to define an optimal strategy for AFP-ISC process monitoring with FBG sensors and also aim to gain knowledge about the AFP-ISC process and its manufacturing stages during the consolidation of each layer.

To date, only an investigation group reported some research activities regarding the implementation of optical sensors within AFP-ISC process, to the best of the author knowledge. Thus, Oromiehie et al. [49,81,83] reported some activities regarding process monitoring of AFP-ISC with FBG sensors. The main difference between both studies resides in the use of different heating sources. In [49,81,83], Oromiehie et al. employed a gas-torch as heating system, while a diode laser was employed in this project. The fact of employing a diode laser as heat source complicated the survival of the sensors, mainly due to direct laser irradiation on the sensor. This involved considerable effort to optimize the embedding strategy.

5.2 Methodologies for embedding FBG sensors in ISC process

In this preliminary section, the investigated methodologies for overcoming the challenging incorporation of optical sensors into AFP-ISC process are presented. The results exposed here determined the embedding approach for the rest of the project.

As previously explained, ISC process and set-up during manufacturing present itself a harsh environment for the embedding of optical sensors in the laminates under fabrication.

The monitoring experimental plan consisted of embedding bare optical fibres (without written FBGs, in order to reduce costs) in thermoplastic samples following different combinations of optical fibre orientation and protective methods. Based on preliminary trials, it was detected that the most harmful cases for the optical fibres consisted of direct irradiation on the fibre causing fibre degradation and the load applied by the roller at the egress point, especially when the laminate thickness increased and the roller may apply shear loads in the fibre causing fibre breakage.

The embedding process consisted of locating the optical fibres on the thermoplastic substrate; and the consecutive location of a tape -or multiple tapes- on the top of the optical fibres. The survival of each optical fibre was evaluated with a fibre-checker, which consisted basically of a source emitting light through the pigtail of the fibre and being visible at the other end of the fibre (if the fibre managed to survive the embedding process).

The investigated optical fibre orientations consisted of:

- 0°: the optical fibres were positioned in the substrate parallel to the incoming tape.
- 90°: the optical fibres were positioned in the substrate perpendicular to the incoming tape.

The investigated protection strategies consisted of:

- No protection: the incoming tape was laminated on the top of the optical fibre without additional protection.
- Egress point protection: The optical fibres were protected only at the egress point. Thus shear forces caused by the compaction roller at the end of the sample were avoided.
- Egress point + protective tow protection: The optical fibres were protected at the egress point, but also a thermoplastic tow was first located manually on the top of the optical fibre, avoiding direct irradiation of the laser over the optical fibre.

The different proposed strategies led to the experimental testing matrix shown in Table 5.1. Several trials were performed following each embedding strategy, in order to obtain reliable results and conclusions. Illustrative descriptions of the embedding trials are given in Figure 5.1.

Table 5.1 Details of optical fibre embedding trials.

Embedding trial	Optical fibre orientation	Protection strategy
OF 1	0°	none
OF 2	90°	none
OF 3	0°	egress point
OF 4	90°	egress point
OF 5	0°	egress point + protective tow
OF 6	90°	egress point + protective tow

The protection of the egress point was performed by adding polyimide adhesive tapes underneath and overhead the optical fibre at the point where the optical fibre comes out the thermoplastic sample. Adhesive kapton tapes were also located in order to supplement the thickness difference

between the egress point and the tooling, when the laminate became thicker. It was observed that shear loads were caused by the compaction roller at those points, which may cause optical fibre breakage.

Regarding the emplacement of a protective tow, the procedure was based on placing a CF/PEEK tape manually on the top of optical fibre. The thermoplastic tow was bonded to the substrate in several points with the aim of a manual welder.

Figure 5.1 Schematic representation of the embedding trials of the optical fibres in ISC process.

A series of representative pictures of the monitoring trials are presented hereunder. The light emitted by the fibre-checker coming out of an optical fibre is shown in Figure 5.2 (i), while the compaction roller above an optical fibre is shown in Figure 5.2(ii). Figure 5.3 (i) shows the placement of optical fibres at 0° and Figure 5.3 (ii) shows the placement of an optical fibre at 90° in relation to the taping direction. Figure 5.4 shows the strategy for protecting the egress point by using kapton adhesive tape. Finally, the procedure for using a protective thermoplastic tape is

shown in Figure 5.5. The following steps can be observed: (i) placement of the protective tow, (ii) alignment of the optical fibre, (iii) bonding of the protective tow to the substrate by means of a manual welder and (iv) the protection of the egress point and connectors with kapton tapes.

Figure 5.2 (i) Light emitted by the fibre checker coming out of an optical fibre for survival checking; and (ii) compaction roller of ISC machine above an optical fibre.

Figure 5.3 Placement of optical fibres during trials at (i) 0° direction aligned with the tape direction; and (ii) 90° direction, perpendicular to the tape direction..

Figure 5.4 Protection of egress point with polyimide adhesive tape

Figure 5.5 Procedure for protective tow emplacement: (i) location of protective tow, (ii) alignment of optical fibre, (iii) bonding of the protective tow to the substrate by means of a manual welder and (iv) protection of egress point and connectors with kapton adhesive tapes.

The results of the performed trials enabled to select an optimal methodology for embedding optical fibres within ISC process. Thus, the direct laser irradiation received by the optical fibres in trials OF1 and OF2 damaged in a critical way the optical fibres. The fibres were broken by the effect of the laser and the compaction roller before being completely embedded in all performed trials under OF2 configuration. Several fibres survived following OF1 configuration in the first instance, but were eventually broken with the lamination of upper layers.

During OF3 trials, optical fibres survived longer times when compared to OF1. Fibre breakage at the egress point was not especially appreciated. However, many of the fibres were also broken at some point during lamination. This suggested that the optical fibre is especially prone to breakage

when direct laser irradiation was applied on the bare optical fibre. Similar results were observed for OF4 trials.

OF5 and OF6 provided the best results, since most of the employed optical fibres survived the embedding process. Hence, at least approximately 80 % of the employed optical fibres managed to survive the embedding process. The combined application of a protective tow with the protection applied to the egress point was considered the optimal strategy.

From the observed results, strategy OF5 (0°, protective tow and protection at egress point) was selected for embedding FBG sensors in AFP-ISC for process monitoring during the rest of the project.

5.3 Experimental methods for real-time process monitoring of ISC with FBG sensors

In this section, the employed material, equipment, FBG sensors and experimental methods are detailed.

5.3.1 Materials and configuration

In this chapter, APC2-AS4 supplied by Solvay-Cytec Group was employed. The material consisted of UD 6.35 mm (1/4") width and 0.135 mm thick tapes. Material specifications can be consulted in previous section 3.2.

All the manufactured samples had a [0] lay-up with a thickness of at least 10 plies, with 3 tows in width and 150 mm length, resulting in final specimen dimensions of 150 × 19 (length × width). Final thickness of the samples depended on the number of laminated layers (thickness per ply was 0.135 mm).

5.3.2 Manufacturing equipment

The thermoplastic samples were manufactured using AFP-ISC machine described in previous chapters 3 and 4. This consisted of a MTorres gantry-style single-tow deposition machine located at FIDAMC facilities (Getafe, Spain), shown in Figure 5.6. The rest of equipment specifications (such as laser type or compaction roller) can be consulted in previous section 3.3.3.

Figure 5.6 Single-tow fibre placement machine with ISC technology situated at FIDAMC facilities.

5.3.3 Interrogation system

A commercial optical interrogator system was purchased for this investigation. The equipment used during the tests was a dynamic Micron Optics SM130, shown in Figure 5.7. The system works in the range of 1500-1600 nm and it analyses the reflected wavelength of the FBG sensors. The system uses a Gaussian algorithm for peak tracking. A computer with installed Micron Optics ENLIGHT software was used for data acquisition during the monitoring experiments.

Figure 5.7 Micron optics SM130 interrogator system during the monitoring experiments.

5.3.4 FBG sensors

A set of different FBG sensors were employed for investigating the different response regarding the different features of the sensors. The set of sensors consisted of a mixture of commercial ones purchased from two different suppliers; and in-house fabricated sensors.

The main difference among sensors consisted of the coating material and the final outer diameter (OD). For the investigation compiled in this chapter, FBG commercial sensors were purchased to ZEUS Industrial products (SC, North America) [261] and FBGS International (Geel, Belgium) [262]. The in-house fabricated FBG sensors were written at the laboratory of the Department of Aerospace Materials and Production of the School of Aeronautical and Space Engineering of the Polytechnic University of Madrid (UPM) [263]. The UPM sensors were written in-house by following the procedure described by Menéndez and can be consulted in his doctoral thesis [171].

A total of three sensors were used and their main characteristics are compiled in Table 5.2, including FBG length, coating material and other FBG typical properties.

Table 5.2 Main features of the FBG sensors employed in this study.

Sensor supplier	Coating	FBG length (mm)	Wavelength range (nm)	Outer Diameter (OD) (μm)	Reflectivity
UPM	none	8	1500-1600	124	80%
FBGS	ORMOCER [®]	8	1500-1600	189	27%
ZEUS	PEEK	8	1500-1600	123	87%

All the sensors were purchased with the same FBG length and working in same wavelength range. The OD differed between suppliers but it was in the same order of magnitude. The reflectivity of UPM and ZEUS sensors were above 70%, considered optimal; whereas FBGS showed a relatively low reflectivity, mainly due to the sensor fabrication process. FBGS usually provide FBG sensors manufactured under Draw Tower Grating (DTG[®]) method, which is based on written FBG sensors at the time when the optical fibre is being vertically extruded. This manufacturing method is associated to a final lower reflectivity, which required an increase of the digital gain of the signal. This led to higher signal to noise ratio during experiments.

Regarding the coating, different materials were investigated since it was thought that the coating material would impact on the quality of the sensor integration. As previously commented, ISC process implies a harsh environment for process monitoring. Hence, high temperature-resistant ORMOCER[®] coating was tested, which was the coating material of the commercial sensors supplied by FBGS International. Also, it was thought that FBG sensors coated with the same material as the matrix of host composite (PEEK in this case) would aim to improving the sensor integration. In this way, PEEK-coated FBG sensors were purchased from ZEUS. Finally, the UPM in-house FBG sensors with no coating were set as baseline for comparison.

As an example, an as-received FBG sensor supplied by ZEUS is shown in Figure 5.8.

Figure 5.8 FBG sensors supplied by ZEUS.

5.3.5 Monitoring tests

An experimental procedure was designed for the investigation of the AFP-ISC process monitoring with FBG sensors. As described in previous section 5.3.1.2, different $[0]_n$ CF/PEEK samples were fabricated. The FBG sensors were embedded within the sample and were used for monitoring the process.

