

École polytechnique de Louvain

Experimental analysis of mechanical characteristics of bio-sourced composites manufactured with Hand lay-up technique

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Abstract

Nowadays, the development of new and better performing materials is becoming essential in certain fields such as aeronautics, the automotive industry but also for medical applications. The use of composite materials is ever growing in these vastly. These combine interesting mechanical and thermal properties with a lower density, making them a good alternative to metals. However, most of the composite materials currently on the market are produced from a petro-sourced matrix and the development of these materials does not fit with the objectives of a more sustainable society. Therefore, new eco-friendly epoxy resins are being developed, as well as natural reinforcing fibres that would reduce the carbon footprint of the final product.

The aim of this work was to determine the static mechanical properties of a composite material based on the partially bio-sourced resin InfuGreen 810 reinforced with flax fibres. To do so, several laminates were first manufactured using the hand layup technique and curing cycles previously developed for InfuGreen 810. Different architectures of flax-based fabrics were used to allow a direct comparison between the final materials. In order to quantify the mechanical properties, several quasi-static tests were carried out on samples such as uniaxial tension, compression, three-points bending and in-plane shear.

These tests highlighted the bilinear behaviour of flax fibre at the composite scale, but also the impact of the manufacturing process on the material. The results generally show better properties but with lower reliability for the composite based on unidirectional reinforcements compared to bidirectional fabrics. Regarding the InfuGreen 810, a poor adhesion with the reinforcements is highly suspected. Moreover, a first comparative study with carbon-reinforced samples showed that the latter had significantly better mechanical performances than the flax-based composites.

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List of abbreviations

ϵ	Strain [-, %, $\mu\epsilon$]
γ_{xy}	Shear strain [-, %, $\mu\epsilon$]
σ	Stress [MPa]
τ_{xy}	Shear stress [MPa]
E	Young's modulus [GPa]
G	Shear Modulus [GPa]
DGEBA	Bisphenol A Diglydyl Ether
FFRP	Flax Fibers Reinforced Polymer
GFRP	Glass Fibers Reinforced Polymer
HV	Vickers Hardness
ILSS	Interlaminar Shear Strength
IPS	In-plane shear
NFRP	Natural Fibers Reinforced Polymer
PAN	Polyacrylonitrile
RT	Room Temperature
RTM	Resin transfer molding
SEM	Scanning Electron Microscope
UD	unidirectional fabric
UTS	Ultimate Tensile Stress
UTS	Ultimate Tensile Stress
V_f	Fiber volume fraction [%]

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Chapter 1

Introduction

Today's society has, in recent years, been increasingly concerned by climate change and is therefore actively seeking sustainable solutions to reduce the impact of human activity on Earth. One of the most studied lead for the industry are polymers and their derived products such as composites. Indeed some of them possess mechanical characteristics coveted in certain fields such as aeronautics and offer other interesting characteristics such as lightweight.

Unfortunately, these materials also have their weak points. In fact, the so-called epoxy resins currently used on an industrial scale as the matrix of these composites are often derived from petroleum and therefore can be considered "non eco-friendly" material. Moreover, the low recycling potential of these epoxy resins adds a consequent weight in the limitation of the use of this material. Regarding the reinforcement fibers, some environmental problems also arise, both in terms of processing and recycling. Today's challenge is therefore to find an alternative to these composites while keeping the properties of current ones.

The Macobio research project aims to find alternatives to these composites and to investigate on materials known as "biosourced". The studied resins are promoted as being greener and therefore could drastically reduce the ecological impact of manufacturing and using composite materials. Some reinforcements of natural origin also show a lot of potential but are still very limited in terms of mechanical and physico-chemical properties and therefore, at the time of writing, they cannot fully replace the current reinforcements as carbon or glass fibers.

The aim of this master thesis will be to characterize the mechanical properties of the biosourced resin InfuGreen 810 marketed by SicominTM in the presence of fibers, whether natural fibers as flax fibres but also synthetic fibers currently used. This work will also enable to highlight the possible limitations that may be encountered in the use of these materials with the processes currently used in industry.

To reach the research goal of this project, this work will be divided into several objectives.

- The first objective is the determination of the quasi-static mechanical properties of Infugreen 810 reinforced with flax fibers. The reinforcements will be found in the form of unidirectional or bidirectional fabrics.
- The second objective is to evaluate the impact of manufacturing parameters such as

fibre drying and degassing, on the mechanical properties of the final product. The used manufacturing technique and its limitations will be also analyzed.

- The last objective is the determination of the tensile properties of the reinforcements at the strand level and its influence on the composite properties.

As an outline, this work is divided into several chapters which together contribute to a more complete overall understanding of the project. First, a state of the art will be given, summarizing the essential aspects of composite materials and their manufacturing processes. Several results from studies about bio-sourced epoxy resins will be provided and in addition, previous works done on Infugreen 810 will be summarized. Studies regarding mechanical properties of flax fiber and its composites will be presented, together with some relevant information on carbon fiber reinforced polymer –CFRP. Goals of this study will be described in a chapter dedicated to the scope of this work. Then, a description of the used materials, the manufacturing the process and the testing methods to achieve the defined objectives will be given. These inevitably lead to the presentation of the results gathered in this manner, according to the characterized property and the used reinforcement. These results will be discussed thoroughly, to provide answers on the initial goals. Ultimately, general conclusion will be given for this work, by summarizing the most important results and related discussion points. At the end of this work, some appendices will provide important complementary data to current results.

Chapter 2

State of the Art

To introduce this work, it is first important to remind the basics of the mechanics of long fibre composites. Then, a brief description of the components used in this work and also results from some related studies will be given.

2.1 Composite material basics

This chapter providing the basis of composites theory is mainly based on the book "composite materials" by Daniel Gay and the textbook "Deformation and fracture of materials" by Prof. Thomas Pardoen [1, 2].

2.1.1 Generalities

A composite material is a material, as its name indicates, composed of two materials of different nature. Reinforced materials are therefore materials consisting of reinforcements embedded in a matrix. Polymeric composites can have two different types of matrix: thermosets or thermoplastic resins [3]. Thermoset resins are often composed of organic components and can be subdivided in several categories, according to their chemical composition: polyesters, vinylesters, epoxydes, phenolic and polyimides resins. All these resins have their own benefits and drawbacks and will be chosen according to the desired application. However, in the contrary of thermoplastic polymer, once they become solid, it is no longer possible to return to the liquid state, as the transformation is irreversible.

Nowadays, the main fibers used at industrial scale are the carbon, the glass and the aramid fibers. These can be in the form of either short or long fibres. For the production of composite materials, the fibers are always assembled to facilitate the preparation. Regarding long fibers, it exists different forms with unidimensional, bidimensional or even tridimensional architecture. This work exploit both unidimensional reinforcements also called unidirectional or even UD and bidimensional ones also called fabrics.

Unidirectional reinforcements

Unidirectional fibers sheet is an assembly of fibers positioned all parallel to each other in only one direction. This kind of tape is commercialized in the form of prepreg – reinforcements already impregnated with non polymerized resin– or dry sheets. For the

latter, the fibers are held together with very thin threads perpendicular to fibers direction. These yarns ensures the transverse cohesion of the main fibres, are expected not to affect the tensile properties in the longitudinal direction. However, This form of assembly is more difficult to drape than fabrics because they present higher stiffness.

Fabrics

Fabrics are any assembly of fibres involving the interweaving of fibres in two directions [3]. These fibres can be differentiated into warp and weft yarns. The difference between these strands is defined during the weaving of the fabric. On the loom, the warp yarns are stretched parallel to each other, while the weft yarns are passed between them according to a very specific weaving pattern. This way of weaving implies a more important crimping of the yarns –especially the weft threads– and directly impacts the properties of the final fabric.

For composite applications, a fabric is defined as balanced when there is the same amount of warp than weft yarns. Fabrics can be characterized by the interweaving mode used during yarns assembling. Plain weave fabrics, as shown in Fig. 2.1, is a mode of weaving where the yarns cross from each other alternatively. It gives products difficult to drape but with a very good flatness.

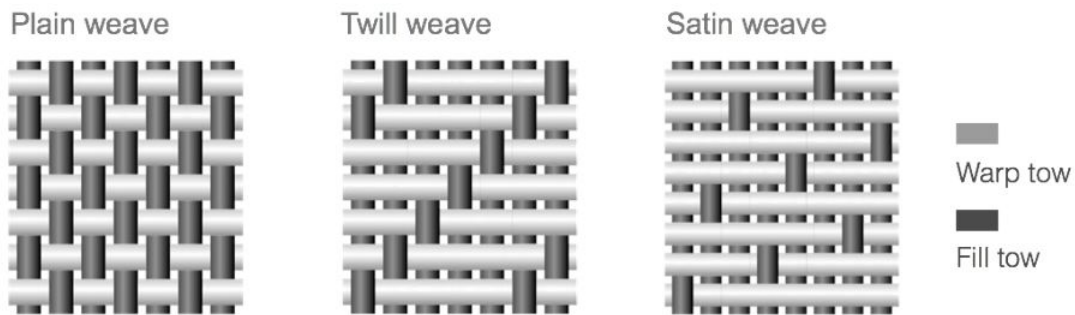


Figure 2.1: Fabrics weave pattern [4].

The twill weave consists of the passage of a warp thread over several weft threads. It is up to the manufacturer whether to make the same pattern in the weft direction or not.

The satin fabric follows the same basic pattern than twill but this time, the weave points for warp yarns do not touch each other. This type of fabric has the main advantage to be the most flexible fabric and is used therefore for the manufacturing of more complex structures. Nevertheless, this benefit can become a drawback since the orientation of fibres is not always accurately known. The main benefits of fabrics over UD are therefore drapability and similar mechanical properties in both directions.

The reinforcements are distributed in an optimal way, which allows the composite to obtain interesting properties which are a mixture of the properties of each component. In most cases, the reinforcements are selected according to their mechanical properties and the matrix has, as its main role, the transfer of forces between the fibres. The resulting materials are very heterogeneous and often have anisotropic behaviour.

Anisotropic material

The isotropy of a material illustrates the invariance of its physical properties in any direction, in other words, the material reacts in the same way to an identical force in several different directions.

If a force is applied, the deformation induced by the occurrence of stresses in the material can be written in matrix form as [1]

$$\begin{Bmatrix} \epsilon_x \\ \epsilon_y \\ \gamma_{xy} \end{Bmatrix} = \begin{bmatrix} \frac{1}{E} & -\frac{\nu}{E} & 0 \\ -\frac{\nu}{E} & \frac{1}{E} & 0 \\ 0 & 0 & \frac{1}{G} \end{bmatrix} \begin{Bmatrix} \sigma_x \\ \sigma_y \\ \tau_{xy} \end{Bmatrix} \quad (2.1)$$

where E is Young's modulus, ν is the poisson's coefficient and G is the shear modulus. There is also a relationship between the last constant and the other two which can be written

$$G = \frac{E}{2(1 + \nu)} \quad (2.2)$$

However, these relationships are no longer valid in the case of an anisotropic material. Indeed, the constants described above are theoretically identical in any direction when the material is isotropic but they are different when the material has anisotropic behaviour. The deformation-stress matrix can be rewritten as

$$\begin{Bmatrix} \epsilon_x \\ \epsilon_y \\ \gamma_{xy} \end{Bmatrix} = \begin{bmatrix} \frac{1}{E_x} & -\frac{\nu_{yx}}{E_y} & 0 \\ -\frac{\nu_{xy}}{E_x} & \frac{1}{E_y} & 0 \\ 0 & 0 & \frac{1}{G_{xy}} \end{bmatrix} \begin{Bmatrix} \sigma_x \\ \sigma_y \\ \tau_{xy} \end{Bmatrix} \quad (2.3)$$

2.1.2 Mechanical properties of plies

For composites, despite their anisotropic behavior and their heterogeneity, elastic properties may be approximated thanks to the rule of mixtures. this rule assumes that the properties of a composite material are a "mixture" of the properties of the constituent materials taken separately according to their proportion within the final material. First, the fiber mass fraction and matrix mass fraction are defined as respectively

$$M_f = \frac{\text{Mass of fiber}}{\text{Total mass}} \quad (2.4) \quad M_m = \frac{\text{Mass of matrix}}{\text{Total mass}} \quad (2.5)$$

with

$$M_m = 1 - M_f \quad (2.6)$$

The same principle may be applied for the fiber volume fraction V_f as

$$V_f = \frac{\text{Volume of fiber}}{\text{Total volume}} \quad (2.7) \quad V_m = \frac{\text{Volume of matrix}}{\text{Total volume}} \quad (2.8)$$

But for the volume fraction, the relation $V_m = 1 - V_f$, even if it is a good approximation, is actually incorrect. Indeed during processing, air can be trapped and creates porosity.

While the mass of this parameter can be neglected, the occupied volume is not. This parameter is sometimes denoted as the void content. The relation becomes

$$Vm = 1 - V_f - V_p \quad (2.9)$$

Knowing the mass and volume fractions, the density of a laminate can be calculated as

$$\rho = \frac{\text{Total mass}}{\text{Total volume}} \quad (2.10)$$

$$= \frac{\text{Mass of fiber}}{\text{Total volume}} + \frac{\text{Mass of matrix}}{\text{Total volume}} \quad (2.11)$$

$$= \frac{\text{Volume of fiber}}{\text{Total volume}} \rho_f + \frac{\text{Volume of matrix}}{\text{Total volume}} \rho_m \quad (2.12)$$

$$= \rho_f V_f + \rho_m V_m \quad (2.13)$$

With these parameters, if there are known in the produced laminate, it is therefore possible to approximate the properties. However, the theoretical values may not always agree the experimental ones because of porosity or interfacial properties for example. In the case of UD ply in the fiber direction as shown in Fig. 2.2a, according to the isostrain hypothesis

$$\epsilon = \epsilon_f = \epsilon_m \quad (2.14)$$

the longitudinal Young's modulus E_l i.e. the modulus in the fiber direction becomes

$$E_l = E_f V_f + E_m V_m \quad (2.15)$$

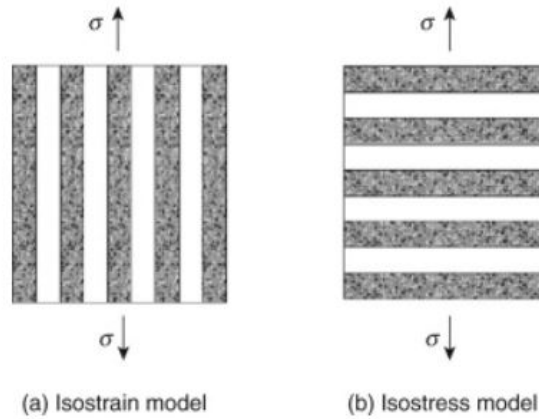


Figure 2.2: Structural models of UD composite laminate [5].

The same approach can be used for transversal modulus but this time, according to the isostress model

$$\sigma = \sigma_f = \sigma_m \quad (2.16)$$

The transversal Young's modulus E_t i.e. the modulus perpendicular to the fiber direction becomes

$$E_t = \frac{E_f E_m}{V_f E_m + V_m E_f} \quad (2.17)$$

Equations 2.15 and 2.17 can express respectively the upper and lower bounds of possible moduli for a UD composite according to the volume fraction –Fig. 2.3. The moduli in

any direction lie between the two curves as the actual moduli. These two bounds are also denoted as Voigt and Reuss bounds respectively for the upper and the lower one [2].

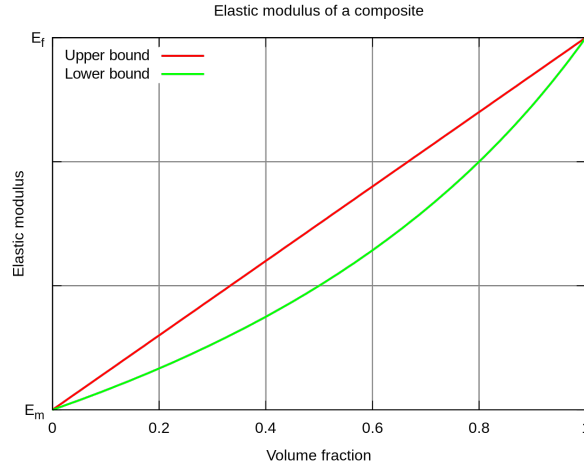


Figure 2.3: Upper and lower bounds on the Young's modulus of a composite as predicted by the rule of mixtures [6].

Woven fabrics

The approximations in the section above are applicable only for UD plies but can be used for woven fabrics with some modifications. Indeed, as explained in section 2.1.1, woven fabrics contain fibers in two directions in the contrary of UD plies. Therefore, the computed longitudinal and transversal moduli are not consistent anymore. A first approximation is to consider a fabric as the stacking of two perpendicular UD plies [1]. Considering e as the total thickness of the fabric and n_1, n_2 number of warp yarns/fill yarns per meter respectively, the thicknesses of the two equivalent UD plies become

$$e_{warp} = e \frac{n_1}{n_1 + n_2} = k e \quad (2.18)$$

$$e_{fill} = e \frac{n_2}{n_1 + n_2} = (1 - k) e \quad (2.19)$$

With these approximated thicknesses, equations 2.15 and 2.17 can be used to compute the elastic moduli of the two plies. An other way is to take the fabric as one single anisotropic layer with a thickness e and $k = n_1 / (n_1 + n_2)$

$$E_x \approx k E_l + (1 - k) E_t \quad (2.20)$$

$$E_y \approx (1 - k) E_l + k E_t \quad (2.21)$$

These approximations may lead to a significant bias between the theoretical values and the experimental ones. Nowadays, more complex and accurate models are existing to better estimate fabric properties.

