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Chapter 10

Advanced Composite Materials by Resin Transfer Molding for Aerospace Applications

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Additional information is available at the end of the chapter

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1. Introduction

Competitiveness drives the aerospace industries to investigate new technology solutions to address market pressure and high-tech demands. The global objective is to reduce to half the amount of fuel by 2020 and at least 70% less by 2025 with respect to the Boeing 777, one of the most efficient aircraft, which is made entirely of carbon fiber. The weight saving to increase payload and the reductions of the cost/time of the production cycle are imperative targets. For these reasons, aerospace companies, which are traditionally based on the use of metal alloys, have been focusing for past decade on composite materials. The main advantages of composites with respect to metals, that are resistance to corrosion and fatigue and high performance/weight ratios, are a set of qualities for winning the current and future aerospace applications. Obviously, this is possible only through the development of economically competitive technologies.

The Resin Transfer Molding (RTM) is one of the most promising technology available today. RTM is capable of making large complex three-dimensional part with high mechanical performance, tight dimensional tolerance and high surface finish. A good design by RTM leads to fabricate three-dimensional near-net-shape complex parts, offering production of cost-effective structural parts in medium-volume quantities using low cost tooling. In addition to these advantages, the problems of the joints, typical of the metal structures, can be eliminated by integration of inserts.

The final performances of a composite depend not only on the choice of the matrix and the fiber but also on the manufacturing process by which they are made. Since the starting of the composite life, the presence of imperfections due to manufacturing must be considered. Such imperfections can be already damage for the manufactured composite piece or lead to the damage quickly. The damage for composites can be defined as a change in the microstructure of the material that causes deterioration in the structural behavior of the
The damage in a composite structure can occur at the level of fibers and matrix as well.

The most common damage to the fibers is the interruption of their continuity. For example, fibers that are subject to load tend to align again inducing states of compression and tension on the matrix. These states may cause, in addition to a local decrease of the properties of the lamina, the breaking of the fibers themselves and the gap between fiber and matrix. In the assumption of an optimal stratification, fiber misalignment is caused during the manufacturing process by a bad balance of the process parameters that leads to the deformation of the fiber bundles. The fibers may also be distributed unevenly in the volume of the composite, and this generates intra-laminar shear stresses under operating conditions.

In the matrix, the damage is essentially correlated to the presence of porosity. The formation of microvoids between fibers and dry spots are potential starting points for propagating cracks or delaminations. The response of the component to delamination depends not only on the compatibility and surface tension between fibers and matrix but mainly on the compaction, and therefore also on the impregnation phases during the manufacturing process.

Comparing RTM with traditional manufacturing process applied in the aerospace industries (i.e. autoclave), the RTM technique results in a suitable alternative to the prepreg approach permitting high finish quality and controlled fiber directions. RTM reduces voids compared to hand lay-up so increasing component mechanical properties. In addition, component design by RTM process can compete with metal one when prepreg cannot be applied to manufacture a product. Hand lay-up requires a low initial investment, but it becomes more expensive (cost per product) than the other techniques due to recurring costs associated with direct labor and material waste. Compared with compression molding (CM), RTM requests lower tooling costs because of the absence of a press system to compact the preform. RTM seems to meet both low cost/high volume requirements of the automotive industry (500 to 50,000 parts per year) and low number/high performances (50 to 500 part per year) of the aerospace industry. In fact, RTM can guarantee the demanded performances for the aeronautical production: reduction of the mass, increase of the operating life, aimed design, reduction of the production times. Today an increasingly number of parts realized using RTM is observed due to the development of new resins and the preforming technology. In addition, such technique is suitable, with only small adjustments, for the realization of large, complex and thick-walled structures for use in infrastructures and military applications.

2. Overview of RTM process

The RTM is a process with a rigid closed mold. Figure 1 summarizes the main steps for a simple case. The lamination sequence (preform) is draped in a half mold, then the mold is closed and the preform compacted. After that, the resin is injected using a positive gradient pressure through the gate points replacing the air entrapped within the preform. Usually,
vacuum is applied at dedicated vents in order to favor the air escape from the mold. When the resin reaches the vents, the gates are clamped and the preform is impregnated. At this point, the cure phase is considered to start. Finally, the mold is opened and the part removed. Especially for aerospace structures, an additional free-mold post-curing phase can be necessary in order to guarantee the polymerization of the matrix and release the internal thermal stress.

The closing mold step is characterized by the compaction of the fiber reinforcement, which permits to reach the desired thickness and design fiber volume fraction. The compaction changes the microstructure and the dimensions of the preform, producing large deformations and nonlinear viscoelastic effects. These effects are accompanied by a change in energy within the material, which causes the residual stresses due to the viscoelastic behavior of the fibers. However, during the impregnation phase a release of stress, probably due to the balance, occurs.

The injection phase must guarantee the complete impregnation of the preform: a bad impregnation of the fibers results in dry spot areas with missing adhesion between the layers, which makes the surface rough and irregular. If partial impregnation occurs in the proximity of a connecting zone among elements, it can cause a bad integration with a consequent loss of mechanical properties.

**MAIN STEPS OF RTM PROCESS**

![Diagram of RTM process]

Figure 1. Sequence of the main steps of RTM process. From left to right: realization of the preform, deposition and draping of the preform in the mold cavity, closing mold, injection phase and curing phase; after the curing phase, the mold is opened and manufactured element is demolded.

**2.1. Process parameters**

The RTM process is governed by variables and parameters that are dependent on each other. Their combination affects the process and the quality of the finished product. Consequently, they need to be carefully determined. The most important parameters, which can not be neglected in the design, are pressure, temperature, viscosity, permeability, volume fraction, and filling time of the process. There are also a multitude of parameters that must be considered independently, such as the angle of attack of the nozzle, the orientation of the fibers, the paths of flow and shear rates, the stratification. In fact, the resin tends to flow more quickly in the fiber direction, thus the flow dynamic depends mainly on the type of fabric used and the number of overlapped layers. Sometimes it may be necessary to have a certain number of skins, not for structural reasons, but to obtain a homogeneous
distribution of the resin. The thickness of the part to be manufactured can also affect the flow progress and the impregnation of the fibers, causing a high percentage of voids and dry spots. The thickness becomes a critical design constraint especially in the case of the inclusion of reinforcements and ribs.

