

# Chapter 4

## Thermoplastic Composites for Aerospace Applications



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### 1 Introduction to Thermoplastic Composites

The use of composite materials in transportation sector has progressively increased over the last decades bringing a breakthrough point in the way vehicles are designed and built. With respect to traditional materials, in theory, composites offer unlimited flexibility in designing complex shapes, higher efficiency in manufacturing processes, reduced operational costs and potential notable weight savings. Looking at the aviation industry, which constantly strives to reduce its impact on the environment, the Advisory Council for Aviation Research and innovation in Europe (ACARE) aims for 75% reduction in CO<sub>2</sub> emissions per passenger kilometer compared to 2000 (Flightpath 2050 Europe's Vision for Aviation n.d.). Therefore, the introduction and development of new materials together with related production processes will be one of the key elements to achieve this goal.

Good evidence of this can be found in the current aerospace programs such as the Airbus A350 (Marsh 2007), Boeing B787 Dreamliner or Bombardier C-Series CS-100 (Marsh 2013) which have about 50–53% of their total structural weight (excluding engines) made of composite materials. The first application of

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thermoplastics on aircraft parts dates back to the US military’s F-22 jet fighter’s landing gear and weapons-bay doors (1980), but today they are not widely used yet, because of some issues related not to the feasibility of the parts, but to the affordability of the manufacturing processes.

Currently flying thermoplastic composite flight-critical parts include the main wing leading edges of the Airbus A380 superjumbo passenger jet and the A340 500/600-series airliners that preceded it, as well as the Gulfstream Aerospace G650’s rudder and elevators (Black 2016). In parallel with aerospace field, BMW is leading a step change in automotive market by starting in 2013 the fabrication of the world’s first volume-produced vehicle (100 cars/day) with a passenger cell made from CFRP, the i3 model, of which about the 10% (about 13.3 kg with respect to the 140 kg of the car body) is thermoplastic matrix based (Starke 2016).

Composites world is in continue evolution and there has been a progressive change in terms of manufacturing processes, passing from standard wet or prepreg manual layup to automated (preforming) technologies, as Automated Tape Laying (ATL) and Automated Fiber Placement, with the objective to increase production rates and make cheaper manufacturing processes (Fig. 4.1) (Red 2014).

With thermoset composites the use of an autoclave for the curing is seen as a bottleneck to progress and main industrial actors are working to develop or improve out-of-autoclave fabrication processes to produce structural composite parts. Within this context, thermoplastics will give significant cost reductions if the potential advantages in processing (as thermoformability and weldability) will be exploited through suitable and automated processing techniques. A specific opportunity is related to the possibility to produce parts by ATL or AFP without autoclave, processes not yet performed with a suitable quality of the fabricated parts.

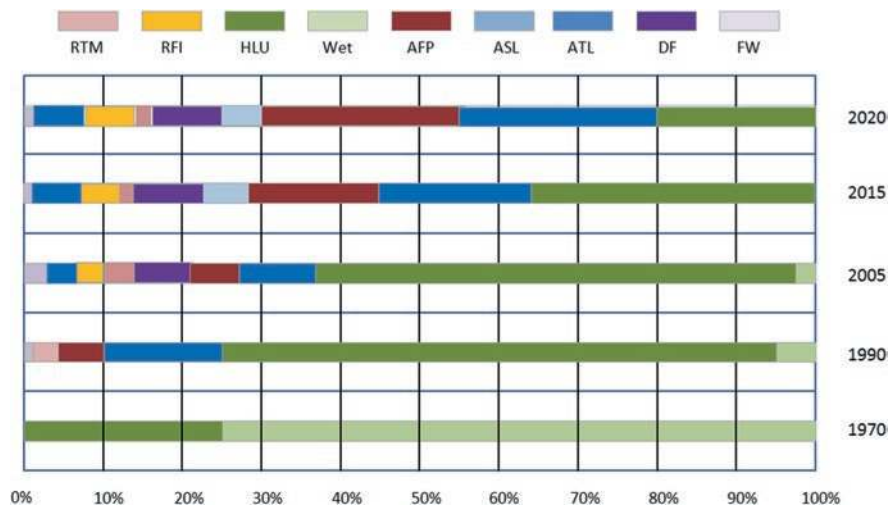


Fig. 4.1 Composites in Aerospace sector, 1970–2020. (Adapted from Red 2014)

This process can give a strong cost reduction which could compensate the high cost of the thermoplastic matrix prepregs. Since thermoplastics are not cross-linked, they are much tougher than thermosetting, hence they are more keen to be used in applications where damage tolerance is the driver. In theory, since thermoplastics are fully reacted, they do not need any reaction during cure so the process is simpler and faster. For this reason thermoplastics can be consolidated in minutes with respect to standard thermoset resin systems which require long curing cycles to build cross-links. Another advantage in terms of economical impact is related to the handling and storage of such materials, since thermoplastic prepregs do not require refrigeration during transportation and storage. Therefore, a wide use of thermoplastics implies a reduction of both investments for storage itself and cost of transportation. Also the material life management is simplified as it is not correlated to the resin expiration having, in theory, an unlimited shelf life. The good thermal stability is important for high-temperature applications (supersonic, high-temperature parts) and the good fire resistance is useful for some specific applications (nacelles, interiors). Figure 4.2 (Wong 2017) reports BMW's manufacturing technology curves highlighting how thermoplastics will impact the production processes and how this transformation will be fast.

The interest for recyclability (discussed in the following) which for thermoplastics is better than for thermosets, is high and it is supposed to increase in the future. On the other side the nature of thermoplastics has some areas of disadvantages respect to the thermosetting. The thermoplastic prepreg is typically supplied in a roll form, stiff and board. As it is fully reacted the material does not have any tack and drapability, resulting in a difficult layup process with respect to the standard

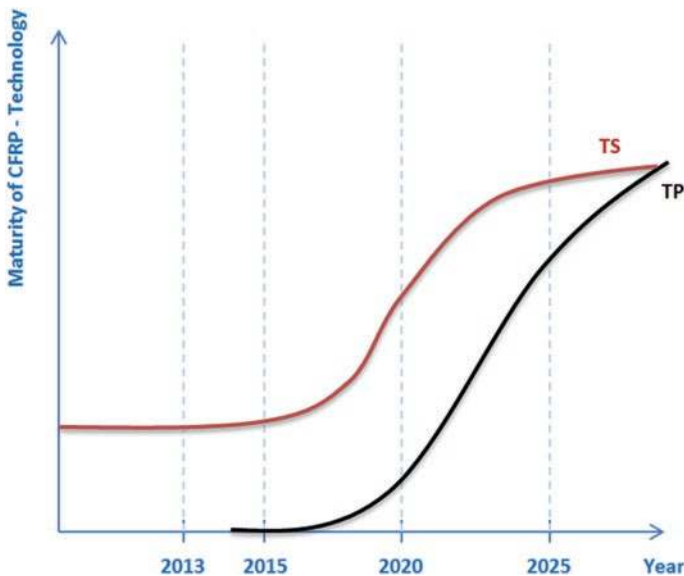


Fig. 4.2 BMW technology curves. (Adapted from Wong 2017)

tacky and drapable thermoset prepreg. In this case, the hand layup process is not a preferred option, especially with complex and contoured shapes. Based on this, the current applications are dedicated to simple geometries even if in the recent years new technologies are focusing on more complex shapes, targeting the standard common applications of epoxy systems. Furthermore, processing temperature of thermoplastics is considerably higher with respect to the standard thermosets. As example a PEEK prepreg composite material should be processed at temperature higher than the melting temperature, around 380 °C, compared to 180 °C of a standard epoxy resin. This means that the equipment to process thermoplastics are more complex, expensive and process toolings with low CTE are needed. Another possible issue is related to resistance to the environment and solvents that is lower than thermosettings (mainly for amorphous thermoplastics).

