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# Unidirectional Carbon Fiber Reinforced Thermoplastic Tape in Automated Tape Placement Process

Svetlana Risteska

## Abstract

Thermoplastic matrix composites are finding new applications in the different industrial areas, thanks to their intrinsic advantages related to environmental compatibility and process-ability. The tape placement process is one of the few techniques that have the potential to continuously process thermoplastic composites in large industrial applications. Fiber-reinforced thermoplastic tapes are subjected to high heating and cooling rates during the tape placement process. The application of laser heating for the tape placement process requires a thorough understanding of the factors involved in the process. Qualitative experimental analysis is presented to identify the important phenomena during the tape placement of carbon (PEEK, PEKK, PAEK PPS) tapes. The present chapter focuses on the input parameters in the process of manufacturing composite parts. The mechanical performance of the final parts depend on a number of parameters. It should be void-free and well consolidated for reliable use in the structure. In the present work, it is becoming increasingly wiser to introduce the production of high-quality laminates, using laser AFP and ATL with quality consolidation during the laying process. The experimental results in this chapter help to better understand the consolidation process during LATP.

**Keywords:** reinforced, fiber, thermoplastic, laser-assisted automated tape placement (LATP), *in situ*

## 1. Introduction

The thermoplastic materials do offer a great advantage over thermoset prepreg and dry fiber materials [1, 2]. For this advantage to be realized, thermoplastics must better compete with thermoset prepreg production for large structural parts in aviation. Today, most thermoplastic research has centered around full *in situ* consolidation to eliminate post-processing for curing. The main restriction of full *in situ* consolidation is on the speed (and productivity) of the LATP/LAFP process, which is limited by the physics or chemistry of thermoplastic materials. This method places all the demands of final part quality (low porosity, proper crystallinity, and autohesion) on the AFP process.

Robotic automated fiber/tape laying processes are used to produce high-performance composite components from unidirectional precast materials. In this

procedure, a robot is most often used to lay the lanes along pre-defined paths, which provides a high degree of freedom in designing the final product. The process of automated laying of strips consists of automated laying, that is, stacking of layers on top of each other on a flat or complex-shaped tool, which is called a mandrel. Pre-layering is associated with the application of heat and pressure. It is important during this procedure to allow the strips to consolidate directly during installation, which will prevent additional time and energy from being spent on post-consolidation [3–7].

Most of the research for automated production is oriented toward the application of thermoplastic prepress materials, although today many responsible and load-bearing composite parts obtained with thermoset materials are in use [5]. The reason for such research is that when processing a thermoplastic prepreg, the material can be bonded on-site directly during layup (placement). However, when processing a thermoplastic prepreg, just like when processing a thermosetting prepreg, many challenging problems occur, such as gaps and overlaps between the tapes, which affects the final characteristics of the material, which requires in-depth research. The research done for the application of robotic processes for automatic laying of fibers/tapes is expected to provide better obtaining of high-quality structures from composite materials with a new process called on-site consolidation (*in situ*) [6–9].

The advantages and benefits of the application of these procedures for automated laying of fibers/tape for obtaining parts of composite materials are the following [4]:

- Higher productivity in production,
- Better quality of the composite,
- Less unused waste material,
- Less labor,
- Shorter production time,
- Reduced qualifications of the professional production staff,
- Accuracy and repeatability of the product,
- Software solutions.