The injection pressure determines the injection velocity of the resin into the mold, the hydraulic pressure and the holding and closing forces of the mold. Consequently, the injection velocity defines the filling time, which should not be too short to ensure an adequate impregnation of the fibers and, at the same time, the filling must be such as to avoid the risk of incurring in premature gelation of the resin. The injection pressure adjusts the distribution of the resin on the preform, which affects the formation of air voids in the matrix, the appearance surface and the mechanical properties of the finished product. Another phenomenon in which this parameter is relevant, together with the viscosity, is the so-called "fiber wash", i.e. the movement of the reinforcement inside the mold during the injection phase. In this case, the surface treatment of the fibers and especially the choice of the binder play a fundamental role. If the binder dissolves too quickly in contact with the resin, then fibers under the injection pressure can move freely.

The temperature is an extremely important process parameter and it is strictly related to the injection pressure and the viscosity of the resin. When the temperature increases, the filling time decreases and the working pressures are lower. When the temperature is low, the viscosity of the resin increases and it is necessary to increase the pressure to ensure the transfer of the resin itself.

3. Preform technology

As previously mentioned, the diffusion of RTM process in the aerospace industries is strictly related to the possibility of building net-shape 3D-complex structures. This possibility is given by the development of the preform technology. Preform is prepared separately and constitutes the skeleton of the final product, greatly simplifying the molding operations and reducing the time and cost of processing. When the production volumes is medium-high, an industrialized preform permits to amortize in a relatively short time the cost of equipment, even in a limited production, as required in the aerospace industry.

The textile methods are numerous. The choice of one method over the other depends on several factors: the processability, the feasibility of the geometry, the desired mechanical properties of the molded part, and the cost of production. Obviously, the preform affects strongly the performance required by the final application. The choice of the architecture of the fiber reinforcement depends on the required performance of the composite structure and the characteristics related to the process, such as permeability, compressibility and drape.

The preforms are formed weaving yarns or rovings. The terms used for the fiber comes from the textile tradition: a single fiber is a filament; a set of fibers produced simultaneously is called strand. Several parallel strands can be rolled up like a ribbon, called "roving" or "tows", or twisted and united as a strand, which is named "yarn". Generally, the yarns are
typically not too complex because the excessive torque reduces the possibility of penetration of the resin in the cavities. Besides, a slight twist of the yarn compacts and raises the formation of composites with high fiber content.

Strands and yarns can be processed as woven roving and cloth. The first type uses effectively the resistance of the fibers, but it can produce composites with high resin content as the roving is not compacted. Woven roving is used to rapidly produce thick composites, because resin can fill them easily. Cloth is slightly less resistant than the roving because of the slight damage that is produced by rolling the fibers and also due to the twisting of the fibers themselves. The cloth is well impregnated from the resin and, as it is compact, high fiber content is obtained.

The weaving process can produce a wide variety of forms. The ratio between the number of filaments in the transverse direction (warp) and the number of filaments in the longitudinal direction (weft) may vary from nearly pure unidirectional, in which the number of filaments of the weft is the minimum required to hold together the fabric, up to the case of equal number of filaments in the warp and weft. In addition, there are hybrid fabrics with carbon fibers in the warp and glass fibers as weft. The weaving method influences the properties of the composite. In flat woven, warp and weft overlap in an alternating manner. In particular, weaving the warp passes through a number of fibers of the weft. These fabrics are better suitable for complex shapes, and their strength and stiffness are slightly higher because the fibers are straight on average. There are also more complex forms of weaving, with an angle of intersection fibers different from 90°. These methods are used to produce biaxial and multi-axial weaves. The complex fiber architectures can be obtained with the weaving method by interlacing and knitting the fibers along the three spatial directions; an example is shown in Figure 2. Different bi-axial layers can be also stitched. The stitching method consists on darning the layers with fibers (Figure 2). These are automated techniques that realize complex shapes and 3-D junctions in place of bolts and rivets. From the mechanical point of view, these methods can increase the resistance to fracture of the structure and reduce the progress of cracks by adopting high strength fibers in the z-direction. On the other hand, the stitching seams can produce local defects induced in the preform as a result of penetration of the wire and the needle. Further, the robotic system is very expensive and sometimes damage due to misalignment of fibers can occur.

The choice of the fiber reinforcement is carried out according to the mechanical requirements of the manufactured component. The types of fibers that can be used are many and with variable characteristics. During machining, the preform can be damaged, particularly when using glass fibers that are fragile and have a high friction coefficient (about 1 in the contact glass on glass). For this reason, immediately after production, the fibers are protected with a coating, "finish" or "size", which performs several functions: it acts as a binder to hold the fibers together, as a lubricant to reduce the coefficient of friction between fibers, and it improves the wettability of the fiber and the adhesion of the resin. Purpose of sizing is also to protect the fibers from the environment, mainly by moisture. These protective sizes are specific to each type of resin, since they must be dissolved from the resin itself. In the case of carbon fibers, the epoxy resin is also used as a protective finish.
Braiding method is another interesting textile process for aerospace structures. The braiding has high level of conformability to complex shapes and it is especially suitable for conical and cylindrical geometry. It regards 2D and 3D preforms. The high level of process automation reduces scrap and labour costs. More advantages of the braiding technique compared to standard tape and fabrics are strength in third dimension, improving fatigue resistance, more efficient distribution of mechanical stresses and the possibility to consider inserts during the preform realization. Despite of these characteristics, the braiding process requires the use of a mandrel and high initial cost for the preform engineering.

An important target of any textile process is obtaining the desired fiber volume fraction that is given by the design. It is very difficult to determine precisely the volume of reinforcement that must match the structural design with the manufacturing aspects. If the fibers are too compacted or their content is excessive, there is no sufficient space for the passage of the resin and the filling time becomes longer. Generally, the optimum volume of fibers is 60% of the product. An increase with respect to this value may cause a bad distribution of the resin and a dramatic drop of the mechanical properties of the manufactured component.

