

## THERMOPLASTIC COMPOSITE STRUCTURES FOR SPACE APPLICATIONS: MANUFACTURING PROCESS SIMULATION

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### Abstract

*Since the sixty's, Airbus Defence & Space has been using more and more composite structures for launchers, satellites and space probes with the objective to provide high performance products. Pushed by cost reduction during the development and qualification phases of new programs dealing with new materials and new processes, process modeling allows to reduce the number of experimental trials and to better anticipate scaling effects. Modeling can also permit to better specify which material properties have to be well mastered and controlled to improve the robustness of the process and finally to avoid non conformances. This paper illustrates the approach followed by Airbus Defence & Space to model on-line consolidation process applied to thermoplastic materials.*

### 1. Introduction

Weight reduction of structures for aerospace aircrafts is still the main key driver even if cost reduction is the main target for designer, manufacturer and procurement people. In this context of where cost reduction is becoming the major factor, the new generation of structures in aluminium alloy is now a serious candidate which is very competitive compared to the current composite structures.

As a matter of fact, if a lot of efforts in research have been made for a long time to better design and justify composite structures, designers still require numerous experimental tests. This is expansive compared to metallic structures because they are facing the lack of universal failure criteria as it has been revealed by the World Wide Failure Exercise [1]. Moreover it is well known that structures made of carbon fiber reinforced plastic (CFRP) are much more sensitive to impact damage than aluminium structure. One of the solutions to overcome this weakest property of CFRP is to improve matrix toughness by incorporating thermoplastic nodules.

One can highlight that new generation of out of autoclave thermoset resins are also good candidate to reduce the cost and this new route with prepreg or infusion techniques is progressing with some applications in the aerospace industry with Vacuum Assisted Process VAP®.

Since 1997 Airbus Group Innovation (former EADS Innovation Work) has been launching several projects on automated tape placement for fiber reinforced thermoplastic, starting with a project where the incoming tape was melt with hot-air heaters. In the years 2000, another

project allowed to improve the understanding of the welding mechanisms with carbon fibers/PEEK matrix and also to identify the appropriated models to simulate the process with hot gas torch under nitrogen environment [2]. The third step was to use the laser technology which has the significant advantage to deliver high energy density and the short response time. The project Flash TP based on the laser-assisted tape placement process was launched in 2005 with the objective of manufacturing complex shape parts with Coriolis robot equipped with Yag laser technology with movable mirror able to better distribute the energy onto the surfaces (Figure 1) [3].

Autoclave and convection oven can be avoided with thermoplastic polymer when using the one-line consolidation process. This process eliminates the need for high temperature of the entire structure thus reducing the cost of tooling and in principle minimizing manufacturing residual stresses.

Up to now consolidation in autoclave of the parts manufactured with this process is required to achieve the best performance of carbon fibers/PEEK material.

## **2. Development thermoplastic structures at Airbus Defence & Space**

In order to meet new requirements in terms of higher service temperature, higher toughness, lower dependence of obsolescence risks due to new international regulations and in-house eco design constraints, Airbus DS has decided in 2007 to launch the development of thermoplastic composite structures and more specifically to manufacture a new generation of wound composite structures with thermoplastic polymers. The project launched in 2010 has the objective to reach the validation of on-line consolidation process at the laboratory level (achievement of level 5 of the Technology Readiness Level scale) for wound structures (Figure 1) [4].