2.1.3 Laminate failure

When the failure occurs, three main mechanisms are possible as illustrated in Fig. 2.4 : fibers failure, matrix failure and delamination. A preferential failure mode takes place according to the nature and the direction of the applied load and the properties of the laminate. Indeed, failure is not the same in a composite with UD in one direction as in a composite where the UD plies are oriented in several directions.

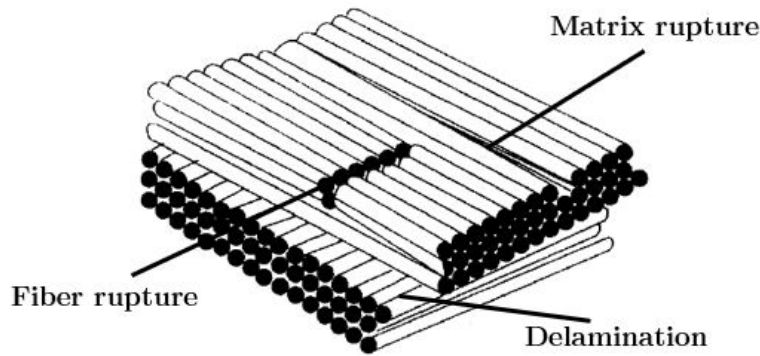


Figure 2.4: Failure modes of a composite laminate [7].

When an UD laminate undergoes longitudinal tensile load, if the reinforcements have a lower deformation at break than the matrix, a rupture of the fibres will be observed [8]. In the opposite case, a transverse rupture of the matrix will occur. But when the crack initiates, two different behaviors can take place depending on the matrix fibre adhesion. When the fiber-matrix adhesion is strong, the crack propagates successively within the fibers and the matrix. However if the adhesion is weak, during the transverse failure of the matrix, as shown in Fig. 2.5, decohesion of the fibers can be observed and this mechanism is called "fiber pull-out".

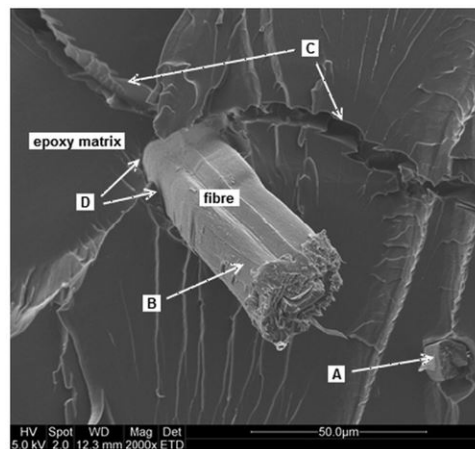


Figure 2.5: "SEM micrograph of typical failure modes of untreated flax fabric reinforced composite in tension. A, failure of fibre; B, fibre pull-out; C, brittle fracture of epoxy matrix and D, fibre debonding. SEM, scanning electron microscopy." [9]

In compression, Fig. 2.6 shows other failure mechanisms also depending of the constituents and the loading mode [10].

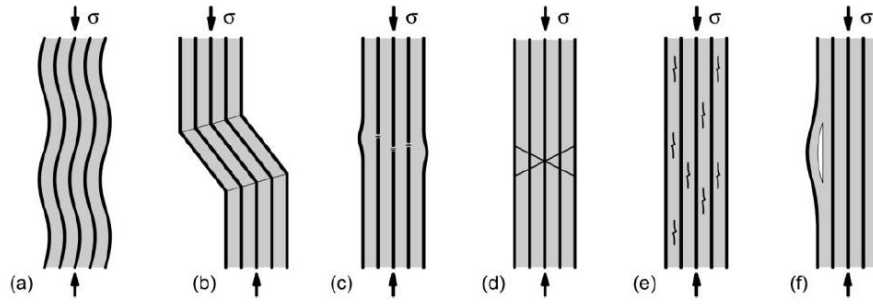


Figure 2.6: *Compressive failure modes of fibre composites. (a) Elastic micro buckling. (b) Fibre kinking. (c) Fibre crushing. (d) Shear band formation. (e) Matrix cracking. (f) Buckle delamination.*[10]

2.2 Manufacturing process

Before reaching the final product, reinforcements and matrix must be assembled by specific process optimized for composites. In this chapter, only the main techniques for the manufacture of long fibre composites will be developed. All the information given in this section are also available in [11].

2.2.1 Hand layup technique

Hand-Layup is one of the oldest and common composite manufacturing technique [11]. It is a molding method which allows the production of wide variety of laminates. This technique offers a simple and low cost method for product with consistent quality. The process is the following: the mold is first coated with release agent to prevent a strong adhesion of the final product on the mold. Then, the reinforcement are manually place on the mold and successively impregnated with the non cured resin. This latter is applied by pouring, brushing, spraying or paint roller; rollers are also useful to release entrapped air during the process.

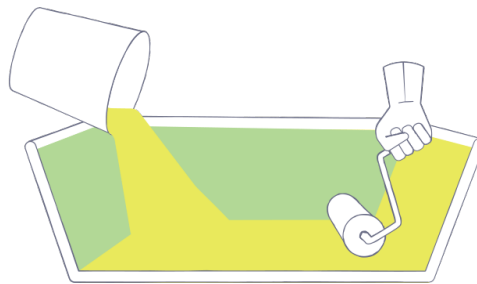


Figure 2.7: Hand Layup process

This technique can also be used with prepreg reinforcements.

2.2.2 Vacuum Bag Technology

Vacuum bag technique is considered as a closed-molding technique and can produce a large volume of material [11]. This method can be either used with dry fabric or with

prepreg. The main benefits of this method comparing to classic hand layup technique is the efficiency to remove entrapped air and for laminates made of dry fabrics, it allows to remove the excess resin.

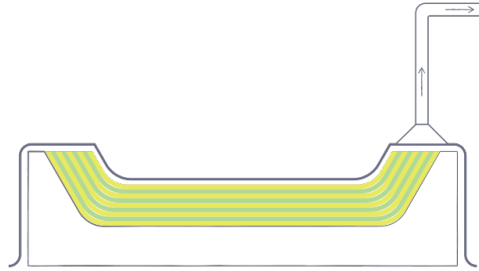


Figure 2.8: Vacuum bag process

When the layup is done, a flexible film is used to compact the product and then remove air voids [11]. Some process modifications are done according to the use of dry fabrics or prepreg. For prepreg, the mold with the plies must be heated or put in an autoclave. Regarding the dry fabrics, some additional layers must be placed before the flexible film such as release film, a bleeder ply to absorb resin and finally the film.

2.2.3 Vacuum infusion processing

In this process, reinforcement plies are manually stacked in the mold as hand layup technique without adding resin [11]. Then, the mold is closed and a lower pressure than atmospheric one is applied within the mold cavity. The injected resin is driven into it because of the vacuum in the mold but resin excess is not possible because laminate is already compacted. Therefore, it allows a higher fiber volume fraction than previously described processes, which leads to enhanced properties.

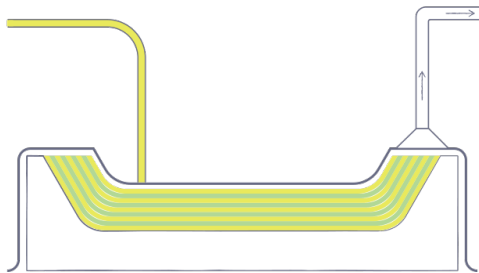


Figure 2.9: Vacuum infusion process

2.2.4 Resin Molding transfer

Resin transfer molding, also called RTM, is a process using dry reinforcements and resin injected under pressure within the closed mold [11]. As other techniques, the reinforcements are laid-up in the mold already coated with adapted release agents. After the stacking of the reinforcements with the desired orientations, the mold is closed, compacts plies. Then the resin is injected and with the pressure, it impregnates the fibers. This process can be used at room temperature but heated molds are also available to achieve fast curing cycles.

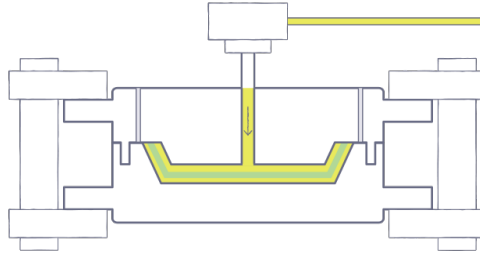


Figure 2.10: Vacuum infusion process

2.2.5 Layup rules

Although the draping of plies may allow some freedom in their arrangement during the production of a laminate, some rules are required to produce a satisfactory end-product. First of all, the mirror symmetry consists of stacking the plies symmetrically around the "neutral fiber" of the laminate. It allows the symmetry of the stresses during the polymerization of the resin and avoid the warping phenomenon. In addition to this, imposing the same proportion of ply in the direction ± 45 enhances the shear robustness of the laminate. To help maintain the laminate integrity in the case of unexpected forces acting, it is recommended to have at least 10% of plies in each direction. Finally, regarding the interlaminar stresses, stacking consecutively more than three plies in the same direction or two plies successively stacked with a direction difference of 90° , may increase the risk of delamination.

2.3 Bio-sourced epoxy resins

2.3.1 Epoxy resin

Epoxy resins are a variety of polymers where the monomers are molecules composed of one or more epoxide precursors. These functional groups are cyclic ether with an oxygen is linked to a C-C bound. The secondary groups in these precursors are quite variable.

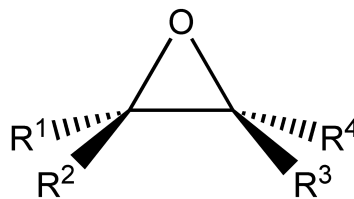


Figure 2.11: Epoxide generic pattern

The polymerisation of these epoxide monomers is a polycondensation directed by a compound called a hardener which allows the formation of a three-dimensional network based on monomer structures. When the cross-linking reaction occurs, it is irreversible and the final product becomes infusible and insoluble.

The most widely used resin in industry nowadays is the blend of two elements: an epoxy precursor DGEBA and a hardener from petro-sourced synthetic amine. DGEBA

compounds is formed from bisphénol-A which reacts with épichlorhydrine in the presence of aqueous sodium hydroxide [12].

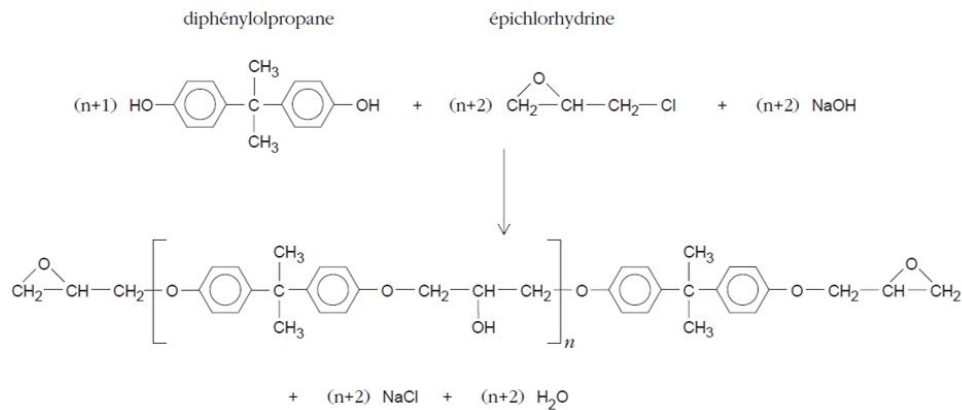


Figure 2.12: "Synthesis of DGEBA diepoxy prepolymers by the Taffy process"[12].

Although these polymers offer quite interesting properties for composite applications, they are all petroleum-based which is seen as a long-term problem today. In recent years, there has been a lot of research into the development of eco-friendly resins, which are partially petroleum-based and where the carbon chains come from other natural resources.

Properties of bio-based epoxies

As already described in the work of *van Innis C.* [13], *Ramon et al.* [14] reported results from different authors about mechanical properties of partially bio-sourced epoxy resins. Fig. 2.13 shows the tensile properties of these bio-based epoxy are very scattered but some of these have similar properties than petroleum-based epoxies

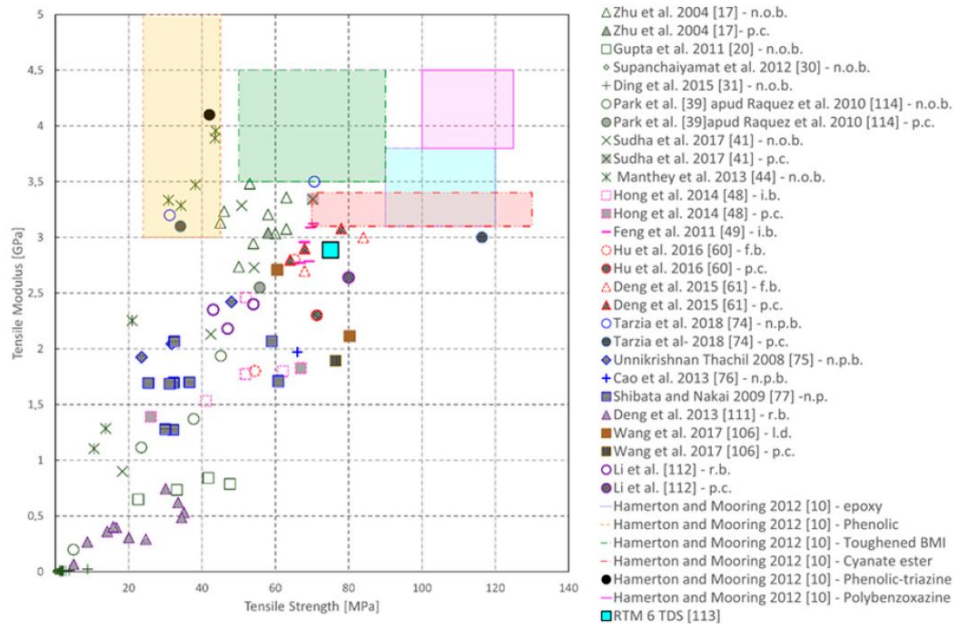


Figure 2.13: "Tensile modulus versus tensile strength of bio-based and petroleum-based resins. *n.o.b.*, *i.b.*, *f.b.*, *n.p.*, *n.r.*, *r.b.*, *l.d.* and *p.c.* stay for natural oil based, isosorbide based, furan based, natural phenolic, epoxidized natural rubber, rosin based, lignin derivatives and petroleum-based, respectively" [14].

The same report was done for flexural properties and as for tensile properties, the results are included in a wide range of values.

Ma et al. [15] assessed different properties of a bio-based epoxy resin synthesized from itaconic acid –IA– and compared it with DGEBA epoxy resin cured with same curing agents. IA is already produced at large scale in industry for other purposes and has as origin glucids fermentation by using fungi. They exploited itaconic acid to synthesize a trifunctional epoxy monomer denoted TEIA. This molecule was synthesized in two steps –Fig. 2.14a: dehydrobromination between itaconic acid and allyl bromide to obtain allylated itaconic acid –AIA– and last, epoxidation of AIA [15]. Regarding the mechanical properties, they performed flexural test by three-point bending to extract properties and compare it with DGEBA blends –Fig. 2.14b.

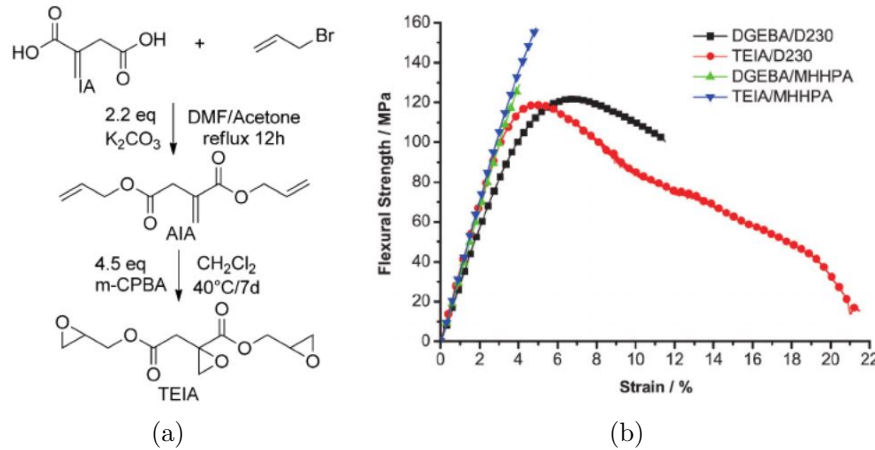


Figure 2.14: (a) Synthesis of trifunctional epoxy resin from itaconic acid (TEIA) (b) Flexural stress-strain curves of the cured epoxy resins reported by [15].

They noted first that the behavior of the produced epoxy resin depends of the chosen curing. For one of them, the resulting epoxy resin exhibited ductile fracture while with the other curing agent, the epoxy resins presented brittle failure.