## 2 Polymer Chemistry

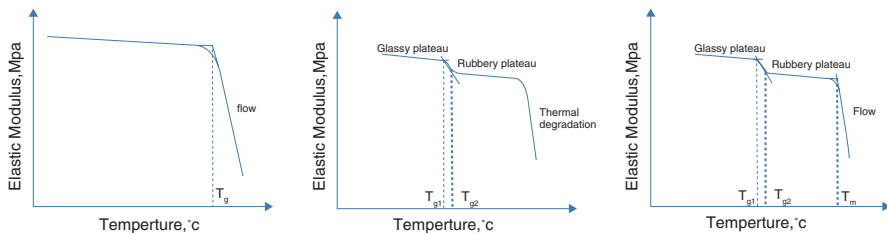
Molecular structure of polymeric materials is characterized by the presence of long chains. This structure determines all the main chemical, physical and mechanical characteristics of polymers. Depending on the way of creating the chains during polymerization, two major families of polymeric materials can be identified, as thermoplastic and thermosetting polymers. In case of *thermoplastic polymers* the polymerization creates long chains without branching; the obtained polymeric material is characterized by the interaction among such chains which are not interconnected by molecular links. The only interactions between the chains are given by low electrostatic attraction (van der Waals forces) and by temporary links (entanglements). When the temperature decreases, causing the reduction of the molecular motions, van der Waals forces are sufficient to take the macromolecules together and the material keeps its volume and shape, as a solid material. At molecular level the macromolecules cannot be organized in an ordered network and the appearance is typical of the liquid state. This status, characterized by a behavior (e.g., own volume and shape) typical of solids with a disordered molecular state that is typical of fluids is called “glassy” state; some characteristic of glasses (e.g., transparency) depend on such structure. When the temperature is increased, above a certain value the molecular kinetic energy is sufficient to overcome the electrostatic attraction and the movements between macromolecules become possible; in this state the polymer behaves as high viscosity liquid.

The transformation from glassy to liquid state (and vice-versa) is called “glass transition”; the glass transition temperature is generally indicated with  $T_g$ . Glass transition is a progressive transition and generally,  $T_g$  is an average value of temperature in a transition zone that can be identified by a starting and an ending temperature. In case of *thermosetting polymers* (thermosets) the polymerization is not only linear, but also branching are created. This type of molecular growth gives also the possibility to make molecular stable links between the molecular chains (cross-links), creating a three-dimensional structure. This structure at low temperatures is

in glassy state (as the one previously described). Above the  $T_g$  molecular motions become possible, but the flow is inhibited by the cross-links that cause a change of properties (e.g., a decrease of the elastic modulus) without flow. This state is called “rubbery state,” because is typical of rubbers, that can be considered a thermosetting material with cross-link (namely with a reduced amount of cross-links). Due to the presence of the cross-links, a polymerized thermosetting polymer does not flow also like cracking and/or oxidation, but flow is not possible anymore. On the basis of the previous description some major differences between thermoplastic and thermosetting properties can be identified: Linear growth of molecular chain (Thermoplastics) vs branching and cross-linking (Thermosets); flow possible above  $T_g$  (Thermoplastics) vs inhibited flow also above  $T_g$  (Thermosets). The behavior of elastic modulus vs temperature for Thermoplastic and Thermosets is shown in Fig. 4.3a, b.

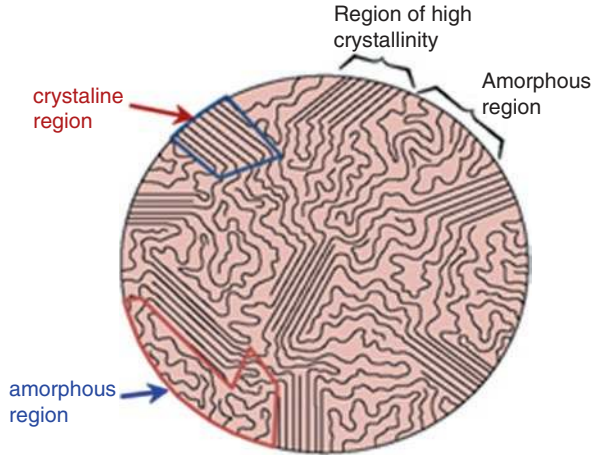
Namely for Thermoplastics the behavior shown is referred to amorphous thermoplastics. In fact in case of molecular flexible chains some portions of macromolecules can reach an arrangement similar to the solid ordered one (crystalline state); in polymers this arrangement can be obtained only for a part of the chains, while the other portions keep a disordered state. A sketch of this structure is shown in Fig. 4.4; the polymeric materials having this structure are called Semicrystalline Thermoplastics and the thermoplastic previously described, without crystalline phase, are called Amorphous Thermoplastics. The crystalline phase is organized like a solid, in crystalline lamellas that frequently are assembled in radial structures (spherulites).

One of the main factors influencing the degree of crystallinity is the rate of cooling during solidification, together with the molecular chemistry of the polymer. It is very difficult that polymers are 100% crystalline, because it is rare to have all chains aligned in all the regions. Heat treating causes crystalline regions to grow and % crystallinity to increase. Degree of crystallinity is usually expressed in % and some physical properties depend on % of crystallinity. The crystalline phase shows also a melting temperature, well above  $T_g$ . The presence of a crystalline structure hinders the flow of the amorphous phase above  $T_g$ , like in the thermosettings. But when the temperature of crystalline melting  $T_m$  is reached, the flow is possible. The behavior



**Fig. 4.3** Typical elastic modulus versus temperature curves for amorphous thermoplastics (a), thermosets (b) and semicrystalline thermoplastics (c)

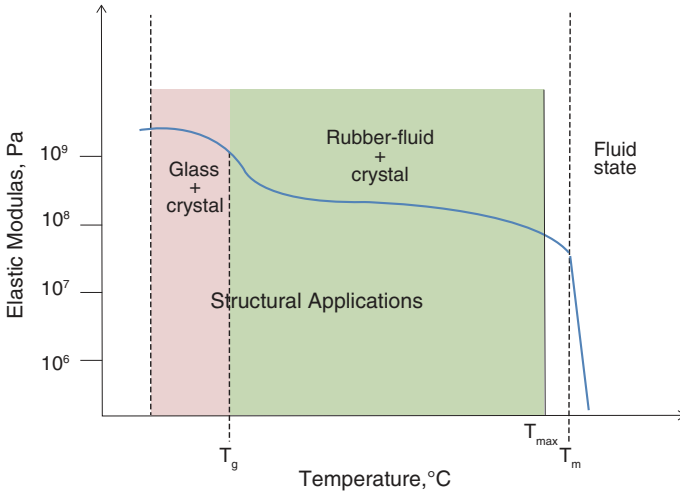
**Fig. 4.4** Sketch of a typical semicrystalline thermoplastics structure. (Adapted from Fig. 14.11 H.W. Hayden, W.G. Moffatt, and J. Wulff, *The Structure and Properties of Materials*, Vol. III, Mechanical Behavior, John Wiley and Sons, Inc., (1965))



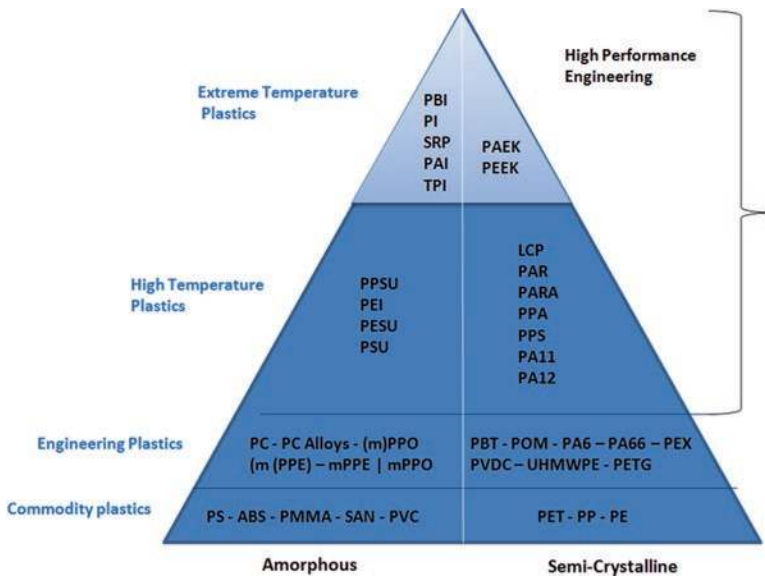
of elastic modulus versus temperature for Semicrystalline Thermoplastics is shown in Fig. 4.3c.

With regard to Fig. 4.3c, in correspondence of the  $T_{g1}$  the reduction of the modulus is lower than what occurs in case of the amorphous polymers. It is noticed that in case of amorphous polymers, in correspondence of the  $T_g$ , there is no change in the arrangement of molecules and only the mobility of the chains changes, which still maintain a disordered state, typical of the amorphous structure. After the  $T_m$  it is not possible to measure the modulus since it is in fluid state. By the point of view of structural properties, for the mentioned materials the Glass Transition Temperature  $T_g$  is a point where such properties decrease; consequently a material for structural applications must show a  $T_g$  suitably above (with a sufficient safety margin) the maximum service temperature, which for commercial aircraft is typically 82 °C. For applications different from the structural ones (e.g., domestic commodities) some semicrystalline thermoplastics (e.g., Poly-ethylene PE or Poly-ethylene terephthalate PET) are used at operating temperature comprised between  $T_g$  and  $T_m$  (Fig. 4.5). They are flexible, due to the reduced stiffness, but keep the shape and are not brittle.