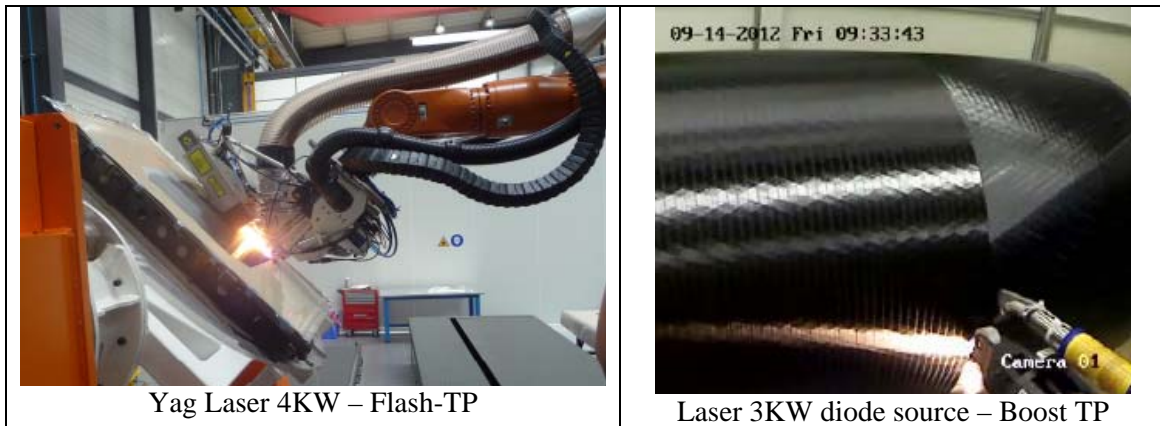
Generally speaking, the filament winding process is a low cost manufacturing technique because it is fully automated and no autoclave is required. Moreover with thermoset resin, the impregnation of the fiber can be done in-situ, known as wet route. In order to achieve a high performance of vessel used for the propulsion (motor case) or for high pressure applications, Airbus Defence & Space (Airbus DS) has been developing since the seventieth's its own pre-impregnated roving (towpreg) with the appropriated rheology of the thermoset resins.

The on-line consolidation process is a non isothermal manufacturing process where the composite experiences a complex thermal history. The composite substrate is repeatedly locally heated and cooled as additional layers are placed (or wound) onto the structure. This thermal cycling can cause microstructural changes that influence the structure's properties. These changes include melting, crystallization, void formation, degradation and consolidation. In the thermoplastic filament winding process, the rate of consolidation depends on the time required to achieve intimate contact and complete auto-adhesive bonding strength at the interface between the tape and the composite substrate.

## **3. Establishment of the process window**

Without understanding the physic and process modelling, the key variables that determine the processing window cannot be properly established and adapted with respect to the design of the structure.

From manufacturing point of view, the key variables are the speed of the head (or winding speed for filament winding), the heat source (mode, intensity), the pressure induced by the roller and cooling down and de-moulding.

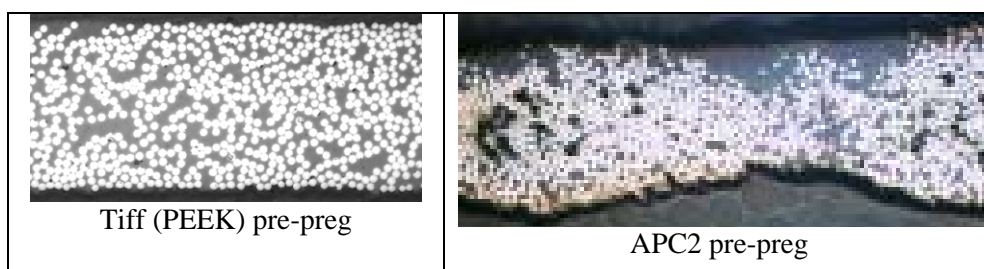


**Figure 1.** Illustration of two devices used to develop carbon reinforced thermoplastic structure a) fiber placement b) filament winding

From material point of view, the quality and the variability of the raw material (tape) in terms of crystallization for semi-crystalline polymers such as PEEK, fiber volume fraction, shape or initial porosity and roughness are also key variables. Figure 2 illustrates the variability of the quality of thermoplastic tape provided by different suppliers.

A lot of research works in modelling the different phases of on-line consolidation process have been doing since the eighties. The reptation and healing theories for amorphous materials in isothermal conditions were developed by G de Gennes [5]. The non isothermal healing model for diffusion bonding and crystallization of carbon/PEEK composites are more recent and the models are not fully reliable [6].

Table 1 summarizes the main phases of the on-line consolidation process which can be described by four key time parameters  $\tau_s$ ,  $\tau_{ic}$ ,  $\tau_{th}$  ( $\tau_{rep}$ ),  $\tau_{cd}$  for semi-crystalline polymer.



**Figure 2.** Illustration of two types of carbon/PEEK pre-preg showing the heterogeneity of the fiber dispersion, initial voids and roughness of APC2 compared to Tiff product [6].