![Figure 2. Examples of 3D weaving and stitching textile processes.](image-url)
4. Resins in aerospace composites

Thermosetting resins are the matrix used in RTM because of their low viscosity during the process. They are very rigid materials consisting of cross-linked polymers in which the motion of polymer chains is strongly limited by the high number of existing crosslinks. During the polymerization phase, thermosetting undergoes irreversible chemical change. The selection of a reactive thermosetting resin for RTM application forces to deal with a large number of choices of chemical engineering, mainly due to a strong relationship between the chemistry and process engineering. In fact, the process parameters, such as temperature and pressure, cannot be selected without considering the chemistry of the resin to be used. Factors to take in consideration for a RTM system can be divided into two broad categories: processing and performance. Initial viscosity and molding life are function of the temperature, and they determine the operational temperature range of a process. The molding time is a function of the rate at which the reaction occurs between the resin and the curing agent and the rate is directly proportional to the temperature. The viscosity depends on the chemical-physical characteristics of the matrix. Viscosity may change over time because of both temperature variations and as consequence of chemical reactions that occur in the liquid state. The knowledge of the rheological behavior of systems is essential for a proper setting of process parameters. In fact, the values of the viscosity during the phases of the process must be such as to guarantee both the simultaneous removal of dissolved gases and moisture entrapped in the matrix, and the compaction of the fibers, before reaching the gel point. However, the viscosity of the resin must not be too high, especially in the case in which the fiber volume fraction is higher than 40-50%.

Within thermosetting resins, several classes are suitable for aerospace applications of RTM, such as epoxy, phenolic, cyanate and bismaleimide. Phenolic resins are produced from the reaction of formaldehyde with phenol to give the condensation products. The reaction is always conducted in the presence of catalysts: these can be both acid and bases and their choice has a determinant role, together with the molar ratio of the reactants, and the nature of the reaction products. There are mainly two types of phenolic resins: resole and the novalacs.
The base resins and formulated systems are available from Borden and Georgia Pacific. Approximately 91,000 tons of molded materials and phenolic are produced every year, but only a relatively small amount is used in composites for structural applications. A large number of industrial applications of phenolic resins are based on the excellent adhesive properties and strength. Especially when combined with appropriate reinforcements, phenolic resins have good chemical and thermal resistance, good dielectric strength and good dimensional stability over a wide range of temperatures. Materials produced with these resins have a very low flammability, are very resistant to creep, have low moisture absorption and have a remarkable resistance to degradation from a variety of lubricating fluids. Their low viscosity and the high high char yield of these materials are used in many structures forming composite carbon/carbon. Phenolic resins are polymers that generate volatiles (mostly aqueous vapor) during the treatment phase. The volatile substances have a strong impact on the processability of these composite materials resulting in structural voids where the content can have a dramatic effect on the mechanical properties of the part. In addition, these polymers are generally brittle, characterized by a low elongation and a low tensile strength. Despite the relatively poor mechanical behavior and the difficulty of processability, these resins are becoming more applied in the structural field due to the low flammability and low smoke production. The only resins comparable to the phenolic for the properties of high fire resistance and low smoke generation are the bismaleimide resins, but they are at least an order of magnitude more expensive. The nature of the RTM process (closed molding) makes it difficult the implementation of this method due to the development of volatile substances typical of phenol in the curing phase. In order to overcome this problem clever methods have been developed. A particularly interesting approach is the use of a catalyst that allows gelation of the resin at temperatures below 100 °C. So if the water is formed during the RTM process, but the temperature is maintained below the boiling point (100 °C), the water will remain in the liquid phase and will act as a plasticizer. If the temperature increases above 100 °C in the initial stage of gelation in the curing phase, the water will evaporate and will leave the part producing a massive degradation. A high temperature cure achieved with a ramp rather slow through the 100 °C will lead the water out from the gelled structure and the cross-linking of the polymer occurs without degradation of the piece. Another issue to consider when phenolic resins are used in RTM is the acidic nature of many catalysts, which can cause corrosion in some materials for molds.

Epoxy resins are well known in the production of composite aerospace materials. The vast variety of epoxy and cure agents makes these systems very versatile in terms of manufacturing process and obtainable physical properties. Although in the last twenty years, a large innovative work in developing new formulations of epoxy resins was done, only a limited marketing of new resins was realized. When liquid epoxy resins such as DGBA and DGBF are used in RTM, they are usually part of a two-component systems. In this case the selection of the curing agent is really important. In the polyester and vinyl ester resins, the catalysts alter the cure time but do not have substantial effect on the viscosity and on the final properties of the polymer. In an epoxy system, the selection of the curing agent is crucial because it determines the thermal and mechanical properties of the matrix and defines the dependence of viscosity from the temperature, thus controlling the processability of the system. Epoxy resins
polymerize with many materials such as polyamines, polyamides, phenol-formaldehyde, urea-formaldehyde, and acid anhydrides. The reactions taking place can be coupling or condensation reactions. DETDA is a liquid aromatic amine which is widely accepted as the primary hardener in many RTM formulations. DETDA is liquid at room temperature and provides good processability in RTM, both in a single-component system and bi-component one. The slow curing of epoxy system with DETDA allows these systems to be processed in a wide temperature range. The inclusion of a catalyst in the formulation of a DETDA significantly increases or decreases the reaction rate and lowers the cure temperature. The DETDA produces a polymer with high glass transition temperature (generally $T_g > 177 \, ^\circ C$) when fully cured. The Young's modulus is usually less than 3.1 Gpa. Therefore, the use of a liquid aromatic amine such as DETDA provides an excellent processability with high glass transition temperature but with a very low value of the elastic modulus for space applications.

