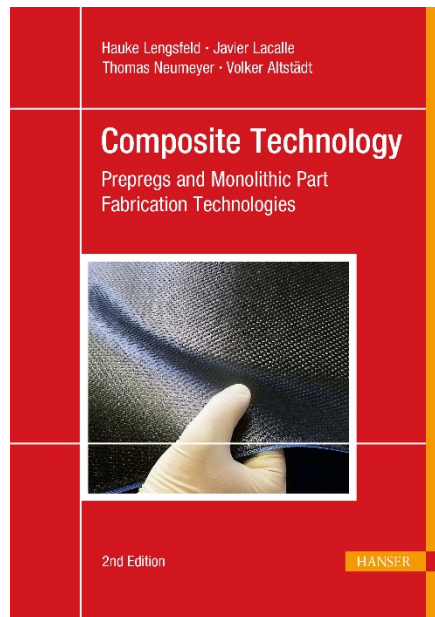


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Preface to the Second Edition

From the early 1970s to the present day, CFRP structural components have largely been manufactured using prepreg technology. Although declared to be dead many times, prepregs are becoming increasingly popular in all segments of the composites industry due to their versatility, high fiber volume content, and wide range of fiber-matrix combinations. This second, updated edition features authors representing perspectives from various fields of industry and applied research. This ensures that the latest trends are incorporated into this new edition. Unidirectional fiber-reinforced semi-finished products with a thermoplastic matrix, such as organo sheets and tapes, have gained considerable importance since thermoplastic composite lightweight construction with organo sheets entered large-scale production. It is therefore time for the authors to provide an up-to-date report in this book on the prospects and trends of these materials as well in the various chapters.

Thermoset prepregs are produced by impregnating reinforcing fibers or textiles with specially formulated, pre-catalyzed resin systems using a dedicated machine technique. The machine technology used ensures intimate contact between fiber and matrix, in combination with a defined surface tack of the prepreg tape. These prepregs are used to produce composite components faster and with less performance degradation than comparable wet impregnation techniques. Covered with a flexible backing paper, prepregs are easy to handle and remain storable for a certain period of time (shelf life) at room temperature.

In contrast to thermoset prepregs, which have been known for around 70 years and have a correspondingly wide distribution, thermoplastic prepregs are a still young material group that has not yet become so widespread. The product group of continuous fiber-reinforced thermoplastics offers both designers and processors new possibilities for combining lightweight construction with design and functionality. Above all, components made from fiber-reinforced thermoplastics are characterized by their significantly higher impact strength compared with that of thermosets, coupled with low density.

Globally, the market for prepregs is expected to grow at a CAGR of approximately 4.6% from 2019 to 2024, reaching approximately US\$6.4 billion in 2024 (according

to Lucintel's Global Prepreg Market: Trends, Forecast and Opportunity Analysis). Although the input costs for prepregs exceed those of traditional materials in many cases, their use often shows significant cost advantages when viewed as a whole, taking into account weight-related properties.

This book discusses important advances in research and development at institutes and in industry laboratories. Fundamental relationships between the structure of the material, its processing, and its properties are shown. Looking to the future, important developments in the field of modern prepreg technology are presented in the book. The book is divided into eight main chapters. After an easy-to-understand introduction to the world of fiber-reinforced composites (Chapter 1), the most important components of prepregs are first presented in Chapter 2. Here one now also finds thermoplastic prepregs as an important addition. This is followed by two chapters describing the manufacture of prepregs and their processing into a preform (Chapters 3 and 4). Chapter 4 pays special attention to automated lay-up technologies, which are updated with the latest developments as well. Chapters 5 and 6 present the curing of the preform to a component in an autoclave, in an oven, with the Quickstep technology, etc., and the associated technologies of the shaping curing tools (toolings) are explained in detail. The cured components must then be tested and in many cases combined to form complete structures in ways that are characteristic of composite materials. Chapters 7 and 8 provide an overview of the specific features that have to be taken into account when using prepreg technology. In the chapters mentioned, the special aspects of thermoplastic components, e. g. in processing, tooling, and testing, are now also discussed.

The aim of the book is to provide the specialist with a comprehensive, application-oriented work that explains both current developments and the path toward them in a comprehensible way.

Bayreuth, March 2021

Volker Altstädt

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Chapter 3 with the participation of Mike Turner and Chapter 6 with the participation of Prof. Dr.-Ing. Hilmar Apmann.