Jost N. [16] assessed mechanical properties of the bio-sourced epoxy resin InfuGreen 810 according to different curing cycles, bio-sourced resin also used in this work. To do so, uniaxial compressive and tensile tests were performed on different samples with different strain rates. Results in Fig. 2.15 show that epoxy resin did not have the same behavior in tension than in compression. First, for tension, depending on the used curing cycle and the used post-curing, the resin did not act in the same way. Indeed, if cured at ambient temperature without post curing, the resin presented more brittle failure, in the contrary of other cured resins with a more ductile behavior.

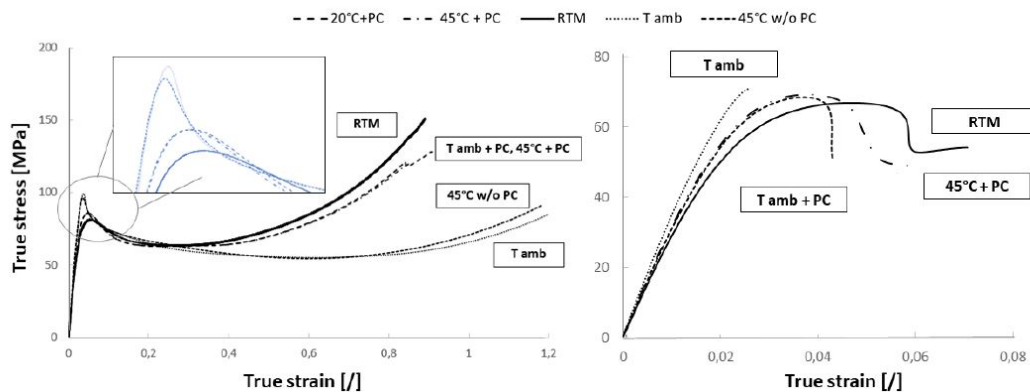


Figure 2.15: "True strain - true stress curves for the different curing cycles at 1mm/min in compression (left) and tension (right)" [16]

For compression, in contrast to tension specimens, all samples presented softening as shown in Fig. 2.15 for the same test speed. Samples with the RTM cycle or with post-curing had stronger hardening than others which could have an impact on resin failure behavior. Jost N. [16] also assessed the variation of the elastic behavior of InfuGreen 810 according to the curing cycle and therefore the curing degree as noticed in Fig. 2.16.

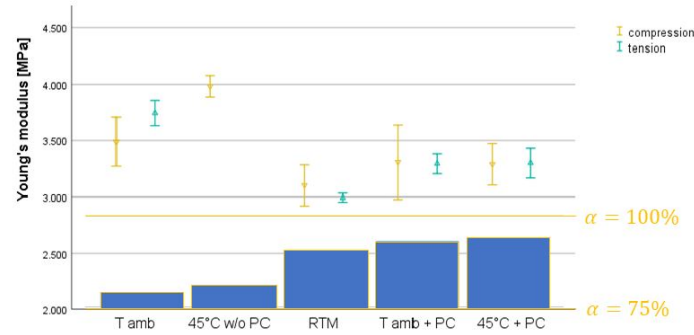


Figure 2.16: "Elastic modulus in tension and compression, tested at room temperature." [16]

At low curing degree, the studied bio-sourced epoxy presented better compressive elastic properties than in tension. Nevertheless, when the polymerization level is higher, elastic properties became approximately equivalent and were around 3.25 GPa.

van Innis C. [13] worked in parallel with two curing cycles studied by *Jost N.* but assessed other mechanical properties as fracture toughness and fatigue failure of InfuGreen 810. Regarding the fracture toughness, despite the low repeatability of the experimental crack, for specimens cured at room temperature with a post curing at 80°C, the stress intensity factor was about 2.81 MPa.m^{0.5} while with the curing at 45°C with the same post-curing gave an intensity factor around 2.43 MPa.m^{0.5}. Finally, a tension-tension fatigue test was performed and an endurance limit of 35.6 MPa was determined for the studied resin.

2.4 Flax fibres

2.4.1 Generalities

Since bio-sourced fibers became an important field of interest for eco-friendly composites, flax has been one of the most used fiber for biocomposites development. This fiber offers many advantages such as low cost, specific stiffness or even recyclability compared to glass fiber [17]. Furthermore, the flax plant has been harvested for a long time which implies a thorough knowledge of the right growing conditions and of the parameters that can impact the properties of the products derived from this plant.

Unlike synthetic fibres such as glass or carbon or even natural fibres such as basalt, flax fibres must be grown, harvested and processed before they can be used as reinforcements.

2.4.2 Growth of the flax plant

Flax does not grow in all conditions and requires a humid climate where the temperature does not exceed 30°C [18]. It is for these reasons that several countries from Europe such as Belgium, Netherlands and France are suitable for the flax harvesting. Growth should start when temperatures reach approximately 10°C, which corresponds to early spring in the countries mentioned above. Then, as many crops, the growth can be divided into several steps as shown in Fig. 2.17. The cycle begins with the sowing and ends at the senescence, which corresponds to the progressive degradation of cells plant.

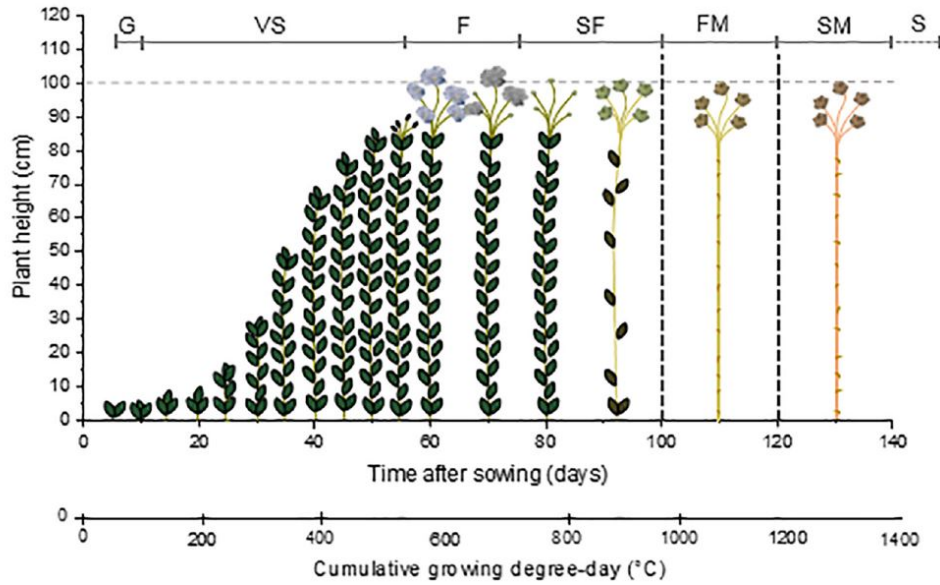


Figure 2.17: Scheme illustrating the growth of the flax plant according to the cumulative growing degree-day GDD [19].

The germination G begins approximately 5 to 10 days after sowing. It is followed by the vegetative stage VS which starts slowly –15cm in 15-20 days– and then accelerates. During the fast development period, the plant may grow up to several centimetres per day to reach a height of 80-90 cm. The plant arrives at its final height of 1 meter during the flowering F –50 days after G. This step is followed by the seed formation SF, the fiber maturation FM, the seed maturity SM and finally the senescence S. The last two steps are not reached regarding industrial flax fibers and these are often pulled out after 100-120 days of the initial cycle of growing.

These stages may be determined not only by the number of days of growth but also in terms of cumulative temperature since sowing. This parameter is called cumulative growing degree day and can be calculated as following [19]

$$GDD_n = \sum_{i=1}^n \frac{T_{max,i} - T_{min,i}}{2} - T_{base} \quad (2.22)$$

where T_{max} is the maximal temperature, T_{min} the minimal one and T_{base} is the base temperature and corresponds to 5°C for flax. Under this temperature, growth is not considered to take place. Therefore, with this new method of calculation, the stages can be associated to several GDD values in addition to the days of growing –Fig. 2.17. However, this diagram only indicates a theoretical evolution and that the cycle can change depending on weather and crop conditions.

2.4.3 Structure of flax

Flax plant can be divided into three main organs: the stem, the root and the leaf. The stem acts as a support but also as a transport of nutrients through the plant. In order to do this, the stem is made up of several elements that differ firstly based on their location but also on their composition as illustrated in Fig. 2.18.

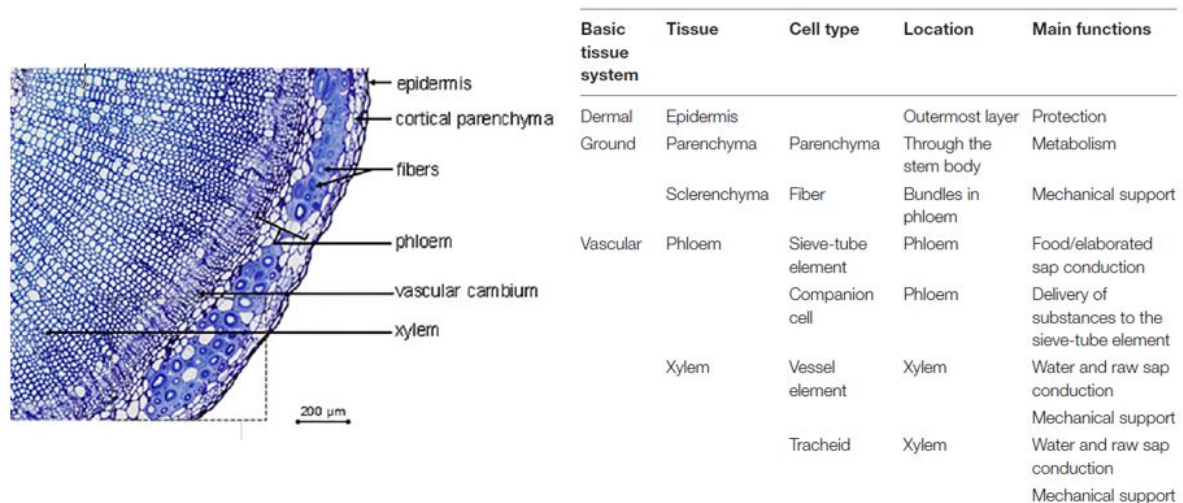


Figure 2.18: Stem structure [18]

The region of interest is located in the periphery of the stem section and consists of fibers bundles –Fig. 2.18. Those are also called "technical" flax fibers and consist of an assembly of 10-40 elementary fibers twisted together with pectin which is a structural polysaccharide of plant origin. These technical fibers may be of plant length [20]. Regarding the elementary fibers constituting these bundles, they have polygonal section shape with sides varying between five and seven but are often reduced to a circular shape as illustrated in Fig. 2.19.

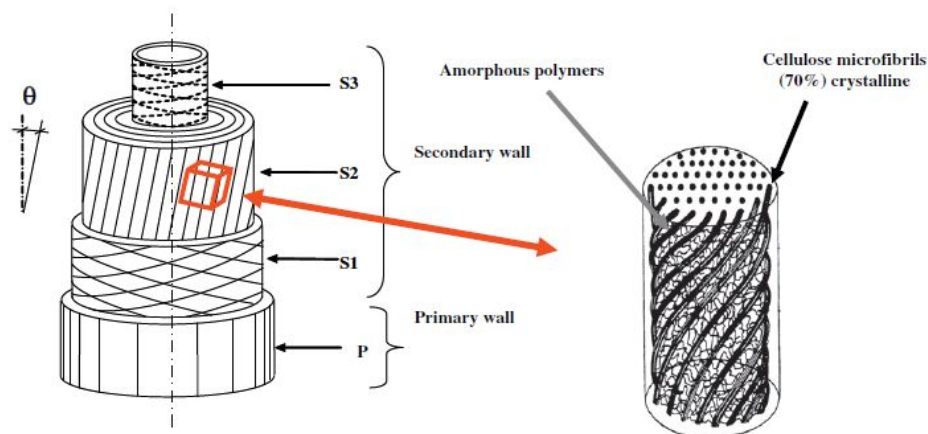


Figure 2.19: flax structure at different scales [21].

The dimensions of these elementary fibers are highly variables and the range are between 5–76 mm and 4–77 μm respectively for the length and width of the fibres. This variability is one of the main characteristic that differs from synthetic fibers.

At microscale, the elementary fiber has a complex structure composed of several cylindrical walls as shown in Fig. 2.19. These layers differ from each other with their thickness and chemical structure. the external wall –0.2 μm – is mainly composed of lignin, pectin and (hemi)cellulose [22]. The microfibrils of cellulose are embedded in the matrix of pectin and hemicellulose, and are wound with a 35° angle to the direction of the elementary fiber [23]. The secondary cell wall is thicker than the external one and can be subdivided into three

layers. The most important one is the S2 because of its thickness and for this layer, the microfibrils are oriented with an angle of 10° . It is this layer that gives the high tensile strength to the flax fiber [22]. In the center of the fiber, the lumen is an empty area that is useful for water and nutrients transportation.

2.4.4 Fiber composition

As noticed in the previous section, the elementary flax fiber is constituted of several chemical molecules in varying proportions that can have an impact on the mechanical properties of the fiber.

Table 2.1: Chemical composition of flax fibers as reported by different authors summarized by [24].

Cellulose (%)	Hemi-cellulose (%)	Pectin (%)	Lignin (%)	Wax (%)	Moisture content (wt.%)
64.1	16.7	1.8	2.0	1.5	10.0
67	11	–	2.0	–	–
73.8	13.7	–	2.9	–	7.9
65	–	–	2.5	–	–
62–72	18.6–20.6	2.3	2–5	1.5–1.7	8–12
71–75	18.6–20.6	2.2	2.2	1.7	10.0

Cellulose

Cellulose is a linear homopolymer composed of D-anhydroglucopyranose units and linked together by glycosidic β -(1,4) bounds. Each bound leads to a 180° rotation of the next unit and therefore the repetition motif becomes a dimer called "cellobiose". This molecule is present in all plant and is the main component of their cellular walls.

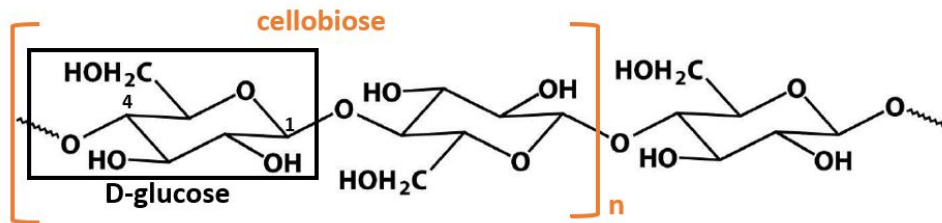


Figure 2.20: Cellulose backbone structure.

For flax fiber, cellulose is gathered between 50 and 100 molecules and constitute the elementary fibrils which in turn, gathered in bundles, constitute the microfibrils. Giving the cellulose is semi-crystalline polysaccharide with a large amount of hydroxyl groups, the flax fiber has a hydrophilic behavior. When this type of fiber is used as reinforcement with hydrophobic matrix, it leads to low adhesion and high moisture absorption [24].

Because of the high amount of cellulose in flax fiber –up to 70%– and being the stiffest and strongest constituent in the fiber, it gives interesting properties to use flax fiber as composite reinforcement.

Hemicellulose

Hemicellulose has as main function to link all cellulose fibrils together by using hydrogen bonds. Despite what its name might suggest, hemicellulose has little in common with cel-

lulose in terms of its chemical structure. This molecule is a branched heteropolysaccharide composed of 3000-5000 units –against 7000-15000 for cellulose– having have backbone made up of β -1,4-linked D-glycans [25].

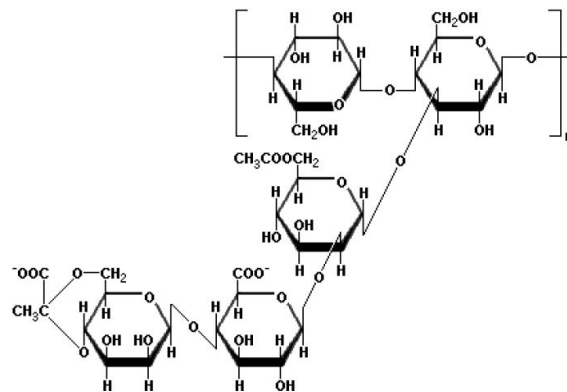


Figure 2.21: Hemicellulose chemical structure [25].

Unlike cellulose, hemicellulose is amorphous and because of the open structure formed by the branches, the hydroxyl and acetyl groups present in the molecules, hemicellulose is soluble in water and promotes water absorption [24].

Pectin

Pectin is also a polysaccharide with a backbone composed of 70-100 units of α -D galacturonic acid and α -L-rhamnose in small quantity. This molecule plays a structural role and connections between pectin chains, which forms a gel allowing the storage of a large quantity of water.