The aliphatic and cycloaliphatic amines are useful agents for treatment of many epoxy resins. These materials are almost all liquids with low viscosity (as DACH, IPDA, PACM, etc.) that are readily soluble in the formulation of epoxy resins. In these cases, the thermal and mechanical properties are inferior to those obtained with the aromatic amine. With these agents, the glass transition temperature is in the range 121 $^\circ C – 177 \, ^\circ C$, and the elastic modulus of the polymer is in the range 2.4 GPa – 3.1 GPa. Anhydrides were widely used for many years as agents for epoxy polymerization in applications of filament winding. This class of catalysts has not received much attention in RTM applications. The liquid nature of most of the commonly used anhydrides (MTHPA, NMA, etc.) and their good solubility with the epoxy indicate that they could be used in RTM monocomponent and bicomponent systems. The availability of a large range of hardeners for epoxy-anhydride makes them available for systems that can meet specific process requirements. Generally, the formed polymers have a glass transition temperature of approximately 140 $^\circ C – 150 \, ^\circ C$ and a modulus of 3.45 GPa, tensile elongation of 3% – 5% and moisture absorption of 1.5%. The curing phase with an anhydride is relatively complex. The mechanism of cure can have several important consequences, such as low moisture absorption and in many cases a high final operative temperature. When anhydrides are exposed to moisture, usually over a long period of storage in a humid environment, they form acids. These acidic components interact with the basic catalysts inhibiting the polymerization, lowering the values of the modulus and the glass transition temperature, and increasing the absorption of moisture. Epoxy systems that have the greatest success in the aerospace market are the mono-component ones. There are substantial reasons for the success of these systems, such as ease of use, quality control of both process and materials and the excellent thermal and mechanical properties. In general, epoxy resins combine incomparable properties of flexibility, adhesion and chemical resistance.

The cyanate resins have a relatively small niche in the production of composites. The very high glass transition temperature and the excellent mechanical properties of the polymer are the primary guides to their use. The relatively high cost of these systems (150 – 500 U.S. $ per kilogram) prevents the entry into any market that is not closely related to their required application. This resin is generally cured with the addition of heat and transition metals as catalysts [Co(III), Cu(II), etc.]. The gelation occurs at about 50% – 60% conversion, similarly to
the chemistry of an epoxy resin. The coefficient of thermal expansion is relatively small, about 50 ppm / °C. Cyanate resins are often formulated with epoxy resins or maleimides in order to modify the processability and properties of the resulting polymer. Generally, the formulations include salts of transition metals and phenolic species as catalysts. There are very few formulations originally formulated for RTM. Usually cyanate resins produce a polymer with high transition temperature, low moisture absorption, good mechanical properties and excellent electrical properties. The use of them in the structures and radomes is driven by the needs of a high transition temperature coupled with a low dielectric constant and low dissipation factor to prevent degradation by high energy radiation transmitted and received through the structure. Some formulations have a flat response for both dielectric characteristics and dissipation in a wide range of temperatures and wavelengths of electromagnetic radiation. For these reasons, cyanate resins are widely used in aerospace fields. In particular, composites made by carbon fibers and cyanate resins are used in satellites for very rigid structures, with a high transition temperature and low absorption of moisture that can withstand to repeated thermal cycles without failure due to internal stresses.

Bismaleimide resins (BMI) currently provide a market niche in the manufacture of composite structures, i.e., for those parts which require a very high glass transition temperature, good stability and thermal oxidation and low flammability. The relative high cost of BMI resins (44 – 260 U.S. $ per kg) limits the applications to advanced ones. The BMI are produced by a reaction of aromatic bi-amine precursor with maleic anhydride. The resulting resins are cured with heat and in general without additional catalysts. The unmodified BMI are fragile materials with failure strain less than 2%. In order to make these systems more resistant, modified mixtures with amines, monomers, vinyl or epoxy resins are formulated. The polymers formed in this way are reasonably resistant, but with the increasing of post-cure temperatures the fragility increases. A typical BMI resin is produced by the reaction of methylene dianiline (MDA) with maleic anhydride, using heat to remove the water produced and push the reaction to completion. A variety of process features can be obtained by changing the properties of the molecular backbone of BMI resin and adding co-reactants. The use of BMI is driven primarily by their exceptional performance at high temperatures, in particular by their ability to maintain mechanical properties at more than 149 °C to below the saturation moisture. The other characteristics that lead to the use of BMI are their good electrical capacity, long term stability to the thermal oxidation for higher temperatures up to 177 °C and the exceptional capacity to not generate smoke when exposed to high heat fluxes. These characteristics at high temperatures push to use these resins in the aerospace field, although at present their main application is the manufacture of electrical circuits in high temperatures. One disadvantage that seems to be common to all BMI resins is the very long time of cure. The desired properties at high temperatures are achieved only by using a high temperature post-cure. The high temperature cure and post-cure cycles lead to somewhat brittle polymers with a significant amount of residual stresses. The high temperatures of curing, accompanied by a significant shrinkage, often lead to the formation of microcracks. To minimize the problems of residual stress, slow processing rates especially during the cooling phase are used.
5. Effects of manufacturing on final product

It is well known that the manufacturing process influences the quality and therefore the performance of the product. For instance, a good surface finish quality plays an important role in the mechanism of composite degradation upon exposure to the operative environment. The surface finish prevents the penetration of elements, such as dust, that produce and enhance micro-cracks within the structure. As consequence, a loss of mechanical properties occurs. Figure 4 shows the stages of the process that influence the behavior of the material and summarizes the relationship among process, material behavior and final performance. The compaction and impregnation phases govern the imperfections due to voids and dry spots. Following sections of this paragraph describe the compaction and impregnation phases.

Figure 4. Relations between manufacturing phases and material behavior.

5.1. Compaction phase

The compaction phase is an important step that occurs in any production process for the manufacture of advanced composite structures. This phase usually takes place through the application of an external pressure, which produces a new arrangement of the fibers and changes of microstructure. In RTM, the compaction occurs when the mold is closed and the preform reaches the expected fiber volume fraction. Compaction produces large deformations and nonlinear viscoelastic effects in the preform. These effects are accompanied by a change in energy within the material. Some authors have modeled the phenomenon, introducing the function of free energy, but with limited success, because this hypothesis does not take into account permanent deformations of the preform. For instance, in case of fabric textile, when a multilayer preform is compacted, the fibers tend to be squashed with other fibers, producing interlayer packing or nesting phenomena depending on the deformation mode and the fibers architecture. If the compression load is removed, fibers tend to not return to their original position inside the preform.
Nesting is a phenomenon that produces irreversible mechanical and geometric changes of
the preform modifying the permeability and mechanical properties of the composite
material. In particular, the nesting influences the rigidity of the piece and even more the
resistance. In particular, the stiffness depends on the positioning of the layers. If the layer is
positioned out of phase without reducing the thickness (interlayer packing), there is no
change of the mechanical properties. On the other hand, if two layers are in phase, i.e. in
case of existing nesting, the difference of stiffness between the two configurations is reduced
approximately of 10-20%. Nesting creates the so-called bridging between layers preventing
delamination and increasing the resistance considerably, up to 10% for the layers out of
phase. Moreover, the compaction induces residual stresses within the material due to the
viscoelastic behavior of the fibers. The stress concentrations, also localized, can act as
nucleation of cracks after the curing phase. However, during the impregnation phase a
release of stress, probably due to the balance, occurs.