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2

Prepregs and Their Precursors

Felipe Wolff-Fabris, Hauke Lengsfeld,
and Johannes Krämer

Over the last 70 years, prepregs have significantly influenced the technological development of high-performance fiber reinforced components. Today, these materials are globally prevalent and are used for the manufacture of composite parts in the aerospace industry, for high-speed trains, cars, boats, and many other applications. More than half of the global carbon fiber production is used to manufacture prepregs [1].

Yet, there are still segments in industry that are discovering the advantages of fiber reinforced composite materials over those of conventional materials, such as metals. Increasingly, combinations of metals and fiber reinforced plastics – so-called hybrids – are introduced.

This chapter will provide an overview of the design of prepregs on the materials typically used to manufacture them. In addition, we will introduce examples of the different generations of prepregs, including the current, modern prepreg systems.

With regard to performance, prepregs claim the undisputed leading position among high-performance composite materials (Figure 2.1). This class of materials allows the manufacture of ultra-lightweight yet highly load-bearing composite components. However, the figure also shows that the advantage in performance comes at the cost of productivity (see also Chapters 4 and 5). Therefore, this technology is mainly used for very small to medium-sized production runs. In these scenarios, component performance and production cost can be easily reconciled using prepreg materials (Figure 2.2). Despite their lower productivity compared to infusion techniques, prepregs excel in terms of their simple and safe processibility.

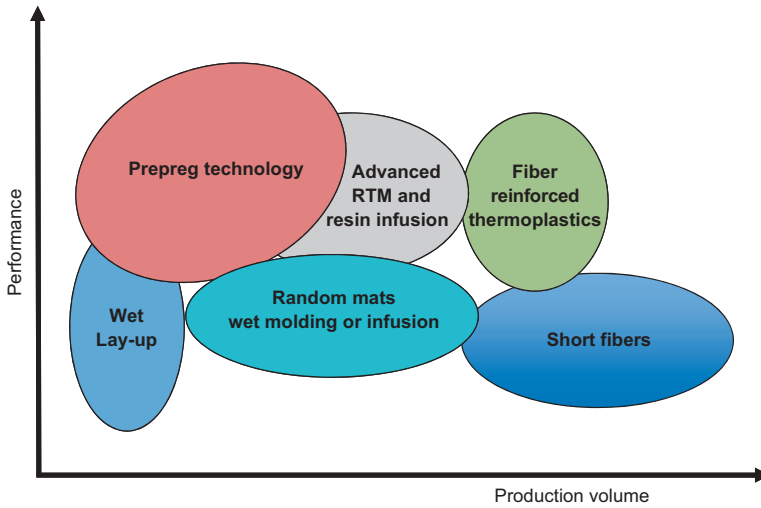


Figure 2.1 Performance and production volume of prepregs compared with other technologies

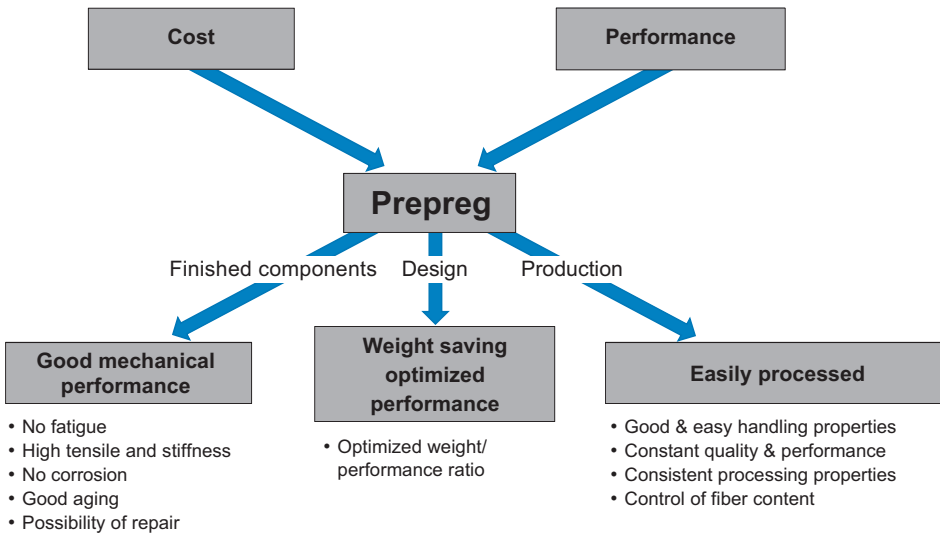


Figure 2.2 Prepregs as key materials for high-performance components

The costs of prepreg materials are determined by the requirements the finished component has to meet. Prepreg costs for the production of aviation components cannot be compared to those for components for wind turbines. The latter application is a good example for the fact that manufacturing and labor costs can be lowered by utilizing prepregs with high areal weight rather than using infusion technology. Using prepregs with areal weights of up to 2000 g/m² allows for high lay-up

efficiency (even by hand) and faster completion of the component. While the matrix is already part of the prepreg and only has to be cured, dry semi-finished products require the additional step of infusion with the matrix resin.