For the correct embedding of the FBG sensors and minimizing any possible operational damage caused on the fibres, a first ply (substrate) was located manually on the lamination tool. After that, a second ply (three tapes in width) was laminated. The FBG sensors were then placed on the top of the central tape of the second layer. The protective tow was then manually located following the methodology described in previous section 5.2, in order to avoid direct laser irradiation on the optical fibre and also avoiding shear loads in the egress points. The protective tow was consolidated by the system without material feeding and the sensor remained securely integrated. A schematic drawing of the procedure is shown in Figure 5.9. It can be observed how the FBG sensor is located aligned with the composite tapes (0° direction) in Figure 5.9 (i) and how the sensor is finally protected and integrated within the sample in Figure 5.9 (ii).

Figure 5.9 Schematic representation of FBG sensor integration into a CF/PEEK sample.

After sensor integration, the sensors were connected to the interrogator system. The lamination of rest of the layers was monitored by the sensors. The process monitoring set-up is represented in Figure 5.10.

The samples were manufactured according to standard AFP-ISC manufacturing procedures, detailed in previous sections. Thus, the selected laser power was adjusted for obtaining a temperature of 400 °C in the nip point. Consolidation force was set at 500 N and the lamination speed was 1 m/min. The information about the monitoring tests is compiled in Table 5.3.

Figure 5.10 Schematic representation of the experimental set-up for process monitoring of ISC process with FBG sensors.

Table 5.3 Detailed parameters of the monitoring tests.

Monitoring test code	FBG sensor applied	Tooling temperature (°C)	Temperature at nip (°C)	Roller force (N)	Lamination speed (m/min)
UPM	UPM				
FBGS	FBGS	25 (RT)	400	500	1
ZEUS	ZEUS				

The experimental procedure for embedding FBG sensors is presented in Figure 5.11. The location of the substrate (first layer) is observed in Figure 5.11 (i). The consolidated set of second layers can be seen in Figure 5.11 (ii). The bonding of the protective tow to the substrate by a welding gun is shown in Figure 5.11 (iii) and finally, the FBG sensors embedded into the samples ready for monitoring can be observed in Figure 5.11(iv).

Figure 5.11 (i) Location of first CF/PEEK layer (substrate); (ii) Set of consolidated second layers; (iii) Bonding of protective tow on the top of the optical fibre sensor and (iv) embedded FBG sensors ready for monitoring the lamination of the remaining layers.

It is important to note that the lamination of the 3rd layer corresponding to the consolidation of the protective tow was also monitored in all tests. Besides, only the lamination of the central tape (out of three tapes in each layer) was monitored and further analysed in this study. The other two tapes of each layer were added for making the tests representative of real manufacturing.

5.4 Results and discussion

In this section, the results derived from all the performed monitoring tests are presented and discussed. The reflected wavelength of each employed FBG sensor was monitored during the tests. FBG are sensors especially sensitive to changes in temperature and strain fields. Thus, the temperature applied by the laser was expected to cause variations in the reflected wavelength. Along with the modification caused by the thermal cycle, some information was expected to be obtained regarding the compaction loads, as well as some information about the residual stresses appearing during AFP-ISC process.

Figure 5.12 shows the monitored results provided by sensor UPM. An extensive discussion is mainly conducted while describing Figure 5.13. However, showing the results of UPM sensor in a first place was considered necessary. This was mainly due to the fact that, as observed in Figure 5.12, UPM sensor did not recover its initial wavelength value after the lamination of a new layer (highlighted in the figure with two red dotted-lines). This effect can be related to the appearance of residual stresses in the composite as the part is being consolidated. Residual stresses are a characteristic phenomenon of the ISC process which has been detailed in the literature which still

has to be sorted out [47,93]. In this way, Oromiehie et al. [49] reported similar results, which were attributed to the residual stresses accumulated during the fabrication, which strained the sensor. This caused the sensor signal not to return to its initial value. However, this phenomenon may also be related to the hysteresis experienced by a FBG sensor which has not been submitted to an annealing process. Annealing is a typical procedure followed in FBG sensing that consists of heating the sensor above the temperatures expected to be monitored, in order to avoid signal hysteresis [192,264,265]. The sensors were not annealed in this study since prolonged exposure to temperatures above 400 °C may have an impact on the sensors and the coatings.

For these reasons, the wavelength shift results are presented for the rest of the chapter by resetting the signal to zero after the lamination of each layer, in order to avoid this effect.

Figure 5.12 Wavelength monitored by sensors UPM during the in-situ consolidation of a CF/PEEK samples containing 10 layers.

A comparison of the results provided by the different employed sensors is shown in Figure 5.13. The wavelength shift (nm) experienced by each sensor during the samples manufacturing is presented. The first peak corresponds to the consolidation of the protective tow. As previously detailed, the sensor was embedded between the second and third layers, but the third layer (protective tape) was first bonded manually and then consolidated by the system without material feed. Thus, the lamination was monitored by the sensor from layer 3 onwards. The wavelength shift was set to zero after the lamination of each new layer, according to the discussed in the previous paragraph.

The results suggest that all the sensors experienced a wavelength shift due to the combined effect of the laser and the compaction roller during the consolidation of each layer. A decrease of the maximum wavelength shift was observed to decrease while increasing the number of layers (thickness of the sample). This was attributed to the increasing distance between the sensors and the external sources with the consolidation of each new layer. Hence, the consolidated thermoplastic material between the sensor and the external perturbations (laser heat and roller pressure) absorbed the effects caused by those sources. Similar conclusions were claimed by Oromiehie et al. [81,83], who reported an inverse correlation between the stacked plies and the

reflected wavelength during the monitoring of the fabrication of glass fibre/HDPE samples. The maximum wavelength shift reported in that case decreased quicker compared to the results showed in this study. The existing differences in the attenuation velocity of the monitored maximum wavelength shift values were mainly attributed to the use of different heat sources. As before commented, a gas torch was employed in [81,83], which is expected to cause less heat penetration through the sample in comparison to a laser source.

The wavelength shift experience during the consolidation of layer 3 presented the most irregular results. As before-mentioned, layer 3 was first positioned manually and then consolidated by the system working without material feeding in order to avoid direct laser irradiation over the FBG sensors. Due to this particular procedure, the monitoring results of the lamination of layer 3 may be the less reliable, especially for ZEUS test, which showed an irregular shape.

FBGS sensor also presented some irregularities during the lamination of layers 3 to 5. Particularly high wavelength shift values were monitored during those layers. This was related to the fact that the sensor was heavily excited during the first layers, where the sensors were very close to the heat source. Besides, the digital gain was increased for proper signal intensity in the case of FBGS sensors, due to their especially low reflectivity. This fact increased the signal-to-noise ratio. Those maximum values corresponded to the moment where sensor is excited by the compaction roller, which may cause a distortion of the signal spectrum, complicating the peak tracking by the system. All the exposed reasons, along with the fast wavelength shift, could take to a mistake in the peak detection during the first layers.

Figure 5.13 Wavelength shift monitored by sensors UPM, ZEUS and FBGS during the consolidation of three different CF/PEEK samples.

The individual maximum wavelength shift values monitored by the sensors during the lamination of each layer are presented in Figure 5.14. In this case, it can be easily detected the irregularities experienced by FBGS sensors during the first layers. As observed in the previous figure, a decrease of the maximum wavelength shift with the thickness was experienced. Hence, the same inverse relation to the reported in [81,83] between maximum wavelength shift and the number of consolidated plies is observed in this study.

The maximum values showed in the figure differ between different sensors, especially for the results derived from lamination of layers 3 to 5. On the other hand, the results matched well from layer 5 onwards. It is remarkable that the values of wavelength shift monitored by the ZEUS sensor were lower compared to the results monitored by the other two sensors. As discussed before, the manufacturing process itself is not perfectly repetitive and the laser power and ultimate thermal history may vary from one process to another. However, other parameters such as the sensor outer diameter and coating material may play a key role in the monitored response of each sensor, which is discussed later in this section.

Figure 5.14 Maximum values of wavelength shift monitored during the lay-up of the CF/PEEK samples.

In order to analyse the results in a deeper way and to understand the phases experienced by the FBG sensors during lamination, the wavelength shift experience during the consolidation of a single layer is shown in Figure 5.15. The monitoring results of the consolidation of layer 6 are presented. Layer 6 was selected due to the fact that is in the middle of the laminate and can be taken as representative of the manufacturing process. The monitored signal was divided into three different regions, corresponding to the different steps taking place during the lamination and consolidation process.

Figure 5.15 Wavelength shift monitored by FBG sensors UPM, ZEUS and FBGS during the consolidation of a single layer. Three different stages can be identified: (a) heating-up by the laser; (b) effect of the compaction roller and (c) and cooling down.

The identified regions consisted of the following phases, highlighted in the figure: (a) heating-up, (b) contribution of compaction roller and (c) cooling down. The identified regions are described below:

- Region (a): Heating-up

The FBG sensors experienced in first place an increase of the reflected wavelength owing to the laser irradiation on the sample, which caused a sudden temperature shift. All sensors started experiencing the same gradient first. However, a different trend was experienced from a certain point by the ZEUS sensor. This may respond to several reasons, including different heat transfer due to material coating or different thermal history in the fabrication of each sample, since the laser created a thermal profile which may vary significantly between cycles.

Before entering into phase (b), the reflected wavelength suffered a short and subtle decrease, which can be clearly observed in the case of FBGS sensor. This short drop was attributed to the so-called “shadow region”, which is a typical phenomenon in AFP-ISC manufacturing. This phenomenon may occur in the moment before the compaction roller passes through a specific point. Thus, the roller itself blocks the laser irradiation on the substrate for a few milliseconds, causing a sudden decrease of the temperature. The shadow region phenomenon has been previously discussed in the literature, and it is commonly agreed that the composite substrate experiences a short and slight temperature decrease moments before the compaction roller passes through [51,70].

- Region (b): Effect of compaction roller

Immediately after the shadow region, the sensors experienced a maximum of wavelength shift. The wavelength shift reaches the maximum level due to the combined effect of heat and strain caused by laser and roller, respectively.

Still during this region, the sensors experienced a decrease on the wavelength shift, suggesting that the sample started to cool down, still under the effect of the compaction roller.

- Region (c): Final cool down

After regions (a) and (b), the material continued cooling down in the denominated region (c). The results suggested that a different cooling rate may take place in this region, compared to the first cooling rate observed in the previous region.