With regard to the molecular weight, intended as the mass of a mole of chains, tensile strength increases with it because longer chains are entangled better. Furthermore, tensile strength and elastic modulus often increase with the percentage of crystallinity. In terms of high-performance materials, as reported in the thermoplastic polymer pyramid (Fig. 4.6), it is worth mentioning the family of Polyaryletherketones (PAEKs) and their derivatives. PAEKs are obtained by joining ether groups with ketone groups and the way these groups are arranged in the final molecule identifies the various constituents of the family. The percentage of ketone groups determines the melting temperature. They are semicrystalline thermoplastic polymers with a wide temperature range, high strength, stiffness and resistance to hydrolysis, which make them suitable for applications demanding extreme conditions.



**Fig. 4.5** Typical operative temperatures for semicrystalline polymers in applications requiring flexibility



**Fig. 4.6** Thermoplastic polymers pyramid

One of the most significant thermoplastic resins used for structural applications in aerospace field is definitely the Polyetheretherketone (PEEK) whose structure is born precisely from the repetition of two ether groups and one ketone. The presence of aromatic rings and ketones confers a certain rigidity to the molecular structure,

**Table 4.1** Thermoplastic resins commonly used in aerospace sector

Chemical name	$T_g$ (°C)	$T_i$ (°C)	Crystallinity	Main characteristics	Cost (€/Kg)	Sector
PEI (Poly Ether Imide)	215	–	Amorphous	Excellent FST properties; limited chemical resistance	15–20	Aerospace interiors
PES (Poly Ether Sulfide)	220	–	Amorphous	Toughness; low density	1–3	LFT; flexible pipes
PEEK (Poly Ether Ether Ketone)	140–145	334–343	Semi	Excellent chemical resistance; high T performance	60–80	Aerospace; medical
PPS (Poly Phenil Sulfide)	85–90	275–290	Semi	Excellent chemical resistance	15–25	Aerospace; industrial

dampened however by the introduction of the ether groups. For this reason, PEEK has a lower melting temperature than other compounds of the same family, such as PEK (Polyetherketone) or PEKK (Polyetherketonketone). Table 4.1 lists main properties and cost of the most commonly used thermoplastic resins in aerospace field, also for structural applications.

At last, in addition to polymers reported in Table 4.1, it is worth to mention the semicrystalline polymer PEKK which is recently emerging for a wider spectrum of applications with respect to PEEK, including the potential production of aerospace primary structures by out-of-autoclave manufacturing processes (i.e., Continuous Compression Molding, In Situ Consolidation).

Different properties of PEEK and PEKK, due to different structures (Fig. 4.7), imply that PEKK has up to 80% greater compression strength than PEEK and wider processing window of parameters than PEEK. With regard to the temperatures, PEKK has a melting temperature comprised in the range 280–390 °C and a glass transition temperature of about 150–165 °C. Chemical structure adjustable gives PEKK superior fusion properties with metal and other polymers, induced interesting applications. PEKK can be located at apex of the PAEK class thanks to unique mechanical, physical and chemical properties.

### 3 Thermoplastic Manufacturing Processes

This paragraph deepens some of the most innovative manufacturing processes already used and under development in aerospace sector to produce structural reinforced thermoplastic-based composites, starting from a brief comparison of the processing aspects with thermosettings.



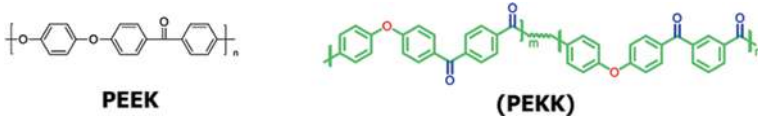


Fig. 4.7 Comparison of PEKK (copolymer) and PEEK (homopolymer) polymer structures

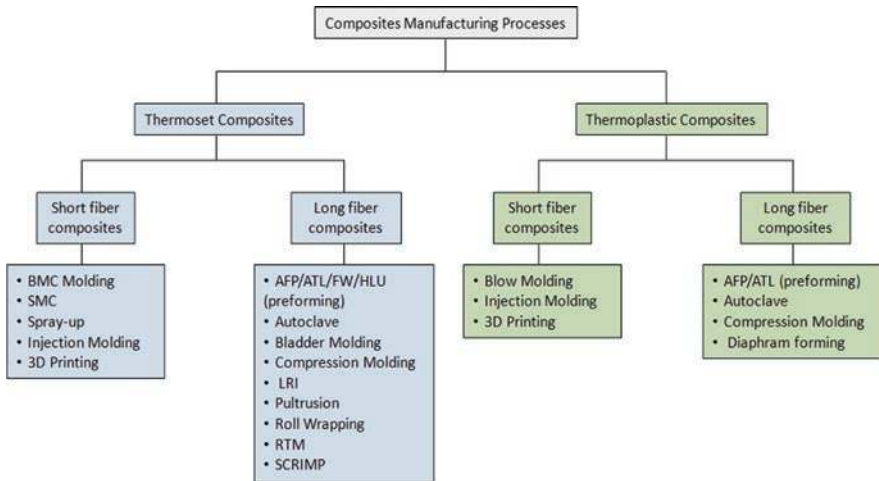
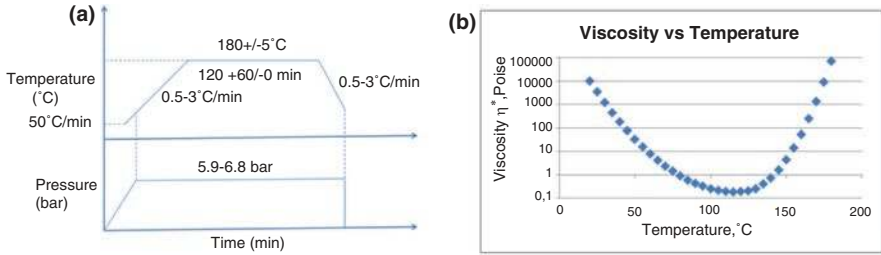


Fig. 4.8 Thermosettings and thermoplastics manufacturing processes

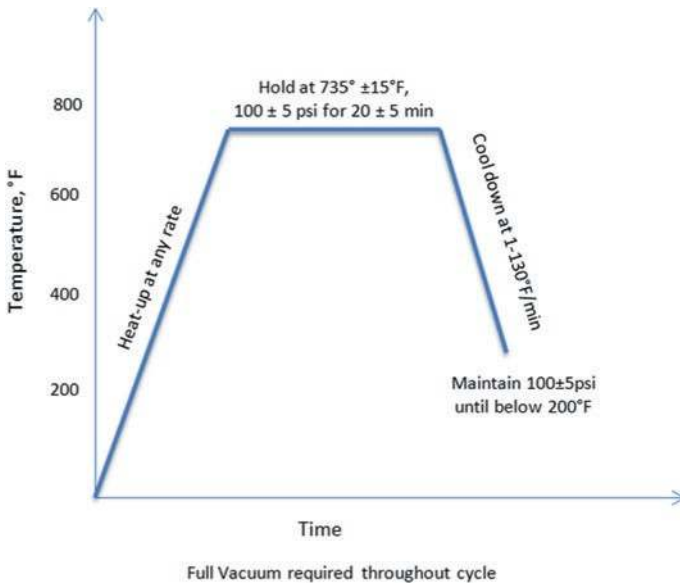
Figure 4.8 summarizes the production processes for thermoset and thermoplastic composites divided by short fiber and continuous long fiber composites.

The most commonly used production process for thermoset composites is based on manual layup of prepregs followed by autoclave curing. This process is based on shaping the part when the prepreg is uncured, consequently tacky and drapable. Afterwards, the material is cured in autoclave with a temperature and pressure cycle (Fig. 4.9a); this cycle requires a controlled heating rate, due to the need to control the curing reaction and the evolution of the resin viscosity with the temperature (rheological curve, Fig. 4.9b). In order to reach a suitable polymerization level, a long period (e.g., from 1 to 2 h) at constant temperature is required before cooling. Generally, the curing cycle is long and requires high pressure, then it is expensive.