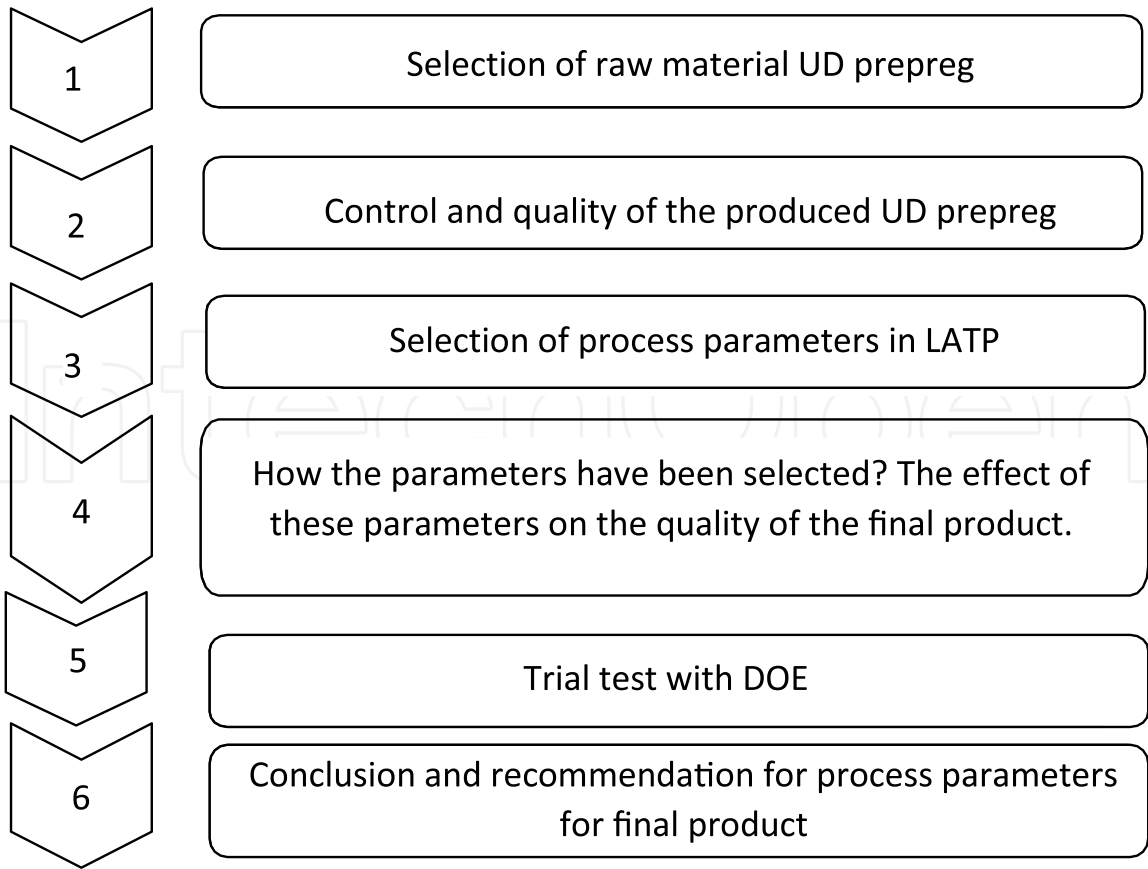
The concept of *in situ* consolidation is simple. The inlet thermoplastic pretape in contact with the substrate or mandrel under sufficient pressure and temperature above the melting point is consolidated and crystallized upon cooling at the controlled or uncontrolled speed [10–25].

## 2. Manufacture of the final product with LATP/LAFP technology

The steps for the production of a quality final product in this process are given in **Figure 1**; in each subchapter, all the steps are elaborated in more detail.

### 2.1 Selection of raw material UD thermoplastic prepreg

A very important process parameter is the material used in LATP/LAFP processes. The production process for obtaining composites from thermoplastic materials mainly depends on the viscosity of the resin and therefore, impregnation is very



**Figure 1.**  
*Procedure for making a quality product with LATP technology.*

important in the processing of thermoplastic materials, that is, the characteristics of the raw material. The characteristics of the thermoplastic prepreg determined by the content of pores, volume fraction of fibers, degree of wetting of the fibers, uniformity, surface roughness, etc. determine the potential of the process. In order to make a suitable and good quality product from thermoplastic prepreg, the first thing that needs to be done is to look for suitable raw material. Thermoplastic tapes, for the purposes of this LATP/LAFP process, consist of unidirectionally aligned carbon fiber tows in widths of up to 12 inches/305 mm, prepregged with a thermoplastic resin. The resins most commonly used in aerospace and other high-performance applications are the following high-performance thermoplastics—polyether ketone (PEKK), polyether ether ketone (PEEK), polyaryl ether ketone (PAEK), polyphenylene sulfide (PPS), and polyetherimide (PEI). Some manufacturers offer tapes prepregged with commodity thermoplastic resins, such as polyamide (PA6), polyether sulfone (PES or PESU), polypropylene (PP), and others, but these are generally considered unsuitable for large aerostructures.

Thermoset tapes are available in widths of up to 60 inches/1524 mm and can go thousands of meters without a defect, while thermoplastic tapes typically top out at 12 inches/305 mm and show as many as 30 defects in just 210 m ft. It is because the thermoplastic UD prepregs rely on a powder-based application process that is more difficult to control and can create resin-rich and dry areas. Such nonuniformity can lead to problematic interplay porosity.

Some materials used in this technology from different manufacturers are given in the **Table 1** and **Figure 2**.