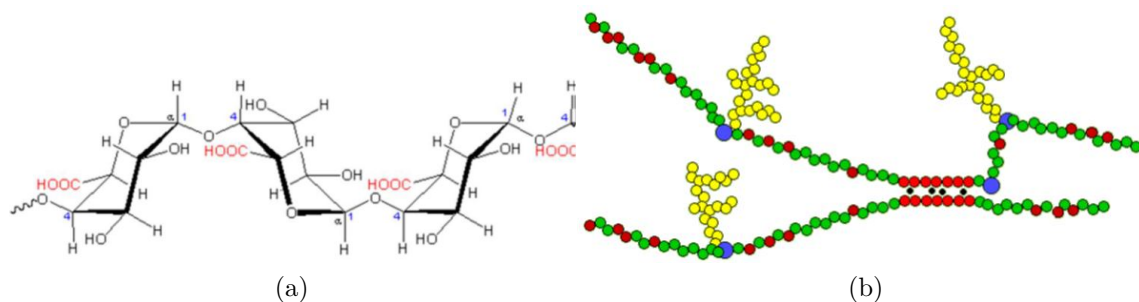


Figure 2.22: Pectin chemical structure (a) polygalacturonic acid chain (b) in red and green, respectively galacturonic acid and methylated galacturonic acid; in blue, rhamnose

Lignin

Lignin is a natural tridimensional polymer made of phenolic groups and present a more hydrophobic behavior than the previous molecules. Its role is structural and gives an impervious layer to avoid water loss. The low amount of lignin in flax fiber does not allow to counteract the global hydrophyl behavior of the fiber [26].

In view of the constituents, several problems may therefore arise. The first one is the moisture absorption. Water in fiber may lead to poor adhesion with a hydrophobic matrix

which in turn can lead to inefficient stress transfer between the matrix and the fiber, to porosity and to more moisture absorption with as result poor mechanical properties [26]. To compensate the affinity of the fiber with water, several techniques of surface treatment have been tested to enhance the adhesion for composites without impacting the initial mechanical properties of fibers as described later on.

The second problem is the low thermal stability of the fibers due to the presence of hemicellulose which degrades at 220°C-280°C. This last temperatures are in temperature range for the process of some thermoset and thermoplastic matrices, which sometimes makes it difficult to use natural fibres such as flax for composites development [27].

2.4.5 Fiber extraction

The extraction of the fibre from the stem is not a simple process and requires several steps which can have an impact on the quality of the fibre and therefore its mechanical properties.

Retting process

The first important step is the retting process. This process takes place during the rainy season just after the harvesting of green stems [28]. The stems are pulled out of the ground then carefully spread evenly on the ground [20]. *"During retting, the flax stems are colonized by fungi enhanced by the combined action of moisture and heat"* [29]. Enzymes released by these fungi degrade pectins present in epidermis, cortical parenchyma and middle lamellae that leads to bundles split [29]. The retting can last from three weeks to a month and a half depending on mainly weather conditions [29]. The retting duration and weather conditions can have a impact on mechanical properties. *Martin et al.* [29] investigated the variability of flax fiber properties according to the degree of retting. For this study, flax plants were harvested and several stems were collected at different times during the retting period as shown in Table 2.2. Sample R6 is considered as well retted and presents the standard quality for textile industry while R1 to R4 samples are assumed as under retted. Single flax fibers were extracted from the central zone of each stem to prevent variability due to the localisation.

Table 2.2: Identification of samples collected and corresponding retting time assessed by [29].

Batch name	R1	R2	R3	R4	R5	R6
Retting time (days)	1	5	9	14	16	19

Martin et al. [29] highlighted several important variations due to the retting time. As expected, retting time influences the quantity of remaining cortex tissue around the fibre bundles. In a second time, water absorption is also impacted by retting. The results obtained in the study show that water uptake increases for under retted samples. Finally, as shown in Table 2.3, the tensile properties as Tensile modulus and stress at break are enhanced by the degree of retting.

Table 2.3: Tensile properties of individual fibers for different retting times assessed by [29].

Batch	Batch diameter (μm)	Tensile test diameter (μm)	E (GPa)	σ (MPa)	A (%)
R1	13.3 ± 4.4	14.0 ± 2.7	38.6 ± 17.3	792 ± 374	2.2 ± 0.7
R3	13.0 ± 4.0	14.5 ± 2.6	48.6 ± 11.8	935 ± 317	2.2 ± 0.7
R6	14.8 ± 4.4	15.9 ± 2.5	55.6 ± 11.8	1036 ± 270	1.9 ± 0.5

Scutching process

This stage is used to remove the woody core from the retted stem and therefore extract flax fibers which correspond to technical fibers already described [28]. With this process, other by-products are created such as flax tows or shives –Fig. 2.23.

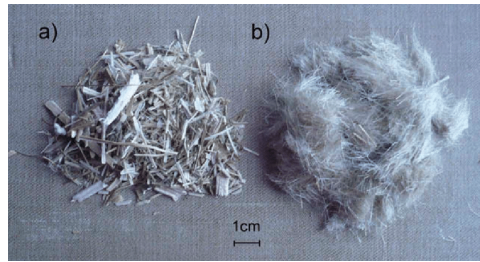


Figure 2.23: (a) flax shives (b) flax tows [30].

The scutching can be done either by striking or by crushing. This step may be crucial for long flax fibers because the scutching parameters may impact the length and mechanical properties of flax bundles and therefore the resulting UD or fabric [20]. For the reinforcements, the technical fibre can be used as is or yarns can be made by twisting several fibres bundles according to the desired textile parameters such as the linear density –in TEX or g/km– or the twist level –number of twist per unit of length.

2.4.6 Mechanical properties of flax fiber

As described in this work, the fibers must have relevant mechanical properties for them to be used as reinforcements in a composite. Flax shows efficient properties in term of specific Young's Modulus and specific tensile strength.

Tensile properties of flax fibers

Charlet et al. [31] analysed monofilament of flax in tension and the resulting curve as illustrated in Fig. 2.24a can be divided in three part as a linear part, a non-linear part and final part until failure. The first linear part can be associated with a global loading in each cell walls –elastic behavior [31]. The non-linear part is explained by the rearrangement of the amorphous components during the alignment of the microfibrils in the direction of the load and can be seen as a elasto-visco-plastic deformation of the fiber [31]. The third part of the curve can be interpreted as the elastic response of the newly aligned fibrils.

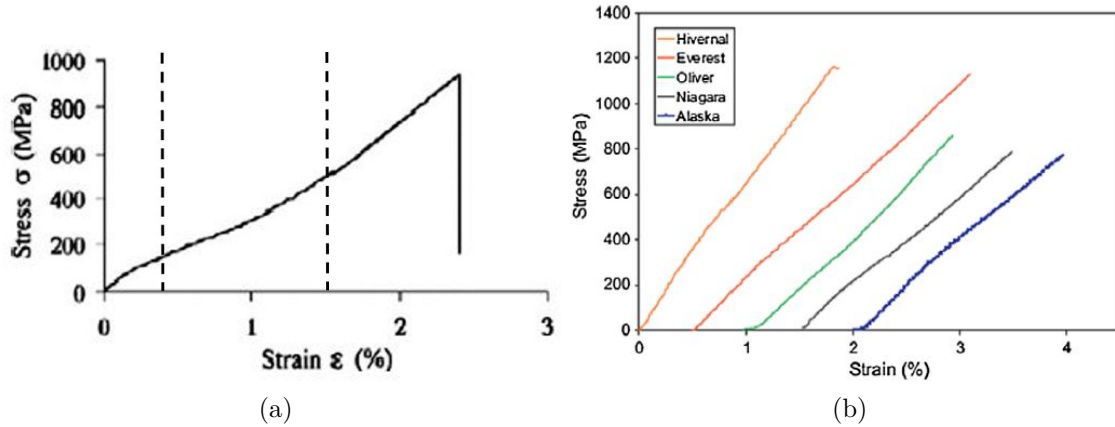


Figure 2.24: Tensile property of elementary flax fiber (a) Typical stress-strain curve adapted from [31] (b) stress-strain curves according to different species of flax fibers reported by [32].

Failure mechanism

Rohmàny *et al.* [33] examined *in situ* the failure mechanism of a technical fiber –i.e. a bundle of elementary fibers– during a tensile test by SEM and acoustic emission. During these tests, they determined that the failure sequence of the fiber followed a particular pattern reflecting the fiber architecture as shown in Fig. 2.25. Indeed, a first axial splitting occurs along the elementary fibers. Then, transverse cracks in elementary fibers appear and finally, the failure of elementary fibers and their microfibrils occurs.

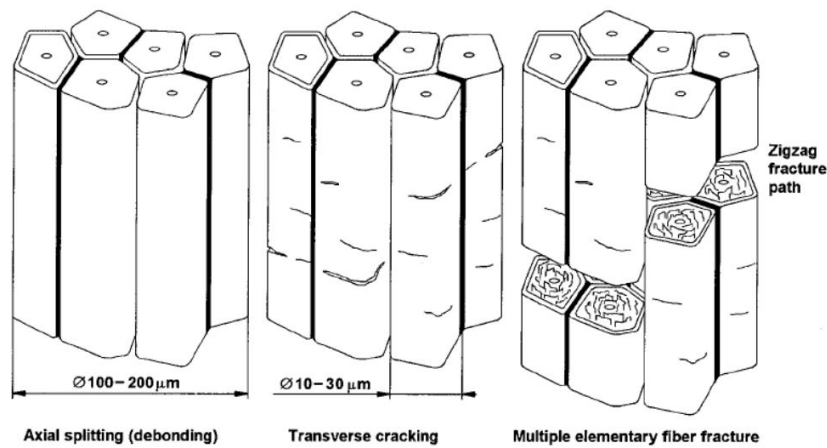


Figure 2.25: Failure sequence of a single flax fiber reported by [33].

Factors affecting tensile properties

Mechanical properties of flax fibers are highly variable considering the weather, growth conditions and the species –Fig. 2.24b. Furthermore, flax properties also depend on the localization in the stem because the composition differs along the position of the fiber [24].

Charlet *et al.* [34] assessed the variation of Agatha flax fiber tensile properties according to the localisation in the stem. They analysed three sets of fibres extracted from the bottom, the middle and the top of the stems. To complete the analysis, they made sugar

and morphological analysis to compare results. They found difference between tensile properties of the three sets and deduced that the mechanical properties were influenced by the biochemical composition of the fibres as shown in Table 2.4 and Fig 2.26.

Table 2.4: Experimental values of parameters as a function of the fiber location in the stem (a) mass of matter released after the extraction series –mg/g of fibre: TSS, TES, cellulose and residue (b) Mechanical parameters –d, diameter; E, Young’s modulus; σ_r , failure stress; Ar, ultimate strain– reported by [34].

(a)					(b)					
	TSS	TES	Cellulose	Residue	Nb. fibres	d (μm)	E (GPa)	σ_r (MPa)	A_r (%)	
Top	55	86	700	159	57	21.5 ± 5.3	51 ± 22	753 ± 353	1.8 ± 0.7	
Middle	41	86	800	73	45	21.3 ± 6.3	57 ± 29	865 ± 413	1.8 ± 0.7	
Bottom	57	59	800	84	59	23.5 ± 7.9	51 ± 26	783 ± 347	2.0 ± 0.9	

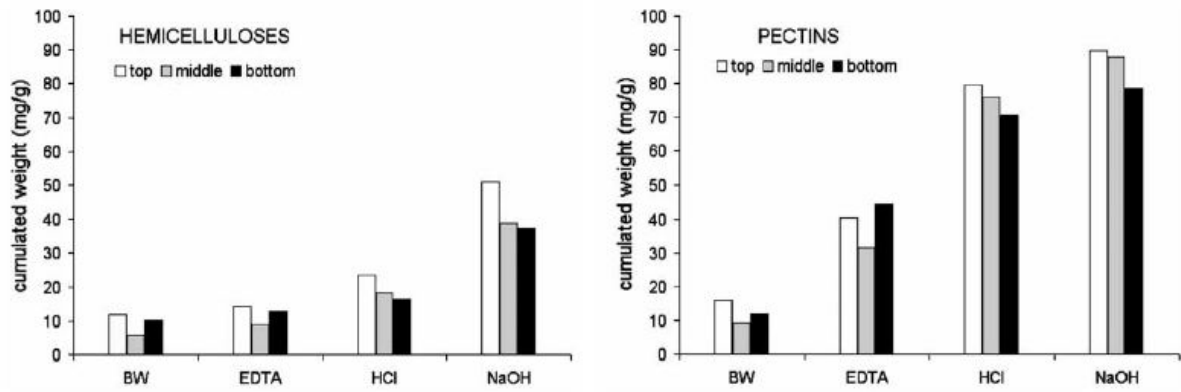


Figure 2.26: Cumulative extracted weights of hemicelluloses and pectins extracted by boiling water –BW–, EDTA, HCl and NaOH according to the location of the fibres in the stem reported by [34].

They defined "Total superficial sugars" TSS as removed constituents extracted by boiling water and EDTA treatment –Fig. 2.26. The sugars extracted by boiling water are mainly originated from the cortical tissues –*cortical parenchyma* in Fig 2.18– and those by EDTA are the compounds holding the fibres in bundles. The amount of these sugars is directly related to the cleanliness of the fibres during extraction and subsequently affects their mechanical properties. The less TSS the fibre contains, the better its tensile properties is as noticed in Table 2.4. On the other hand, "Total encrusting sugars" TES extracted with HCl and NaOH –Fig. 2.26– are determined as the matrix embedding the cellulose microfibrils in the secondary wall.

S. Y. Nayak et al. [26] analyzed the variation in tensile strength of single fiber with chemical treatment with different duration.

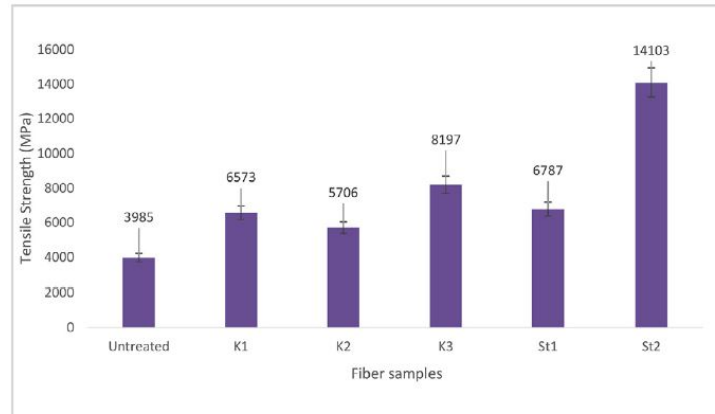


Figure 2.27: Graphical representation of tensile strength for a single fiber. (K1,K2,K3: KMnO_4 treatment during respectively 10, 20 and 30 min; St1, St2, stearic acid treatment during respectively 24h and 36h reported by [26]).

As shown in Fig. 2.27, these treatments increase the mechanical properties by removing surface irregularities. The difference for results between the two treatments is the KMnO_4 is much more aggressive than stearic acid and led to a deterioration of fiber surface.

Baley et al. [21] assessed the influence of drying on flax fiber properties since this step can be crucial for using flax fiber as reinforcement for composite. Indeed, when fibers are not dried, water contained in fibers becomes an issue if evaporates during composite manufacturing when a high process temperature is needed. Therefore, they wanted to determine if drying cycle induce a drop of the mechanical properties of a single fiber. For their work, they first dried for 14h at 105°C and immediately tested after this period to avoid reabsorption.

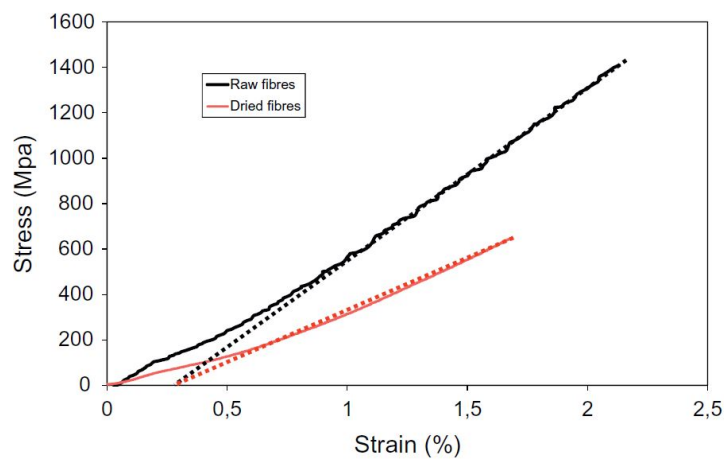


Figure 2.28: Tensile stress-strain curve of single fiber, untreated fiber in black and dried fiber in red reported by [21]. As explained by the authors, the dotted lines are just drawn to show the non-linearity of the curve.

They observed that tensile modulus was not significantly influenced by the drying process. They explained this invariability by the fact that the stiffness of the fiber depends mainly on the cellulose volume fraction which is not impacted by the drying. Indeed, the cellulose

only degrades above around 230°C. Nevertheless, their work shows once again the decrease of the stiffness of the fiber in the second part of the curve due to the rearrangement of the microfibrils. The end of the microfibrils reorientation is supposed occur at the change of slope of the curve and then, an hardening of the fiber appears. As it is noticed in Fig. 2.28, the decrease of the the slope after the first linear part was higher for dried fibers compared to untreated fibers. Then the slope increased again for both. And finally, the dried fibers broke at lower stress than untreated fibers. They explained these differences by the fact that water removal has an impact on the interactions between the fibrils and water acting as a plasticizer, its removal embrittles constituents [21]. Drying could also induce internal stresses within the fiber because of the difference in behavior of all the components during drying. The consequence of this stress is the decrease of mechanical properties.