From a manufacturing point of view, it is evident that compaction changes the spatial
arrangement of the fiber bundles, modifying substantially the morphology of the porosity. These variations alter the permeability of the preform and so the impregnation phase.
Furthermore, compaction affects the adhesion between the layers, the material failure modes
and the magnitude interlaminar shear stress more than the impregnation.

5.2. Impregnation phase

In RTM applications, the defects induced by the resin flow such as voids and dry spots are
known as the biggest source of problems for production quality and reproducibility. The
damage of the matrix is essentially related to the presence of porosity. The formation of
micro-voids among fibers and dry spots are potential starting points for the propagation of
cracks and delamination.

The resin flow front faces two resistance levels through the preform: the resistance between
the fiber bundles and the resistance inside the fiber bundles. This means that the preforms
are characterized by two different permeabilities: the permeability between the fiber
bundles and the permeability inside the fiber bundles. These can be considered as a resin
flow in macro-scale and micro-scale. The type of flow scale determines the potential void
formation.

In the macro-scale, the formation of macro-voids can be formed when the air displaced from
the resin remains trapped, or when pressure is insufficient to overcome the resistance of the
preform or the viscosity is too high. In micro-scale, the porosity is given by micro-voids.
Figure 5 shows the mechanism of void formation in the fiber bundle: the flow front runs
around the tows and continues to impregnate the tow even after the passage of the front. If
the air is not evacuated at the beginning of the filling process, it remains trapped in the tow
and micro-voids occur at the center. Micro-voids formation can be also due to the
macroscopic pressure drop. The pressure drop produces an apparent change in the
permeability along the preform. The pressure drop can be explained by assuming the sink
effect: the fiber bundles act as fluid sinks. The concept is based on the dual scale porous media. The individual fibers of the bundle are separated by a distance much smaller than that existing between two bundles. Consequently, the resin flows more easily between the fiber bundles rather than inside one of them. Therefore the liquid continues to impregnate the bundle even when the front part of the flow has passed. This means that a part of the injected fluid penetrates in a package of fiber, rather than push forward the front face of the flow. The result is that the pressure profile is influenced by the relative flow rate of the resin inside a bundle relative to the flow between bundles.

Figure 5. Mechanism of void formation in the intra-bundle.

6. Modeling of RTM process

In the RTM process, the resin flow is usually modeled with the assumption of isothermal flow, ignoring the exothermic nature of the thermosetting resin. Basing on this hypothesis, the resin flow is described by the conservation equation of mass and momentum with the boundary conditions. In order to complete the equation system, the constitutive equations of the fluid must be considered. The following section concerns the continuity and momentum equations of a resin flow within a region consisting of fibers and porosity (Figure 6).

Figure 6. Control volume of porous media.

6.1. Flow model

The fiber reinforcement is a dual-scale porous material and to apply the conservation of momentum is an unrealistic approach. In fact, determining the pressure profile during the impregnation means to apply the Navier-Stocks equation for each channel of fibers network and solve the equation systems in their surroundings. This would allow to know the flow
rate and the pressure within each channel. Anyway, a simple piece of preform may have millions of channels. This approach is impractical in a real case. Furthermore we have no interest in knowing the exact value of the pressure during the manufacturing process, but we need to know the relationship between pressure drop and flow rate during the progress of the flow front in the preform at a macroscopic level. For these reasons, the resin flow front is modeled by the Darcy law, considering that Reynolds number relative to the pore size is $\leq 1$. Darcy equation (Eq. 1) describes the relationship between the flow and the pressure gradient that drives the flow through the porous medium, using the permeability parameter. This parameter, as discussed after, characterizes the fluidity of the resin through the average porosity of the fibers.

$$\mathbf{u} = -\frac{K \nabla P}{\mu}$$

where $\mathbf{u}$ is the speed mediated on the volume, $\mu$ is the viscosity of the fluid, $\nabla P$ is the pressure gradient and $K$ is the permeability tensor of the preform (according to the fact that fiber arrangement generally is anisotropic in porous medium).

Rewriting Eq. 1 in expanded form:

$$
\begin{pmatrix}
u_x \\
u_y \\
u_z
\end{pmatrix} = -\frac{1}{\mu} \begin{pmatrix} K_{xx} & K_{xy} & K_{xz} \\
K_{yx} & K_{yy} & K_{yz} \\
K_{zx} & K_{zy} & K_{zz}
\end{pmatrix} \begin{pmatrix} \frac{\partial P}{\partial x} \\
\frac{\partial P}{\partial y} \\
\frac{\partial P}{\partial z}
\end{pmatrix}
$$

In most RTM applications, the flow can be approximated in two-dimensional domain, since the dimensions in the plane are two or three orders of magnitude larger than the thickness. The approximation of a 3D geometry with a 2D one gives significant saves in terms of computational time and reduces the number of material parameters that need to be measured or calculated. This assumption can be considered valid since the values of permeability in the plane of the preform (in the plane in which the layers are stacked to form the thickness) are approximately of the same order of magnitude. Boundary conditions are imposed on three segments:

- injection gates
- free surface of the resin flow (i.e. on the flow front)
- walls of the mold.

The resin is injected into the mold at constant pressure or at constant flow rate, for which the gate conditions are

$$P = \text{constant} = P_{\text{inj}}$$

or

$$A \left( -\frac{K \nabla P}{\mu} \right) \cdot \mathbf{n} = -Q_{\text{inj}} = \text{constant}$$
Some injection systems allow to vary the values of pressure or flow rate during the same injection. In such case, $P_{inj}$ and $Q_{inj}$ must be considered as a function of the time. This change does not alter the technique of solution, because the problem is solved as a stationary one for each time.