The key point is that high-quality tapes, with low void content, are an enabler of fast, automated processing of high-quality composites. Tapes with high levels of voids will require longer consolidation cycles to produce high-quality parts. Today there are several manufacturers of thermoplastic UD tapes, such as Suprem,

Matrix	Supplier	Type	PAW (g/m <sup>2</sup> )	Carbon fiber	Thickness (mm)
PPS	Ten Cate	UD TC1100 PPS/AS4UD	220	Standard	0.14
	Suprem	Suprem T 60% AS4 / PPS-214	220	Standard	0.14
	Barrday	C/PEEK TU200–145-HM63–37-12	220	IM	0.14
	Barrday	TU0110–60% PPS DIC MA520/AS-4D	220	Standard	0.14
PEEK	Ten Cate	TU0200 PEEK VICTREX 150-AS4D	220	Standard	0.14
	Ten Cate	TU0200 PEEK /H800	220	IM	0.14
	Ten Cate	TU0200 PEEK /IM7	220	IM	0.14
	Suprem	Suprem T 60% AS4 / PEEK-150	220	Standard	0.14
	Suprem	Suprem T 60% AS4 / PEEK-151	220	IM7	0.14
	Barrday	TU0200-PEEK VICTREX 150-AS4D	220	Standard	0.14
	Barrday	TU0200-PEEK VICTREX 150-IM7	220	IM	0.14
	Barrday	MC0000-KG is C/PPS TU0110–145-HM63–37-12	220	IM	0.14
	Tejlin (T.Tenax)	TPUD PEEK-HTS45	220	Standard	0.14
	Tejlin (T.Tenax)	TPUD PEEK-6-34-IMS65 P12 24 K-UD	220	IM	0.14
	Solvay	Solvay’s APC 2 (PEEK-FC)	220	IM7	0.14
	Solvay	Solvay’s APC 2 (PEEK-FC)	220	Standard	0.14
PEKK	Ten Cate	Ten Cate’UD TC1320 PEKK/ AS4	220	Standard	0.14
	Suprem	Suprem T 60% AS4 / PEKK-7002	220	Standard	0.14
	Suprem	Suprem T 60% IM7/ PEKK-7003	220	IM	0.14
	Suprem	Suprem T 60% AS4 / PEKK-7002	300	Standard	0.19
	Barrday	TU300 AS-4D PEKK ARKEMA 7003	220	Standard	0.14
	Solvay	Solvay’s APC (PEKK-FC)	220	Standard	0.14
PAEK	Ten Cate	TC1225 Cetex® TC1225 LM PAEK, AS4D	220	Standard	0.14
	Tejlin (T.Tenax)	PAEK/CF Tejlin-UD	220	Standard	0.14

**Table 1.**  
*UD tape thermoplastic prepreg from different suppliers for LATP/LAFP.*

Barrday, Tejlin (Toho Tenax), Toray advanced composite (Tan Cate), Solvay, Celstrane, etc. They all offer different thermoplastic tapes as mentioned above with different matrices and of course with different carbon fibers (**Figure 2**). So, before