With respect to thermosets, thermoplastics are generally purchased in a polymerized form; they are not tacky and their processing requires to reach a temperature sufficient to have the resin in a fluid state (above  $T_g$  for amorphous thermoplastics and above  $T_m$  for the semicrystalline ones). The holding time at high temperature is only the one needed to shape and consolidate the plies, not for polymerization (the resin is already polymerized), consequently can be short enough. Also cooling can be fast, except for taking in account thermal deformation and (for semicrystalline thermoplastics) the effects on crystallinity of the cooling rate. As a consequence of



**Fig. 4.9** Typical curing cycle of thermoset (epoxy) composites (a) and typical viscosity curve of epoxy resin during cure heating (b)



**Fig. 4.10** Recommended autoclave cure cycle for APC-2-PEEK Thermoplastic Polymer. (Source: Solvay Cytec website)

the previous considerations, processing of thermoplastics is potentially cheaper and faster than the one of thermosettings, if peculiar fabrication techniques (as thermoforming and assembly through welding) are developed. As example, recommended autoclave consolidation cycle for APC-2-PEEK Thermoplastic Polymer from Solvay Cytec is reported in Fig. 4.10.

### 3.1 Thermoforming Process

The fabrication process for thermoplastic composites requires three main steps: heating to melt the polymer matrix; pressurization to consolidate and shape; cooling to bring the polymer to the solid state. When forming and consolidating thermoplastic composite materials, there are three main mechanisms that allow the user to fabricate the parts:

- Polymer percolation (polymer flow through layers)
- Transverse flow (fibers spreading under pressure)
- Intraply shear (ply movement during forming)

Thermoforming process is a rapid manufacturing process used to form a 2D panel into a 3D shape. It is an attractive process thanks to the relative low cycle time. Typically, it requires the use of a semi manufactured item. In order to optimize the process, the thermoforming step is fed using a pre-consolidated sheet.

The pre-consolidated sheet is manufactured by using the thermoplastic composite material, starting from a roll form. The material is cut in a certain number of plies and the stacking sequence is made by holding them together with soldering iron on the edge of the preform. Most recently this activity has been made automatic using numerical control cutting equipment and pick and place stations, where the plies are laid up automatically to get the final panel. The consolidation can be performed with autoclave or press. In any case the temperature to be reached should be higher than the melting point (see Table 4.1). As example, PEKK autoclave consolidation requires a cure cycle with holding phase at 380 °C for 20–30 min and a pressure of 7–10 bar, depending on final part application. The holding time depends on the panel's thickness and it has to be measured on the lagging thermocouple. Experimental studies (Offringa 1996; Scherer and Friedrich 1991) demonstrate that the effect of pre-consolidation has a direct impact on final part quality after the thermoforming process. Free void laminates at this stage can generate good part quality after the final forming. After the pre-consolidation the panel can be stored, shipped, and trimmed. This is the typical part used for the final forming. This stage requires two different equipment. The first one is the *I/R* oven which allows the panel to become “soft” and achieve the melting point. Then the part moves to the press where the tooling is heated. The main phases of the thermoforming process are reported in Fig. 4.11. The transfer time from the *I/R* heater to the press is a key parameter to reach the right crystallization rate. For PEKK the tool should be set at

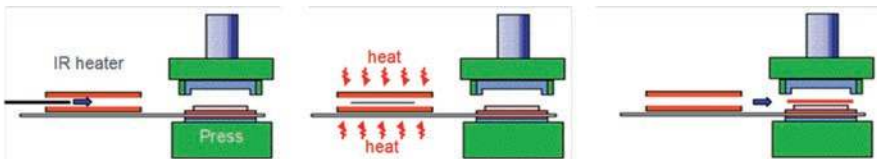


Fig. 4.11 Sketch of thermoforming process

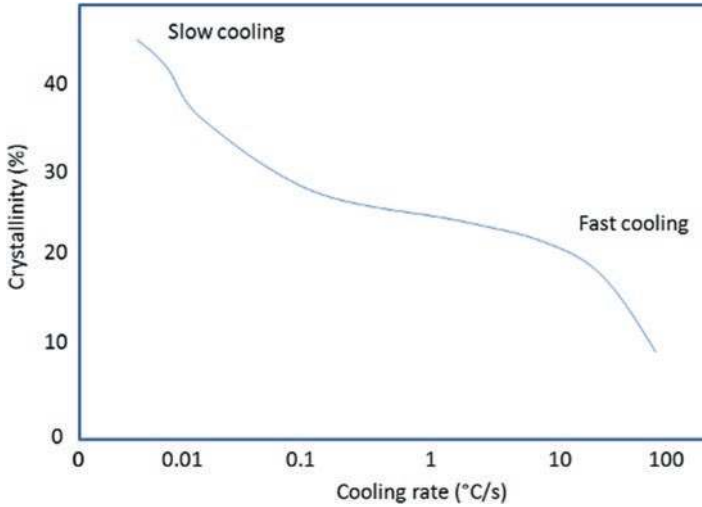


Fig. 4.12 PEEK/CF processing window: relation between degree of crystallinity and cooling rate

about 240° and its function is to anneal the part and obtain the right crystallization rate. The pressure typically reaches 20–30 bar and the time depends on the geometry.

The quality of the final formed parts includes several properties like porosity level, crystallinity and interlaminar bond strength. All the properties depend on local thermal and pressure influence.

While it has been demonstrated by experimental study (McCool et al. 2012; Wakeman and Blanchard 2005) that the porosity level is influenced by the quality of the pre-consolidated sheet, the crystallization rate depends on the cooling rate during the forming process. The polymer morphology depends on the thermal history created during the manufacturing process. To better understand the effect of parameters as temperature and pressure on the mechanical properties of a part produced by thermoforming process, taking as reference a reinforced PEEK-based composite material and related processing window (Fig. 4.12), it was shown (Mallon et al. 1998; O’Bradaigh and Mallon 1989; Davies et al. 1989) that laminates consolidated in the temperature range comprised between 360 and 390 °C have a more uniform impregnation of the matrix and an increased flexural failure.

Furthermore, high cooling rates upon forming decrease the degree of crystallinity of the polymer and increase the void content (Manson et al. 1990; Wakeman and Blanchard 2005). With regard to the fracture toughness, it was demonstrated (Ye and Beehag 1996) that the pressure effect during the cooling returns an increment of the mode II with a higher cooling pressures; with regard to the effect of the temperature, both mode I and mode II fracture toughness of laminates formed between 380 and 400 °C, because of the improved composite interfacial strength, are considerably higher than the ones formed at 360 °C. Despite of these results, it does not mean that increasing the temperature improve properties.

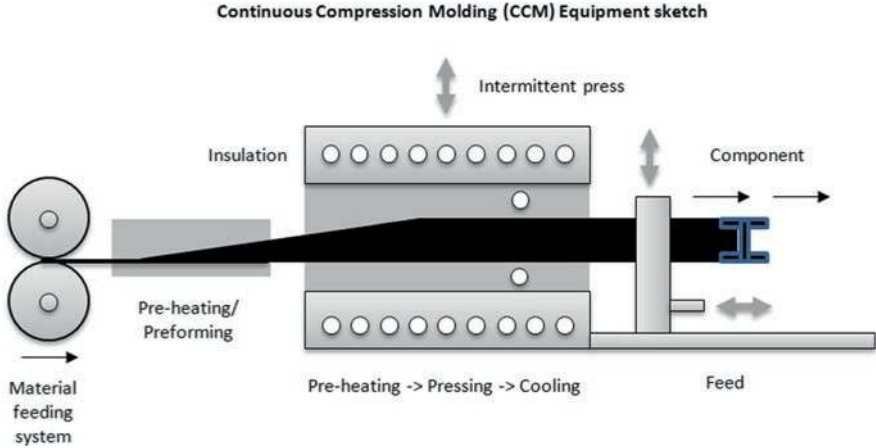
In conclusion, a numerical tool can be used for the design of the mold and in order to predict the effect of processing parameters on the part quality. For instance one of the most important parameters is the formed sheet temperature, but its distribution is also important because it impacts the wall thickness distribution of the part (Pepliński and Mozer 2011).