The observed change in the cooling rate was attributed to the fact that the sample first started cooling down in direct contact with the compaction roller, which acted as a good heat conductor causing a fast cooling. Once the roller passed away, the material kept cooling down but at a slower rate, since the sample was then surrounded by air, which acted as a thermal isolator. Other studies reached similar results [48,87]; which were attributed to the different thermal conductivities of the compaction roller silicone and the air (air $\sim 0.024 \text{ W/m} \cdot \text{K}$; silicone $\sim 0.22 \text{ W/m} \cdot \text{K}$).

This results matched in a good agreement with the results reported by Oromiehie [82], who identified same regions to the ones reported in this study. However, Oromiehie claimed that the initial contact of the compaction roller on the sample and the take-off of the roller from the sample at the beginning and the end of the lamination of a tape were detected by the employed FBG.

In order to study the response of the sensors in other single layers, the monitored wavelength shifts during the lamination of layers 4, 6, 8 and 10 are presented in Figure 5.16. Same three regions (heating up (a), compaction roller (b) and cooling down (c)) can be perfectly distinguished also in those monitoring results.

Figure 5.16 Wavelength shift data monitored by the FBG sensors during the lamination of different plies, including layer 4, 6, 8 and 10.

It is notable that the cooling down rate experienced by the sensors decreased with the number of layers. This suggests that the consolidated layers on the top of the sensor acted as a heat retainer, making the heat dissipation harder. This phenomenon is related to the void rebound effect commented in previous chapters, and also reported by Comer et al. [48]. The void rebound effect is a phenomenon which can take place during the ISC process. Thus, during the lamination of upper layers (when the thermoplastic substrate thickness is significant), voids that were already compressed by the compaction roller may expand again after the roller has passed due to thermal expansion, only if the composite matrix remained at a high enough temperature. Thus, when the substrate thickness is significant, it acts as a heat retainer and may be related to the accumulation of voids in the upper layers of ISC structures.

The sensor supplied by ZEUS (PEEK coated) experienced a lower wavelength shift during the tests. Both FBGS and UPM sensor behaved in a similar manner, except in the case of the layer 4, where FBGS showed two sharp shifts, probably due to the irregularity before discussed.

In order to study the existing differences between the sensors response, the samples were cut and some coupons were extracted for 2D microscopic analysis. Figure 5.17 shows the micrographs corresponding to the samples ZEUS (left) and FBGS (right). Both sensors showed a good level of integration since voids were not found around the sensor. The core and the coating of FBGS sensor can be clearly distinguished, while the coating of ZEUS sensor (PEEK) was fully integrated to the composite matrix (also PEEK). The material of the coating may play a key role in the transfer of temperature and strains to the sensor.

Figure 5.17 Cross section of FBG sensor coated with PEEK supplied by Zeus (left) and FBG sensor coated with ORMOCER® supplied by FBGS (right).

5.5 Conclusions

In this chapter, the first steps taken in the investigation regarding the feasibility of thermoplastic AFP-ISC process monitoring with FBG sensors have been presented and discussed.

First of all, a methodology for embedding the optical sensors in the ISC manufacturing set-up was investigated. Laser-assisted AFP-ISC process represents itself as an aggressive and harmful environment for embedding brittle optical fibre sensors, mainly due to the high temperatures caused by laser irradiation and the high forces applied by a compaction roller. Thus, defining a strategy which ensured the survival of the optical fibres was mandatory for this study. The proposed embedding strategies consisted of different options, including different fibre orientations different protections at the egress point and the use of a protective tape on the top of the sensor. The results revealed that most of the sensors were damaged when they were not (or barely) protected before lamination. A successful embedding strategy was found, where most of the optical fibres survived. That strategy was based on positioning the optical fibre aligned with the incoming thermoplastic tape (0° direction), protecting the egress points with kapton (polyimide) adhesive tape and protecting the optical fibre with the protective tow. The protective tow was placed by manually bonding the thermoplastic tow on the top of the optical fibre with the aim of a welding gun, and later consolidating the tow with the automated system with no material feeding.

After finding a successful embedding strategy, an experimental monitoring plan was defined. A set of $[0]_{10}$ CF/PEEK samples were manufactured by AFP-ISC and the lamination was monitored by different FBG sensors with different features. The set of employed sensors consisted of sensors purchased from two different suppliers (ZEUS and FBGS), along with the in-house manufactured at the Polytechnic University of Madrid. Different coating materials were investigated, including PEEK and ORMOCER®. The sensors were mainly purchased from different suppliers in order to investigate the different sensors response.

The sensors were embedded between the 2nd and 3rd layers of each sample and the rest of the lamination was monitored. The system was able to record the reflected wavelength of each sensor during the process. The wavelength shifts experienced by all sensors were presented and analysed.

The tests revealed very valuable information. A cyclical pattern was observed during the lamination of each layer. Three main manufacturing stages were identified in the monitored profiles of each layer. Those regions corresponded to (a) heating up, (b) effect of compaction roller and (c) final cooling down. During the heating up, an increase of the wavelength shift was observed up to a certain point where a decrease was suddenly observed, corresponding to the denominated shadow region. The shadow region consists of the compaction roller blocking the laser irradiation over the sample for few milliseconds. After that point, the maximum wavelength shift was reached, due to the effect of the compaction roller. Cooling down started when the roller was still on the sample and it continued when the compaction roller passed. In that way, two different cooling rates were easily observed, corresponding to the cooling through the roller (silicone) followed by cooling through the air.

A decrease of the maximum wavelength shift was observed while increasing the number of layers. This was mainly attributed to the fact that more material is between the external sources and the sensor with the lamination of a new layer, causing an attenuation of the effects. The results matched well with Oromiehie et al. [81,83], who reported an inverse relation between the wavelength shift and the thickness of the fabricated laminate.

It was also found that the cooling rates decreased with the thickness of the substrate, since the composite on the top of the sensor acted as heat retainer. This phenomenon was related to the hypothesis formulated by Comer et al. [48] about void rebounds, who said that void rebound may occur due to the fact that the substrate is still at high temperature when the compaction roller passed. This may cause voids rebound due to thermal expansion, which can be related to the accumulation of voids in upper layers observed in previous chapters.

Regarding the comparison between the responses of the employed FBG sensors, the results revealed that the monitored signal of all sensors differed during the first layers (3 to 5), whereas the monitored data matched well from the fifth layer onwards. The results provided by FBGS sensor showed some irregularities in the first layers. This was mainly attributed to the fact that the digital gain was increased for FBGS sensors due to their low reflectivity. In this way, the fast wavelength shift experience due to laser irradiation, along with the higher signal-to-noise level may lead to mistakes in the peak tracking system. Sensors supplied by ZEUS (PEEK coated) experienced lower maximum values of wavelength shift in each layer. It was discussed that this may be related to different coating materials and therefore different thermal distribution to the sensors, or due to the current low repeatability of the process since the laser power was manually adjusted. In any case, those reasons are part of the discussion and more experiments are suggested regarding the effect of the coating material.

In summary, a methodology for embedding optical sensors within the AFP-ISC process has been satisfactory presented. Also, a set of monitoring tests was performed based on the process-monitoring of the fabrication of several thermoplastic samples. The results of the monitored data allowed gaining valuable information about the process. Next chapter is focused on the strategy for the thermal history monitoring during AFP-ISC lamination.

Chapter 6

Dual temperature and strain monitoring of AFP-ISC process with FBG sensors

6.1 Introduction

The embedding of FBG sensors within AFP-ISC thermoplastic samples was successfully investigated in the previous chapter. In this chapter, the methodology for monitoring the thermal profiles by means of FBG sensors is investigated, along with the initial stages for strain monitoring during process and post-residual strain assessment.

As previously described, thermoplastic AFP-ISC processing may lead to cost and time reductions since no further post-consolidations steps are required, such as oven or autoclave consolidation. However, an optimization of the process has still to be achieved, mainly driven by a further process understanding. During AFP-ISC process, the main phenomena experienced by the thermoplastic matrix are mainly governed by the thermal history and the applied pressure. Those phenomena consist of melting the resin of the substrate and the incoming tape, inter-diffusion on

the polymer chains, intimate contact and final crystallization [70,79]. Even though pressure (by means of the load applied by the compaction roller) is also a key parameter, it is clear that the thermal history (given by the laser) will eventually govern the process since it is the most relevant parameter in those phenomena. In addition to that, it was concluded in previous chapter that the applied processing temperature will also have a great impact on the final mechanical performance and the quality of the fabricated part. In fact, the applied temperature could not only lead to poor degree of intimate contact or consolidation, but also may lead to matrix degradation of the temperatures are above a certain limit. Within this context, the thermal history of the cycle must be known for being able to investigate the process and therefore to reach process optimization.

As described in the previous chapter, the processing temperature is nowadays monitored by non-contact IR thermographic cameras that are limited to surface measurements. Some studies regarding the use of IR cameras were published, as the performed by Stokes-Griffin and Compston [266] and Grouve et al. [86], who concluded that the measurements could be overestimated due to the multiple reflections of the emitted flux. Another studies investigated the use of contact devices, such as thermocouples [48,51,267].

In this chapter, a step forward is given in the use of FBG sensors for AFP-ISC process monitoring, especially after optimizing an embedding strategy in the previous chapter. In this way, the FBG sensors are mainly applied for obtaining the thermal history of the process, which is one of the main challenges of this PhD thesis.

Several strategies are known for using FBG sensors as temperature probes, which make the sensor being isolated from external mechanical interactions. In this study, the approach based on encapsulating the sensor within a metallic capillary was selected [205,209–214,268]. Thus, the sensor were expected to be isolated from the loads generated by the compaction roller and to be only sensitive to thermal shifts during laser irradiation and later cooling down. Apart from the monitoring of the thermal history, it is also investigated the strain profile taking place in the process. For this purpose, temperature compensation was also performed, in order to investigate the feasibility of monitoring the strains generated in the process. Finally, the post-consolidation residual strains were assessed by the FBG sensors.

The experimental thermal history was compared with a thermal model simulation. Also, a FEM simulation was created in order to compare the results obtained regarding the strain shifts.

6.2 Experimental methods

In this section, the experimental procedures for the investigation of dual temperature and strain monitoring of the ISC manufacturing process by FBG sensors are presented.

6.2.1 Materials and configuration

Similar procedures to the described in section 5.3.1 were applied for this chapter. Same material APC2-AS4 consisting of CF/PEEK UD 6.35 mm (1/4") width and 0.135 mm thick tapes was employed for the samples manufacturing. The manufactured samples for this chapter also consisted of $[0]_n$ specimens with 3 tows in width and 150 mm length, resulting in final specimen dimensions of 150×19 mm (length $\times$ width); and at least 10 plies in thickness.