The pressure at the resin front flow $P_{ff}$ can assume the value of the atmospheric and vacuum depends on the type of process, generally:

$$P_{ff} = P_{vent}$$

(5)

Inserts or multiple injection points can form several different resin flow fronts. When these fronts meet and they are not connected to a vent, the air moved by them remains trapped in the regions surrounded by the resin flow fronts. In this case, the boundary conditions on the front can be modified to consider that the pressure increases on the front due to the void pressure. The trapped air can be considered as an ideal gas for which the following relation applies:

$$\frac{P_{void} V_{void}}{\rho} = \text{constant}$$

(6)

However, this would require knowing a plot of the vacuum created during the impregnation of the fronts and calculate the volume at each instant of filling. In the real case, a part of this air is dissolved in the resin, reducing the pressure of the vacuum. In order to take into account this phenomenon, the system may include an equation of the resin diffusing at the mass-to-air interface and the diffusion coefficient has to be determined. But this is not a significant factor and researchers ignore this aspect and assume that the air will dissolve in the resin.

The boundary condition on the wall of the mold is the no-slip condition:

$$u_n = -\frac{1}{\mu} \left( K_{nn} \frac{\partial p}{\partial n} + K_{nt} \frac{\partial p}{\partial t} \right) = 0$$

(7)

where $n$ and $t$ are the directions normal and tangential to the mold wall.

The continuity equation of resin flow through fibrous arrangement can be determined assuming that:

- fibers are incompressible
- variations of resin velocity and stress-strain are small in the volume control
- process is quasi-steady
- body forces, such as weight, are negligible.

The derivation is quite similar to that for any fluid within a region, except for the expression of the resin density $\rho_b$ that is the mass of the resin divided the control volume containing both resin and fiber. In this case, $u$ is the interstitial resin velocity within the composite. Within a fibrous region partially filled is $\rho_b = \epsilon \rho_p$ where $\epsilon$ is the porosity of the control volume available to be occupied by the resin, $\epsilon = (\text{pore volume/volume control})$, which can be expressed as $\epsilon = 1 - V_f$, where $V_f$ is the volume fraction of the fiber. $S$ is the fraction of
saturation of a porous space filled by the resin, and \( \rho \) is the true density of the resin (mass of the resin/volume of resin). The pore volume within the volume control is completely empty when \( S = 0 \), and completely full when \( S = 1 \).

The mass balance for the volume control can be expressed as:

\[
\text{(net flow of mass)} = \text{(inflow)} - \text{(outflow)} - \text{(losses)}
\]  

(8)

The term of the losses is due to an internal volume \( V \) that absorbs fluid mass in a quantity equal to \( (x, y, z, t) \) per unit volume and unit time (i.e. the fiber volume). Dividing both sides by \( \Delta x, \Delta y \) and \( \Delta z \) and then doing the limit for \( \Delta x, \Delta y \) and \( \Delta z \rightarrow 0 \), the mass balance becomes:

\[
\frac{\partial (\varepsilon \rho)}{\partial t} + U \cdot \nabla (\varepsilon \rho) + (\varepsilon \rho) \nabla \cdot U = s
\]  

(9)

It is noted that even if the resin density \( \rho \) is constant, the porosity or saturation of the fibrous medium may not be constant within the whole region. Therefore, it cannot be zero everywhere.

7. Experimental determination of permeability

The permeability \( K \) is in general a symmetric tensor, which for an isotropic material, as random mat, is a scalar number. For a given stationary porous medium, it is necessary to know 6 scalar values \( K_{ij} \) to completely determine the tensor \( K \). If the selected directions of the reference system are along the principal directions of the preform, the matrix becomes diagonal. Therefore, choosing the coordinate system along the main axes of the preform, the principal values of the permeability can be measured. Then, not diagonal terms can be calculated using a coordinate transformation system.

\[
K = \begin{pmatrix}
K_{xx} & K_{xy} & K_{xz} \\
K_{yx} & K_{yy} & K_{yz} \\
K_{zx} & K_{zy} & K_{zz}
\end{pmatrix}
\]  

(10)

Permeability must be determined experimentally. There are basically two methods for this: radial flow and linear flow methods. The simplest way to determine the permeability is the use of 1D version of Darcy’s equation (linear method). For a 1D flow in the direction of the axis (assumed to be the x-axis) Darcy’s equation can be written as

\[
\frac{dx}{dt} = -\frac{K_x}{\Phi \mu} \frac{\partial P}{\partial x}
\]  

(11)

Where \( \Phi = 1 - V_i \) is the porosity of the material and \( V_i \) is the fiber volume fraction, \( K_x \) is the permeability in the \( x \) direction, \( \mu \) is the fluid viscosity and \( \frac{\partial P}{\partial x} \) is the pressure gradient between the injection point and the flow front. Since the injection pressure and the vent pressure are constant, we can transform the partial derivative in a finite difference, so we have:
Integrating between the position of the injection point \((x = 0)\) at time \(t = 0\) and the position of the flow front \((x = x)\) at time \(t = t\), the expression that allows to determine the permeability along a main direction of the preform is:

\[
\frac{dx}{dt} = -\frac{K_x \Delta P}{\mu x} \tag{12}
\]

The slope of the line is calculated by plotting the position of the flow front at different times; the pressure gradient is equal to the injection pressure. In the case of orthotropic laminates, the permeability has different values in the two principal directions, and then two experiments sets must be performed.

For typical composites, the thickness is on average 2 – 3 mm. As a consequence, the transverse permeability \(K_z\) is considered negligible. This means to assume that the resin flow advances uniformly on the two external surfaces (top and bottom) and that flow velocity is an average through the thickness. However, as the thickness increases, the gradient along \(z\) (i.e. the thickness) becomes greater, due to the fact that the resin flows quickly on the upper surface, while it continues to impregnate the lower levels. Advani et al. have developed a theoretical model to calculate an average effective permeability for flat rectangular preform with thickness \(H\) formed by \(n\) layers.

\[
K_x = \frac{\mu \text{slope of } x^2 f(t_i)}{2AP} \tag{13}
\]

\[
\bar{K} = \frac{1}{H} \sum_{j=1}^{n} \frac{K_j h_j}{l_j} \tag{14}
\]

\[
\bar{l} = \frac{1}{H} \sum_{j=1}^{n} \frac{l_j h_j}{n} \tag{15}
\]

where \(h_j\) is the thickness of the single layer, \(l_j\) is the length of the impregnation of the single layer in the injection direction, \(\bar{l}\) is the average length of impregnation in the entire laminate, \(K_j\) the permeability of the single layer.