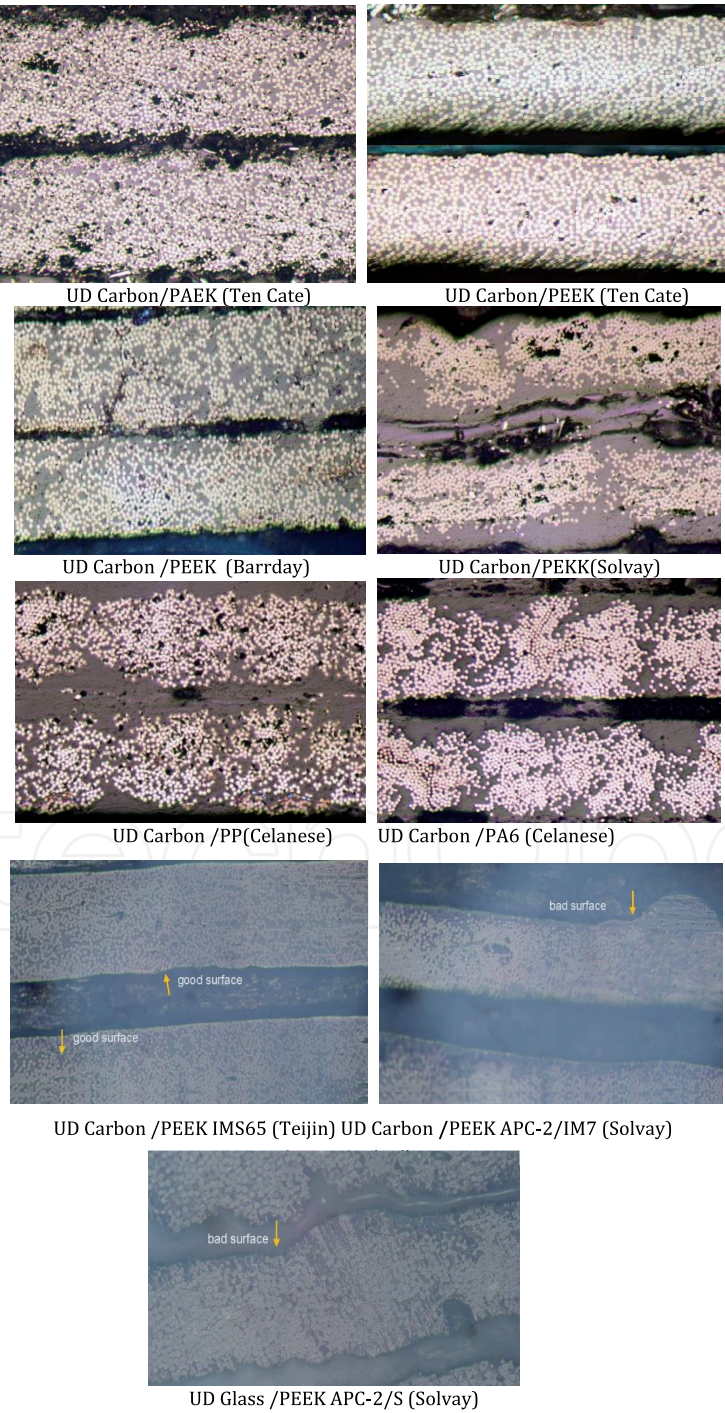


- d.Preparing the report of the raw material examination.
- e. Decision of raw material acceptance/rejection.

The thermoplastic tapes are typically not as consistent as thermoset materials, the uneven resin application can produce regions of resin richness, which can be both helpful and detrimental. In order to see a homogeneous distribution of the resin in the raw material, microphotographs of the procured raw materials are made with the help of an optical microscope (**Figure 3**).

The following figure shows some optical images from more raw materials UD thermoplastic prepreg:

In LATP/LAFP process, the **surface roughness** of the raw material has the biggest impact. Complete intimate contact is achieved if all the unevenness is the same



**Figure 3.**  
*Microscope images of some raw material (UD thermoplastic prepreg).*

size, that is if all the unevenness is pressed to the same order [19]. The **presence of voids** in the starting material requires a longer heating time and greater pressure during the process of laying a thermoplastic strip with a laser. It is, therefore, necessary to know the percentage of voids on the tape that will be used in the process.

The **resin content** in the thermoplastic band has a great influence on the mechanical properties of the finish, but it also has an effect on the percentage of crystallinity and the heat balance for the LATP/LAFP process.

Determining the crystallinity of the raw material for the LATP / LAFP process is important to determine the temperature of the tool and to compare it with the crystallinity of the final product.

2.3 Selection of process parameters in LATP/LAFP

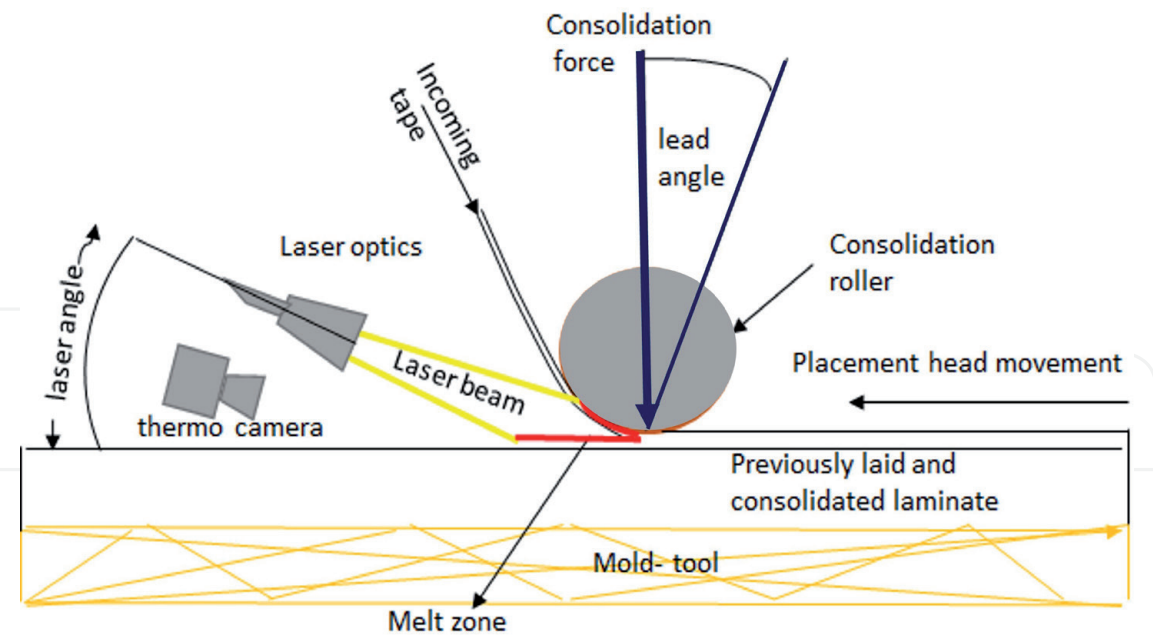
In general, the LATP/LAFP process using laser heating involves many parameters that affect the results of the process. To obtain quality parts, the combined effect of the main process parameters must be studied and analyzed. **Table 2** and **Figure 4** show the factors that affect the output of the automated fiber/tape laying process by using a laser heater. In the same table is entered a part where it is said which factor how much they influenced the final product and at what.