6.2.2 Manufacturing equipment

MTorres gantry-style single-tow deposition machine located at the FIDAMC facilities (Getafe, Spain) was employed for the samples manufacturing, as described in previous section 3.3.3.

6.2.3 Interrogator system

Dynamic Micron Optics SM130 equipment introduced in previous chapter was employed for this investigation.

6.2.4 FBG sensors

In this case, FBG sensors supplied by FBGS and in-house manufactured by the UPM were employed. The sensors had similar characteristics to the ones described in previous section 5.3.4. Those features consisted of sensors in the 1500-1600 nm wavelength range, 8 mm of FBG length, outer diameter of approximately 125 μm and an optimal reflectivity of more than 80 %. As described in the previous chapter, the sensors supplied by the UPM were in-house written at the Department of Aerospace Materials and Production of the UPM, according to the procedures described by Menéndez [171].

In this case, a sensor construction which allowed the thermal history monitoring and temperature compensation was proposed, in order to being able of distinguish the effects of temperature and strain experienced by the optical sensors. Several strategies for discriminating temperature and strain can be found in the literature. In summary, methods as twisted FBG sensors [209], encapsulation of FBGs [210,212,213] or FBG sensors laid with a certain angle have been investigated [83].

In this investigation, an approach based on sensor encapsulation was selected for the discrimination between temperature and strain effects. In the case of UPM sensors, the procedure was in-house carried out according to the described by Fernandez-Lopez et al. [210]. Thus, a FBG sensor was written at the end of an optical fibre and consequently encapsulated inside a metallic capillary tube. This procedure made the FBG sensor being isolated from external mechanical variations and therefore the sensor can be considered as a temperature probe. A schematic representation of an encapsulated FBG is shown in Figure 6.1

Figure 6.1 Schematic representation of an optical fibre with a FBG array formed by a bare FBG sensor and an encapsulated sensor in a metallic capillary tube [210].

The equation which determines the wavelength shift of a FBG sensor caused by temperature and strain is given in equation (6.1), where $\Delta\lambda_b$ is the wavelength shift, κ_T and κ_ϵ are the sensitivity coefficients of temperature and strain, respectively, and ΔT and $\Delta\epsilon$ are the temperature and strain

shifts, respectively. The equation is a simplified version of more complex expressions (which can be consulted in Chapter 2), where the thermal and strain sensitivity coefficients are expressed as a function of other physical parameters such as the photo-elastic coefficient of the fibre core material or the thermal expansion coefficient of the fibre.

$$\Delta\lambda_b = \kappa_T \cdot \Delta T + \kappa_\varepsilon \cdot \Delta\varepsilon \quad (6.1)$$

In the case when a FBG sensor is isolated from external mechanical variations, one of the terms can be eliminated from the equation and the wavelength shift of the encapsulated sensor can be then rewritten, as defined in equation (6.2), where $\Delta\lambda_b^{encaps}$ is the reflected wavelength shift of the encapsulated FBG sensor.

$$\Delta\lambda_b^{encaps} = \kappa_T \cdot \Delta T \quad (6.2)$$

In this way, when a FBG is encapsulated as depicted in Figure 6.1, it is only subjected to thermal variations and can be used for monitoring the thermal history and for performing the temperature compensation of a non-encapsulated sensor. Temperature compensation is a well investigated methodology in which the temperature contribution is monitored by a temperature probe (the encapsulated FBG sensor in this case) and it is inserted in previous equation (6.1) of a non-encapsulated FBG sensor, so the contribution of the mechanical variations can be assessed.

The employed encapsulated sensors purchased from FBGS and UPM mainly differed in the OD. The sensor purchased from FBGS consisted of a commercial sensor with code TP-01. The sensor supplied by the UPM had a final OD around 0.75 mm; meanwhile sensor TP-01 had an OD of 1.7 mm. A code was given to each encapsulated sensors. In this way, FBGS-T and UPM-T codes were assigned to sensors purchased from FBGS and UPM, respectively. The different encapsulated FBG sensors were employed in the monitoring tests described in the next subsection.

6.2.5 Monitoring tests

An experimental monitoring plan was defined in order to monitor the thermal history of the AFP-ISC manufacturing process. The lamination and in-situ consolidation of the mentioned $[0]_n$ CF/PEEK samples were monitored by means of the encapsulated FBG optical sensors. After monitoring, the results obtained by the encapsulated sensors were compared to the results obtained by the non-encapsulated sensors of the previous chapter. The sensors were embedded between the 2nd and 3rd layers of each sample, using the protective tow and egress point protection procedures. The schematic representation of sensor embedding is depicted in Figure 6.2. In Figure 6.2 (i), the location of the FBG on the top of the middle tow of the 2nd layers is shown, while the embedded sensor after applying the protective tow and egress point protection methodology is shown in Figure 6.2 (ii).

Figure 6.2 Schematic representation of an encapsulated FBG sensor integration into a CF/PEEK sample: (i) placement of the FBG on the 2nd layer; and (ii) embedding of the encapsulated FBG by applying the protective tow and egress point methods.

The set-up of the monitoring tests is depicted in Figure 6.3. The figure shows the lamination of a thermoplastic sample which is being monitored by the encapsulated FBG sensor connected to the interrogator system and the data acquisition computer.

Figure 6.3 Schematic representation of the experimental set-up for process monitoring of AFP-ISC process with an encapsulated FBG sensors.

As did in the previous chapter, the samples were manufactured by means of laser heating at an approximate temperature of 400 °C at the nip point, consolidation force of 500 N and lamination speed of 1 m/min. The information about the monitoring tests is compiled in Table 6.1. In this case, two different tests were proposed including the different sensors.

Table 6.1 Detailed parameters of the monitoring trials.

Monitoring test code	FBG sensor applied	Tooling temperature (°C)	Temperature at nip point (°C)	Roller force (N)	Lamination speed (m/min)
UPM-T	UPM-T	20 (RT)	400	500	1
FBGS-T	FBGS-T				

6.2.6 Experimental determination of thermal and strain sensitivity coefficients

Once the monitoring tests were performed, the manufactured samples with embedded FBG sensors were demoulded from the fabrication tool in order to determine their thermal and strain sensitivity coefficients. It is well known that the strain and thermal sensitivity of the FBG is different between free space, surface bonded or embedded FBG sensors [210,269]. Thus, the values of the strain and thermal sensitivity coefficients of the embedded sensors employed in this study were determined experimentally for the assessment of the thermal history and later temperature compensation.

The samples with embedded sensors were subjected to tensile mechanical test and to a thermal cycle. Regarding the application of tensile loads, the employed testing machine consisted of a universal MTS testing machine model 370.10. The coupons with the embedded sensors were located in the clamp and an Epsilon extensometer was located in the middle of sample for strain monitoring. The sample was subjected to tensile loads at a constant rate of 0.5 mm/min above 1000 $\mu\epsilon$. The wavelength was monitored during the test. An image of the set-up is shown in Figure 6.4.

Figure 6.4 Set-up for the experimental determination of the strain sensitivity coefficient of the embedded sensors.

After the application of tensile loads, the coupons were located in a climatic chamber at the laboratory and subjected to thermal cycles up to 200 °C in order to assess the thermal sensitivity.

The thermal cycle was monitored by means of K-type thermocouples and a data logger, while the reflected wavelength of the sensors was also monitored. The set-up of the procedure is presented in Figure 6.5.

Figure 6.5 Set-up for the experimental determination of the thermal sensitivity coefficient of the embedded sensors.

6.3 Process simulation

In order to compare the obtained experimental results, a simulation approach was followed. The thermal history results were compared with a thermal model of the AFP-ISC manufacturing process, while a FE model was created for investigating the strains which take place during consolidation.

6.3.1 Thermal modelling

The thermal history results derived from FBG process monitoring were compared to the results obtained from a thermal model created by Martín [70], who provided the thermal model and collaborated in the comparison of the results. The model was created by using the heat transference module in COMSOL Multiphysics. The model was based on the resolution of the heat transfer equation, shown in expression (6.3), where ρ represents the material density, C_p is the specific heat capacity, k the thermal conductivity and Q represents possible heat sources.

$$\rho C_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) + Q \quad (6.3)$$

For modelling the heat transference in ISC process, a bi-dimensional geometry composed by rectangles was created in COMSOL Multiphysics. The geometry took into account the tow length in the lamination direction and also in its thickness. The employed tooling was represented as a geometrical component in the simulation. The thermal conduction through the roller was considered by a boundary condition but not a physical element with a defined geometry. The laser heating was simulated by considering power per area unit. The whole geometry was meshed by using a mapped strategy, using at least three elements in the thickness for each layer.

Heat equation (6.3) was solved following a series the boundary conditions including adiabatic tooling in both sides (6.4), laser heating source (6.5), the presence of a thermal resistive layer caused by the air trapped among the layers (6.6), heat conduction through the roller (6.7) and heat transference through the surrounding air (6.8).

$$-\mathbf{n} \cdot (-k \nabla T) = 0 \quad (6.4)$$

$$-\mathbf{n} \cdot (-k \nabla T) = Q_b$$

$$Q_b = \frac{P_{b,tot}}{A} \quad (6.5)$$

$$-\mathbf{n}_d \cdot (-k_d \nabla T_d) = k_s \frac{T_u - T_d}{d_s}$$

$$-\mathbf{n}_u \cdot (-k_u \nabla T_u) = k_s \frac{T_d - T_u}{d_s} \quad (6.6)$$

$$-\mathbf{n}_u \cdot (-k_u \nabla T_u) = h_{roller} \cdot (T_{ext} - T) \quad (6.7)$$

$$-\mathbf{n}_u \cdot (-k_u \nabla T_u) = h_{air} \cdot (T_{ext} - T) \quad (6.8)$$

For simulating the AFP-ISC head movement during the process, the boundary conditions of laser heating, interactions with the roller and air were included by means of a mathematical function that changed its position with time (and therefore with the lamination speed). The generated geometry remained inactive until it took part in the lamination process.

The inputs included in the simulation are compiled in Table 6.2.

Table 6.2 Inputs for the thermal model simulation.

Parameter	Value
Lamination speed [m/min]	1
Layer thickness [mm]	0.135
Density [kg/m ³]	1600
Heat capacity [J/kg K]	1000
h_{air} [W/m ² K]	10
Compaction roller width [mm]	12
h_{roller} [W/m ² K]	800
$h_{towtooling}$ [W/m ² K]	1000
h_{towtow} [W/m ² K]	100000

6.3.2 FEM strain modelling

For the comparison of the monitored strains, a FEM model was created using ABAQUS (v 6.14). This helped to understand the induced strains during the AFP-ISC process. The combined effects

of the loads generated by the compaction roller and the thermal stresses caused by the thermal history of the process were taken into consideration in the model.