Mastering these parameters will give to manufacturers enough confidence to supply good products with low porosity, high toughness and with finally limited distortion of the structure. Thermoplastic tape must require: a good fiber-matrix adhesion, a good fiber-matrix distribution, a good wetting of the fibres at the surface of the tape, no porosity inside the tape. This is why a close collaboration must be settled between the supplier and the end-user who has to specify the required characteristics of raw material regarding the applications. In addition, mandrel design is also important because of its influence to residual curing strain and then its capability to be easily removed.

		<p><b>Non degradation of the matrix</b></p> <p><b>Porosity reduction:</b>          -inter layer → during intime contact          -intra layer → during the applied pressure</p> <p><b>Residual strain reduction</b>          -high thermal gradient          -roller indentation (fiber weaving)          -thermal expansion of the polymer          -chemical shrinkage          -relaxation</p>	
<p><b><math>\tau_s</math></b> Time of heating depends on:          -mode of heating of the surfaces          -surface state          -lay-up orientation          -required temperature depending on crystalline of the material without polymer degradation</p>	<p><b><math>\tau_{ic}</math></b> Time of Intimate contact depends on:          -Rugosity          -Viscosity          -Pressure distribution          -Temperature (higher than <math>T_g</math> temperature or close to <math>T_f</math> for semi-crystalline polymers)</p>	<p><b><math>\tau_h</math></b> Time of healing depends on          -<math>\tau_{rep}</math> ; time of reptation          :function of molecular weight and polymer structure</p>	<p><b><math>\tau_{cd}</math></b> Time to cool down          -germination and crystallization</p>

**Table 1.** Synthesis of the main multi-physic mechanisms encountered in the on-line consolidation process

The heating time  $\tau_s$  is crucial especially for semi-crystalline polymer because it is necessary to choose the processing temperature high enough to melt all the crystals without reaching the degradation of the polymer. The crystalline parts (spherulites) as well fiber reduce the mobility of the long chains of molecule which will contribute to reinforce the interface between the two layers.

The determination and control of the temperature during  $\tau_s$  and  $\tau_{ic}$  times are not obvious. Airbus DS has chosen to heat up the two surfaces with Laser diode source technology which brings flexibility regarding the energy which can be tuned with power and its position with respect to the geometry of the piece and the variation of the motion of the compaction head. When laser light strikes a fiber reinforced composite, a part of its initial intensity will be reflected, some absorbed and some transmitted. The amount of light that is reflected, absorbed or transmitted depends on a number of factors, such as material properties (which includes fiber distribution, matrix) and the laser wavelength. The heating problem is not to simply modify the power of the Laser but to know the effective energy received by the material. Airbus DS in collaboration with Ecole Centrale de Nantes has launched a research program to better predict the thermal history of the interface which governs the interdiffusion and crystallization phenomena. A model will be developed to calculate the incident light distribution on the laminate and the tape for various incident laser angles. The laser wave length needs to be matched to the optical material properties of the tape and the laminate [7].

The second critical issue is the time needed to get an intimate contact ( $\tau_{ic}$ ) at the interface which depends on the roughness of the tape. A rough material introduces a thermal contact resistance which increases the time of healing. This parameter is difficult to estimate and can vary significantly. We know from literature and our own experience that roughness can significantly modify the condition of a good adhesion between the layers. If the conditions of  $\tau_{ic}$  and  $\tau_h$  are achieved, the pressure must be applied at a temperature higher than the glass transition temperature ( $T_g$ ) as illustrated in Table 1. If it is not necessary to apply the pressure for a long time to get good adhesion for bulk resin, it could be longer with composite. Care must be taken to minimize the loss of temperature due to heat exchange with the environment (potential heat sink brought by the roller and the composite). From practical point of view, it is difficult to measure the temperature of the faces in contact under the roller during the process. Again we need an accurate thermal model which has to be validated by measuring the gradient of temperature in the composite with the same boundaries conditions met in the industrial machine.

Once two adjacent interfaces come into contact, the mechanism controlling interply bond formation is admitted to be autohesion based on reptation theory. Researchers have established a relation between the degree of autohesion with the bond strength interply. Bond strength is directly proportional to time ( $t$ ) to the power  $\frac{1}{4}$  [5]. If the time is equal to  $\tau_{rep}$  which is a strong function of temperature, the maximum toughness of bulk polymer is then achieved.