■ 2.1 Structure and Preparation

A prepreg (*pre-impregnated fibers*) is a semi-finished product consisting of a highly viscous matrix and continuous reinforcing fibers. The material is typically wound on rolls and comes in two variations:

- Unidirectional (UD) prepreg (fiber reinforcement in only one direction)
- Fabric prepreg (orthogonal fiber reinforcement)

In order to manufacture a prepreg, the fibers are wetted with the matrix material, resulting in a pre-impregnated semi-finished product (see also Chapter 3). The choice of matrix material includes thermosetting resin systems (TS) as well as thermoplastics (TP); however, thermosetting systems are more commonly used.

The cross section in Figure 2.3 shows the typical composition of a thermoset prepreg as it is delivered between a polyethylene film (PE film) and a release paper, ready for further processing. Some processing techniques, such as automatic tape laying (ATL, see Chapter 4), require a carrier material only on one side of the prepreg. Both the PE film and the release paper protect the prepreg from contamination, prevent sticking, and facilitate handling, e.g., during cutting. In the case of thermoplastic prepreps (TP prepreps), these separating and carrier films or papers are not necessary because the material does not bond to itself (see Figure 2.4).

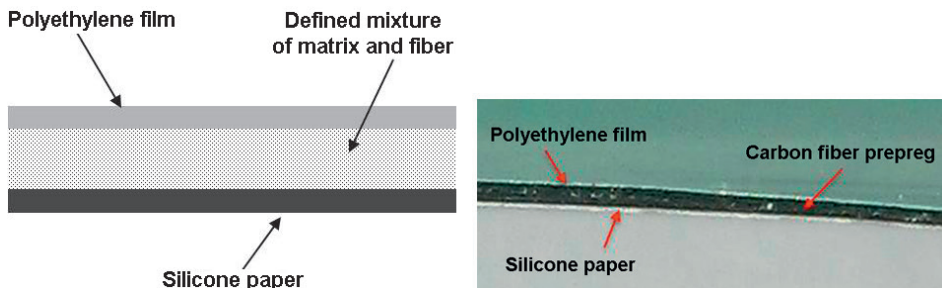


Figure 2.3 Typical thermoset prepreg cross section

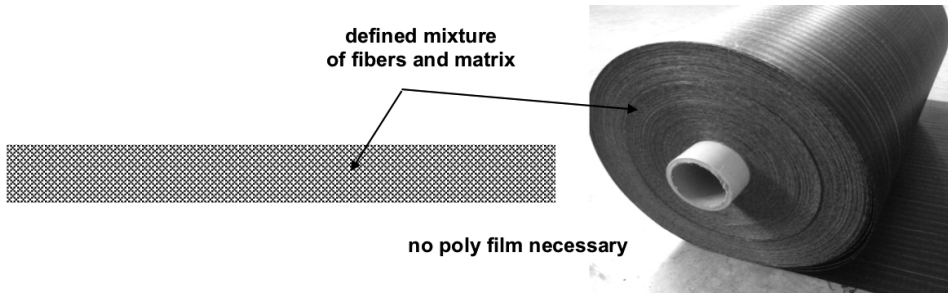


Figure 2.4 Typical thermoplastic prepreg cross section

The dimensions of the semi-finished product vary depending on application and type of material (UD or fabric prepreg). The roll width also depends on the width of the production line. Common roll widths and lengths are shown in Table 2.1 and Table 2.2.