The association of semicrystalline thermoplastic tapes with the LATP/LAFP process has been analyzed in many literatures [6, 7]. Many studies have been done to determine the temperature distribution near the connection zone in order to determine the optimal angle of incidence of the laser beam to achieve a good interface between the layers [10–38]. Recent studies show that automated thermoplastic laminating processes with a velocity even greater than 200 mm/s produce composite

1. Raw material (UD tape)	
1.1. Tape impregnation quality (X)	1.3.Tape Surface Roughness (R) (\$) (@)
1.2. Tape thickness (ttape)	1.4.Tape Tension ( $\tau$ )
2. Mandrel	
2.1. Mandrel Curvature (DM) (@)	2.3.Mandrel Thermal Conductivity (kM)
2.2. Mandrel Temperature (TM) (#)	
3. Compaction roller	
3.1. Compaction Force (F) (@)	3.4. Roller Diameter (Dr)
3.2. Compaction Area (S)	3.5. Roller Thermal Conductivity (kr)
3.3. Roller interface Temperature (Tr)	
4. Heat source (laser)	
4.1. Beam intensity (I) (\$)	4.4. Beam angle ( $\beta$ ) (\$)
4.2. Beam profile (Y) (\$)	4.5. Incoming tape angle ( $\alpha$ ) (\$)
4.3. Beam size and shape (W) (\$)	
5. Process parameters	
5.1. Heat Transfer Coefficient (h)	5.3. Substrate Temperature (Ts) (@)
5.2. Placement Rate (v) (@) (#)	5.4. Tape Temperature (Tt) (@)
(@) has a high impact on the final product for voids. (#) has a high impact on the final product for the degree of crystallinity. (\$) has a high impact for (Tt) and (Ts)	

**Table 2.**  
Parameters that affect the quality of the final product of the process.





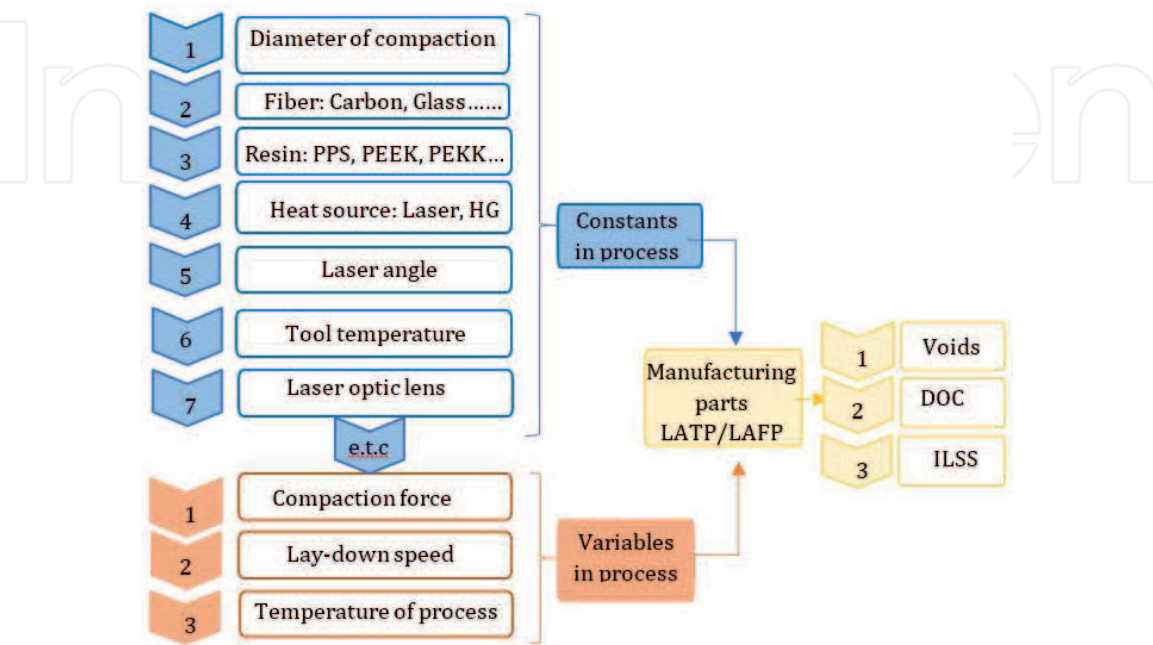
**Figure 4.**  
*Factors affecting the automated application process of laser heating.*

flat slabs with properties close to those of the resulting flat slabs when consolidated in an autoclave [13–17].