### 3.2 *Continuous Compression Molding*

Continuous Compression Molding (CCM) is an innovative fabrication technique aimed to obtain parts by using a press to overcome some problems of thermal history management that can be experienced in “traditional” thermoforming. Specifically, thermoforming process is made with hot press and thermoplastic matrix composites forming is performed at a temperature above  $T_g$  for amorphous thermoplastics, above  $T_m$  for the semicrystalline ones. After forming process, the press cannot be opened before cooling up to a temperature where the material is solid. This process then requires long times (each cycle requires heating and cooling) and high energy consumption. Starting from these considerations, a cheaper process can be performed by the heating of pre-consolidated layers, followed by the forming in a press with cold tool. Sometimes, in order to reduce the internal stresses induced by fast cooling and in some cases to prevent amorphization by fast cooling, preheated tools (temperature below the preheating one and above room temperature) are utilized. Both the described processes can produce items with a maximum size limited by the available tool and press. Instead CCM is capable to produce highly shaped profiles or flat panels of effectively unlimited length. Operable by one person, the computer-controlled process yields product at speeds approaching those quoted for pultrusion—as high as 40 m/h (131 ft/h) for shaped profiles and up to 91 m/h (300 ft/h) for flat panels (Gardiner n.d.).

If the need is to produce an angle ply laminate (i.e., quasi-isotropic layup), strips of woven or UD material may be cut and welded together to form a multilayer unconsolidated panel with a specific layup which is used as input material for the continuous press. This sort of preform could also be produced by AFP/ATL machines reaching in this way a very high level of automation of whole manufacturing process. Continuous Compression Molding is based on the utilization of a press having a temperature decreasing from one side to the other; the part to be fabricated can be moved along the platens of the press with an alternate opening/closing cycle, moving the part ahead during the opening. Figure 4.13 shows a sketch of the equipment needed for the CCM process. This process makes possible to obtain a progressive cooling of the part thanks to its movement in the direction of lower temperature zones.

The cooling rate of the material running along the plates of the press depends on the change of temperature along the press ( $^{\circ}\text{C}/\text{min}$ ) and on the medium speed of the part along the press (m/s); the combination of the two changes gives cooling rate ( $^{\circ}\text{C}/\text{s}$ ) which can be set through the two abovementioned process parameters. As a

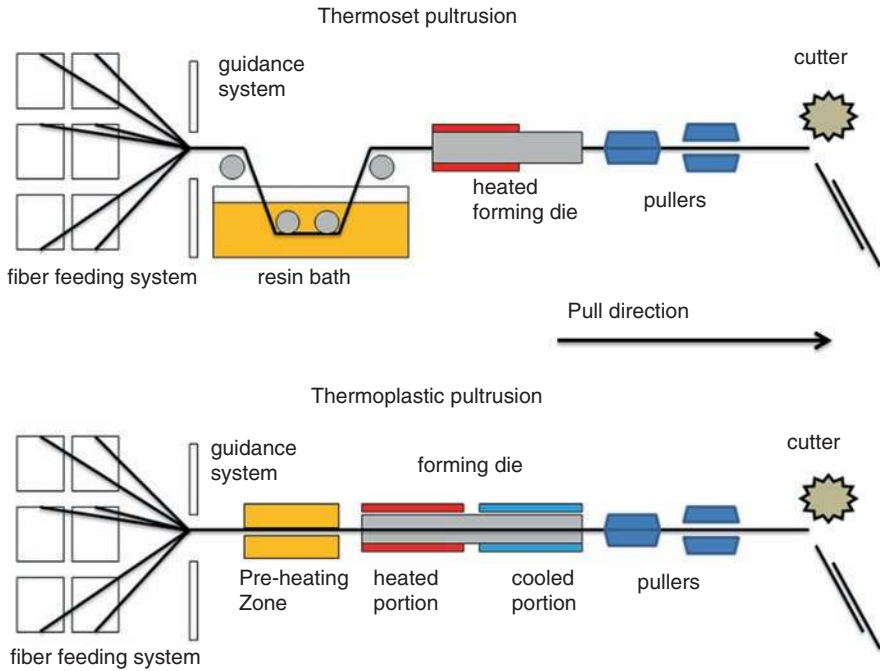


**Fig. 4.13** A sketch of the CCM process

consequence of that, the first advantage of CCM is the capability to control cooling rate; a further advantage is given by working with a press at a temperature which is variable through the length but is kept constant in time, with a significant energy saving with respect to the standard thermoforming process. The geometry of the parts that can be fabricated with this technique is characterized by constant thickness and layup, as well as constant section and theoretically unlimited length. The parts are straight, but modifications to obtain constant radius curved parts seem possible. For these parts (straight or constant radius curved, constant section and layup) this technique is very cost effective. Changes in thickness and curvature radius are not presently possible, but they are being presently studied. One of the key features of the CCM process is that a part layup can be modified by adding/reducing layers without stopping the machine, and just adequately replacing the feeding material.

### 3.3 Pultrusion

Pultrusion is a continuous and automated technology able to produce constant section and rectilinear axis for high rate productions. Thermoset (epoxy, vinyl ester, polyesters) pultrusion is a mature process consisting in four main steps, as: continuous fibers (mat and rovings) are pulled through a guidance system; fibers enter a resin bath carrying out the impregnation step; fibers enter a heated die where exceeding resin/air are removed and starts the curing process (continuous cross-linking); finally, composite profile is pulled and cut at the desired length. A sketch of pultrusion line for thermoset and thermoplastic prepreg composites is reported in Fig. 4.14.



**Fig. 4.14** A sketch of pultrusion line for thermoset- and thermoplastic-based composites (Novo et al. 2013)

Thermoplastic pultrusion is a relatively new process and limited research has been done up today. This is because the process has been mainly applied for thermosetting matrices as there are inherent difficulties associated with the thermoplastic matrices, such as high-processing temperatures and high melt viscosities (2 or 3 order of magnitude higher). Despite of this, with recent developments, the use of preforms as pre-consolidated tapes, commingled yarns and towpregs, allowed the thermoplastic pultrusion to gain a great interest (Nguyen-Chung et al. 2007). For the production of thermoplastic based composite profiles, pre-impregnated materials are guided into the preheating furnace to be heated up to the required temperature. Then, they enter in the pultrusion heated die to be heated and consolidated to the required size and, after that cooled down in the cooling die to solidify. Several variables are involved in the impregnation process. The fiber bundles are pulled through the die with an imposed force or velocity. For a given geometrical configuration, if we increase the pulling velocity, the contact time decreases and impregnation can become poor. Conversely, if one decreases the pulling velocity, the process efficiency can become too low and will have no economical interest. As a consequence, a thorough understanding of the pultrusion physics is crucial for a rigorous prediction of impregnation in order to reduce the time and the cost of fabrication (Novo et al. 2013).

### **3.4 *Laser-Assisted Automated Fiber Placement (AFP)/ Automated Tape Laying (ATL) and In Situ Consolidation (ISC) Process***

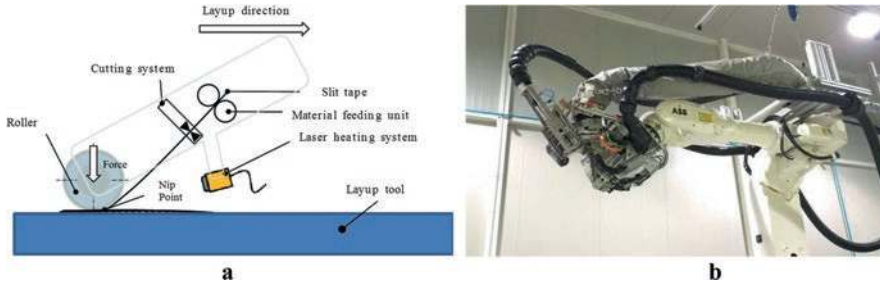
AFP/ATL technology has been introduced in aerospace field in order to reduce the production cost-flows of primary and secondary structures. The improvement is made possible thanks to the higher deposition rate with respect to manual layup (about 4:1); increased buy-to-fly ratios; a seamless transition between design and manufacturing (usage of advanced numerical tools); high repeatability of the process. The base concept of the AFP/ATL technology is quite the same, since the end-effector typically includes a tape feeding system with a cutting mechanism, a compaction roller and a heating source. The usage of AFP or ATL depends on specific needs concerning the geometry and layup of the composite part. More in detail, ATL is faster than AFP if parts have minimal contoured surfaces and exhibit little complexity or surface irregularities. In case of curved parts with complex contours, which require the placement of reinforcements in specific locations, AFP is more effective, because it counts on tapes (slit tape format, from 1/8"), more easily managed than the single tape of an ATL (Barile et al. 2017). Currently, the standard thermoset prepreg-based AFP/ATL process is utilised to produce fuselage sections of Boeing 787 and Airbus A350 XWB. Most recently the AFP/ATL machines are used also to manufacture engineered pre-consolidated sheets in order to reduce the scrap rate with respect to the standard hand layup process.