Table 2.1 Examples of TS-Prepreg Dimensions and Roll Weights

	UD prepreg	Fabric prepreg
Width in mm	6.35 (1/4") to 1500 (59")	900 to 1500
Length in m	up to 800	up to 200
Roll weight in kg	up to 80	up to 200

Table 2.2 Examples for Dimensions and Roll Weights of Thermoplastic Prepregs (TP)

	UD prepreg	Fabric prepreg (laminates)
Width in mm	6.35 (1/4") to 304.8 (12")	300 to 1000 mm
Length in m (UD) or mm (fabrics)	100 to 1000	ca. 40 to 60
Roll weight in kg	up to 250	up to 150

The ratio of fiber to matrix content is precisely defined for each prepreg, with narrow tolerances that depend on application. The tolerances for nominal resin content (RC) range from $\pm 2.5\%$ to as little as $\pm 1\%$. However, the resin content may be adjustable within certain limits, depending on application-specific requirements. A common choice is a combination of fiber areal weight and resin content that allows for a fiber volume content of approx. 60 wt.% after curing of the material. In many cases, resin is lost during curing in vacuum processes, and therefore the resin content in the semi-finished product is often adjusted to a higher value. Depending on resin content in the prepreg and/or in the composite component, the theoretical value of the cured ply thickness (CPT) changes. Excessive resin loss during the curing process may also lead to a smaller CPT than theoretically calcu-

renewed interest in research and development projects. Within a short time period, the carbon fiber manufacturing and processing industries have evolved into sophisticated technologies. Starting from a few selected materials and fiber composites, which were produced in very labor intensive, manual processes, today a variety of industries, including the aerospace and automotive industries, rely on low-weight, stiff, high-performance composites that could not be produced if it were not for prepreg materials and fully automated processes.

Thermoset prepregs were joined over time by thermoplastic-based prepregs. The latter's development and broad industrial use is more complex than that of thermoset prepregs due to the more complex processes involved.

■ 3.2 Introduction: Manufacturing Methods

Prepregs, as semi-finished products containing pre-impregnated fibers (Figure 3.1), are a prerequisite for high quality and load-optimized lightweight fiber composites. The current manufacturing techniques and processes guarantee consistent quality of the prepregs, e.g., in terms of FAW and resin content, at a very high level. On the one hand, this simplifies processing for the manufacturer, and on the other it allows the reproducible production of high-quality components.

Prepregs always consist of a combination of a typically highly viscous matrix and a fiber reinforcement. Once the reinforcing material has been pre-impregnated with a matrix, it is considered a prepreg material. Both thermoplastic and thermosetting materials (reactive resin systems) can be used as matrices (see also Chapter 2) [2]. In the following, we will discuss the production of thermoset prepregs.

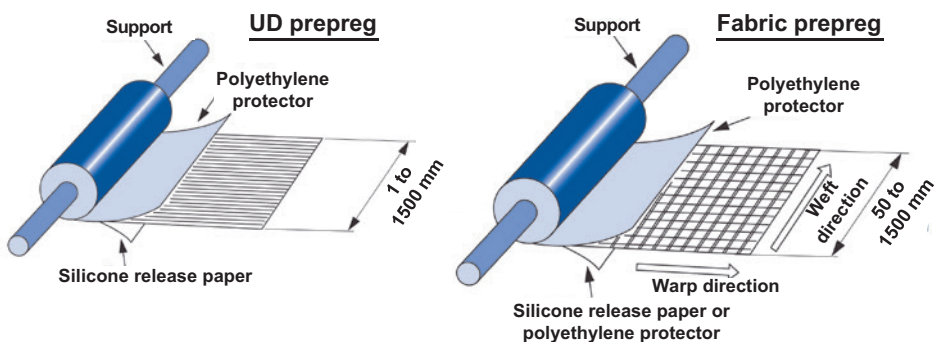


Figure 3.1 Delivery form of UD prepreg (left) and fabric prepreg (right) on supports [Courtesy: © Hexcel Corporation]

Fiber reinforcements are supplied in a number of different forms:

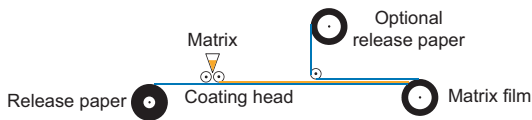
- uni-, bi-, and multidirectional fabrics
- fabrics
- non-wovens and random fiber mats

Depending on the reinforcing structure, processing techniques include creel sets (for rovings) and roll unwinders for fabrics.