**2.4 How the parameters have been selected? The effect of these parameters on the quality of the final product**

In research, some of these factors (**Table 2**) are taken as constants and some as variables. Most often, the selection of the variable parameters for the research within the tests and the analysis of their impact on the automated LATP/LAFP processes when laying the thermoplastic tape is given in **Figure 5**.

*Raw material (UD tape)* - The influence of the characteristics of the raw material was previously explained.



**Figure 5.**  
*Constants and variables for the LATP process.*

*Mandrel* - In the process of passing this technology, important factors that affect the final product of the mandrel are as follows:

- **Mandrel Curvature** – Depending on the shape of the mandrel should
  - to determine the laying path - choice of lane width, for complicated and small curves the lane width should be smaller,
  - selection of the angle of the laser - depending on the shape of the mandrel, it is chosen how to position the laser so that we have even heating of the tape and the substrate
- **Mandrel temperature** – Numerous studies [20–26] have shown that heating the mandrel helps to release the stresses of the forces that occur in the cooling band so that there is no curvature of the flat parts. Mandrel heating is also important for the percentage of crystals in the final product. Experimental studies from many literatures have shown that the best mandrel temperature is if it is used 10–30 °C above the glass transition temperature of the matrix used in the process.

**Compact roller** - The dimensions of this roller (diameter, width and thickness of the siliconized layer) are important because they affect the intimate contact between the layers in the final product.

The development of intimate contact between the layers, which is a prerequisite for connection, consists of equalizing the unevenness of the tape and the laminate. The initial surface unevenness is deformed under the action of heat and pressure. The time required to achieve intimate contact depends on the unevenness of the surfaces, the applied pressure, and the viscosity of the matrix which depends of course on the temperature. Due to the temperature dependence of the viscosity, the increase in temperature facilitates the development of contact. In regions where intimate contact is achieved, interdiffusion of polymer chains occurs due to accidental thermal movement. The interdiffusion process is generally explained by the mobility of the polymer chains. The polymer matrix consists of intertwined chains that have limited movement. Their mobility, and thus the degree of diffusion, increases with increasing temperature. In the case of semicrystalline polymers, the presence of crystalline regions can severely inhibit the interdiffusion process. Based on this, it can be noticed that the interface is one of the important parameters, and therefore the thermal aspects when laying the tapes are very important.

When processing thermoplastic tapes, the connection is the result of intimate contact and autohesion (direct-bonding or self-bonding). *Autohesion* begins after the onset of intimate contact. Different connection models have been proposed in the literature depending on the type of process [18, 30–35]. Some processes require a long processing time to complete the layers. However, for LATP/LAFP processes, the temperature of the polymer rises to the melting point before the start of intimate contact [35]. Stokes-Griffin and Compston [34] later found that the bonding of carbon fiber-based thermoplastics and PEEK using LATP/LAFP takes place at temperatures below the melting point and above the glassing temperature. They found that PEEK polymers are very amorphous during consolidation and require short processing times with LAFP or LATP processes using extreme cooling rates (1000°C/s), and the polymer is heated above the melting point before starting intimate contact.

#### **Heat source (laser)**

Today, industries and research institutions do a lot of research on the application of lasers in automated fiber/tape laying procedures. Laser systems with high power

up to 10 kW are commercially available, and even more, if needed. Consider these variables when evaluating AFP diode laser sources:

**Beam intensity (I) Power:** The power of a diode laser module can be customized from just a few watts to 10s, even 100 s of kilowatts. For an AFP application, typically 1 kilowatt or less per tow is sufficient. The exact power depends mostly on the material and the processing speed.

**Beam profile (Y) Beam homogeneity:** The amount of heat, needs to be uniform across a designated target area.

**Beam size and shape (W) Size and weight:** A compact diode laser system allows for a more compact AFP head that can maneuver more intricate structures.

**Beam angle ( $\beta$ ) Incoming tape angle ( $\alpha$ ):** When the laser beam is positioned toward the composite beam, part of the initial intensity ( $I_0$ ) will be reflected ( $I_r$ ), partly absorbed ( $I_a$ ), and partly transmitted ( $I_t$ ). The amount of beam that is reflected, absorbed, or transmitted depends on a number of factors such as the characteristics of the material or strain, the wavelength of the beam, and the distribution of the fibers [4]. Ideally, the surface of the fiber-reinforced strip is covered with a thin and ideally flat layer of the thermoplastic matrix that aids in bonding. But in practice, such a layer rarely exists. The conical angle at which the radial rays fall on the material which is simultaneously under the pressure of the roller strikes the wedge-shaped cavity formed by the substrate and the thrust roller and causes different levels of reflection. The laser beam can be reflected from the thermoplastic matrix and also from the fibers on the surface of the tape. The part of the beam that is reflected from the surface should be as small as possible for laser heating to be effective. Any beam that is not reflected is transmitted or absorbed by the matrix and the fibers. It is actually the absorption of the beam that helps to heat the material.