The scrap rate for a semi-complex part can be somewhere in between 20% and 50% when laid up by traditional Hand Layup. This scrap rate can be brought down to a value below 5% by the usage of ATL/AFP process. The main parameters which affect the quality of the laminate are the heating source power (and so the temperature reaches at the nip point during the deposition), the roller pressure, and the speed of the robot, while the clean room humidity and temperature, as well as the creel setup have no influence, as instead it happens in case of thermoset composites. AFP/ATL used for processing thermoset materials differ from the ones used for thermoplastics mainly for the heating source used. Due to the need to achieve higher temperatures to melt the polymer and allow the ply–ply adhesion, for thermoplastics usually a diode laser is installed on the end-effector in the place of I/R heater, which is commonly used in case of thermoset prepreps.

Furthermore, for the deposition of thermoplastic composites, an infrared camera is used to set up the material heating law and for monitoring the temperature at the nip point, which is at the interface between roller and mold surface (Fig. 4.15a). The data can be used in an open or closed loop system. Figure 4.15a shows the Laser-Assisted AFP head concept and as example, Fig. 4.15b shows its application on a standard anthropomorphic robot (AFP cell) developed by Coriolis Composites (France) and available at NOVOTECH Aerospace Advanced Technology (Italy).

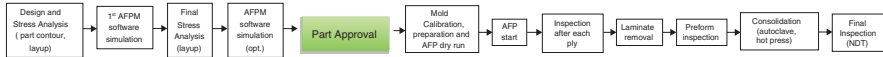
The first step, before fiber placement starts, is to put in relation the tooling with machine head by means of a calibration procedure. After the calibration is per-





**Fig. 4.15** AFP head scheme (a); Coriolis Laser assisted AFP robot at NOVOTECH Aerospace Advanced Technology (Italy) (b)

formed and the Part Program (layup, contours) is approved by the Design office, before starting fiber placement process, a dry run is performed by the AFPM operators to check the layup strategy (e.g., collisions, etc.). Afterwards, an amorphous layer or a release film is applied on the mold. When the fiber placement process starts, a band of parallel tapes are fed to the AFP head, passed through the cutting system, and placed on the mold according to geodetic curves set via AFP dedicated programming software. When tapes reach the nip point, at the same time they are subjected to the compaction force of the roller together with heating of the laser needed to melt the resin lightly in order to produce the right tackiness for the band deposition. Being able to adapt the width of the deposited band, AFPM is able to eliminate the excessive overlaps between adjacent courses. At the end of each trajectory, remaining tapes are cut so as to respect the contour of the part. Each tape is cut at  $90^\circ$  angle. The head is then positioned at the beginning of the next band. During the deposition of a band each tape is placed with its own speed so as to ensure that each other is in conformity with the others on the surface of the part. Therefore, the fibers can satisfy specific design requests. The action of the compaction roller enables to remove the air pockets. The positioning of the bands is previously set by a dedicated programming software associated with the fiber placement machine. The software enables to set the ply boundaries and orientations, the material parameters, gap/overlap, and other tolerances to be considered during the deposition. Basically, main technology parameters are: layup speed, roller's compaction force and heating source power (temperature at nip point). The software can also analyse fiber direction and perform specific simulations useful to optimize the deposition strategy. Specifically, it can include several possible analyses useful to assess the quality of the part that will be produced (i.e., steering analysis, angular deviation, etc.) and avoid collisions of the robot with mandrel. Figure 4.16 shows more in detail the Laser-Assisted Automated Fiber Placement process flowchart, in case of thermoplastic prepreg materials. At the current state of art, the main issue to fabricate thermoplastic-based composite aerostructures is not in the feasibility of the parts, but in the affordability of the production process. In particular, in case of primary structures, for sizes for which hot press is not suitable, the AFP/ATL



**Fig. 4.16** State of art for Thermoplastic composite structures produced by automated preforming and next consolidation step

deposition requires a consolidation in autoclave at high temperatures with high energy costs and the usage of very expensive “not flying materials.” In such context there is a technology which targets to have a consolidation during the layup phase known as In Situ Consolidation (ISC) by AFP process, mainly driven by a lower cost of part fabrication thanks to the reduced process flow. The concept of the ISC is depicted in Fig. 4.17. The consolidation temperature must be higher than the  $T_g$  for amorphous polymers and higher than  $T_m$  for semicrystalline polymers.

Generally, because of the higher viscosity, the temperature shall be at least 150–200 °C higher than the  $T_g$  in amorphous polymers and up to 100 °C higher than  $T_m$  in semicrystalline polymers. Complete ISC foresees the thermoplastic tape melted and consolidation during the machine lay up in order to not require any further process like consolidation in autoclave. The material quality plays an important role in this process as the resin-rich side improves the bonding and consolidation. Furthermore due to the nature of instantaneous consolidation and low amount of time to remove the porosity, the ply should be intraply void free, resulting fully impregnated.

Basically, sources of voids have to be searched in an insufficient level of intimate contact between the layers of a laminated. In particular, voids can migrate from the prepreg tape or can be created by the roughness of the tape and presence of gaps/overlaps during the placement. In order to improve the material adhesion this can be preheated but then the melting point can be reached at the interface between the new material layer and the previous one. The intimate contact occurs in fraction of seconds and the consolidation occurs only using the roller force application. Cooling rate is driven by the mandrel temperature and ambient temperature. Current limitations to commercial adoption of ISC processes in aircraft structures include: lack of proven data on materials and part performance with these new processing techniques; the need to demonstrate robustness process parameters for integral stiffened panels. If the applied pressure is less than a critical value, de-consolidation will occur and if it is higher than critical value, re-consolidation will happen. Also de-compaction behavior of the fiber and applied external pressure determine final de-consolidation state while resin melt viscosity which depends on processing temperature affects the time for de-consolidation to reach a stable state. Therefore, the general perception to avoid a two-phase process (AFP preforming process followed by autoclave or press consolidation) and obtain parts with mechanical properties comparable to the ones of parts produced with autoclave/press consolidation, is to optimize the machine parameters and material behavior when subjected to heating.

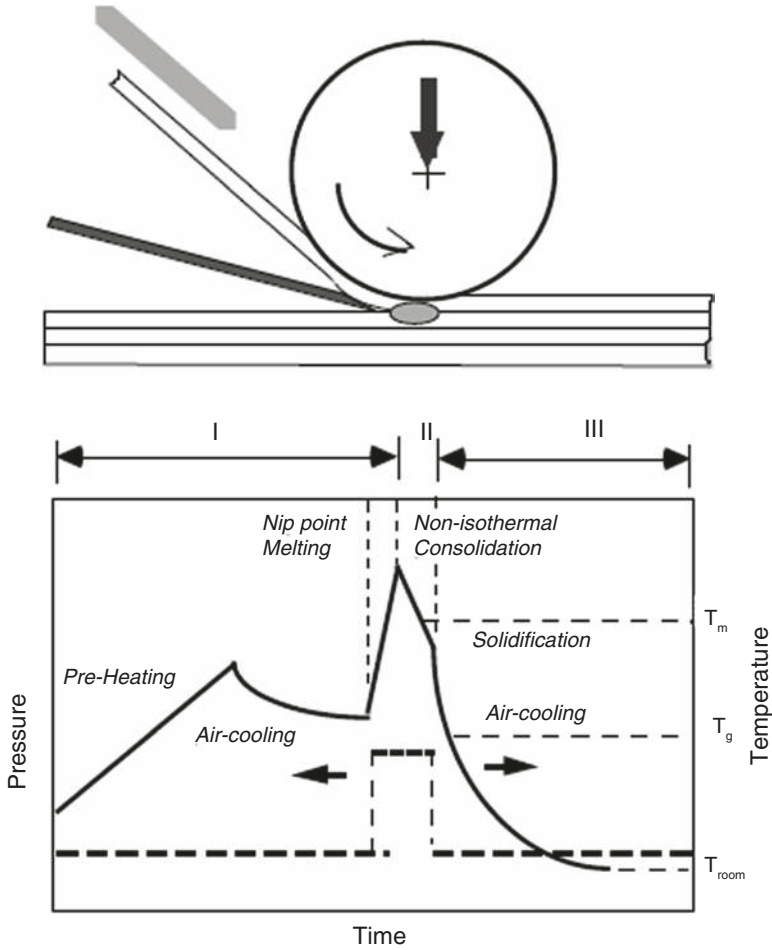


Fig. 4.17 Concept of the in situ consolidation by AFP process

## 4 Joining Processes

The choice of a material for an aerospace application depends most on the time of assembling procedures of the final structure (Vodicka 1996; MIL-HDBK-17-3F n.d.). This consideration is even more relevant for thermoplastic composites since their most important advantages are high damage tolerance and fast process rate (Biron 2007): for these materials the development of fast and reliable assembling procedures is a need coming from their requested production rate. In addition, it is important to consider that for thermoplastic composites higher limits in terms of complexity of the geometries that can be manufactured are experienced and that often a solution of the designer is the development of structures made of several

subcomponents with easier geometries. Thermoplastic composites, differently than thermosets, can be melted and reformed. In this way delaminations and other damages may be re-welded and repaired heating the matrix and applying a consolidation pressure. At the same time a complex component can be assembled through the fusion of some portion of the subcomponents to be jointed.