A plane strain approach along the width of the laminate with distributed loads and temperature was chosen in order to simulate the manufacturing process and to represent the longitudinal strains caused by the compaction roller and the thermal expansion of the material. Longitudinal strains were mainly analysed since it is well known that FBG sensors are especially sensitive in the axial direction. Even though the author is aware that the manufacturing process may induce non-linear strains including three dimensional strains, only the longitudinal strains were analysed in this study, and a future work including a more complex simulation is proposed.

The analysis consisted in a planar geometry representing the $[0]_{10}$ CF/PEEK specimens. The geometry was divided into partitions that represented the plies of the samples and the incoming tow at the moment of lamination. The fingerprint of the compaction roller was also included.

A homogeneous lamina material was created with temperature-dependent elastic and expansion behaviour. The CF/PEEK lamina properties depending on the temperature and the coefficient of thermal expansion up to a temperature of 200°C were obtained from the manufactured Cytec data sheet and an Automated Dynamics technical report [259,270]. Extrapolation was applied to the properties at higher temperatures. The needed data was found in the literature, where Díez-Pascual et al. [271] reported the evolution of E modulus of pure PEEK polymer and Lu et al. [272] reported the evolution of the CTE of PEEK. Table 6.3 shows the elastic properties applied to the model whereas Table 6.4 compiles the temperature dependent values of the coefficient of thermal expansion (CTE).

Table 6.3 Temperature-dependent elastic properties given to the material in the FEA

E1 (GPa)	E2 (GPa)	ν_{12}	G_{12} (GPa)	G_{13} (GPa)	G_{23} (GPa)	Temp (°C)
138	10,1	0,25	5,4	5,4	4	25
138	8,2	0,25	4	4	3,3	165
138	2,1	0,25	0,9	0,9	0,8	200
138	1	0,25	0,5	0,5	0,4	225
138	0,9	0,25	0,4	0,4	0,4	525

Table 6.4 Temperature-dependent CTE given to the material in the FEA

α_{11} (°C⁻¹)	α_{22} (°C⁻¹)	α_{33} (°C⁻¹)	Temp (°C)
-1,40E-06	2,70E-05	2,70E-05	25
-1,40E-06	3,10E-05	3,10E-05	50
-1,40E-06	4,30E-05	4,30E-05	150
-1,40E-06	7,50E-05	7,50E-05	250
-1,40E-06	6,00E-05	6,00E-05	525

A fine mesh with 0.065 mm element size was used so the thickness of each ply was defined by partitions formed by at least three different elements. S4R Quads elements were selected for the

mesh. Two loads were applied which simulated the effects of the compaction force of the roller and the pre-tensile force applied by the spools on the incoming tow. The pressure made by the roller was estimated as a parabolic function. A maximum value of 5.2 MPa compaction pressure was applied in the nip point. The tensile force of 6 N applied to the incoming tow by a spools system in the real process was also incorporated to the model. Figure 6.6 shows the detail of the applied loads to the model. $U1=U2=0$ were applied as boundary conditions to the bottom surface which represented the attachment of the specimens to the manufacturing tooling, so a sticking between the laminate and the tool with no slipping was applied. Finally, a thermal field was added to the model in order to simulate the laser irradiation during the process. The thermal field was defined by a double negative exponential equation, in both directions x and y . The equation represented the evolution of the laser irradiation in the specimen in both directions, representing the heat attenuation in both x and y directions. The applied thermal field is depicted in Figure 6.7.

Figure 6.6 Detail of the applied load simulating the compaction roller force (left) and the load simulating the pre-tensile force acting on the incoming tape by the spool system (right).

Figure 6.7 Detail of the applied thermal field.

6.4 Results and discussion

In this section, the results derived from all the performed monitoring tests are presented and discussed. The reflected wavelength of each employed FBG sensor was monitored during the trials. As previously described, the encapsulated sensors employed in this chapter were isolated from external mechanical variations and therefore were acting as temperature probes.

For a thorough analysis of the results, the results provided by each monitoring test are first discussed individually.

The test UPM-T consisted of the monitoring of the lamination process of a $[0]_{11}$ CF/PEEK sample with the UPM-T sensor embedded between the 2nd and 3rd layers following the protective methodology. Figure 6.8 shows the detail of the embedding process of the encapsulated FBG sensor UPM-T. In Figure 6.8 (i, ii), the placement of the sensor on the top of the middle tow of the 2nd layer can be observed. Figure 6.8 (iii) shows the sample after the consolidation of the protective tow. It can be observed in the figure that the sensor was not perfectly embedded after the lamination of the 3rd layer since the sensor remained exposed to the ambient. This may be caused due to the significant external diameter of the used capillary. The uncomplete embedding of the sensor at that stage suggested that direct laser irradiation took place during the lamination of the 4th layer. In Figure 6.8 (iv), the sample is shown after the lamination of the 4th layer, where sensor UPM-T was then fully embedded into the specimen.

Figure 6.8 Embedding process of UPM-T sensor: (i) Placement of UPM-T sensor on the middle tow of 2nd layer, (ii) detail of sensor positioned on the 2nd layer, (iii) sensor partially uncovered after the consolidation of 3rd layer; and (iv) detail of sensor completely embedded after the lamination of 4th layer.

Figure 6.9 shows the monitored reflected wavelength experienced by sensor UPM-T during the lamination of the sample. Some interesting observations were deducted from the results. First of all, the sensor experienced a smaller maximum wavelength value during the lamination of the 3rd layer compared to the monitored values in the lamination of 4th layer. This fact was attributed to the incorrect embedding of the sensor during the lamination of 3rd layer, shown in previous Figure 6.8 (iii). Thus, the incomplete embedding was associated to an inadequate thermal transfer to the sensor which may lead to incorrect wavelength measurements.

According to the results discussed in the previous chapter, the special conditions of lamination of the 3rd layer (due to the sensor protective strategy) may lead in some cases to not completely reliable monitored results in that specific layer. On the other hand, the wavelength shift experienced during the lamination of 4th layer may be oversized due to possible direct irradiation of the laser over the sensor, leading to possible overestimations.

Another important finding observed is related to the baseline level of the reflected wavelength after the consolidation of each layer.

In the previous chapter, the results of monitoring test named UPM were especially discussed. A progressive variation from the baseline level of the reflected wavelength after the lamination of each layer was mainly attributed to two possible reasons consisting of residual stresses and hysteresis experienced by the sensor. During UPM-T monitoring trial, the encapsulated sensor was isolated from external strains (including those causes by residual stresses) so the mentioned variation in the baseline level caused by residual stresses was not expected. However, a variation of the baseline level of the reflected wavelength was observed during the lamination of the first layers of the sample. This effect has been highlighted in the figure with a red-dotted square in the Figure 6.9. After 5th layer, the baseline level did not vary significantly after the consolidation of the sub-sequent layers. This fact was attributed to hysteresis experienced by the sensor (highlighted in the figure) since annealing was not conducted on the sensors. As described in Chapter 5, the annealing of a FBG sensor consists of a previous heat treatment that is commonly performed in FBG applications. Thus, if the monitoring thermal and strain conditions of a system are known, it is suggested to perform an annealing to the sensors exceeding the monitoring conditions, in order to avoid the hysteresis effects.

Thus, if the operational temperature of the sensors was expected to reach up to 400°C, it is recommended to heat the sensors up to a temperature above to the working temperature in order to minimise the hysteresis effect [171,264,273,274]. Unfortunately, annealing treatment was not performed in this study. This was mainly due to the fact that commercial sensors were purchased with coating materials which could be altered during annealing at those temperatures. If a thermal cycling had been performed for long period in the case of PEEK coating of ZEUS sensors, the coating could have been lost. Hence, it was decided to do not subject any sensor to annealing if any of them could be subjected. However, some conclusions can be derived from the fact that the baseline level remained stable after the lamination of the 5th layer. Firstly, this suggests that the thermal cycles experienced by the sensor during the lamination of layers 3 to 5 acted as an annealing process themselves, since further hysteresis was not observed. Also, this also suggests that the sensor acted successfully as a temperature probe, due to the fact that the baseline level did not changed after 5th layer due to residual stresses.

Figure 6.9 Monitored reflected wavelength during monitoring trial UPM-T.

The effect of the hysteresis was digitally corrected. In this way, the wavelength shift was reset to zero after the lamination of each new tape. The results after the hysteresis correction are presented in Figure 6.10.

Figure 6.10 Wavelength shift of sensor UPM-T after hysteresis correction.

After the monitoring trial, the sample was demoulded and subjected to the thermal cycle in order to determine the thermal sensitivity coefficient of the embedded sensor as depicted in chapter 6.2.6. The results of the monitored thermal cycle are presented in Figure 6.11, where a linear fitting revealed that the sensor UPM-T had a thermal sensitivity coefficient of 83.3 pm/°C.

Figure 6.11 Experimental results for determining the thermal sensitivity coefficient for sensor UPM-T

The corrected results provided by the sensor UPM-T, along with the experimental thermal sensitivity coefficient allowed determining the thermal history of the process, according with expression (6.9), which is derived from expression (6.2).

$$\Delta T = \frac{\Delta \lambda_b^{UPM-T}}{k_T^{UPM-T}} \quad (6.9)$$

The results of UPM-T test after performing the conversion to temperature is shown in Figure 6.12. The thermal history is presented in Figure 6.12 (i) and the maximum temperature values in the lamination of each layer are shown in Figure 6.12 (ii). Dotted lines were added to the graphs indicating the melting point, T_g and room temperatures.

Many important findings were derived from these results. First of all; and according to the experimental data shown in previous chapter 5, an inverse relation can be observed between the maximum monitored temperature and the number of layers (composite thickness). This relation was observed for all the layers except for the case of the 3rd layer, due to the irregularity before discussed. One of the most important findings is based in the fact that with the lamination of each layer, at least the two subsequent layers are heated above the PEEK melting point. Thus, the sensor (located between 2nd and 3rd layers) still experienced a maximum temperature of 354 °C during the lamination of the 5th layer.