## 2.5 Process parameters

Lee and Springer in their research [18] presented a model for the production of composites from thermoplastic composite tapes based on PEEK and carbon fiber using LATP/LAFP. They conclude that the process consists of three main steps—impregnation, consolidation, and crystallinity [18]. Impregnation is a parameter that cannot be controlled by automatic fiber/tape laying while consolidation and crystallinity can be controlled. Consolidation consists of two subprocesses—intimate contact and autohesion, that is direct connection or just connection (autohesion, healing). Autohesion is the formation of bonds between two surfaces of the identical polymer at elevated temperatures usually slightly above  $T_g$ . It is a new technique for precise bonding by self-bonding polymers without the need for adhesives. To achieve better final characteristics of the thermoplastic composite it is necessary to achieve a good degree of intimate contact and good self-connection.

Agrawal et al. [17] indicate that better on-site thermoplastic consolidation is achieved when a unified thermal model is applied by applying a wider beam with higher laser power. More authors [7, 38] also studied the effect of laser power, pressure, and deposition rate on the strength of the bond between layers. They found that the power of the laser had a dominant effect on the quality of the connection, and the force of the roller pressing had a minimal effect. Grove [36] in his research worked on the modeling of on-site consolidation processes and from experiments he concluded that the resulting poor crystallinity is due to the high cooling rate. Yousefpour and Ghasemi Nejhad [37] later proposed preheating the strip below the  $T_g$  glazing temperature to achieve better laminate consolidation.

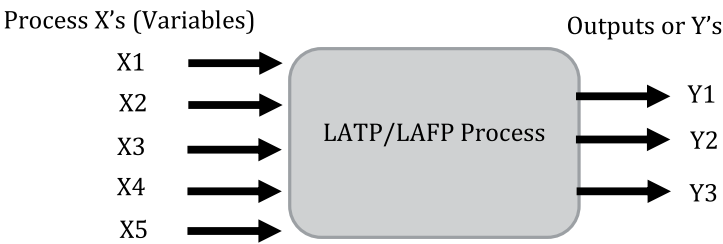
2.6 Trial test with DOE

More and more factors have an influence on effectiveness and efficiency in industrial processes and systems. To find the optimum in control of the processes there are often a lot of experiments to realize—practical and theoretical ones. Design of experiments involve designing a set of experiments, in which all relevant factors are varied systematically (**Figure 6**). When the results of these experiments are analyzed, they help to identify optimal conditions.

In the previous chapter, we talked about some input parameters in LATP/LAFP process, and here in this chapter will talk about the three main output parameters that are determined after each experiment (trial test with DOE) in this process and that are—voids and crystallinity ILSS (**Figures 4 and 5**).

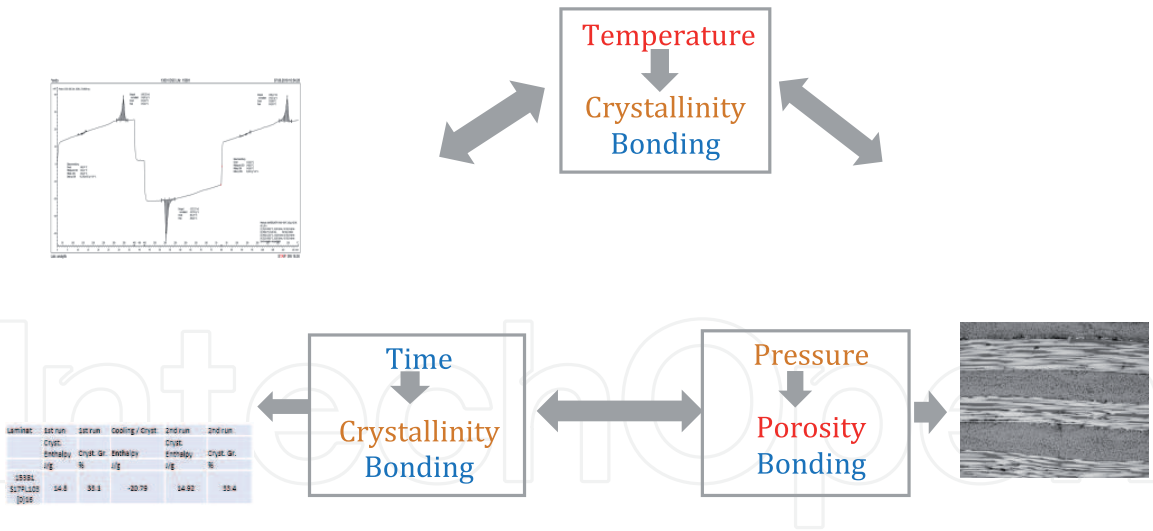
**Voids:** There are two types of voids that are inherent in the on-site consolidation process—intralaminar and interlaminar. The intralaminar cavity occurs during the impregnation of the tape, while, the interlaminar cavity is mainly the result of the process of laying the tape. The intralaminar cavity is embedded in the tape.