Today, a number of different methods and machine concepts are available for the production of coating lines. They are able to use different fiber materials and combine them with a wide selection of matrix resins. State-of-the-art technologies include:

- solution coating processes (also called dipping (solvent) processes) (Figure 3.2 and Figure 3.3)
- hot melt processes (Figure 3.2)
- knife systems
- powder scattering (Figure 3.3)
- extrusion / slot die systems (Figure 3.3)

Step 1 – Film production



Step 2 – Film transfer

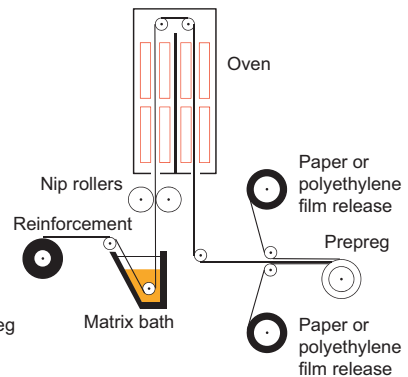
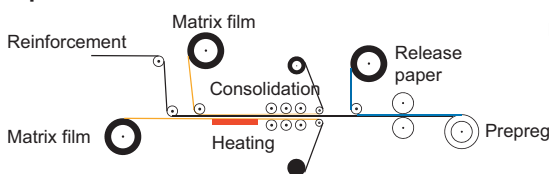


Figure 3.2 Manufacturing methods for thermoset prepregs, left: hot melt process; right: solvent process

Some processes, such as powder scattering, were initially developed for thermoplastic matrices, but later adapted to thermoset processing. However, these processes have not found widespread industrial-scale use. Today, there are two major impregnating methods in industrial practice: the dipping (solvent) process and the hot melt process [2].

4

Prepregs: Processing Technology

Hauke Lengsfeld, Javier Lacalle,
and Thomas Neumeyer

■ 4.1 Introduction

In this chapter, we will describe the different technologies used to process prepregs and to transform them into prepreg components. We will introduce both manual and automated deposition methods as well as methods to cut and form prepreg materials.

One of the advantages of fiber reinforced materials is the fact that the fiber reinforcement can be strategically placed in the component to optimize the relationship between mechanical properties and weight.

Manufacturing methods using prepreg are particularly efficient because they achieve highest quality and most accurate fiber placement in the component. This also ensures optimum fiber volume content in the component, because the ratio of resin to fiber has already been coordinated in the prepreg. The layer structure may be deposited by hand or by automated processes.

In general, the processes used to manufacture composites are laminating and deposition processes that use flat, semi-finished products (e. g., prepregs) and deposit them in a specific sequence and in a defined orientation and shape on a mold or tool.

Today, there is a wide variety of processes available. There are two different approaches to classify these processes. First, considering the pressure applied during forming and curing leads to the classification of the most common processes as shown in Figure 4.1.