This fact may be related with the void rebound phenomenon, which refers to the thermal expansion of the voids after the pass of the compaction roller if the resin of the top layers is still at a considerable temperature. Thus, this observed fact is also related with the accumulation of voids in upper layers discussed in chapter 3. According to these results, a modification in the lamination strategy of AFP-ISC is recommended. For instance, a manufacturing strategy that may potentially address this phenomenon would consist of the use of a second compaction roller, which could aim to keep pressure for longer times until the resin is cooled down. The results were in a strong agreement with the results reported by other authors. In [227,275], the authors declared that voids can appear in upper layers of ISC-manufactured structures due to a partial de-consolidation effect when consolidating the last plies. Also, Comer et al. [48] discussed the possibility of the void rebound effect due to the thermal history of the process.

As described in Chapters 2 and 5, only one research investigation was found related to thermal history monitoring of ISC process by means of FBG sensors. In that study, Oromiehie et al. [83] reported similar results to those reached in this chapter. As previously indicated, the main difference among the investigations resided in the employed heat sources. Their results exhibited a quicker attenuation of the maximum temperatures while increasing the number of layers. Thus, the results reported here are; to the best of the author knowledge, the first time that the thermal history of a laser-assisted AFP-ISC process is monitored by means of FBG sensors.

Figure 6.12 Thermal history of laser-assisted AFP-ISC process: (i) temperature monitored by sensor UPM-T; and (ii) Maximum temperature values reached during the lamination of each layer.

For a close analysis, the monitored thermal history of several layers is shown in Figure 6.13. In this case, the thermal histories monitored during layers 4, 7 and 10 are overlapped presented. The main lamination stages can be easily distinguished. It is important to mention that the cooling rates were again observed to decrease while increasing the thickness of the sample.

Figure 6.13 Thermal profile monitored by sensor UPM-T during the lamination of layers 4, 7 and 10.

In the case of the FBGS-T monitoring test, the employed sensor was formed by a FBG sensor coated with ORMOCER and encapsulated within a metallic capillary. The diameter of the capillary was significantly larger than the one used for encapsulating sensor UPM-T. The sensor was also supplied with a PTFE capillary installed at the end of the metallic capillary for protection. The location of the sensor FBGS-T on the top of the 2nd layer is shown in Figure 6.14 (i). Due to the large external diameter of the capillary, the sensor was not correctly embedded during the lamination of the 3rd layer, and the sensor remained exposed. This can be observed in Figure 6.14 (ii). This fact may have an impact on the monitored data leading to oversized measurements, which is discussed later in this section.

Figure 6.14 Embedding procedure of sensor FBGS-T in a sample: (i) placement of the sensor on the top of 2nd layer; and (ii) sensor not correctly embedded after lamination of 3rd layer due to large external diameter of the capillary.

The monitored reflected wavelength during test FBGS-T is shown in Figure 6.15. In this case, the monitored data during the lamination of 3rd layer presented some irregularities. The monitored signal can be observed as irregular, and also smaller if compared to the subsequent layers. In addition to the irregularities on the 3rd layer, it can be also detected that the maximum peak of 4th layer is especially high. Both effects were attributed to the incorrect embedding of the sensor in those layers, as shown in Figure 6.14, especially the high maximum peak during 4th layer, where the capillary was exposed to direct laser irradiation.

Figure 6.15 Monitored reflected wavelength during monitoring test FBGS-T.

In order to convert the reflect wavelength data into the thermal profile, the sample was demoulded and subjected to a thermal cycle as before detailed, for the determination of the thermal sensitivity coefficient of the embedded sensor. The results of the monitored cycle are presented in Figure 6.16, where a linear fit was applied, revealing a coefficient of 12.2 pm/°C.

Figure 6.16 Experimental results for determining the thermal sensitivity coefficient of sensor FBGS-T.

Once the coefficient was determined, the thermal profile was directly calculated by using equation (6.10), where $\Delta\lambda_b^{FBGS-T}$ is the wavelength shift and k_T^{FBGS-T} is the thermal sensitivity coefficient of sensor FBGS-T.

$$\Delta T = \frac{\Delta\lambda_b^{FBGS-T}}{k_T^{FBGS-T}} \quad (6.10)$$

The monitored thermal profile is shown in Figure 6.17 (i), whereas the maximum temperature monitored in each layer is shown in Figure 6.17 (ii). The inverse correlation regarding the maximum temperature and the number of layers can be observed. Also, the temperatures monitored during the lamination of layers 3rd and 4th seemed not to be reliable since the embedding problematic discussed before.

Figure 6.17 Thermal history of laser-assisted AFP-ISC process: (i) Temperature monitored by the sensor FBGS-T and (ii) Maximum temperature values reached during the lamination of each layer.

Figure 6.18 shows a comparison of the thermal histories obtained by both sensors UPM-T and FBGS-T. Both monitored thermal profiles are shown in Figure 6.18 (i), whereas a comparison of the maximum temperature monitored values is shown in Figure 6.18 (ii). Several findings can be derived from this comparison. First of all, both sensors monitored questionable temperature values during the lamination of layers 3 and 4. It has been previously attributed to the difficulties experienced during the embedding of the encapsulated sensors due to the large OD. Both capillaries remained exposed to direct laser irradiation during the lamination of the 4th layer, which could eventually lead to overestimated temperature measurement during the consolidation of the layer. In the same way, the measurements during consolidation of layer 3 may be underestimated, due to incorrect embedding. From 4th layer onwards, both sensors experienced a decrease in the maximum monitored temperature. However, it is important to remark that FBGS-T sensor monitored higher temperature values systematically during the lamination of the rest of the layers. This effect was mainly attributed to the existing differences between the OD of both sensors. Two cross sections of the samples containing UPM-T and FBGS-T sensors are presented in Figure 6.19. It can be observed that the diameter of the encapsulation system is comparable with the thickness of several plies. The external diameter of both sensors was measured experimentally with the microscope and the results were 0.794 mm for UPM-T and 1.690 mm for FBGS-T. The observed differences between the monitored thermal histories was attributed to the differences between the sensor ODs. Thus, a system with larger OD is closer to the external heat source, therefore experiencing higher temperature measurements. In addition to that, a larger OD may cause a larger bulky area in the sample surface when laminating. The bulky area itself may eventually cause an increment in the temperature due to unexpected laser reflections since the surface is not flat. This may involves higher temperatures over the sensor.

Figure 6.18 A comparison of the monitoring trials with UPM-T and FBGS-T sensors. (i) Thermal histories monitored and (ii) Maximum temperature values reached during the lamination of each layer.

Figure 6.19 Cross-sections of samples containing encapsulated FBG sensors: (right) UPM-T sensor and (left) FBGS-T sensor.

The thermal histories obtained with both sensors were finally compared to the thermal history provided by the Martín thermal model [70]. The comparison is shown in Figure 6.20.

Overall, a good correlation was observed between experimental and simulated thermal histories. Both results showed a progressive decrease of the maximum temperature with the lamination of each new layer. However, there are some discrepancies between the simulated and experimental results. The maximum temperature values matched very well during the first layers but they diverged in upper layers.

The mismatch of the reached temperatures in upper layers was attributed to a possible overestimation of the temperature measurements due to the OD of the employed sensors. This may explain the higher maximum temperatures monitored by sensor FBGS-T (OD $\sim 1690 \mu\text{m}$) compared to sensor UPM-T (OD $\sim 794 \mu\text{m}$). Thus, when the simulation is predicting a certain temperature when the laser is at a given temperature; the experimental data provided by the sensors may be overestimated because the OD of the sensor causes that the sensor itself is closer to the external source and monitors a higher measured temperature. In order to address this issue, the use of a correction factor related to the OD of an encapsulated sensor is proposed. Further research is suggested in this investigation line.

On the other hand, cooling rates also presented some variations for the case of the simulated results. This was associated to an underestimation of the parameter h_{roller} that controls the thermal interchange between the roller and the thermoplastic composite, according to Martín [70].

Figure 6.20 Comparison of thermal profiles obtained from experiments and COMSOL simulation.

The fact of overestimated temperature measurements with encapsulated FBG sensors complicated the temperature compensation procedure for extracting the strain field. As mentioned before, an encapsulated FBG is isolated from external mechanical variations, so the wavelength shift is expected to be lower when compared to a bare FBG sensor (which is sensitive to both thermal and strain variations). However, the OD of the encapsulated sensors was causing an overestimation in the wavelength shift which can eventually cause a higher monitored signal when compared to the bare sensor, therefore complicating the temperature compensation procedure.

The wavelength shift profile obtained with the encapsulated sensor UPM-T (Figure 6.12) was compared to the monitored profile of its counterpart bare sensor (UPM) investigated in the previous chapter. Thus, the temperature compensation was carried out in order to separate the effect of temperature and strains in the bare UPM sensor. It was found that the monitoring responses of UPM and UPM-T sensors in the lamination of 5th layer were almost identical, with a slight difference in the maximum peak, where the maximum wavelength shift of sensor UPM was higher than UPM-T, as theoretically expected. Thus, Figure 6.21 shows the comparison of both monitored wavelength shifts during the lamination of the 5th layer by UPM and UPM-T sensors. Layer 5 was considered representative of the process. The variation between the sensors response has been also added to the figure. The main difference between the monitored signals is found in the phase when the compaction roller applied the force (highlighted in the figure). Due to the sensor configurations, the variation between the monitored data is only related to strain changes.

Figure 6.21 Comparison of wavelength shift monitored during the lamination of a single tape with a bare sensor (UPM) and encapsulated sensor (UPM-T). The variation between the sensors is also plotted in the graph.

Temperature compensation was performed using equation (6.11), where $\Delta\epsilon$ is the strain variation, $\Delta\lambda_{UPM}$ is the wavelength shift of bare sensor UPM, $\Delta\lambda_{UPM-T}$ is the wavelength shift of the encapsulated sensor UPM-T and κ_{ϵ}^{UPM} is the strain sensitivity coefficient of the bare sensor. This coefficient was measured experimentally, according to the details compiled in previous section 6.2.6. The results revealed a coefficient of 1.8 pm/ $\mu\epsilon$ for the UPM sensor.

$$\Delta\epsilon = \frac{(\Delta\lambda_{UPM} - \Delta\lambda_{UPM-T})}{\kappa_{\epsilon}^{UPM}} \quad (6.11)$$

Figure 6.22 shows the strain shift derived from temperature compensation. The figure consists of a zoom into the region of interest where the compaction force was applied. The approximate time when the compaction force acted has been highlighted. It is important to remark that in this study, the monitored strains obtained after temperature compensation were considered longitudinal strains, due to the fact that the FBG sensors are especially sensitive to longitudinal strains. The author is aware that during AFP-ISC process also non-longitudinal strains may take place, which may cause distortion of the FBG spectrum. In this way, further research is proposed with the use of more complex sensors and techniques which can distinguish among longitudinal and transverse loads; such as MOFBG sensors.