Amroune et al. [35] investigated the influence of technical fiber length on tensile properties for flax. To do so, 400 samples were tested with 10 different gauge length between 10 and 500 mm. Obtained results showed first a relationship between stress and tensile modulus. With increasing stress, the tensile modulus follows the same pattern. A significant dispersion is noted for the properties and one of the hypothesis of the authors is the presence of internal defects in the tested fibers. The results also show a dependence between the tensile modulus and the UTS with the gauge length as shown in Fig. 2.29.

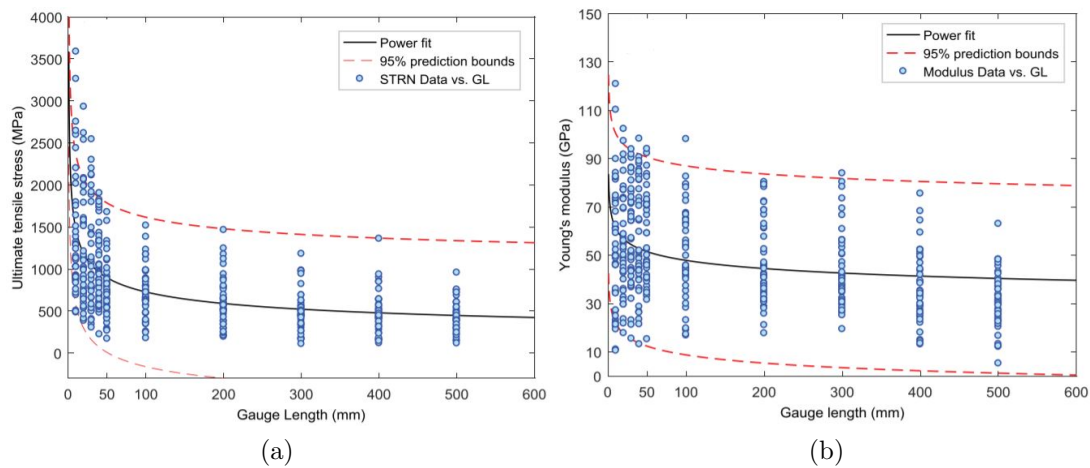


Figure 2.29: Tensile mechanical properties according to gauge lengths of the single flax fibers reported by [35].

S. Y. Nayak et al [26] assessed different chemical treatment for flax fibers and their impact on different properties as density, tensile strength or interfacial adhesion strength. First of all, they noticed an increase of the fiber density with chemical treatment. They explained this variation by the simple fact that when the fibre is chemically treated, constituents such as pectin and hemicellulose are removed from the initial fibre, which are light molecules but occupy a large volume. These constituents are also hydrophilic and therefore, with chemical treatment, adhesion can be enhanced with the matrix. Chemical treatment as Potassium permanganate KMnO_4 treatment can also help to remove some impurities present on the surface of the fibers.

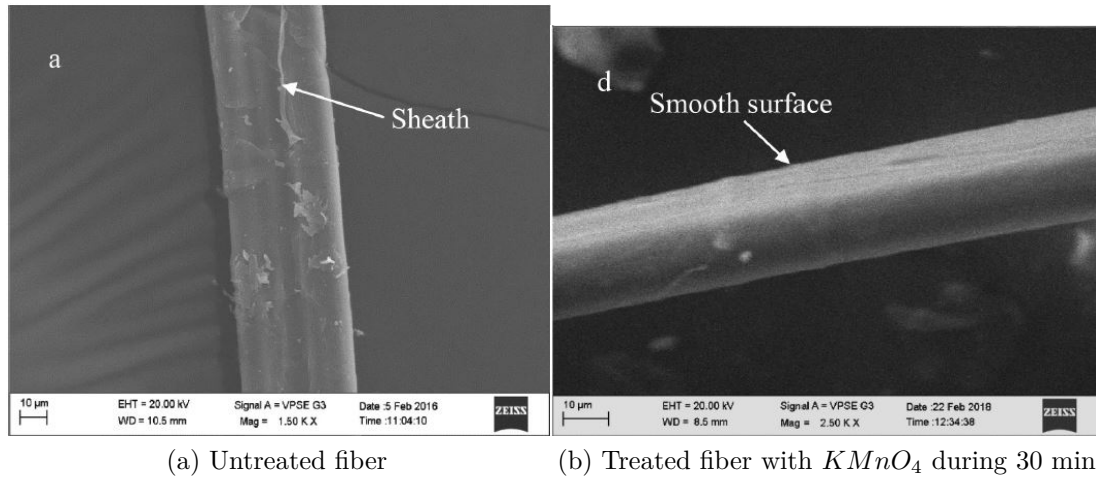


Figure 2.30: SEM images for untreated and treated fibers reported by [26].

Compressive properties

Nowadays, very few papers about compressive properties of flax fibres are available.

Bos et al. [36] assessed the mechanical properties of single flax fiber and investigated the influence of isolation technique on strength properties of fiber. Compressive strength was measured using the elastica loop technique, as shown in Fig. 2.31a. The tests were performed on elementary fibers containing small kink bands, defects induced by classical decortication process –Fig. 2.31b–, and on elementary fibers free of kink bands decorticated by hand.

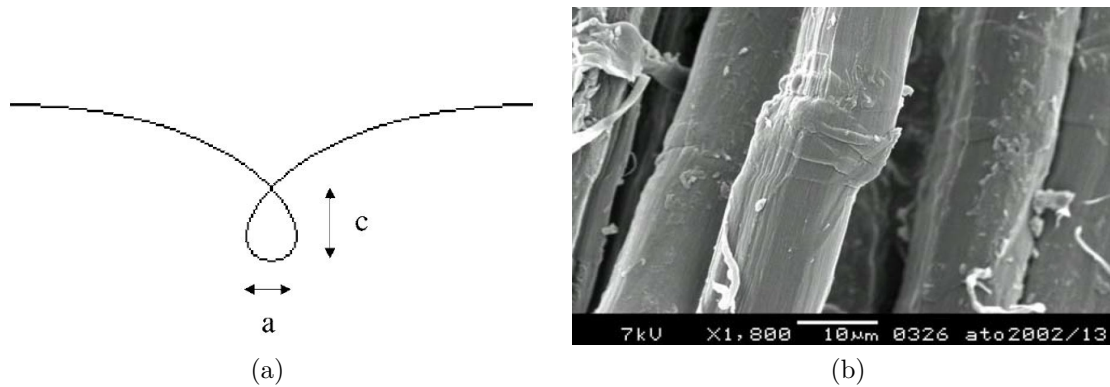


Figure 2.31: (a) elastica loop technique used for compression testing in [36] (b) Kink band in a flax fiber reported by [37] –same author.

The compressive strength of fibers measured by *Bos et al.* [36] was approximately 80% of the their tensile strength. This difference was explained by the structure of the secondary cell wall which is therefore more sensitive to kink bands.

2.4.7 Flax fiber and its composites

Mahboob et al. [38] determined tensile, compressive and shear properties of flax fiber reinforced epoxy composite. To do so, several laminates with different UD staking

were prepared with hand layup technique following heated-platen compression moulding procedure.

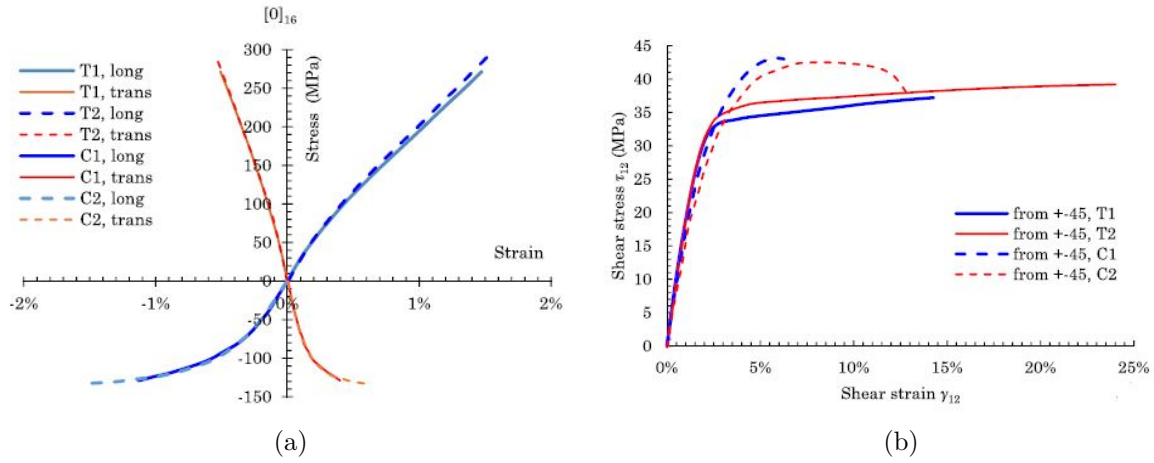


Figure 2.32: Response plots (a) for specimen $[0]_{16}$ in tension and compression in both directions (b) derived in-plane shear response reported by [38].

As shown in Fig. 2.32a, the given laminate did not response in the same way according to the loading mode or the direction of loading. The Young's modulus and the tensile strength in the direction of the fibres were respectively 31 GPa and 287 MPa while that in the direction transverse to the fibres were 5.6 GPa 34 MPa. Regarding the compressive properties, the Young's modulus and compressive strength were 30 GPa and 127 MPa in the longitudinal direction and 5.7 GPa and 80 MPa in transversal direction. Regarding the general shape of the curves, a variation in the modulus for the tensile tests was observed between 0.2-0.25% strain. They explained this change by the behaviour of the fibre itself. They determined that the variation of modulus was explained by the reorientation of the microfibrils in the fibers. The same behavior was seen for the compression curve but this time, the inflection region was between 0.2% and 0.6% strain. The In-plane shear response of the composite was derived from angle cross-ply $[\pm 45]_{4S}$ tension specimens –Fig. 2.32b. They determined that shear modulus and shear strength were 2.1 GPa and 37 MPa, respectively. They noticed that the curve in this case had also a bilinear behavior where the inflection region was about 3% strain.

Liang *et al.* [39] assessed mechanical properties of UD flax/epoxy composites with different stacking and compared it with GFRP laminates. The used fabrics consisted to two UD plies oriented perpendicular to each other and attached together with a cotton thread. Laminates were first impregnated by hand layup and then, pressed and cured in a hydraulic press equipped with heating plates. The resulting laminates and their characteristics are shown in Table 2.5.

Table 2.5: Characteristics of flax/epoxy and glass/epoxy laminates reported by [39].

Notation	Fibre	Fabric	m_{area} (g/m ²)	Stacking sequence	V_f (%)	V_p (%)	ρ_c (kg/m ³)	t (mm)
FE_0	Hermès	UD	140 (6)	[0] ₁₂	43.9	0.48	1310 (10)	2.55
FE_90	flax			[90] ₁₂	(1.5)	(0.02)		(0.12)
FE_090		Non-	235 (16)	[0/90] _{3S}	43.1	3.1	1280 (10)	2.18
FE_45		crimp		[±45] _{3S}	(0.6)	(0.3)		(0.07)
GE_090	E-glass	Non-	430 (5)	[0/90] _{3S}	42.5	5.9	1730 (30)	2.33
GE_45		crimp		[±45] _{3S}	(1.0)	(2.2)		(0.04)

In the work of *Liang et al.* [39], the volume fraction was computed by Eq. 2.23 where n , m_{area} , ρ_f and t are the number of plies, the fabric areal weight, the fiber density and the thickness respectively.

$$V_f = \frac{n \times m_{area}}{\rho_f \times t} \quad (2.23)$$

The tensile, compressive and shear properties were assessed according to ISO 527, ISO 14126 and ISO 14129 standards respectively. The results reported by *Liang et al.* [39] are available in Table 2.6.

Table 2.6: Mechanical properties reported by [39]. "The superscripts +, -, UTS, UCS and USS refer to tension or compression loading, ultimate tensile, compression and shear stresses respectively."

	E_x^+ (GPa)	ν_{xy}	σ_x^{UTS} (MPa)	ϵ_x^{UTS} (%)	E_x^- (GPa)	σ_x^{UCS} (MPa)	ϵ_x^{UCS} (%)	G_{12} (GPa)	τ_{12}^{USS} (MPa)	γ_{12}^{USS} (%)
FE_0	22.8 (1.0)	0.434 (0.084)	318 (12)	1.65 (0.05)	24.7 (0.6)	136 (2)	2.41 (0.27)	–	–	–
FE_90	4.52 (0.18)	0.084 (0.011)	26.1 (0.6)	0.622 (0.036)	5.93 (0.27)	100 (4)	3.27 (0.12)	–	–	–
FE_090	14.5 (0.8)	0.153 (0.045)	170 (20)	1.72 (0.13)	13.0 (1.2)	98 (7)	2.12 (0.20)	–	–	–
FE_45	6.48 (0.66)	0.748 (0.035)	79.5 (6.6)	3.85 (0.62)	–	–	–	1.96 (0.17)	39.7 (3.3)	6.23 (1.08)
GE_090	21.9 (1.1)	0.142 (0.004)	380 (26)	2.16 (0.16)	27.3 (2.1)	405 (16)	1.63 (0.11)	–	–	–
GE_45	11.1 (1.1)	0.483 (0.083)	103 (8)	16.1 (2.1)	–	–	–	3.44 (0.26)	51.4 (4.2)	23.84 (3.17)

Liang et al. [39] determined lower value for tensile and compression moduli than *Mahboob et al.* [38] but presented similar values for strength properties and the established stress-strain curves exhibited the same non-linear behavior. Nevertheless, the non linear behavior of specimens in compression suggested the collapse by buckling prior to material failure in the two tested directions [39]. Furthermore, this buckling could be favored by the presence of waviness in the fibers within the untested specimens, characteristic which was not found in the GFRP samples. Regarding the In-plane shear properties, they determined that the non-linear stage before failure was induced by the matrix. In their study, *Liang et al.* also assessed the variability of the results, knowing that properties of natural fibers present high scattering of mechanical properties. They measured low dispersion of results and highlighted that variability at the scale level of UD composite was lower than single fibers [39]. This decrease could be explained by the fact that composite properties are an averaging of all properties of fibers reinforcing the laminate. Therefore, fibers with high mechanical properties compensate the behavior of the weaker ones and induce a decrease in variability at laminate scale [39].

Van Vuure et al. [40] also assessed tensile and compressive properties of different natural fibers including flax. The UD laminates were done via vacuum assisted resin infusion and gave specimens with a volume fraction of 40%. Tensile and compressive tests were done according to ASTM 3039 and ASTM 3410 standards, respectively. In contrast to *Mahboob*

et al. [38] and *Liang et al.* [39], they determined lower modulus in compression than in tension for flax composite, the ratio between both being approximately 60 %.

Table 2.7: Comparison of tensile and compressive properties for flax and bamboo UD composites with equal fibre volume fractions of 40% reported by [40]

	Flax Tensile properties	Flax Compressive properties	Bamboo Tensile properties	Bamboo Compressive properties
Strength (MPa)	$222,9 \pm 6,1$	$136,9 \pm 5,5$	$254,7 \pm 18,3$	$149,5 \pm 8,9$
Modulus (GPa)	$23,9 \pm 1,9$	$15,1 \pm 2,5$	$19,5 \pm 1,0$	$15,5 \pm 2,7$

Moudood et al. [41] in addition to tensile properties investigated the flexural properties of UD flax/bio-epoxy composites. Unidirectional laminates were prepared with vacuum infusion technique. They were cured at room temperature during 24h and then post cured at 60° during 8h. The fiber volume fraction was estimated to 50% with the same equation used by *Liang et al.* [39]. The flexural properties were measured by three-point bending according to ASTM D790-03 standard. The results determined by *Moudood et al.* [41] show a flexural strength and flexural modulus around 225 MPa and 20 GPa respectively against around 360 MPa and 28GPa for tensile strength and modulus.

Factors affecting the composite properties

Baley et al [20] reported published values by several authors of tensile properties to evaluate the correlation between the fiber volume fraction and tensile properties. In Fig. 2.33a, it can be noticed that the correlation between the Young's modulus and the volume fraction is reasonable despite some uncertainties according to used flax fibers. However, the same reporting as been done and shown in Fig. 2.33b with the tensile strength and the correlation with the volume fraction is less obvious. This higher scattering could show a greater sensitivity for strength to variations in flax fibers and composites manufacturing than tensile modulus.

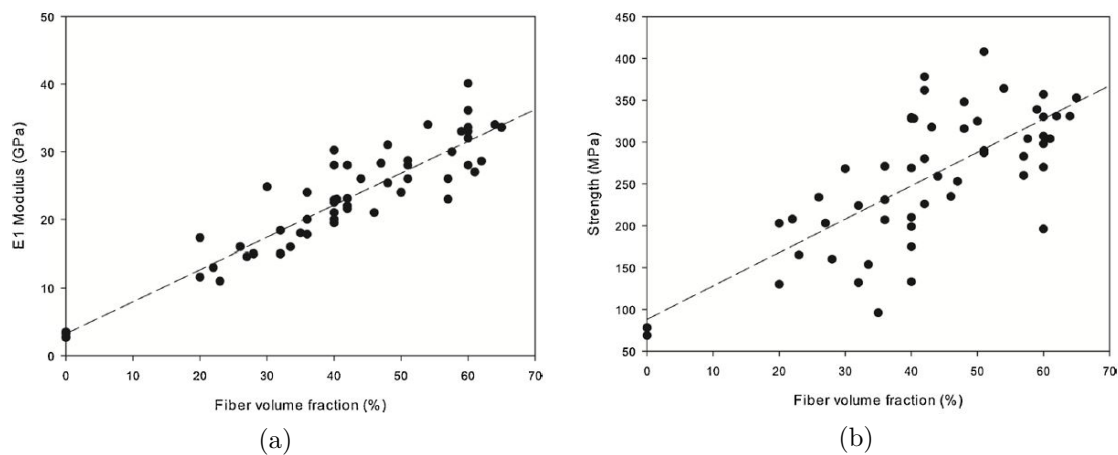


Figure 2.33: Tensile properties versus fiber volume fraction for UD flax/thermoset composites reported by [20] (a) Tensile modulus versus V_f (b) Tensile strength versus V_f .