The measure of the strain energy release rate of specific samples will be a useful tool for material engineers to identify the process window. It has been shown by some authors that bond strengths for inner layers, closest to the mandrel could be lower than intermediate layers in the composite [8].

The last phase of the on-line consolidation process concerns the crystallisation of the polymer ( $\tau_{cd}$ ). It has been shown that the mechanical properties of thermoplastic matrix composites can be related directly to the crystallinity of the polymer. Several authors have studying the crystallisation behaviour in the tape placement process. The rate of crystallisation is governed by the cooling rate but also by winding speed and temperature of the heaters. Preheating the substrate yields better consolidation and higher crystallinity and hence improves the bond strength [6, 8].

In addition to these fundamental parameters which govern the toughness of the material, the design of the fiber placement robot or winding machine have to be adapted to this process which requires more accuracy of the kinematics of the winding head in comparison of those used for thermoset prepreg. Ideally tapes should be deposited such that neither gaps nor overlaps are created. However the tapes deform and spread under the application of the heat and pressure. Because overlapping of the tapes is not suitable for the final health of the structure (performance penalty) it is again important to control the tolerance of the width of the tape and its deformation. The friction coefficient used in the squeeze flow model allows taking into account the dependence on the tape to substrate orientation. In case of parallel deposition, nesting of the fibers will result in a high friction coefficient whereas in the case of an orthogonal deposition the fibres will slide more easily [9].

Last but not least, manufacturing of thermoplastic composites is inevitably accompanied by the development of residual stresses that cause shape distortions of the final structure.

Table 2 presents a synthesis of the main tests that are required to determine the parameters of the models and also to validate the process window with structures tests carried out at the industrial level to validate the window process. From practical point of view this process is iterative at the lab environment scale level (achievement of level 5 of the Technology Readiness Level scale). From an experimental point of view, the main problem is the lack of relevant methods to measure the surface temperatures.

<b>Process validation at lab environment scale – manufacturing activity</b>		
 <p style="text-align: center;">φ800mm vessel</p>		Validation of the process window with the integration of the kinematics of the industrial winding machine  *Instrumented full scale test *Behaviour analysis and material expertise
 <p style="text-align: center;">φ304 mm vessel</p>		Validation of the process window with representative test samples  *Full scale test *Material expertise
<b>Material characterization for process control – lab activity</b>		
 <p style="text-align: center;">Peeling test</p>	 <p style="text-align: center;">GIC of curved sample</p>	Mechanical tests to determine or to control the process parameters based on the materials properties and the outcome from modelling
<b>Means:</b> DSC,DMA Mechanical characterization		Material characterization to control the product
<b>Determination of the parameters of the models - research activity</b>		
<b>Means:</b> *DSC for model of crystallinity) *DMA, *Rheometer (viscosity) for reptation model, *Optical characterization (reflectance, absorbance) for Laser monitoring *Squeeze flow device for tape deformation		Material characterization to identify the material parameter linked to the physic and chemistry laws

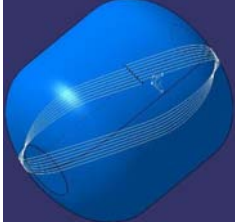
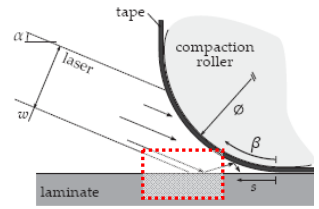
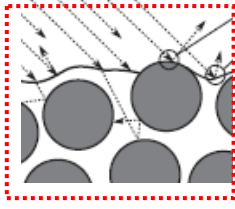
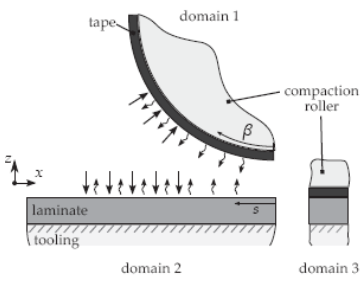
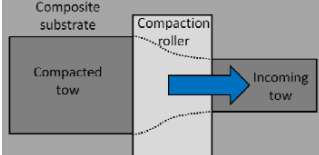
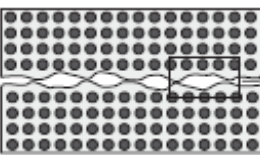
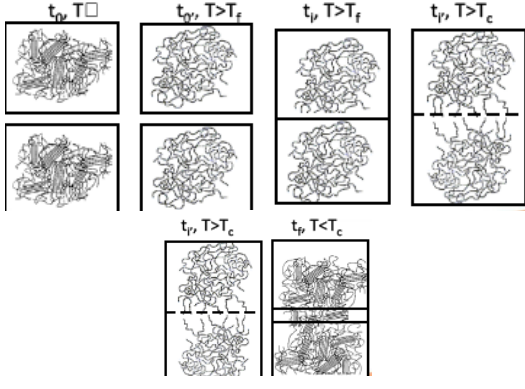
**Table 2.** Synthesis of the main tests performed from research to industrial development performed at Airbus DS