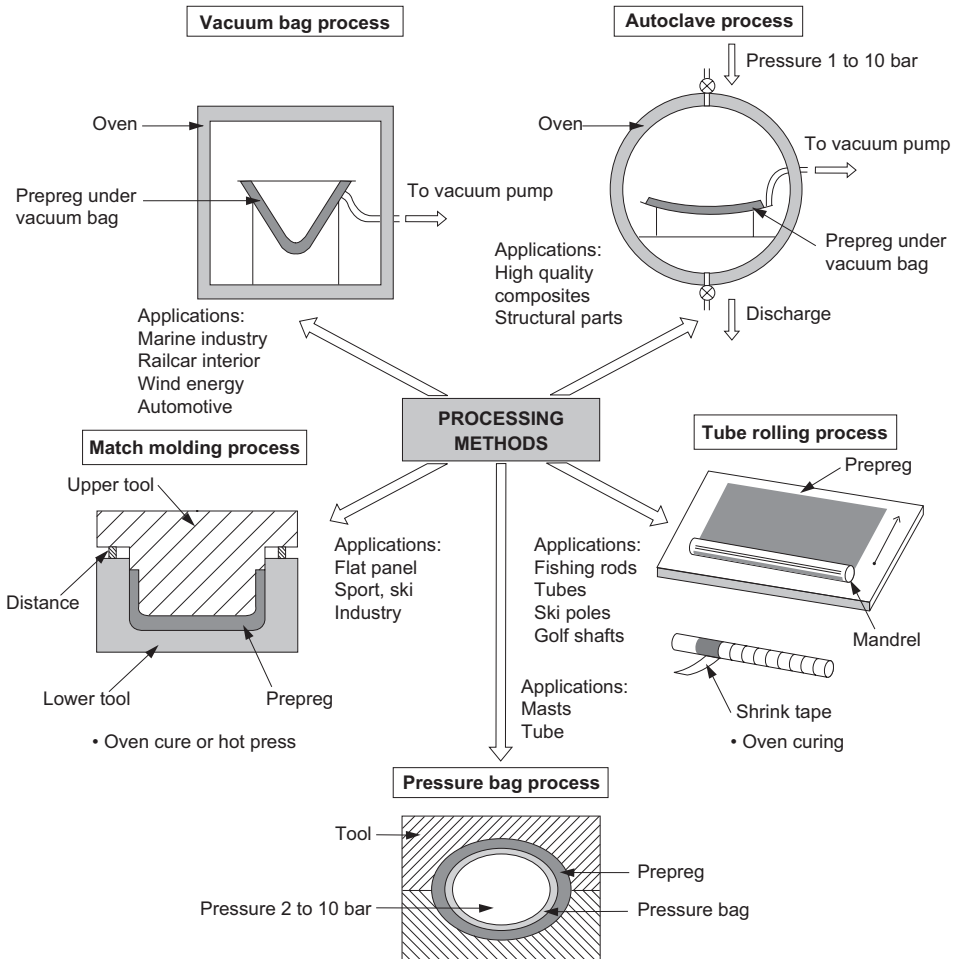


Figure 4.1 Curing and forming processes for prepregs [Courtesy: © Hexcel Corporation]

An alternative perspective considers the deposition and processing technologies that are used to process prepregs (Figure 4.2). This approach also includes the subsequent process of manufacturing a fiber reinforced composite part and provides a differentiated classification in the various technologies. While the pressure-oriented approach (see Figure 4.1) often includes the curing process, it is deliberately separated from the deposition and processing technologies. Curing is considered a separate step and thus includes the pressure processes shown in Figure 4.1.

During hand lay-up, the cuts are typically aligned using templates, leading edges, or markings on the edge of the mold to achieve sufficient deposition accuracy (positional tolerance) of the individual plies and to avoid incorrect deposition. Direct labeling of the finished cuts during cutting (whether manual or by cutter) prevents the interchange of cuttings during lamination. In addition, nowadays laser positioning systems are used that project the exact position and form of individual cuts on the lay-up area. These systems also prevent turning of cuttings and lay-up in the wrong orientation. The disadvantage of using templates becomes apparent when many different-sized cuts require the same high number of templates. With larger cuts, templates tend to become rather unwieldy. Another problem with hand lay-up of prepreg cuts is the sheer number of prepreg cuts necessary for the manufacture of complex components, which can reach several hundreds. This results in an additional logistics problem, especially when the cuts cannot be stored at room temperature but have to be kitted and frozen for storage.

Several parameters, including size, production rate, and the required accuracy and repeatability of the prepreg lay-up (form and positional tolerances) determine whether a component will be manufactured by hand lay-up or by an automated process. On the one hand, hand lay-up processes are time and personnel intensive; on the other hand, the high capital and operating costs of automated processes have to be considered.

In the past (until approx. 2004), large components, such as the vertical tail plane of the Airbus A320 and A330 series, were manufactured by hand lay-up of unidirectional tapes. For large components, the handling, exact positioning, and laying of long prepreg cuts without entrapping air is extremely difficult, and defects in the composite part are therefore hard to avoid. Therefore, the prepreg structure of such large components is typically manufactured using automated systems.

■ 4.4 Automated Lay-up Technologies: Automated Tape Laying (ATL) and Automated Fiber Placement (AFP)

4.4.1 Introduction

Today, the automated laying of pre-impregnated fiber materials is a key technology for the manufacture of large composite components in the aerospace industry. For a number of years, automated tape laying has been used in conjunction with other technologies, such as hot-forming, to manufacture vertical tail panels, wing struc-

tures, stringers, spars, etc. Thermoset materials have traditionally been the basis for the production of composite parts.

However, the fast growing use of composite components both in aerospace and automotive applications, together with the increasing complexity of these components, has triggered the continuous growth in use and research of automated and highly efficient laying technologies, and the further development and additional formats of materials. Figure 4.16 and Figure 4.17 show the geometric complexity as well as the size of composite parts.

Also the increased use for productive, serial applications of other materials such as thermoplastics has opened a new development branch in the materials and processes.