Bensdown F. [42] studied the influence of flax fabric architecture and the twist angle of the yarns on several mechanical properties of the resulting composites.

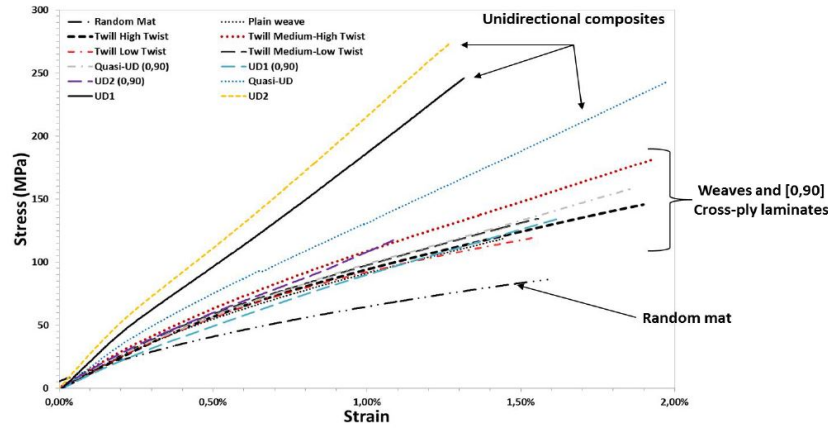


Figure 2.34: Tensile stress-strain curves for flax/epoxy composites for all studied architectures by [42].

As shown in Fig. 2.34, UD laminates studied in this work presented higher stiffness and tensile strength compared to other architectures and the same behavior was reported for flexural properties. The weaves and [0,90] cross-ply laminates are rather close to each other for tensile properties [42]. However, better flexural properties were reported for [0,90] cross-ply laminates comparing to woven fabrics. The difference between the results shows that crimp could have an impact on quasi-static mechanical properties especially flexural properties.

Yan et al. [9] assessed the improving of mechanical properties of FFRP by applying alkali treatment on the fibers. They firstly evaluated the influence of alkali treatment on the fiber itself then they did the same for the associated composite. Regarding the single fibre, they extracted a single yarn from a commercially available fabric. They washed it with fresh water to remove contaminants and then dried the yarn during 48h. The yarn was next washed in 5wt% NaOH solution[9]. The same procedure was done on fabrics. This treatment had the aim to disrupt hydrogen bonding and increase the surface roughness. This treatment also removes constituents as lignin and waxes. Furthermore the apparition of hydrogen groups could enhance the compatibility of the fabrics with the epoxy matrix. The composites were fabricated with vacuum bag technique and then first cured at room temperature during 24h followed by a second curing at 65°C for 7h [9]. Regarding the single-strand yarn, they noticed a decrease of tensile properties for treated flax fibers except for the elongation at break which is a bit higher. They also observed that flax yarns and flax monofilament did not have the same failure stress. They explained this distinction by the difference of mechanism failure between the both. While the fibre followed the sequence already explained in section 2.4.6, the tensile failure of yarn was a set of fibre slippage and fibre breakage [9]. They justified the tensile properties decrease of the yarns by the fact that alkali treatment removes constituents which have the role of being the cement of the fibre. Indeed, the removal of these, the stress transfer between the cellulose microfibrils is less efficient than before [9]. Regarding the composite products, the first benefit of alkali treatment was the rougher surface of the fibers. Indeed, this modification allowed a better adhesion between the fibers and the epoxy matrix [9].

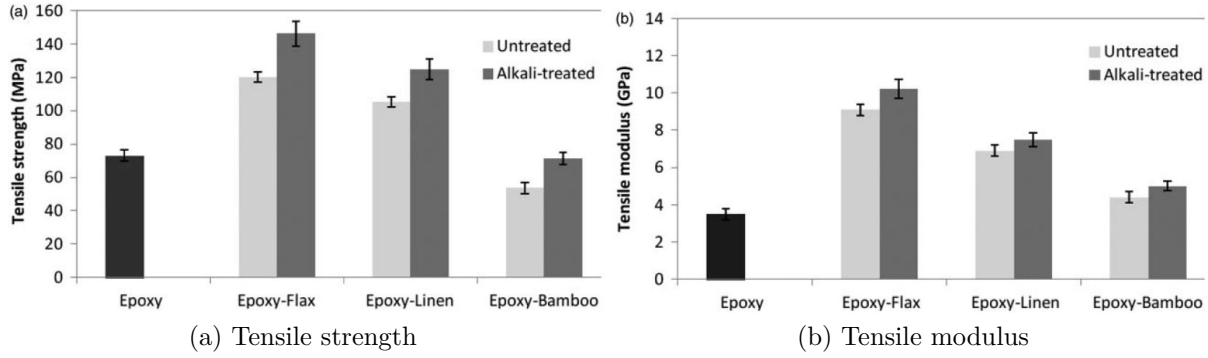


Figure 2.35: Tensile properties of untreated/alkali-treated flax and other natural fabric reinforced composites compared to epoxy resin [9].

Figs. 2.35a and 2.35b show the variations of tensile strength and tensile modulus for the different studied NFRP, respectively. As expected, they obtained better properties with the addition of fibers compared to epoxy alone and in the contrary of single yarns, the tensile properties of the composites are improved with alkali treatment for all type of fibers. They also compared experimental values for untreated fibers/epoxy with theoretical values by applying rule-of-mixtures. They noted that experimental properties are lower than the theoretical ones. They explained this difference by the overestimation of the theoretical values. Indeed, the rule-of-mixture does not take into account defects that can appears during the preparation of composites, such as the misalignment of the fibers, the adhesion fibers/matrix and the presence of transversal yarns [9].

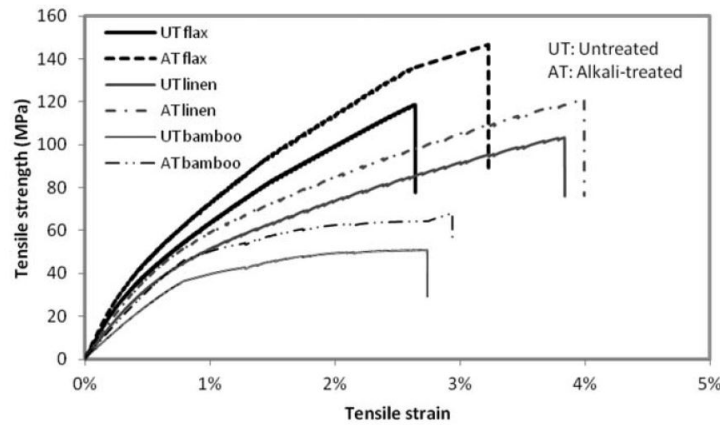


Figure 2.36: Typical tensile stress-strain curves for untreated/alkali-treated flax, linen and bamboo fabric reinforced composites reported by [9].

Fig. 2.36 allows the comparison of the tensile stress-strain curves of the different composite products [9] where all curves follow the same tendency. Furthermore, by comparing the results of *Yan et al.* [9] with the results of *Mahboob et al.* [38], although the curve has the same shape, the leading values are not identical. It may be explained by the used matrix, the weave mode of fibers plies or even the flax species. For results presented by *Yan et al.* [9] in Fig. 2.36, the curve ends with a brutal drop of the stress which indicates a brittle failure of the composite.

Baley et al. [21], in addition to the evaluation of the impact of drying on a single fiber have also assessed the tensile properties of UD composites with dried fibers and epoxy. Specimens were prepared by compression molding and first cured at room temperature to avoid water removal within the undried fibers and then post-cured for 12h at 65°C. Specimens with both fibers were tested in tension according to ISO 527.

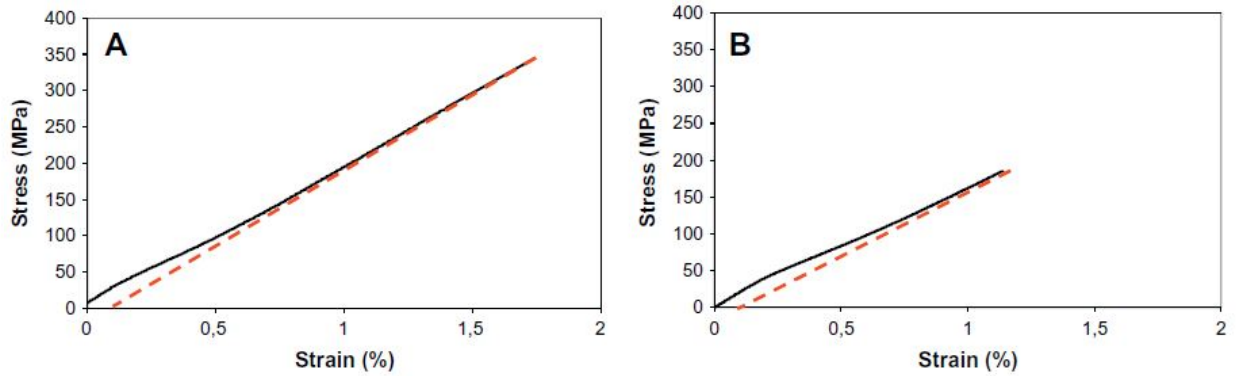


Figure 2.37: Tensile stress-strain curves for Epoxy UD-composites reinforced by (A) untreated fibers (B) dried fibers reported by [21].

Fig. 2.37 shows the results obtained by *Baley et al.* [21]. As in other studies [9, 38], UD composites presented the same tensile behavior than single fiber. With a similar fiber volume fraction of 40% for both specimens series, they obtained the same tensile modulus around 22.5 GPa for "as-received" and dried fibers. As for single fibers, the drying treatment did not affect stiffness of the final product. Nevertheless, they noted a significant degradation of the properties at failure with an UTS of 210 MPa for dried fibers against 330 MPa for untreated fibers. They determined that this degradation of the mechanical properties at failure of the fibers during drying was transferred to the UD composite. However, they also specified that because of the complex structure of the flax fibers, other damage mechanisms such as cracks and fibers aggregation specific to the natural fiber may influence the general behavior of the composite [21].

Li et al. [43] assessed the influence of porosity on mechanical properties of FFRPs by varying several processing parameters. Before the curing of the composites, flax UD were dried at 120°C for 2h. Each ply was brushed separately with epoxy resin and then dried at 60°C during 30 min to obtain prepegs. The used curing cycle in the hot press is shown in Fig. 2.38 with the set parameters for the six prepared laminates in Table 2.8 [43].

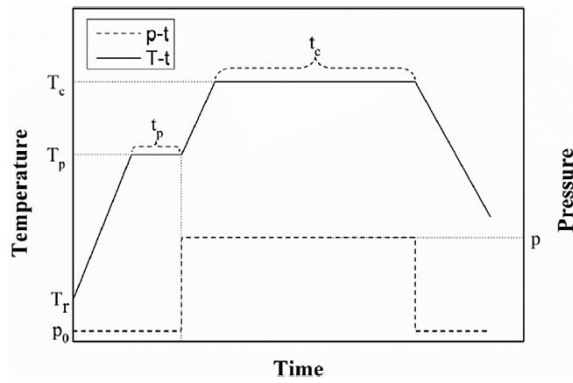


Figure 2.38: curing cycle reported by [43].

Table 2.8: Processing parameters for manufacturing FFRPs laminates by hot press reported by [43].

Laminate number	T_p ($^{\circ}\text{C}$)	t_p (min)	p (MPa)	T_c ($^{\circ}\text{C}$)	t_c (h)
1	90	30	0.5	125	2
2	90	30	1.0	125	2
3	90	30	1.5	125	2
4	90	20	1.0	125	2
5	90	40	1.0	125	2
6	100	20	1.0	125	2

First of all, *Li et al.* [43] established that void formation in the flax composite differed from CFRPs and GFRPs because of the rough surface and the twisted structure of the flax yarns. The void may be localized at the interface between the fiber and the matrix, between the yarns and within the yarn itself and may impact mechanical properties and the failure mechanism. They also determined that increasing the curing pressure allowed to decrease the void content and laminates with low void content had high tensile strength and interlaminar shear strength as shown in Table 2.9[43]. Furthermore, the localization of the voids changed also with curing pressure and they were delocalized from interface and between yarns to inside the yarns that may impact failure mode. *Li et al.* [43] also evaluated that properties of FFRPs were more sensitive to void content than carbon and glass.

Table 2.9: The mechanical properties of specimens obtained from several laminates with different curing parameters reported by [43].

Laminate number	1	2	3
ν_f (%) (By Microscope)	2.42 ± 0.11	1.53 ± 0.05	1.51 ± 0.04
Tensile strength σ_t (MPa)	172.08 ± 17.69	205.56 ± 8.14	218.45 ± 7.87
Tensile modulus E_t (GPa)	22.25 ± 1.29	22.67 ± 0.47	23.52 ± 0.30
Interlaminar shear strength σ_{ILSS} (MPa)	29.99 ± 6.36	32.89 ± 3.93	33.18 ± 3.26

El Sawi et al. [44] studied the influence of the manufacturing process on mechanical properties of FFRPs. To do so, laminates were manufactured with compressive molding or autoclave processes with adapted curing cycles. Then tensile, bending and in-plane shear tests were performed on samples. In addition, authors conducted moisture absorption test. Regarding the results, a difference in porosity was noticed between compression molding with two different curing cycles and autoclave process. In fact, autoclave process allowed a lower porosity because of the vacuum which helped evacuating the entrapped air. This void content also impacted the moisture absorption which was higher for samples manufactured with compression molding. However, composite materials showed similar properties despite the different manufacturing processes except for in-plane shear for which the autoclaved samples had higher stiffness. But all specimens were experiencing a drop in their properties after water absorption. In conclusion, FFRPs are highly hydrophilic

and the water uptake can be up to 18% for compression molding, behavior which is not desirable for many applications.

2.4.8 Flax with InfuGreen 810 by Sicomin

Amaro J. [45] studied the effects of several surface treatments to optimize the adhesion and compatibility between flax fibers and InfuGreen 810. To obtain data, different tests were performed on untreated fibers, aminosilane-treated or washed with one or two different treatments selected from literature [45]. First, Wilhelmy technique was used to investigate the changes in physicochemical interactions for flax fibers. In addition, XPS analysis was done to examine the changes of surface composition. The microbond tests were performed to characterize the influence of these parameters on the adhesion fiber-matrix. Only amino-treated fibers showed better interface properties and washing methods seemed to worsen interface strength [45].

2.5 Carbon fibers

2.5.1 Generalities

Today, carbon fibre is the most widely used fibre in applications requiring high performance materials such as the aeronautical sector. This fiber has the benefit to provide high strength to weight ratio and begins to replace more conventional materials such as metals. Other advantages are the low density, high corrosion and fatigue resistances [46].

2.5.2 Manufacturing process

Depending on the given application, the carbon fiber can be manufactured in several ways. This work will only focus on carbon fiber for reinforcement applications. Regarding the fiber production, it exists two types of precursor: mesophase-pitch carbon fiber and polyacrylonitrile –PAN– precursor [47].

Mesophase pitch precursor

Mesophase pitch is firstly obtained by thermal treatment of isotropic pitch. The treatment leads to the condensation of the small polyaromatic molecules constituting the isotropic pitch. This reaction must take place under inert atmosphere at temperature between 350 and 450°C [47]. There is therefore a growth of the mesophase within the isotropic pitch until it becomes the main phase of the medium. The obtained mesophase pitch can provide fibers with high tensile modulus.

After chemical transformation into mesophase pitch, this latter one can be spinning and stretching. these steps have the aim to orient the structure and to obtain fibers. The main difficulty associated to this step is the viscosity of the mesophase. Indeed, the viscosity of the product after thermal treatment is proportional to the amount of mesophase and for high performance fibers. A high amount of mesophase is preferred, but a high viscosity leads to a difficult stretching. To overcome this, it is possible to work with higher temperature but there is a risk of product transformation. Therefore, the pitch is heated between 350 and 450 °C and pass trough a spinneret in cone shape with a diameter

between 0.1 and 0.4 mm at the outlet. Then the filament is stretched to a size of 300% to 500% of its initial size, and finally spooled [47].

After the spinning and stretching, there is oxidation step to make the mesophase infusible. This step is done in presence of oxygen at temperature between 150 and 250°C.

After the precursor production, the fibers are carbonised under inert conditions at 1500°C to obtain fiber carbon [47]. An additional graphitisation step can be done with temperature higher than 2000°C to obtain higher properties.