**Crystallinity:** Crystallinity also affects the mechanical properties of the final product and depends on the thermal cycle and cooling rate of the thermoplastic. In most LATP/LAFP applications, the cooling rate is as high as 1000°C per minute. Parts made of thermoplastic prepress tape based on PEEK and carbon fiber have a degree of crystallinity from 20 to 35% [18]. The two subprocesses—intimate contact and autohesia are a function of temperature, pressure, and consolidation time (passing speed), and these are the three most important parameters in any process. At a given pressure, a higher temperature and longer time are required to achieve the optimal degree of intimate contact and self-connection. Crystallinity affects the mechanical properties of thermoplastic composites. Higher crystallinity increases strength and rigidity [20]. On the other hand, the lower the degree of crystallinity increases the impact resistance and breaking strength [21–25]. The degree of crystallinity depends on the history of thermal processes. Low cooling rates result in a higher degree of crystallinity and vice versa. Hence, the degree of crystallinity achieved through on-site consolidation of the laying strip is limited due to the extremely high cooling rates. Kumar et al. [26] measured the degree of crystallinity of PEEK as well as the growth rate of spherulites (supermolecular forms of semicrystalline polymers) for samples heated to a melting point of 380°C and 420°C, then cooled to a crystallization temperature of 300°C or 320°C at a speed of about 3°C/s [26]. They concluded that the degree of crystallinity and the size of the crystals were higher for samples cooled by higher melting temperatures. In other words, at a constant cooling rate, the size of the polymer spherulites depends on the maximum processing temperature. Sonmez and Hahn [27] found that the degree of crystallinity of thermoplastic laminate obtained from one-way carbon fiber-based strips and REEK is between 25 and 35% in a single-site consolidation process. Similar degrees of crystallinity have been obtained in other studies indicating that PEEK is not sensitive to the cooling rates involved in the LATP/LAFP process [28, 29].



**Figure 6.**  
*Model of design of experiments.*





**Figure 7.** Preliminary quality verification for final flat parts: Low porosity + Hight crystallinity + good bonding = good mechanical properties (tensile, flexural strength, ILSS, NOL test).

**Mechanical properties:** The most common mechanical tests performed to determine the process parameters are 3pbT and ILSS. These tests make it easier to see if the interlayers are well bonded during this process. If necessary, additional mechanical tests are performed. The ILSS survey is a well-established and fast calculation to be observed as a process (input parameters affect the final product).

2.7 Conclusion and recommendation for process parameters for the final product

Achieving complete consolidation by applying robotic processes requires the realization of additional research related to the optimization of many parameters for obtaining composite parts of different sizes and shapes. The interrelationship between the process parameters, the properties of the material, and the strength of the bond between the interlaminar layers has been investigated and shown in **Figure 7**.

Process parameters:

- Temperature (laser temperature, laser angle, mandrel’s temperature)
- Pressure (compaction force of the roller)
- Time (layup speed) automatic tape laying process using a laser heating

3. Conclusion

- Robotic tape laying processes are very suitable for the efficient production of carbon fiber reinforced parts, especially for use in aerospace and other industries.
- The application of laser heating has some advantages over alternative heat sources, such as hot gas. In the hot gas torch (HGT) process, the thermal flux is difficult to control, while in the laser heating process, the laser beams are controlled spatially and temporally. Therefore, the obtained parts of composite materials with automated processes using laser application are expected to be of uniform quality, compared to those obtained by the HGT process. The two most important advantages of laser heating application are the high energy

density (accumulated energy per unit system per unit volume [ $\text{J}/\text{m}^3$ ]) and the short response time. The first allows the application of higher speeds when laying lanes, while the second allows laying lanes of complicated geometries, including large variations in laying speed. However, the application of lasers also has its drawbacks. The cost of the equipment is high compared to conventional heat sources, and also the laser always requires the equipment to be housed in a protected environment. In addition, the application of laser heating requires a thorough understanding of the interaction of light with fiber-reinforced thermoplastic tapes.

- When the speed of laying the thermoplastic tapes is high, then the heat that enters from the laser source to the entrance to the material is negligible and insignificant. Therefore, the researchers concluded that a good model of heating of the thermoplastic material should be made in correlation with the angle at which the radial rays fall on it and the speed of placement of the laser in order to successfully connect the layers.
- Experiments [20–23] have shown that excellent interlaminar quality, and thus good properties of the final parts, can be obtained when the laying of the tape is at low speeds, higher compaction force, and laser beam in the case when it is primarily aimed at the tape.

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