Figure 4.16 Carbon fiber reinforced composite fuselage structures
[Courtesy: Airbus Operations GmbH]

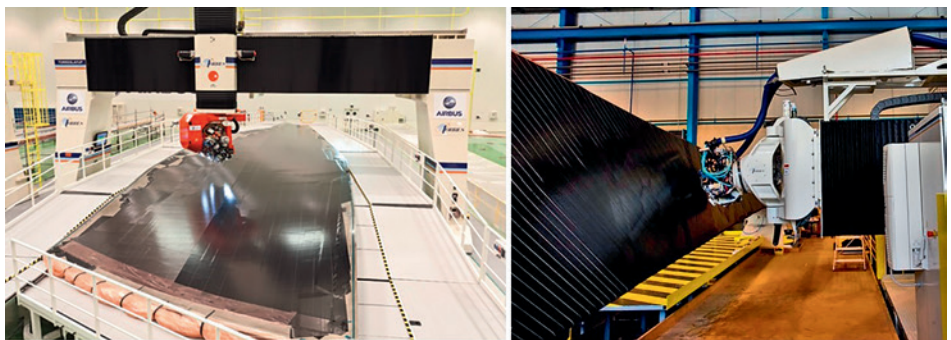


Figure 4.17 Automated lay-up processes: ATL (left), AFP (right)
[Courtesy: M.Torres Diseños Industriales S. A. U.]

■ 6.2 Tooling Materials

A variety of different materials is available for the manufacture of tools for the forming and curing operations of prepregs materials; these materials can also be combined, depending on the area of application.

Table 6.2 provides an overview of common tooling materials that will be described in more detail in the following.

Table 6.2 Examples of Tooling Materials

Material	Description	Type (examples)
Metals	<ul style="list-style-type: none"> ▪ Steel ▪ Aluminum ▪ Nickel-steel alloy (Ni-36) 	<ul style="list-style-type: none"> ▪ S235 JR, S355 JR ▪ ALMG 3/3.3535 ▪ INVAR 36, Pernifer 1.3912
CFRP (epoxy resin)	<ul style="list-style-type: none"> ▪ Fabric prepreg ▪ Quasi-isotropic prepreg mat 	<ul style="list-style-type: none"> ▪ Cycom® 7620, ▪ HexTOOL® M81
CFRP (BMI or BOX resin)	<ul style="list-style-type: none"> ▪ Fabric prepreg ▪ Quasi-isotropic prepreg mat 	<ul style="list-style-type: none"> ▪ Duratool® 5270 ▪ HexTOOL® M61 ▪ Toolmaster BetaPreg
CFRP foam	<ul style="list-style-type: none"> ▪ Carbon foam 	<ul style="list-style-type: none"> ▪ Touchstone CFoam® 20
GFRP	<ul style="list-style-type: none"> ▪ Dry fabric + resin, processed by resin infusion or hand lay-up 	
Other materials	<ul style="list-style-type: none"> ▪ Wood ▪ Epoxy tooling boards ▪ Cellular concrete ▪ Sand casting compounds 	<ul style="list-style-type: none"> ▪ OBO-Plywood, RETIstab ▪ Necuron, Rampf WB700, ▪ OBO-Modulan, TB650 Series ▪ Ytong ▪ Polymer bonded quartz sand

6.2.1 Metals

Metals are the easiest and most commonly used class of materials for the construction of tools for prepreg processing. This is due to their easy availability as well as their high load capacity. In addition, metal curing tools stand out for their robustness and thus high structural durability so that they can be utilized without difficulty for more than 1000 cycles. The surface of these tools is resistant to organic solvents and release agents, and even damage, such as scratches and dents, can typically be repaired easily.

Steel, aluminum, and ferronickel alloys are generally utilized, with steel and aluminum being the materials of choice because of their price and durability. Both materials exhibit a high coefficient of thermal expansion (Table 6.4) that needs to be taken into consideration during design of the tooling, in particular for high curing temperatures, in order to ensure its dimensional accuracy. Depending on tooling and application, thermal expansion may be desired, e. g., in order to facilitate for the finished component to shrink off the tooling during cooling (Chapter 8). Contour accuracy of 0.3 to 0.4 mm, even with large toolings (e. g., 20 × 5 m), can be achieved with metal tools at room temperature.



Figure 6.3 Steel tooling for the manufacture of rotor blades
[Courtesy: Premium Aerotec GmbH]

A disadvantage of steel is its high weight, which complicates handling and impedes heat transfer. Metal curing tools are generally manufactured by contour milling, but also by preforming of sheet metal. In order to ensure the created geometry, in particular for a large tooling (e. g., thick profiled sheet), even at high temperatures and/or during handling (e. g., by crane), the geometry is often stabilized by a stiff sub-structure (“egg carton” structure). This sub-structure also facilitates safe handling and installation, e. g., of vacuum lines and hoisting points to lift the tooling via crane.