PAN

As cited at the beginning of this chapter, this technique include polyacrylonitril chain as precursor. First, the fibers are heated up to 300°C to induce the cyclisation of carbonitril groups –Fig. 2.39a. After the cyclisation, the obtained reactant is once again heated but this time at higher temperature to transform the cycles into pyridine aromatic groups due to the loss of a hydrogen on a carbon –Fig. 2.39b. It occurs during this step a release of dihydrogen –Fig. 2.39c. Then, the temperature increase between 400 and 600°C to combine adjacent chains and form ribbon. Once again, there is a release of dihydrogen during this step.

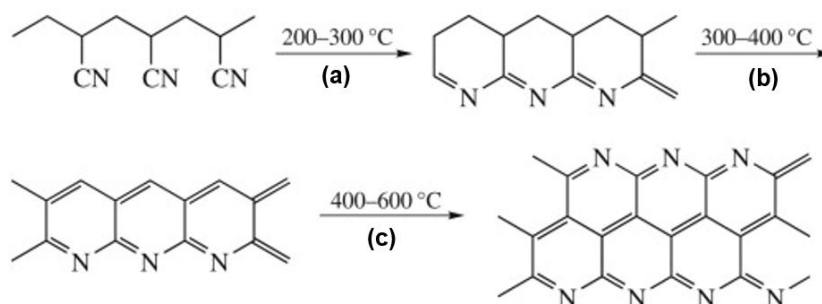


Figure 2.39: Graphical abstract of the manufacturing process for carbon fiber from PAN precursor [47].

To enhance the production by this technique, PAN precursor can be stabilized by oxidation step to allow the thermal stabilisation of the product. This step convert the PAN fiber into an infusible polymer by converting $\text{C}\equiv\text{N}$ into $\text{C}=\text{N}$ [48].

2.5.3 Carbon fibers and its composites

Paiva et al. [49] evaluated and compared some mechanical properties of four different Carbon/Epoxy composites. To do so, the authors used two epoxy matrices, a typical one –F155– and an other toughened with thermoplastic –F584. Regarding the fibers, two fabrics were used, a plain weave fabric –PW– and a, "Eight harness satin" –8HS. Laminates were manufactured by autoclave with different curing cycles depending on the used matrix. Then flexural, shear and compressive tests were performed according to ASTM D790, ASTM D2344 and ASTM D3410, respectively. The results obtained by the authors for flexural and interlaminar shear strengths are available in Table 2.10 and compressive properties in Table 2.11.

Table 2.10: (a) Flexural strength (b) Shear strength of manufactured laminates reported by [49].

(a)		(b)	
Composite	Flexural Strength (MPa)	Composite	Interlaminar Shear Strength (MPa)
F155 / PW	837.9 $\pm$ 23.5	F155 / PW	63.9 $\pm$ 1.8
F155 / 8HS	792.4 $\pm$ 30.8	F155 / 8HS	63.5 $\pm$ 3.5
F584 / PW	1288.5 $\pm$ 45.4	F584 / PW	84.8 $\pm$ 5.2
F584 / 8HS	1119.1 $\pm$ 43.2	F584 / 8HS	72.2 $\pm$ 4.8

Table 2.11: Compressive strength of manufactured laminates reported by [49].

Composite	Compressive Strength (MPa)
F155 / PW	533.6 $\pm$ 29.5
F155 / 8HS	545.5 $\pm$ 26.2
F584 / 8HS	728.8 $\pm$ 30.5

Authors noted flexural and shear properties were more impacted by the used matrix than the fabric. F584 composites showed higher properties than F155 ones because of the presence of thermoplastic toughener which better prevents crack propagation. The same behavior was present for compressive properties.

Rahmani et al. [50] investigated the effects of some parameters as the resin type, the number of plies and the orientations on carbon/epoxy composites. For this experiment, they used UD carbon with 3 types of epoxy resin and varied the orientation between 0°, 35°, 45° and 90°. The results showed tensile properties were highly dependant of laminate orientation and less of matrix type. Regarding flexural properties, the authors noted a difference in properties according to the number of plies but not significant.

Eksi S. and Genel K. [51] compared mechanical properties of composites reinforced by UD and woven carbon fibers. First, laminates were manufactured with hand lay-up process and then cured during 24h at room temperature before a post-curing at 75°C. Tensile, compression and shear tests were assessed according to ASTM D3039, ASTM D3410 and ASTM D3518, respectively. Results are given in Table 2.12.

Table 2.12: Mechanical properties for laminates determined by [51].

Reinforcement type	Volume of fiber V_f	Density [g/cm ³]	Elasticity module [MPa]	Shear module [MPa]	Poisson ratio [-]	Tensile strength [MPa]	Tensile strength/density	Shear strength [MPa]	Compression strength [MPa]	Elongation at break
Woven glass	30	1.55	14 352	4728	0.24	220	141.9	119	96	0.016
Woven aramid	30	1.2	19 087	2585	0.38	357	297.5	53	64	0.019
Woven carbon	30	1.31	42 000	12350	0.32	340	259.5	180	118	0.009
Unidir. glass (0°)	30	1.55	18 300	3895	0.25	432	278.7	30	71	0.028
Unidir. glass (90°)	30	1.55	7 940	3895	0.17	52	33.5	30	16	0.0096
Unidir. carbon (0°)	30	1.31	78 715	2195	0.4	826	630.5	20	118	0.0100
Unidir. carbon (90°)	30	1.31	4 930	2195	0.25	37	28.2	20	27	0.0130

In accordance to other studies, results showed the higher performance of UD carbon laminates compared to woven carbon except for shear properties. In addition, carbon presents better properties than other synthetic fibers as glass and aramide.

Chapter 3

Scope of the work

In this chapter, the main objectives of this work will be defined and will allow to introduce the next sections such as the used materials and methods to reach the results described later on.

This work is part of the MACOBIO project and is a continuation of the work produced by Charline Van Innis and Noah Jost, whose aim was to characterise the InfuGreen 810 epoxy resin marketed by SicominTM. MACOBIO is a project that is part of the FEDER projects, more specifically with the "Low Carbon Footprint Materials" project that aims to develop innovative bio-sourced 100% materials. The current research conducted by the universities and research centres associated with the project aims to transfer this knowledge to industry to enable them to move towards the use and manufacture of low carbon footprint materials in the future.

As described in the MACOBIO project sheet, the use of polymers such as composites is constantly growing in many fields and it is essential to introduce the development of sustainable and ecological materials in view of the increasing scarcity of oil.

The development of bio-sourced composites has focused on the study of natural reinforcements such as flax, hemp or basalt. The objective is to find an alternative to the reinforcements currently used, which manufacture and use are harmful to the environment and sometimes to human health. However, many problems still arise with the use of bio-sourced reinforcements because of their poor mechanical and physico-chemical properties such as their hydrophilic character or their low thermal stability.

In order to advance the MACOBIO project, this work will essentially focus on the introduction of bio-sourced reinforcements into the InfuGreen 810 resin previously studied and to extract certain mechanical properties from the composite produced. The fibres studied will essentially be flax fibres, presented in several different fabrics. A more succinct study will also be made on carbon fibres. Furthermore, this work is in close collaboration with the work of Cyril Vander Borgh [52] whose studied the same properties but for basalt fibers. Although the carbon reinforcement is not part of the current project itself, these fibres are the most widely used in the industry today, it is therefore interesting to make a comparison with them.

This work will therefore be divided into several stages in order to achieve the objectives set. The first step is the manufacture of the plates using the selected fibres and the

InfuGreen 810 resin. During this one, several parameters will be modified in order to see their influence on the manufacturing process itself but also on the properties of the final product. The materials and methods used will be described in Chapter 4.

Then, several tests will be started on extracted specimens from the raw manufactured laminates to characterize the composite material. Selected tests were chosen according to their relevance and the time that was allotted, a prioritisation had to be made in order to be able to recover the most interesting properties first. As the process methods, all the steps for these tests are described in Chapter 4.

To conclude this work, a final discussion will be done to go through all the results obtained and to compare them with the previous work carried out on Sicomin resin and to compare the different reinforcements studied with each other. A short discussion on the processes used will also be made to compare them with the processes currently used in industry.

Chapter 4

Materials and methods

4.1 Materials

4.1.1 Bio-sourced epoxy resin

The used resin in this work is the epoxy resin InfuGreen 810 commercialized by SicominTM with the hardening SD4471 extra slow. As for *Van Innis C.* [13] and *Jost N.* [16], the used proportion of the two products are 77.5% and 22.5% for resin and hardening respectively. Although this resin is defined as biosourced, there is less than 30% of natural carbon in the final product. Main specifications are presented in Table 4.1.

Table 4.1: SR InfuGreen 810 system technical data.

	InfuGreen 810	SD 4471
	<i>Epoxy resin</i>	<i>Hardener - Extra slow</i>
Aspect	Clear liquid	Light yellow liquid
Viscosity (20°C)	200	11
Carbon green content	38%	/
Density (20°C)	1.16	0.944
Storage Stability	24 months	24 months

4.1.2 Fibers

Flax fibers

For laminates with flax fibers, three different reinforcements were used in this work. First, because of the lack of materials, two different types of UD were used as named in this work as "UD OLD" and "UD NEW". These two UD had the same surface density of 150 g/m² but did not have the same fiber yarns structure as shown in Fig. 4.1a.

Used flax fabrics were in the form of 2/2 twill as illustrated in Fig. 4.1b and had the same surface density as the UD reinforcements. The fabrics cam from the same supplier as the "UD new" roll and had the same yarn type as the latter.



Figure 4.1: Used flax reinforcements for this work (a) strand structure for both UD reinforcements: "UD OLD" strand is an assembly of non-twisted fibers while "UD NEW" strand is a twisted yarn (b) flax twill fabric structure. The strands are twisted yarns as well.

Carbon fibers

Carbon fibers for this work were provided by ISOMATEX S.A. . These fibers are presented in plies of 200x200 mm² woven in the form of satin 5. Main specifications are available in table 4.2.

Table 4.2: Main characteristics of carbon fabrics (G0926).

Type of yarns	Nominal weight	Nominal construction	Weight distribution	Thickness
TENAX E HTA 40 E13 6K	375 g/m ²	4.6 yarns/cm	warp/weft 50%/50%	0.38mm

4.2 Samples preparation

In this work, different processes took place for samples preparation with different parameters. The first plates will be named as "small" laminates in the following part of this work. The second ones are called "RTM" plates. All laminates were stacked with the hand layup technique but with a different curing cycles because of temperature and pressure conditions.

4.2.1 Small laminates

For the small plates, two molds were used with different dimensions, 10x10 cm² and 20x20cm². These molds consisted of an assembly of movable parts composing the bottom of the mold and an additional counter plate to close it and apply pressure during curing.

Before laminate manufacturing, the mold is treated first with acetone to remove dust and resin remains. Then all parts of the molds are coated two times with FREKOTE release agent with an interval of 15 minutes.

The plies are one by one impregnated with the blend Infugreen 810/SD 4771 using a brush –Fig.4.2a. If necessary, a final pass with a roller is made to homogenize the impregnation

and extract the visible air bubbles –Fig. 4.2b.



Figure 4.2: Impregnation of small laminates using the hand layup technique.

When the stacking is complete with the desired plies orientation, the product is first covered by a teflon sheet and finally the mold is closed and a pressure is maintain with clamps to remove the resin excess.

Ambient curing

For this work, one of the cycles studied by *van Innis C.* [13] and *Jost N.* [16] was used for the curing of small laminates. It consists to cure the composite at ambient temperature during at least 36h up to atmospheric pressure. Then, a post-curing is performed at 80°C during 4h with a temperature ramp of 0.5°C/min to avoid stress apparition in the product. Before the post-curing step, the manufactured plate was removed from the mold.

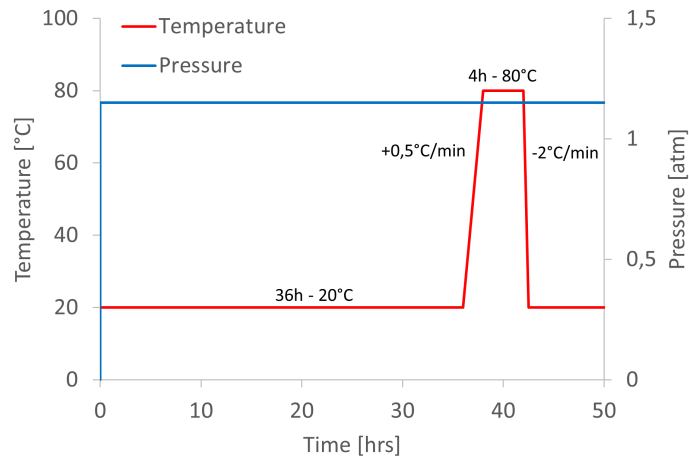


Figure 4.3: Scheme of the used curing cycle for small laminate samples 1) curing step at room temperature for at least 36h 2) Heating step with an increase of 1.5°C/min 3) Post-curing at 80°C during at least 4h 4) gradual return to room temperature. A pressure up to 1 atm is applied on the mold.

Additional steps

For the comparison of the final specimens not only according to the used reinforcement, the idea was to vary some parameters during the preparation of the laminates. The first one was to dry the flax plies before the impregnation. To do so, the plies were dried at temperature of 70°C during 2-3 hours. A second parameter was the degassing of the resin. The first step was to degas the resin itself during 15 minutes and after the laminate preparation, degassing the overall plate during 15 minutes before closing the mold.

4.2.2 RTM laminates

Because of the lack of knowledge about the InfuGreen 810 and to avoid damage to the injection piston, the second method consisted to create a hybrid technique between the RTM and the hand layup techniques. Indeed the mold was used to produced 50x50cm² but the resin injection machining was not exploited. Therefore, the plies were stacked and impregnated manually on the treated mold. Nevertheless, in addition to being able to manufacture larger plates, the benefit to use RTM machining was the access to air extraction process and the possibility to apply 9 bars pressure on the laminate during the curing.



(a)



(b)

Figure 4.4: (a) Impregnation of RTM laminates using the hand layup technique (b) RTM device.

Before laminate manufacturing, the mold was treated to enhance the surface of the mold. First of all, all parts in contact with the fibers and resin were cleaned with acetone to remove impurities such as dust or resin residues. Then the following steps were done:

1. The parts were cleaned with WaterCleanTM marketed by Zyvac®, rinsed with water and dried to remove stubborn residues from previous uses;
2. Parts were coated with SealProofTM pmarketed by Zyvac®, a first layer is applied and after 30 minutes, a second one was done. This step had the aim to enhance the surface quality of RTM parts.
3. One hour after the previous step, 2 layers of release agent from Zyvac® were applied with 15 minutes between the both.

For this process, the flax plies were systematically dried before impregnation. For the curing of RTM plates, the used cycle with its parameters is shown in Fig. 4.5. The extraction of the resin excess was done during the first part of the curing cycle and arbitrary stopped when the quantity of the removed resin was enough.

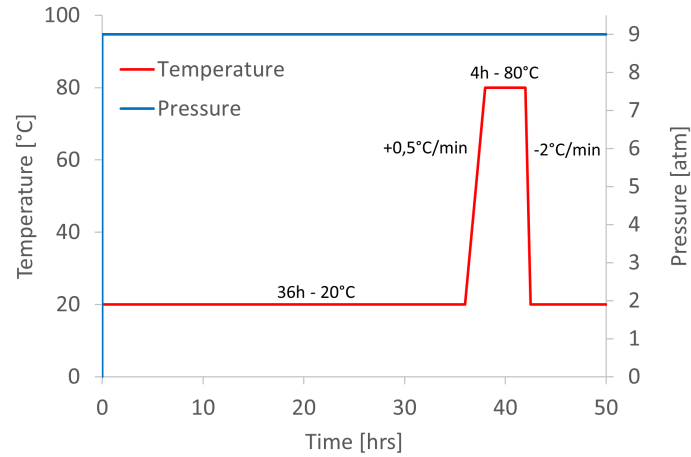


Figure 4.5: Scheme of the used curing cycle for small laminate samples 1) curing step at room temperature for at least 36h 2) Heating step with an increase of 1.5°C/min 3) Post-curing at 80°C during at least 4h 4) gradual return to room temperature. A pressure of 9 atm is applied on the mold.

4.2.3 Laminates ID

In order to facilitate the identification of the specimens during mechanical tests, all laminates were identified according to their size, their reinforcement material, their number of plies and the reinforcement architecture. A descriptive example is available in Fig. 4.6. All identified manufactured laminates with their parameters will be summarized at the beginning of the chapter.

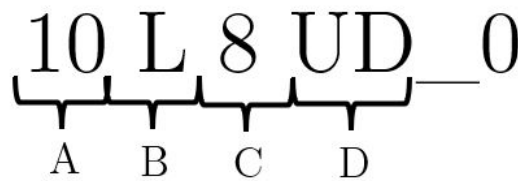


Figure 4.6: Manufactured laminates designation (A) Identification of the used mold. For small laminates this number can be either 10 or 20 depending of the mold area. For laminates manufactured with the RTM mold, the used code is 'RTM' (B) Identification of the reinforcement material. The letters 'L' and 'C' are used for flax and carbon, respectively. This letter is not present for RTM laminates (C) Number of plies in the laminate (D) Reinforcement architecture. 'UD', 'S' and 'G' are for UD fabric, twill fabric and G0926 fabric, respectively. The last digit is incremented if several laminates present the same characteristics.