The process window for on-line consolidation is narrow and challenging to determine without a multi-physic modelling approach because the key parameters of the process cannot be all monitored one line. To aid in this process, simulation tools will help to limit the trial-error procedures with involve to manufacture and to test a lot of samples.

The process window for wound structures with semi-cristalline tapes like carbon fiber /PEEK is too narrow to be achieved with the current quality of the tapes provided by the suppliers and with the lack of accurate measures of the process parameters especially the surface temperatures.

We can conclude that modelling the on-line consolidation process is a difficult challenge due to the combination of several problems which have to be addressed as summarized in table 3. Moreover the current models are not fully validated for industrial parts.



	<p>Kinematics model to control the tape deposition (gaps and overlaps)  <b>Status:</b> Work in progress, linked with the maturity of the squeeze flow model</p>
	 <p>Optical model for the energy deposition onto the interface when using laser sources  <b>Status:</b> In research phase</p>
	$q_z''(t) = k_z \frac{\partial T}{\partial z} =$ <p style="text-align: center;">convection  <math>-h(T - T_\infty)</math></p> <p style="text-align: center;">radiation  <math>-\epsilon\sigma(T^4 - T_\infty^4)</math></p> <p style="text-align: center;">laser  <math>+ Q(t)</math></p> <p>Thermal model for non isotherm heat exchange  <b>Status:</b> Work in progress, linked with the maturity of the energy deposition and interface resistance contact (domain 3)</p>
	 <p>Squeeze flow models for the deformation of the tape [9]          Intimate contact model  <b>Status:</b> Work in progress</p>
	<p>Healing model          Crystallisation model for semi crystalline polymer: Differential Avrami model or equivalent (Ozawa)  <b>Status:</b> Work in progress for model validation</p>
<p>The residual stresses begin to build up at :          -glass transition temperature for amorphous matrix          -at the beginning of crystallization for semicrystalline matrix [10]</p>	<p>Residual stress model accounting for visco-elasticity behaviour of the material  <b>Status:</b> Work in progress ; similar methodology used as thermoset composites</p>

**Table 3.** Synthesis of the models which described the physics involved in on-line consolidation process and status of their maturities at Airbus DS.

All the models have not the same level of maturity and research works are still needed. From the industrial side, these models must be in fine robust and simplified to be implemented in industrial software. A significant breakthrough in computation has to be achieved to allow coupling the models and then to reduce CPU time. The POD and PGD methods are the solutions investigated by Airbus DS in collaboration with Ecole Centrale de Nantes and already used to solve the mechanical and thermal problems [3, 11].

#### 4. Conclusion

Simulations of fiber reinforced plastic composites processes are more and more used in the aerospace industry. Several commercial codes are available to simulate Resin Transfer Moulding process, autoclave curing process for thermoset resin. The one-line consolidation process with thermoplastic polymer offers new potential applications of manufacturing large and complex structures without oven or autoclave and therefore with a lower cost. The high performance of the structures cannot be achieved without mastering the key parameters that govern the complex phenomena generated by the non-isothermal conditions. These difficulties met with this challenging technology breakthrough cannot be overcome without establishing multi-physical models. Lot of efforts and research works are presently done at Airbus DS to strengthen these models that help the manufacturer in processing window determination.

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