According to Figure 6.22, a tensile effect was first experienced by the sensor, followed by an immediate compression. The figure also shows the prediction of the longitudinal strain shift obtained from the created FEM model, which qualitative matched the experimental results. Thus, the model predicted also a tensile peak followed by compression. The results are in a good agreement with the results achieved by Oromiehie [82], who reported a maximum compressive strain in the consolidation of each new ply.

The deviation observed between the experimental and simulated results was mainly attributed to the simplicity of the model in contrast with the high complexity of the manufacturing process itself. Further work is suggested for creating a more thorough model. In [82], the author also attributed the deviation between experimental and simulated results to the complexity of ISC process simulation.

Figure 6.22 Strain shift monitored by the FBG sensor (UPM), obtained after performing temperature compensation.

In order to extract more valuable information about the use of the FBG sensors for residual strain monitoring of the ISC process, an additional test was carried out. Due to the fact that residual stresses are one of the main challenges to overcome of the AFP-ISC process, the FBG sensors are an adequate tool for determining the accumulated post-residual stress in a thermoplastic part after fabrication. With this aim, a monitoring trial was performed based on the monitoring of the demoulding activities. For the trial, two FBG sensors were embedded at the bottom and top plies of a $[0]_{10}$ CF/PEEK sample. In this way, if the sample experiences a deformation after manufacturing and demoulding due to the residual stresses of the process, the FBG sensors would be capable of measure the final strains accumulated. The employed sensors were supplied by the UPM and had similar features to the other FBG sensors in-house manufactured by the UPM employed in this investigation.

A schematic representation of the monitoring set-up is shown in Figure 6.23, where the location of the FBGs in the upper and lower layers can be observed. Another schematic representation of the expected deformation behaviour of the sample after demoulding due to the accumulated residual stresses is depicted in Figure 6.24

Figure 6.23 Schematic representation of the set-up for the demoulding monitoring with FBG sensors.

Figure 6.24 Schematic representation of the deformation of a thermoplastic sample after demoulding due to the accumulated residual stresses.

The results derived from the demoulding monitoring are shown in Figure 6.25. It was expected that the sample suffered a deformation after fabrication, where the upper layers experienced compressive effects meanwhile the bottom layers experienced tensile effects. These predicted effects were validated experimentally since the monitored strains by the FBG sensors revealed $176.1 \mu\epsilon$ derived from tensile loads in the bottom layers meanwhile the sensor in the upper layers revealed $-194.4 \mu\epsilon$ corresponding to the compressive loads experienced in the upper layers. The deformation of the sample is presented in Figure 6.26.

This monitoring trial consisted of a proof of concept where the FBG sensors were validated for monitoring the final strains accumulated in a thermoplastic sample due to residual stresses.

Figure 6.25 Microstrains experienced by the employed FBG sensors during the demoulding of a thermoplastic sample

Figure 6.26 Deformation of the thermoplastic sample after demoulding.

6.5 Conclusions

In this section, the main conclusions obtained after investigating the use of FBG sensor for monitoring the thermal history and the strain field during AFP-ISC process were presented.

An experimental monitoring plan was designed. For the thermal history monitoring, an approach based on the use of encapsulated FBG sensor was employed. The encapsulation of FBG sensors within metallic capillaries is a known methodology for isolating FBG sensors from external mechanical effects [205,209–214,268]. In this way, an encapsulated sensor acts as a temperature probe. Two different encapsulated sensors were applied in this investigation, consisting of sensors supplied by the UPM (in-house manufactured) and commercial sensors supplied by FBGS International. The sensors had similar features with the exception of the OD, which was considerable larger in the case of the FBGS sensor (OD~1690 µm) compared to UPM sensor (OD~794 µm). The temperature sensitivity coefficients were obtained experimentally.

Both sensors successfully monitored the thermal history of the AFP-ISC manufacturing process and those results consisted of the first of its kind being published related to the process monitoring

of a laser-assisted ISC manufacturing process. Both monitored thermal histories were compared. The results revealed that the FBGS-T sensor monitored higher maximum temperature values in the lamination of each ply. The slightly higher temperature values monitored by FBGS-T were attributed to the difference in OD. Thus, a sensor encapsulated in a capillary with larger diameter may be closer to the external heat source and therefore may be monitoring overestimated temperature profiles. These results suggested that the final diameter of the employed probe is decisive for reliable temperature measurements. Future work is proposed with the use of small diameter capillaries or with other monitoring strategies such as angled FBGs.

The results were compared with a thermal model developed by Martín [70], which is based on the resolution of the heat transfer equation. The comparison of experimental and simulated results revealed that the thermal profiles monitored by both sensors were higher than the simulation. Those results are in line with the previous claim about the diameter of the capillary being a key parameter to take into account which eventually affects the final monitored thermal history.

The possible overestimation of the monitored temperature data complicated the acquisition of the strain field obtained after temperature compensation. Theoretically, the response of an encapsulated sensor should be smaller when compared to the response of a bare sensor, since the encapsulated sensor is isolated from external strains. However, the encapsulated sensor may monitor a higher signal when compared to a bare sensor. Nevertheless, it was found several cases where the signal of the encapsulated sensor was smaller than the bare sensor, especially in the area where the compaction roller applies the load, according to the theoretical hypotheses. The profiles were compared and the difference between signals was associated to the strain experienced by the sample during the consolidation. The strain shift suggested that the sample first experienced a tensile peak followed by a compressive peak. Those results were in-line with the predicted by the performed FEM simulation. However, some differences were appreciated between the experimental results and the simulation, mainly attributed to the simplicity of the created FEM model in contrast with the high complexity of the process. The results were in line with those reported by Oromiehie [82], where maximum compressive strains were reported during the lamination of each layer. In their study, the high complexity of simulating the ISC process was also declared. The monitored results related to the strain field consisted of an important step forward for the use of FBG sensors for dual temperature and strain process monitoring.

A final monitoring trial was performed based on the strain monitoring during the demoulding of a sample, in order to investigate the post-deformation experienced by a thermoplastic part due to residual stresses accumulated during lamination. The results confirmed that FBG sensors can determine the final strains caused by the residual stresses.

It is also important to note that the investigation performed in this chapter along with the previous chapter determined a successful strategy for embedding FBG sensors (bare or encapsulated) into ISC-manufactured parts. This opened the opportunity to sensor integration during manufacturing for future SHM activities of thermoplastic composite structures.

Chapter 7

Concluding remarks

According to the results obtained in this research work, this chapter compiles all the main achieved conclusions and milestones.

Overall, this thesis has contributed to gain knowledge regarding laser-assisted AFP of thermoplastic composites with in-situ consolidation technology. Several issues were identified regarding the manufacturing process which led to start this investigation. The main issues were centred on the drop of mechanical properties of ISC laminates, the lack of interrelation between porosity, ultrasonic testing and mechanical properties, and the interest in developing a process monitoring technique, especially focused on the thermal history monitoring.

The main conclusions are listed and discussed hereunder in this section.

- **Characterization of the mechanical performance of different OoA manufactured thermoplastic composites**

As before mentioned, one of the main detected issues which justified this investigation was the drop of mechanical properties of thermoplastic laminates when they are fabricated by ISC process. In this way, a set of thermoplastic laminates were manufactured by using three different OoA manufacturing techniques, including hot-press, vacuum bag-only (VBO) and ISC. Among

the technologies, VBO was selected as the reference technique. A set of mechanical tests which combined fibre-dominated and matrix-dominated tests were selected for the mechanical characterization, including tensile, flexure, ILSS and IPSS. According to the applied standards, the lay-up of the laminates subjected to tensile, ILSS and flexure tests was $[0]_{16}$, whereas $[45/-45]_{48}$ was the lay-up for IPSS tests. The results revealed that laminates manufactured by VBO and hot-press techniques behaved in a broadly similar way; meanwhile ISC laminates experienced a drop of properties. The drop of the strength of the different studied properties for ISC laminates compared with the reference consisted of 33.8 % for ILSS, 30 % for IPSS, 16.9 % for flexure and 13.2% for tensile. It was detected that, as expected, the drop of properties was more evident in the matrix-dominated properties. The investigation of these phenomena was performed encompassing different perspectives, such as void content, void size and distribution, and degree of crystallinity. The results revealed that hot-press and VBO laminates were around 1 % of void content, while ISC laminates presented a void content around 3 %. Besides void content, void size and distribution were investigated by means of X-ray micro-CT (an individual point is given to those results later on this section) and the results revealed a different void morphology and distribution for the ISC laminates, which can be one of the main reasons causing the drop of properties. DSC results revealed that the drop of crystallinity of ISC-manufactured laminates was not as sharp as expected; if compared with similar studied. The main reason behind the high level of crystallinity found in ISC laminates was attributed to the use of self-heating laminating tooling, which were heated up to 200 °C during fabrication.

▪ Analysis of the voids size and distribution of ISC thermoplastic composites

As mentioned in the previous point, 3D X-Ray micro-CT was performed in this investigation for analysing the voids size and distribution of thermoplastic laminates, including samples manufactured in the three OoA methods. Regarding void distribution, it was found that voids are mainly accumulated at interlaminar regions (between plies) in ISC laminates; meanwhile they are homogeneously distributed in VBO and hot-press laminates. It was also found that voids can remain especially occluded in upper layers of the samples. A hypothesis was formulated at that point (and validated later in the study), which related those results to the void re-bond effect. This effect was based on the voids expanding after a compaction roller pass due to the still very high temperatures caused by the substrate acting as a heat retainer. Regarding the study of voids size, it was revealed that laminates manufactured by ISC contained more and larger voids than those laminates manufactured by VBO and hot-press. In addition to that, projections of the stacking of XY slides of the sample were created, which allowed visualizing the micro-CT results in the same way as ultrasonic c-scan. This was a first step in a possible further investigation which may correlate tomography and ultrasonic inspection.

It was deduced from the results that not only the average void content should be analysed in a production line of composites structures, since voids distribution and size may be a critical factor leading to a significant drop of mechanical properties or even to a critical early failure, and could not be detected by standard NDT methods.

To date, this study is the first investigation regarding the study of voids size and distribution of ISC-manufactured thermoplastic laminates

- **Definition of a manufacturing methodology of porosity-induced thermoplastic laminates**

As before mentioned, one of the main challenges of this investigation was to elaborate interrelations between porosity, ultrasonic NDT and mechanical response. A lot of information can be found in the literature about thermoset composites but little information is found regarding thermoplastic composites. For this investigation, it was necessary to manufacture a set of thermoplastic laminates in a wide range of porosity. With this aim, different manufacturing strategies were investigated for producing laminates with different levels of porosity. One of the main challenges while doing research about porosity is the high complexity of producing samples with porosity, since void dynamics mechanism are driven by different factors and preferential distribution channels (where voids can move) are mainly random.