Several joining techniques are available for composites and some of these have been specifically developed for thermoplastics. All these techniques can be divided into three classes:

- **Mechanical fastening:** Bolted joints are characterized by easier manufacturing, lower cost, and ease of inspection. Anyway holes weaken the structure since they act as stress concentrators (Thoppul et al. 2009).
- **Adhesive bonding:** Adhesive bonding requires controlled and delicate procedures for surface treatment and adhesive application method. Thermoplastic matrices are characterized by lower surface energies when compared to thermosets which makes difficult for adhesives to wet the thermoplastic adherend surfaces and creates a bond with high mechanical strength (Hart-Smith 1973).
- **Fusion bonding or welding:** In this case bonding is obtained by heating the region above melting temperature of thermoplastic matrix under applied pressure (Ageorges et al. 2001).

Joining by welding can provide a significant step in the further exploitation of thermoplastic composites in aerospace sector. In fact the welded joining of thermoplastic composites are often characterized by outstanding mechanical performance (high single lap shear strength and resistance to peel stress). Moreover the welding techniques can be easily automated, thus allowing the development of joining procedures characterized by fast production rate.

#### ***4.1 Fusion Bonding or Welding***

In Fusion bonding or welding the property of thermoplastics to be melted and reformed is used to create a joining. There are different welding techniques, depending on the way to apply heat for matrix melting:

- **Frictional heating** (*spin welding, vibration welding, ultrasonic welding*): In this case matrix heating is obtained using the friction between the two adherends. In frictional heating the motion of the substrates may cause deterioration of the microstructure, such as fiber breaking. Moreover, there is a limit with this heating principle regarding the area that can be welded. Ultrasonic welding seems to be less affected by these disadvantages, making this technique of interest for aerospace sector.
- **Hot plate welding:** In this technique the heating device can be a hot plate that has to be removed before welding of adherends. This technique has limitations on size of the component that can be welded since the whole joining surface must be heated in a single step.

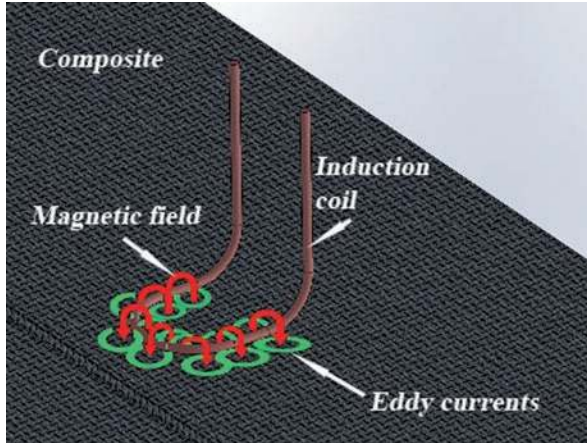


Fig. 4.18 Working principle of induction welding

- **Resistance welding:** In this technique the matrix is heated thanks to the Joule heat produced by a resistance placed between the adherend surfaces. The limits of resistance welding are that the resistance remains embedded into the joining (causing a decrease of mechanical performance) and the possible current leakages for carbon composites.
- **Induction welding:** In this process the material is heated by means of a high frequency alternate magnetic field (from 100 kHz to 1 MHz for plastic materials) generated by an induction coil. The magnetic field produces eddy currents within the conductive composite material, which heat the material due to Joule effect (Fig. 4.18). Joining consolidation is usually obtained thanks to the pressure applied with a consolidation cylinder. Among the materials than can be welded with this technique there are carbon-reinforced composites, but also glass-reinforced composites in which conductive layers were embedded near the joining surface (like, for example, metallic meshes or carbon fabric layers).

In this chapter special focus will be given to Induction Welding Technology, since this technique has a high potential of exploitation for different aerospace applications.

## 4.2 Induction Welding (IW)

Induction welding is one of the most promising techniques to join thermoplastic composites (Rudolf et al. 2000; Ahmed et al. 2006), for different reasons:

- High mechanical properties of the joining, for both static and dynamic loads: Single lap shear strengths between 30 and 50 MPa can be achieved with advanced

thermoplastic composites (like PEEK, PEKK, PPS and PEI carbon composites).

- High reliability and process control: Induction heat transfer phenomenon is one of the most efficient in the nature, so it is possible to heat the material fast and with high precision.
- Possibility of full automation of the welding process: There are different examples of Induction Welding equipment able to carry out automated continuous welding of complex geometries thanks to the use of robotic arms (Pappadà et al. 2015).

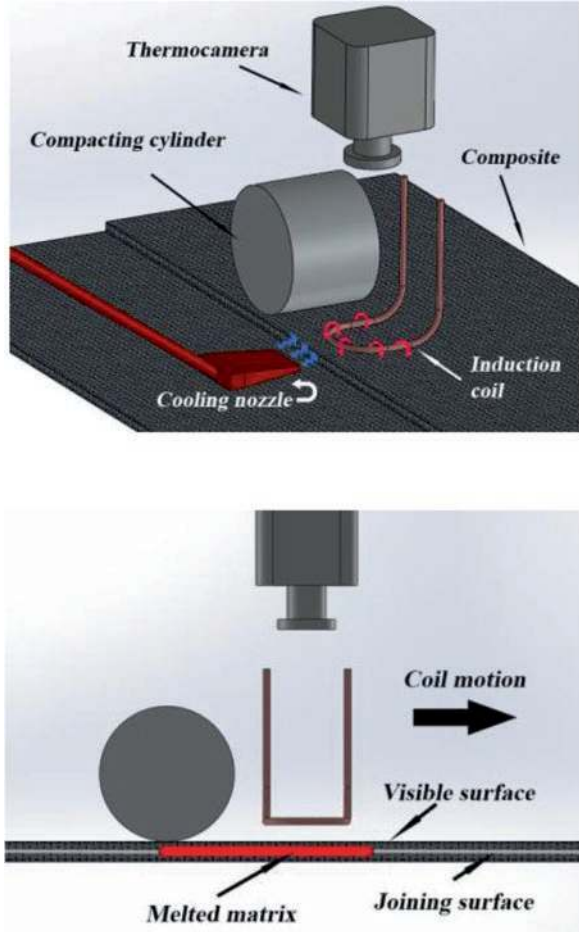
The main disadvantage of the induction welding of thermoplastic composite is related to the difficulty to optimize the temperature distribution through the thickness of the joining, avoiding, for example, too high temperatures on the surface directly exposed to the induction coil (the electromagnetic field decreases with the square of the distance, so in traditional Induction Heating systems the temperature under the induction coil is higher than the temperature in the joining interface) and near the edges of the joining (for the currents concentration due to the “point effect”). Anyway different approaches were developed to avoid this effect, like the use of metallic mesh in the joining interface for heat concentration, the use of low-melting film in the surface to be welded, or the introduction of cooling systems to decrease the temperature under the induction coil. In Fig. 4.19 the working principle of the induction welding machine developed by CETMA is described (Pappadà et al. 2015; Patent EP3017931A1 2014). In addition to the induction coil for material heating one cooling nozzle is introduced, to direct an air flux to cool the composite on its surface or near the edges, in order to optimize the thermal gradient during the welding. The cooling is adjusted by software, and the temperature on the upper surface of the joining is controlled by means of a thermo-camera. This cooling system makes possible to melt the composite matrix just near the joining surface, avoiding the melting of the same matrix in the upper surface (Fig. 4.19b) and its excessive heating near the borders.