8. Numerical analysis approach

The advantage of the numerical simulations is to help the process engineer to understand the behavior of the resin inside the mold, especially when the part geometry is complex and presents variations in permeability and fiber volume fraction. This knowledge improves the design tools and optimization of the injection scheme. General method is to use a simplified model of the resin flow reducing the problem from 3-D to 2-D and using a finite element approach and control volume (FE/CV) that does not require a re-meshing at each step. The geometry is discretized as a thin shell using triangular elements or rectangular ones. The material properties such as thickness, permeability and volume fraction of fibers can be
assigned individually to each element, such as the non-uniform material properties and thickness. A linear pressure profile is assumed among the nodes of an element:

$$
P^e = \sum_{i=1}^{n} N_i P_i$$  \hspace{0.5cm} (16)

where $n$ is the number of nodes of the element, $P^e$ is the pressure inside the element, and $P_i$ is the nodal pressure that is unknown and a function of interpolation. The Galerkin finite element method can be used to convert the governing equations to the partial derivatives in a system of algebraic equations that minimizes the error at the nodes. It can be expressed as a linear system of equations

$$[[S^*]][P] = [f]$$  \hspace{0.5cm} (17)

where $S$ is the stiffness matrix whose components are:

$$S_{ij} = \int_{\Omega} \left( \frac{k_{xx}}{\mu} \frac{\partial N_i}{\partial x} \frac{\partial N_j}{\partial x} + \frac{k_{yx}}{\mu} \frac{\partial N_i}{\partial y} \frac{\partial N_j}{\partial y} + \frac{k_{xy}}{\mu} \frac{\partial N_j}{\partial x} \frac{\partial N_i}{\partial y} + \frac{k_{yy}}{\mu} \frac{\partial N_j}{\partial y} \frac{\partial N_j}{\partial x} \right) d\Omega$$  \hspace{0.5cm} (18)

This set of equations can be solved in every moment during the filling process. The $i$-th vector component is the amount of mass generated per unit time at the $i$-th node. The pressures are calculated at nodes by solving the set of algebraic equations described by the system.

The next step is the determination of the resin flow front progress. The approach by the control volume consists of dividing the geometry under consideration into control volumes first, and then associating one of them to each node. The flow between two control volumes is calculated by multiplying the average speed for the area connected between two control volumes. For example, the equation of the flow associated with the node $i$ and the one associated to node $j$ is

$$q_{ij} = -\int_{s_{ij}} \left( \frac{n}{\mu} \cdot [K] \cdot \nabla P \right) ds$$  \hspace{0.5cm} (19)

where $s_{ij}$ is the boundary between two control volumes, $h$ is the thickness of the mold cavity at the contour and $n$ is the normal to the contour of the plane lying of the preform.

The nodal fill factor is used to track the movement of the flow front. This factor is associated with each node and represents the fraction of the volume occupied by the control fluid. The pressures are therefore determined only at the filled nodes and empty nodes are ignored. The nodes are considered partially filled close to the flow front. The progress of the flow front is considered at any instant of time $t$, updating the fill factor of the control volume by means of the flow between the connected nodes. Consequently, the amount of the masses is strictly taken into account. This technique can simulate the resin flow in thin cavities with high geometric complexity in 3-D. Usually the simulation process is assumed to be isothermal, because of the disadvantages in conducting non-isothermal numerical simulations. First of all, the computational efficiency and speed of convergence decrease drastically when the equations that govern the flow are solved simultaneously with the energy equation, especially with the increasing of the number of nodes. Research in the simplification of non-isothermal analysis reduced the time, but it still requires a CPU time of
two or three orders of magnitude higher than the resolution of isothermal problems. Moreover, to obtain the results non-isothermal analysis requires a large number of material parameters, such as dispersion coefficient, thermal conductivity and kinetics of resin cure. For high temperature resins, such as in case of aerospace application, a good compromise would be to fill the mold under isothermal conditions, and at least considering the kinetics of cure. From a numerical point of view, this solution is very efficient because diffusive and convective problems are not faced.

9. Aerospace applications

Developing a new composite product requires a synergy among different disciplines and sometimes entities, in other terms a Concurrent Engineering (CE) approach. This section shows some aerospace components manufactured by RTM process in CE method. The prototypes presented in the following sections are the results of years of collaboration between the School of Aerospace Engineering (currently DIAEE) of Sapienza University of Rome and Italian aerospace industries (AgustaWestland and Aermacchi).

9.1. Concurrent engineering approach

The need of high quality production, due to market pressure and high-tech demands of the aerospace field, drives to the faster and most effective design of the product. To this aim, engineers adopt new methods based on concurrent approaches to optimize the part before manufacturing the final piece. Especially from 1990s, with the explosion of the composite materials market in several industrial fields, involved people from industry and universities are working to determine the best way to apply the concurrent engineering as a systematic approach for product development. Some works are addressed to improve the management team and communication among members; others go toward building a virtual environment based on CAD systems.

The software systems for automation design tools are powerful. They can help any company to develop products of superior quality, faster and at lower cost. This implies clear advantages in terms of competitiveness. The empirical methods “test and fix” still dominate the field of molding technologies for composite materials, but this approach produces deleterious effects in terms of cost and time. Simulation studies lead to the complete optimization of the product before creating the prototype. The objective is to establish the optimal characteristics of the preform and tooling, optimizing the process parameters.

The targets that can be obtained with the use of a code are:

- reduction of design time
- reduction of the costs and consumption of materials
- optimization of the design parameters in relation to the process
- reduction of the times of modification and tuning of the molds
- integration between production process and optimization of the component with respect to constraint and loading given by specification.
About the entities involved in a project, concurrent-development teams typically exhibit the following characteristics:

- they include no more ten members
- members choose to serve on the team
- members serve from the beginning to the end of the project
- members participate in the team fulltime
- members report solely to the team leader and the leader reports to general management
- key functions - at least marketing, engineering, and manufacturing - are included in the team
- members are co-located within conversational distance of each other

The plan should give specific information about the qualification programs regarding all the entities involved in the RTM process: materials, process, tooling, tests.