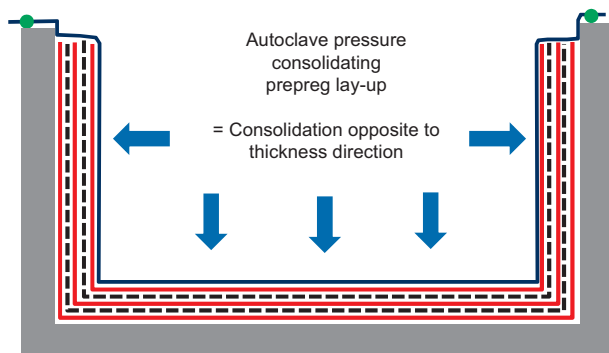


Figure 8.29 Consolidation and compaction inside tooling

Thus, the inner plies are forced to expand or to perform a relative movement in order to balance the change in length caused by the consolidation. However, they may not always be able to make these adjustments. The applied autoclave pressure (typically 7 to 10 bar) and the bonding between the plies by the prepreg resin render relative movement of individual plies impossible. Because the autoclave pressure is more effective on the plane than on the radii, the inner prepreg plies will bridge the radius to a certain degree, thus changing force progression and load capacity of the component (Figure 8.30). The continuous, inner line marks the actual, the dotted line the required fiber orientation.

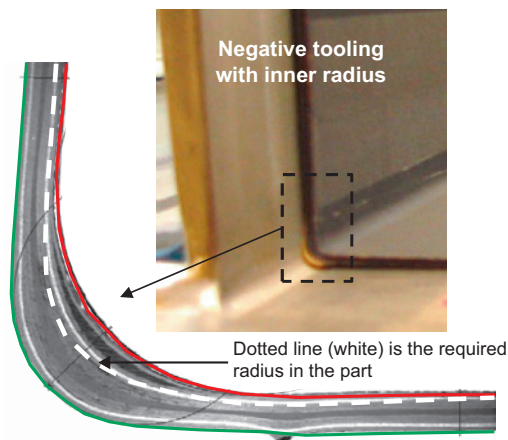


Figure 8.30 Micrograph of radius area showing the bridging effect of inner plies

This problem can be mitigated by one of several measures. For one, using a prepreg with a higher level of impregnation may minimize the consolidation. Another approach is the manufacture of a flat prepreg stack via ATL (rather than hand lay-up) and subsequent hot forming that would facilitate a pre-compaction of the stack prior to the curing process. Yet another, although somehow controversial,

method is the use of pressure strips (rubber or silicone corner profiles) in the radii. It is possible to effectively increase the autoclave pressure in this area using pressure intensifiers in the form of round cords. However, they often cause deep undesired indentations in the laminate or the formation of beads in the border area of the pressure strips.

Interrelations: Example of a Sandwich Structure

The interrelations between component design, material, and curing process and their implications will be described using a sandwich structure as an example.

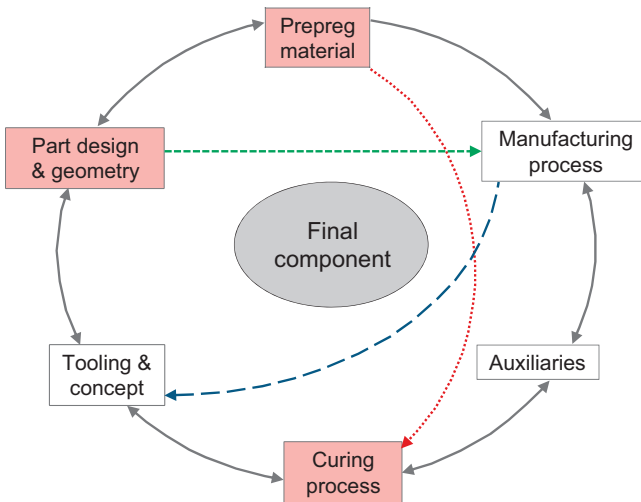


Figure 8.31 Effects of design, material, and curing process on component

PMI rigid foams (Evonik) can be used for the manufacture of Omega stringers (Figure 8.32). Typically, the formed foam remains in the component after curing of the prepreg, allowing for one-step curing of stringer and skin segment to be reinforced.



Figure 8.32

Example of sandwich design: Omega-stringer with foam core

The cell walls of certain rigid foams do not exhibit sufficient dimensional stability under the temperatures (typically 180 °C) and pressures (7–10 bar) required for prepreg curing. Therefore, the pressure on the component is reduced to, e.g., 3 bar absolute.

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