The investigated manufacturing methodologies consisted of (i) sealing the edges of the thermoplastic preforms before consolidation to avoid void extraction; (ii) submitting to the thermoplastic prepreg to moisture absorption before consolidation to induce porosity in the final part; and (iii) modification of process key parameters in the consolidation cycle in order to cause porosity.

In an earlier stage, it was demonstrated that methodologies (i) and (iii) were able to produce laminates in a different porosity range; meanwhile methodology (ii) was cancelled due to the low moisture absorption observed in the thermoplastic prepreg. A final decision was taken and the investigation continued by following strategy (iii), since it was the only strategy that can be extrapolated to the three manufacturing techniques under research.

Thus, a manufacturing plan was designed by modifying the key manufacturing parameters of the manufacturing process. Processing temperature was modified in the three manufacturing techniques, while pressure was only modified in hot-press and ISC manufacturing, by means of the hydraulic press pressure and the load generated by compaction roller, respectively.

Finally, a set of CF/PEEK thermoplastic was manufactured in a range of porosity from 0 to 19 %.

- **Correlation between void content, ultrasonic attenuation and mechanical response of thermoplastic composites**

Once the set of thermoplastic laminates with induced porosity was manufactured, an experimental characterization programme was designed based on void content assessment, ultrasonic NDT and mechanical tests. The results allowed obtaining interrelation curves between those magnitudes.

These curves consisted of combining the ultrasonic absorption coefficient α and the void content leading to a bilinear fitting which suggested that attenuation (in dB) increases drastically beyond a certain value of void content. Also, the relation combining void content and mechanical properties suggested that the laminate strength started to decrease beyond a certain level of void content. That void content level was denominated critical void content and it was assessed for each manufacturing technique, which showed different results. Thus, different manufacturing technologies showed different behaviours, indicating that each of the manufacturing method should have its own quality control criteria. This evaluation can be very useful in engineering applications of thermoplastic composites and the creation of NDT acceptance criteria.

To date, this study presented the first interrelation of its kind.

- **Definition of a strategy for embedding FBG sensors within ISC-manufactured thermoplastic parts**

The first step performed towards the ISC process monitoring by means of FBG optical sensors consisted in investigating an embedding strategy of the sensors themselves in the structures under fabrication in the ISC process. The typical manufacturing conditions given during ISC manufacturing involve a harsh environment for the implementation of FBG sensors; what made necessary the study of the sensors survival through a specific embedding strategy. The main issues which were addressed consisted of the direct irradiation of the laser over the optical fibre that may cause damage on the fibre, the very high temperatures employed, typically around 400 °C, the compaction loads applied by a compaction roller and the inlet/egress points where the compaction roller may induce shear loads which could be decisive for the sensor breakage.

A test plan was designed by trying different embedding strategies or configurations. Those configurations were defined by changing the optical fibre orientation (parallel or perpendicular to the lamination direction), the protection of the egress points and the application of a protective tow.

After investigating the different configurations, it was detected that one of the strategies was successful in most of the trials. That strategy consisted of combining the following operations: laying the optical fibre in the taping direction (0°, parallel to the fibres), protecting the inlet and egress points with kapton (polyimide) adhesive tape and locating a protective CF/PEEK tow on the top of the sensor in order to avoid direct laser irradiation over the sensor which could eventually deteriorate the sensor and the optical fibre. The placement of the protective tow was performed manually, and the tape was bonded to the substrate with the aid of a manual welding gun.

The developed embedding strategy allowed extending the investigation related to ISC process monitoring with FBG optical sensors.

- **Real time monitoring of the ISC process with FBG sensors**

Once that the embedding strategy of FBG sensors in the composite substrate for ISC process was optimized, a set of FBG sensors were employed for investigating the feasibility of ISC process monitoring by means of FBG sensors. For this purpose, different FBG sensors were used for monitoring the automated lamination and consolidation of different thermoplastic samples.

Different sensor configurations were investigated mainly focused in different external coating material. The employed FBG sensors were purchased from different suppliers, including ZEUS, FBGS and in-house written sensors in the UPM. The studied coating materials consisted of PEEK, ORMOCER® and standard acrylate, respectively.

The monitoring test demonstrated the feasibility of ISC process monitoring with FBG sensors. The results helped to obtain valuable information about the process. The wavelength shift experienced by the sensors was monitored during the lamination of the samples. The main phases of the lamination of each new layer were identified and consisted of: (a) heating up, (b) effect of compaction roller and (c) final cooling down. During the heating stage, an increase of the signal was detected until a sudden and short decrease, corresponding to the denominated shadow region,

where the roller blocked the laser irradiation over the sample causing a rapid decrease of the temperature. After that, a maximum was observed, corresponding to the effect of the compaction loads. Another finding was reached, based on two observed different cooling rates. Those cooling rates corresponded to the cooling through direct contact with the roller and later cooling through air once the roller passed by.

Also, an inverse correlation of the maximum wavelength shift with the number of layers was found. This relation was attributed to the increasing material thickness between the laser source and the sensor. In addition to that, it was found that the cooling rate decreased with the number of layers, meaning that the composite substrate may act as a heat retainer when the number of layers increases. This fact may be related to the void rebound phenomenon and may explain the accumulation of voids in upper layers in some ISC thermoplastic laminates.

▪ **AFP ISC thermal history monitoring with FBG sensors**

One of the main milestones achieved during this study was the monitoring of the thermal history of the ISC process by means of FBG optical sensors. As commented in several occasions in the document, the current state of ISC technology has a non-contact temperature monitoring system based in an IR thermal camera, which is limited to surface measurements and may also lead to overestimated measurements. In this study, a monitoring approach using encapsulated FBG sensors was followed. By encapsulating a FBG sensor, the sensor is isolated from external mechanical variations and acts as a temperature probe. Encapsulated in-house UPM sensors were employed for the thermal history monitoring, along with encapsulated sensors purchased to FBGS. The results suggested that the laser irradiation applied for the consolidation of a new tape may melt up to 4-5 plies below the one being consolidated. The comparison among sensors revealed that the OD of the encapsulation plays a key role on the thermal history monitoring. It was observed that sensor with a large OD monitored higher maximum temperature peaks in each new layer, suggesting that the measurements could be overestimated.

The experimental thermal histories achieved were compared to a thermal simulation performed by Martín [70]. The experimental results presented higher maximum temperature values when compared to the simulation. This also suggested that the OD of the encapsulation system may have an impact on the final accuracy of the monitored results.

The results presented in this study consisted in the first time that the thermal history of a laser-assisted ISC process has been monitored by means of FBG sensors.

▪ **Dual temperature and strain monitoring of the AFP-ISC process with FBG sensors**

The proposed encapsulating system for FBG sensors allowed monitoring the thermal history of the AFP-ISC process. This thermal history was used for performing temperature compensation in the signal monitored by a bare FBG sensor which was excited by both temperature and strain, in order to separate the effects of the experienced temperature and strain during consolidation.

The monitored strain shift was compared to a created FEM model. Both experimental and simulated results suggested that the substrate experiences tensile loads followed by compressive loads.

This was a major step for the dual temperature and strain monitoring of the ISC process by means of FBG sensors. However, the monitoring process should be still optimized.

Chapter 8

Future work

The analysis of the results generated in this PhD project allowed suggesting new research lines which may continue this research work.

The main suggested research lines are presented hereunder in this chapter.

- To increase the study of the impact of the processing parameters in the final void content of the thermoplastic laminates by adding the dwell time as one of the key manufacturing parameters. Some studies have reported in the past the optimal consolidation dwell times for thermoplastic consolidation.
- To investigate the rheological behaviour of the thermoplastic prepreg depending on the applied consolidation temperature. In this study, different consolidation temperatures were applied for obtaining porosity laminates based on the assumption that the thermoplastic matrix would prevent the voids evacuation if the resin viscosity is high enough. However, rheological experimental work was not performed in this work and therefore it is proposed as future work.

- To perform numerical simulation regarding the effect of defects in thermoplastic composites. In this work, the impact of the porosity was evaluated in an experimental way. An interesting continuation of this work could be based in the simulation of different defects (mainly voids) including different defects content, size and distribution; and the further simulation of the mechanical testing.
The present study could be used for validation of the numerical simulations, which can consist of an interesting tool for reducing costs and times in future characterization and certification activities. This research is commonly known as “virtual testing”.
- To study the morphology of the crystalline part of the thermoplastic composites after manufacturing. In this study, only the degree of crystallinity of the manufactured laminates was analysed by means of DSC testing. Future work may investigate not only the degree of crystallinity, but also the morphology of the crystals depending on the manufacturing process and the employed manufacturing parameters. For this purpose, techniques such as wide-angle X-Ray diffraction (WXRd) are recommended.
- To develop a machine learning tool based on the comparison of ultrasonic c-scans and micro-CT analysis. Thus, a c-scan may offer more information (regarding void distribution or size) than the current state of the technology does if a thorough correlation is performed between the results provided by ultrasonic c-scan and micro-CT for a considerable sample population.
- To study the impact of more processing parameters in the final mechanical behaviour of ISC-manufactured laminates. In this aspect, there are more parameters that could be studied than those investigated in this work, which may result of high interest. Parameters such as lamination speed, roller geometry or roller material could have also an impact in the final structural behaviour.
- To investigate different manufacturing concepts for the AFP-ISC process. New manufacturing concepts or strategies were proposed in several sections of this study. Those strategies may help to overcome some current issues, such as the drop of mechanical properties of the laminates or the accumulation of voids in the upper layers. Thus, manufacturing strategies such as the use of a second compaction roller are susceptible of being investigated.
- To investigate the residual stresses which appear in the ISC-manufactured thermoplastic structures. An investigation based on the analytical study of the residual stresses is proposed, along with an experimental plan to relieve the final stresses in the parts.
- To extend the monitoring activities of ISC process by monitoring with distributed optical fibre sensors. In this study, FBG sensors were employed for monitoring the ISC process. Even though it was a major step towards ISC process monitoring, FBG sensors can only offer local information. The use of distributed optical sensors may improve the presented study.

- To create a close-loop system for the laser power control. In this study, the laser power is the ISC equipment was adjusted manually while manufacturing all the samples. A future work based on the development of a close-loop system which may adjust the laser power automatically from a input provided by embedded optical sensors is proposed.

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