Figure 7 shows the flowchart related to the CE approach set by the DIAEE of Sapienza University of Rome during the development of several RTM helicopter components. The flowchart highlights the structural and manufacturing couplings. Process parameters have been considered at the early stage of the design as a discrimination of the material selection. The resin flow behavior and the filling time of the mold were the process criteria. These parameters are directly dependent on the preform characteristics such as the textile structure, the laminate orientation and the fiber volume fraction. The preform characteristics are the first features that determine the mechanical performance of the component. In this sense, the optimized project is a compromise between structural and process optimizations and requires a concurrent engineering design and manufacturing.

### 9.2. Strake

The strake is a typical aerodynamic surface used in both supersonic and subsonic vehicles to improve the stability and/or reduce the drag. The thickness of the structure is around 2-3 mm and usually made in aluminum alloy. In this case, the selected resin was a mono-component benzoxazine polymer (Henkel Epsilon 99110). The recommended process conditions were typical infusion temperature at 90 °C and curing at 180 °C for 90 min. The fiber reinforcement used was a carbon textile HEXCEL G0926 5H Satin. The preforms were realized manually overlaying several layers adopting a predefined lamination sequences. The lamination sequences were determined by structural analysis. The injection equipment was a commercial Hyperject machine for mono-component polymers. The Hyperject injects the resin at constant pressure and it is provided with heated dispensing resin. The resin was loaded, degassed and heated inside the Hyperject. When both the temperatures of the resin and the mold were reached, the resin was pumped inside the mold until the resin came out from the vent. Molds were studied to be multi-components in order to make easily the demolding.
The numerical analysis was performed by commercial codes based on a finite element analysis-control volume (FEM-CV). Modelling of the flow was to allow investigating the resin impregnation process and strategically designing gates and vents and injection scheme in order to optimally fill the composite part without any dry spot. The analysis took into account the most relevant process parameters such as injection pressure and material features like the resin viscosity and permeability of the preform. The FEM-CV analysis was isothermal and provided detailed information about the pressure field, flow front patterns, and strategic injection scheme. The different choice of gate positions affects very much the filling time and the quality of the finished part. For these reasons, different injection schemes were considered. All simulations were performed using the resin viscosity value at the mold temperature.
Figure 8 shows the simulation result of the best injection scheme. In particular, the simulation represents the trend of the filling time. Figure 9 shows the prototype of the strake after demolding.

Figure 8. Injection scheme of the mold as determined by numerical simulation. On the top: filling time trend through the preform. On the bottom at left: carbon fiber reinforcement before closing mold.

Figure 9. Strake of Aermacchi nacelles after demolding.
9.3. Inboard flaperon

The aim of the work was to realize a primary structure of a helicopter by RTM process in order to compare its properties with those of the parts made with classical hand lay-up technique. The chosen component was the inboard flaperon of the BA609 (Bell Agusta), which is an aerodynamic control surface that presents different critical features. In fact, the inboard flaperon is a primary component so the reliability must be nearly absolute. Further, it is a large part and involved a complicated lamination sequence that can affect the manufacturing process. It is evident in Figure 10 the presence of three different lamination sequences that correspond to different permeability, which was determined experimentally.

![Finite element model of the inboard flaperon with a particular on a rib (red component on the right). The different colors individuate the different permeability values associated to the lamination sequence.](image)

<table>
<thead>
<tr>
<th>Lay-up Sequence</th>
<th>Permeability</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.124E-11 m²</td>
</tr>
<tr>
<td>2</td>
<td>0.5365E-11 m²</td>
</tr>
<tr>
<td>3</td>
<td>0.744E-11 m²</td>
</tr>
</tbody>
</table>

In the first scheme, the injection points were placed on the two leading edge borders of the skin in correspondence of the rib reinforces and the venting points on the trailing edge line. As shown in Figure 11, the total filling time given by this configuration is very long (more than 1.5 h) and comparable with the resin pot life limit.

A second scheme has been analyzed. In this case five injection points were placed in the middle of each skin side and between the rib reinforces. The venting points were located both on the trailing and leading edge lines. With this second scheme the filling time was considerably reduced down to 20 min in good agreement with the resin processability. From the results of the previous simulation it was noted that much time was needed to fill a very small area near the outbord side. For this reason a further optimization was carried out including a sixth injection point in this area. With this simple modification the total filling resulted to be less than half of the previous simulation (Figure 12).
Figure 11. Filling time with the first injection scheme and predicted resin flow rate during the injection with the first scheme.
Figure 12. Filling time trend for the injection scheme selected for the mold.

The dry fabric plies for the construction of the preform were cut with the GFM ultrasonic cutting machine using a multilayer method. In this way some areas of the preform were cut directly in the final lamination sequence allowing for considerable time saving in the preform assembly phase. The different plies and sequences were sticked together on a flat surface using an adequate agent (binder). It was used the same cut program and operative cycle of the original part. In Figure 13, the glass layers used to replace the syntactic foam can be seen. The use of the glass mat strongly improves the permeability of the preform allowing very much shorter injection times. Further, the syntactic foam was incompatible with the RTM process.

Figure 13. Particular of preform preparation. On the right: glass layers used to replace the syntactic foam of the original component.
9.4. Bracket

The bracket is a secondary element that is present in large numbers in a helicopter. Usually it is made by metal alloy. The aim to re-design the bracket in composite materials is to reduce the weight and time of production. In this case, a consistent saving can be reached adopting an automatic textile process, which avoids the long time due to the lamination. The stitching was the selected preform technique. Figure 15 shows the cut of the layers and the preform after the assembling. Figure 16 shows the realization of the component during the impregnation phase and the part demoded after the curing step. In this case, the use of RTM process permits to save 30% weight with respect to its equivalent in metal alloy and reduces drastically the production cycle.
Figure 15. Realization of the preform: cut and stitching.

Figure 16. Realization of the bracket by RTM process: resin injection (left), bracket demolded (right).

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