The key parameters to understand the potential of Induction Welding techniques are reported below:

- Time to heat a carbon composite panel from room temperature up to 400 °C: 3–5 s
- Maximum welding velocities for common aerospace structures (skin and stringer for example): 2–5 mm/s
- Maximum working power: 3.3 kWh
- Maximum single-lap shear strength: 30–50 MPa depending on thermoplastic matrix
- Cost of a fully automated induction welding equipment (induction head equipped on a robotic arm): 200–300 k€

It is important to consider that since in the Induction Welding joining can be obtained thanks to a consolidation cylinder, the cost of the tools required for welding are low, since they are required just to keep subcomponents in the right position (tools to apply pressure during welding are not necessary).

**Fig. 4.19** Working scheme of the induction welding process in the induction welding technology developed by CETMA



The most important aerospace companies are currently carrying out research and development activities on different applications regarding Induction Welding, from TRL3 up to TRL9. Different approaches are investigated to optimize the thermal gradient within joining thickness during welding, in order to maximize performance and minimize defects (like voids and delaminations). Regardless of the followed approach, the fundamental aim is always the same: to develop reliable and efficient Induction Welding procedures, able to ensure high performance for welding of most used aerospace structures. According to the opinion of the author, this fundamental step for thermoplastic composites could be achieved through a realistic finite element modeling of the continuous welding process, and thanks to the definition of “Design for Assembling” procedures, able to optimize the design of the components to be welded, thus facilitating the development of the joining process.

## 5 Thermoplastics and Thermosets Recycling

The recycling of composite materials is a very important topic, which thanks to the potential of thermoplastics, has become even more attractive in the last decades. Nowadays, the majority of waste components produced with thermosetting resins are sent to landfill. This is an unsatisfactory solution from the point of view of the environmental, legislative, resource management and potential economic opportunity, linked to the recycling of waste. Taking into account that companies involved in production of composites for aerospace sector must be compliant with Aerospace Standard 9100, which includes also specific requirements of quality and safety aspects. It is evident that for an actual recycling there is a need to carry out suitable testing campaign useful to rewrite standards for products for their acceptance with recycled content. From the technical point of view, the thermoset-based composites have to face the problem of recycling cross-linked resins, which have been transformed in a nonreversible way. Consequently recycling requires dissolution of the matrix by high-temperature acids or thermal degradation of the resin, and only the fibers can be (partially) recovered. In case of thermoplastic-based composites recycling is much simpler, because the resin can be remelted by simple heating. Consequently the only problem to be solved is to transform the produced part, at the end of life, to obtain a different product. A possible recycling is performed by grinding the long fiber-reinforced aeronautical part to obtain pellets, which are composed by thermoplastic resin with short fiber reinforcement. Pellets can be used to produce new items by processing techniques suitable for plastic pellets (e.g., injection molding or thermoforming) just using the right temperature, but basically without major concerns. By the work environment point of view thermoset give some problem due to the presence of reacting components (e.g., epoxy groups, amine, and catalyzers) that can give health damages if they come in contact with the workers. Instead thermoplastics are already completely polymerized, thus no unreacted component is contained within them and handling of them is much safer. Furthermore, thermosetting processing requires a consolidated and cross-linking reaction, which is strongly exothermic and related to the chemo-rheology of the resin. Therefore, a very long process is needed, requiring slow heating (about 2 °C/min) and a long isothermal curing (generally about 2 h); this process is very demanding for energy consumption and, consequently, for CO<sub>2</sub> generation. Instead for thermoplastic heating is only required to make the part workable (no limits for heating rate) and the holding period is only needed for consolidation, and a short time is sufficient.

## 6 Conclusions and Future Perspectives

This chapter is aimed to give an overview on the recent advancements and applications in thermoplastic composites, focusing on the reasons why for aerospace sector, they are increasingly representing a more viable option for structural components.



Starting from thermoplastics polymer structure and difference with respect to thermoset-based composites, the most promising automated and continuous out-of-autoclave manufacturing concepts and processes, including assembling methods, have been deepened and some test cases have been illustrated. At last, an overview on different recycling concepts related to thermoplastics and thermoset composites has been provided. Current barriers to a wide application of thermoplastic composites can be overcome by the responding to the following needs:

- Development of affordable automated and fast processes to manufacture thermoplastic complex structures (aerospace sector)
- Development of efficient joining thermoplastic composite structures

At European level, Airbus in parallel with a variety of national aerospace consortia/programs are supporting the thermoplastic development roadmap. These activities are possible also thanks to the public–private partnership programs part of the Clean Sky, the largest European research program funded by EU's Horizon 2020 program (2014–2021) aimed to develop innovative, cutting-edge technology aimed at reducing CO<sub>2</sub>, gas emissions and noise levels for the next generation of aircrafts. As previously stated one of the problems affecting a larger diffusion of thermoplastic-based composites for aeronautical applications is the need to process the material at very high temperature (above  $T_m$  for semicrystalline thermoplastics); these needs cause the increase of processing costs, both in the phase of prepreg and part fabrication. Furthermore, also cooling rate must be controlled to be in a window (e.g., for PEEK a cooling rate between 10 and 300 °C/min) (Kenny et al. 1989). A possible evolution aimed to solve this problem is the utilization of thermoplastics with lower melting temperature and more forgiving cooling rate requirements, as PEKK (Poly-Ether-Ketone-Ketone), material with a molecular structure very close to the PEEK one (except than the unit sequence is modified). PEKK shows some modification like the ones previously envisaged.  $T_m$  of PEKK is 337 °C, the one of PEEK is 343 °C;  $T_g$  of PEKK 160 °C is higher than the one of PEEK 145 °C. In general these slight changes can help, but the changes are minor. Major advancements are expected by more radical changes, like the utilization of uncured polymers which develop chains without cross-links (“curing” thermoplastic).

Another possible solution is the use of hybrid semicrystalline/amorphous polymers. Based on this, a new concept was developed (Preimpregnated materials with semi-crystalline matrix and amorphous surface layers 2011) to keep the advantages of working on the amorphous state without negative effects on crystallinity: the amorphous-semicrystalline thermoplastic matrix prepreg. This is obtained by making a sandwich with a PEEK matrix carbon prepreg (Solvay APC-2) between two amorphous films, and consolidating it in a process which gives full consolidation and controlled cooling rate, in order to obtain the right PEEK crystallinity (both amount and morphology). The good integration of the two abovementioned materials is obtained through a transition zone with different percentages of the two materials, progressively ranging from 100–0% to 0–100%, which is possible due to the capability of the two materials to make blends (Maffezzoli et al. 1989). A multinational research program was activated with the aim to develop fuselage

thermoplastic composite demonstrators by means of hybrid prepreg manufacturing process; skin panel fabrication by Automated Fiber Placement In Situ Consolidation; stringers production by Continuous Compression Moulding; final aeronautical structure assembly by Induction Welding. This research project named NHYTE (New Hybrid Thermoplastic Composite Aerostructures manufactured by Out of Autoclave Continuous Automated Technologies) is in progress, supported by the European Commission (Barile et al. 2018; NHYTE Project [n.d.](#)).

With regard to future perspectives, a fabrication manufacturing process known as “Additive Manufacturing” or “3D Printing” deserves mention, by which digital 3D design data is used to build up a component in layers by depositing material. There are different additive manufacturing processes, but they have as common characteristics the capability to produce complex shapes by addition of melt layers; consequently this technique is used for metals and for thermoplastic polymers. The points that need to be improved are productivity, in terms of mass per time, and maximum size of the producible parts.

The thermoplastic parts can be produced both by amorphous and semicrystalline resins; in case of semicrystalline the cooling rate issue and its effect on final crystallinity must be considered. Continuous fiber composites cannot be processed by additive manufacturing; discontinuous fiber composite is under investigation, as well as the utilization of different resins, including the recycled ones. Injection molding is a relatively old technique; additive manufacturing is a new technique being developed, but both produce small and complex shapes. The main difference between these two techniques is in the tooling: injection molding requires expensive female close mold tools, with injection path and demolding pins, and additive manufacturing requires very simple tools or no tool at all. Production rate is presently much higher for injection molding, but additive manufacturing productivity could be improved in the future. Both for injection molding and additive manufacturing cannot be used continuous fiber composites. For injection molding a technique has been invented based on insertion of a continuous fiber composite part inside the injection molding shape, and then injecting the short fiber reinforced resin inside the mold. This technique is called “overmolding,” and allows to combine the fabrication of complex shapes with the partial utilization of continuous fibers. A similar technique (additive molding on continuous formed consolidate parts) can be considered also for additive manufacturing. In conclusion, the continue research on thermoplastics will be the key point for the development of the next-generation transportation systems only if automated, fast, and reliable manufacturing methods will be improved according to the chemical of the materials and the specific application field.

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