4.2.4 Samples cutting

To obtain the specimens needed for testing from the manufactured laminates, a cutting step was necessary. Two methods were available: milling was used for small UD laminates while water cutting was necessary to cut carbon. RTM plates were also cut with water because of their sizes and twill laminates as well just for organisational reasons. Milling was performed on site but water cutting was out-sourced by the company M-Kut, located in Taminés.

To assess the reliability of laminate properties, three to four samples for the same test were extracted from each laminate.

4.3 Mechanical testing

In this section, the selected mechanical tests to characterize the laminates are described with the extracted properties according to the associated standard.

4.3.1 Samples preparation

Before the tests, it is important to classify each specimens according to the original plate and the test. To do so, each sample must be clearly nominated and separated for the next steps to avoid any confusion between tests or batches. One set contains the samples for a single type of test and from only one laminate.

After nomination, the sample dimensions are encoded by taking the average of 3 measurements for thickness and width. It is needed for the test set up but it also gives a first appreciation of manufacturing and cutting quality by measuring the dimensions variation.

4.3.2 Tensile test

For tensile properties, when it was possible, ASTM D3039 standard requirements was followed for testing. The required dimensions were possible only for RTM laminates and not for small laminates. For polymer reinforced with continuous fibers, the samples shape is rectangular and the dimensions varied as following according to the laminate dimensions.

Table 4.3: Samples dimensions for uniaxial tensile test.

Laminate	length L [mm]	width [mm]
10x10	80	12
20x20	190	15
RTM	250	15

Due to the lack of time, even if it is highly recommended, no tabs were set on specimens.

For testing, different parameters must be set such as the speed, the initial gage length, the position of the grips and the pre-load. This last parameter is important to remove any backlash in the assembly before the start of the loading to avoid to avoid distorting the results at the beginning of the curve.

Table 4.4: Parameters for uniaxial tensile test according to ASTM D3039.

Specimen	Gauge length [mm]	Pre-Load [N]	Speed [mm/min]
10x10	20	10	2
20x20	50	30	2
RTM	75 or 90	30	2

The tensile testing machine was a ZwickRoell 50kN for all samples except for carbon specimens from the RTM plate which were tested with a ZwickRoell 250kN. The temperature and humidity conditions were set at 23°C and approximately 40% , respectively and are similar for the other tests.



Figure 4.7: Installation for tensile test. The sample is maintained by the grips and each extensometer arm is touching sample surface at one point during loading.

For the results, stress and strain must be computed according to the following equations:

$$\sigma_i [MPa] = \frac{F_i}{A} \quad (4.1)$$

$$\epsilon_i [-] = \frac{\delta_i}{L_g} \quad (4.2)$$

where F_i is the standard force in newtons, A is the cross-section in mm^2 , δ_i is the extensometer displacement in mm and L_g is the initial extensometer gage length in mm. The tensile modulus is computed between the two strain values $\epsilon_1=1000\mu\epsilon$ and $\epsilon_2=3000\mu\epsilon$ following:

$$E [GPa] = \frac{\sigma_2 - \sigma_1}{\epsilon_2 - \epsilon_1} \quad (4.3)$$

Furthermore, knowing the thickness of each samples and the number of plies, an estimation of volume fraction will be made first according to Eq. 2.23.

4.3.3 Compression test

For compression tests, these were made according to ASTM D6641 with the ZwickRoell 50kN. Because of the needed assembly parts and the risk of buckling, it was not possible to extract specimens from 10x10 laminates and neither 20x20 laminates of 4 plies. For

compression, the required geometry is also rectangular and the dimensions are 140 mm and 12 mm for length and width, respectively.

For compression samples, gauges were glued to the specimens to allow the recording of the deformation data during the test. They must overlap several warp and weft fibers to obtain a mean value over the surface. For this work, simple gauges were used instead of double and therefore, the poisson's coefficient was not recorded. Once the gauges were installed and connected, the sample was carefully placed in the assembly which was tighten using a torque spanner to apply a load of 2.7 N to each screw. The parameters were set as indicated in Table 4.5. The gauge length is defined as the length let free between the grips.

Table 4.5: Parameters compression test according to ASTM D6641

Specimen	Gauge length [mm]	Pre-Load [N]	Speed [mm/min]
All	13	5	1.3

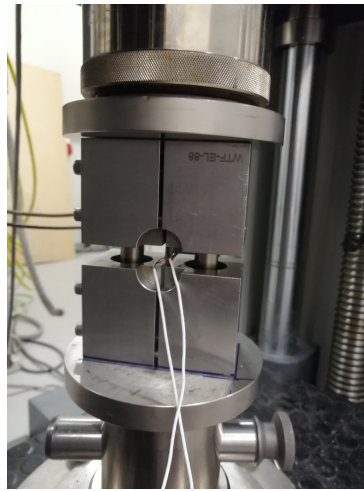


Figure 4.8: Installation for compression test. The samples is maintained in the mounting blocked and connected to the computer with the two gauges. The upper part of the testing device is moving and set under load the sample.

Regarding the results, stress and strain were computed according to the following equations:

$$\sigma_i [MPa] = \frac{F_i}{A} \quad (4.4) \quad \epsilon_i [-] = \frac{\epsilon_{g1i} + \epsilon_{g2i}}{2} \quad (4.5)$$

where F_i is the standard force in newtons, A is the cross-section in mm^2 , ϵ_{g1} and ϵ_{g2} are the signal deformation in $\mu\text{m}/\text{m}$ given by the gauges.

The tensile modulus is computed between the the two strain values $\epsilon_1=1000\mu\epsilon$ and $\epsilon_2=3000\mu\epsilon$ following:

$$E [GPa] = \frac{\sigma_2 - \sigma_1}{\epsilon_2 - \epsilon_1} \quad (4.6)$$

Modulus in compression must be validated with the help of the following equation:

$$B_y [\%] = \frac{\epsilon_{g1} - \epsilon_{g2}}{\epsilon_{g1} + \epsilon_{g2}} * 100 \quad (4.7)$$

This parameter allows to check if the bending of the sample is not excessive and must not be above 10% to validate the resulting modulus.

4.3.4 Bending test

Specimens were tested by three-point bending following the standard ISO 178 for all samples. Those were cut to the dimensions $L_s = 40mm$ and $w = 25mm$. When possible, the span between the supports respected the ratio $L = (16 \pm 1)h$ where L is the span length and h the thickness. The test speed was set to 1mm/min.

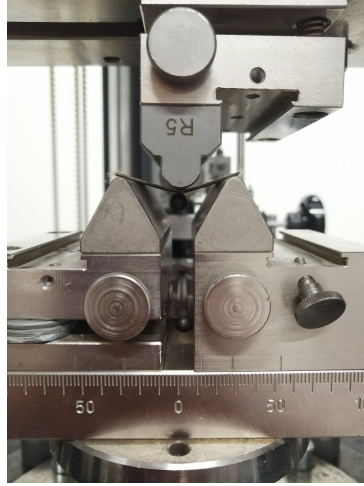


Figure 4.9: Installation for three-point bending test. The bottom part with the two cylinders is stationary while the upper cylinder load the sample.

For the results, the stress and strain were computed according to the following equations:

$$\sigma_i[MPa] = \frac{3F_i L}{2wh^2} \quad (4.8) \quad \epsilon_i[-] = \frac{6\delta_i h}{L^2} \quad (4.9)$$

where δ is the deflection of the sample in mm. The flexural modulus is computed between $\epsilon_{f1} = 500\mu\epsilon$ and $\epsilon_{f2} = 2500\mu\epsilon$ following:

$$E_f[GPa] = \frac{\sigma_{f2} - \sigma_{f1}}{\epsilon_{f2} - \epsilon_{f1}} \quad (4.10)$$

4.3.5 Interlaminar shear stress test

First tests were assessed according to the standard NF EN 2377 for flax samples and EN 2563 for carbon samples. The two standards follow the same test method but with different support diameters. All specimens were cut to the dimensions $L_s = 20mm$ and $w = 10mm$ and the test speed was set to 1mm/min.

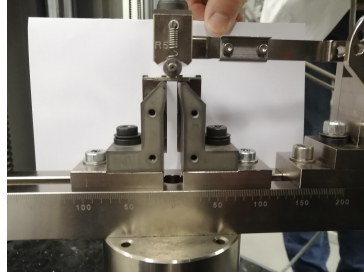


Figure 4.10: Installation for interlaminar shear strength test. The bottom part with the two cylinders is stationary while the upper cylinder load the sample.

For this test, the apparent interlaminar shear strength can be computed according to the following equation:

$$\tau[MPa] = \frac{3F_{max}}{4wh} \quad (4.11)$$

where F_{max} is the maximal load at the first failure in Newtons and h the thickness in mm.

For this test, the failure mechanism is needed to validate the results. If the specimen breaks in single or multiple shear, the apparent stress calculated with Eq. 4.11 is correct, but if the specimen has plastic deformation or breaks in bending, the apparent stress is not admissible. To help to differentiate the failure mechanisms, EN 2563 standard gives the typical curve for each type of failure.

4.3.6 In-plane shear test

IPS test was relevant only on specimens manufactured with RTM process with carbon and flax twill fabrics. This test is performed on $\pm 45^\circ$ laminates following ASTM D3518 but for the method, it is in accordance with the ASTM D3039 standard already described. All specimens were of length $L=250\text{mm}$ and width $w=25\text{mm}$. For this test, specific gauges with longitudinal and transverse sensor were placed on each side of the specimens to record displacements in both direction. According to ASTM D3518, shear stress σ_{xy} and shear strain γ_{xy} can be computed respectively

$$\tau_{xy_i}[MPa] = \frac{F_i}{2A} \quad (4.12) \quad \gamma_{xy_i}[-] = \epsilon_{x_i} - \epsilon_{y_i} \quad (4.13)$$

where F is the load in Newtons, A is the cross-section in mm^2 , ϵ_x and ϵ_y are longitudinal and transverse strains respectively. The shear modulus is computed between the strain values $\gamma_1=2000\mu\epsilon$ and $\gamma_2=6000\mu\epsilon$ following:

$$G_{xy}[GPa] = \frac{\tau_2 - \tau_1}{\gamma_2 - \gamma_1} \quad (4.14)$$

To compute the maximal shear stress, standard determines it as the stress at 5% strain if the specimen does not break at lower deformation.

4.3.7 Micro-hardening

For this experiment, samples of two UD laminates were selected to investigate the hardness of the composites in the fiber and transverse directions. To do so, laminate drops after

cutting were preserved and embedded in an epoxy resin. For the both laminates, two samples in fiber direction and in the transverse one were kept. After the curing at room temperature, the embedded samples underwent several grinding (G) and polishing (P) steps, as shown in Table 4.6.

Table 4.6: Grinding and polishing steps parameters

Steps	G1	G2	G3	P1	P2	P3
Support	SIC Paper	SIC Paper	SIC Paper	MD-Plan	MD-Mol	MD-Nap
Abrasive	SIC	SIC	SIC	DP-Spray	DP-Spray	DP-Spray
Grain size	320	800	1200	9 μ m	3 μ m	1 μ m
Lubricant	water	water	water	ethanol	ethanol	ethanol
Speed [rot/min]	150-200	150-200	150-200	150	150	150
Force [N]	Manual	Manual	Manual	Manual	Manual	Manual
Time [min]	4.00	3.00	2.00	6.00	5.00	1.00

Between each step, samples were carefully cleaned with cotton and water, then rinsed with ethanol and finally dried. They were also analyzed under a microscope to evaluate the surface quality. After samples treatment, a Vickers hardness test was performed and 30 indentations were made on each sample. Knowing the load applied on the sample and the diagonals of the print after the test, the Hardness can be computed with the following equation:

$$HV \approx 1.854 \frac{F}{d^2} \quad (4.15)$$

4.3.8 Single flax strand tensile test

To obtain complementary data regarding the properties of the used flax reinforcements, tensile tests on flax strands were performed. For this part, the same principle used by *Amaro J.* [45] was set up. In this work, several yarns were extracted from the different fabrics, glued on paper template and then tested with the universal tribometer device UMT TriboLab®. The working principle of this experiment is illustrated in Fig. 4.11.

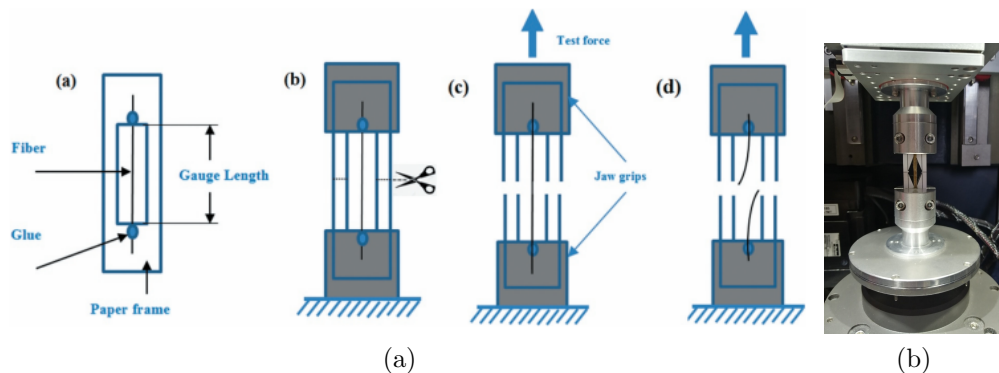


Figure 4.11: (a) Working principle of tensile test on single fiber [35] (b) Current test setup.

To check the reproducibility of the properties, each batch contained 5 samples. Then for each extracted yarn, the dimensions were measured 3 times in 3 different locations

along the fiber with optical microscope. For the old UD –see section 4.1–, cross-section was estimated as rectangular while for other yarns from new UD and fabrics, cross-section was assumed as circular.

For all samples, tensile properties were measured for four different lengths $GL=25,50,70,90$ mm. Extensometer being not available on the used device, the displacement is directly associated with the displacement of the load cell. For the results treatment Eq. 4.1 and Eq. 4.2 were used.

4.3.9 Scanning electron microscopy

For this work, Scanning electron microscopy was used for two purposes:

1. To qualitatively analyse the inner structure of Flax/Infugreen 810 specimens
2. To investigate the failure behavior of composites after tensile loading.

For the first one, SEM was performed on 20L4UD_0 sample dedicated to micro-hardness testing. The polished surface of these specimen allowed to clearly see the distribution of fibers within the InfuGreen 810.

The composite materials being not conductive, the samples were first connected to the ground plate by painting a conductive silver pattern on their surface. To enhance the conductivity, they were glued to the ground plate with silver lac as well. After the silver connection path drying, the surfaces to study were coated with gold particles to avoid the charging of samples during imaging.

Chapter 5

Results

In this chapter, all the results for the different mechanical tests are shown and the average of the main properties for each laminates are extracted. In Table 5.1, all the manufactured plates are summarized with their main parameters. Only those used in this master thesis are present, unusable plates are not shown.

Table 5.1: Manufactured plates characteristics

ID	Size	Fibers		Stacking	Curing			
		Material	Weaving		Drying	Degazing	T[°C]	P[bar]
10L8UD_0	10x10	flax	UD OLD	8	/	/	RT	clamps
10L8UD_1	10x10	flax	UD OLD	8	/	/	RT	clamps
10L8UD_2	10x10	flax	UD OLD	8	YES	/	RT	clamps
20L4UD_1	20x20	flax	UD OLD	4	/	/	RT	clamps
20L4UD_2	20x20	flax	UD OLD	4	YES	/	RT	clamps
20L8UD_0	20x20	flax	UD OLD	4	/	/	RT	clamps
20L8UD_2	20x20	flax	UD NEW	8	YES	YES	RT	clamps
20L12S_0	20x20	flax	Twill	12	/	/	RT	clamps
20L12S_1	20x20	flax	Twill	12	YES	/	RT	clamps
20L12S_2	20x20	flax	Twill	12	YES	YES	RT	clamps
20C4G_0	20x20	Carbon	Satin 5	4	/	/	RT	clamps
20C8G_0	20x20	Carbon	Satin 5	8	/	/	RT	clamps
20C12G_0	20x20	Carbon	Satin 5	12	/	/	RT	clamps
RTM6UD_0	50x50	flax	UD	6	YES	YES	RT	9
RTM12S_0	50x50	flax	Twill	12	YES	YES	RT	9
RTM8G_0	50x50	Carbon	Satin 5	8	YES	YES	RT	9

This chapter will first focus on properties at the composite scale and then, the additional tests performed on the strands are presented –See chapter 4. For all mechanical tests on composites, each result is presented according to the nature or the architecture of the reinforcements, the matrix being the same for all.

5.1 Tensile properties

5.1.1 UD laminates

In this section, UD specimens tested in the fiber direction are presented following by the results in the transverse direction.

In the fiber direction

All computed stress-strain curves following ASTM D3039 are shown in Fig. 5.1. First of all, a bilinear behavior can be seen for all samples with a knee joint at approximately 0.2% strain. This knee joint for all curves is present before the range defined by ASTM D3039 for the tensile modulus, therefore a new range was defined to prevent the underestimation of the modulus. For this test, curves are scattered and it is confirmed by the results available in Table 5.2. However, specimens from RTM6UD_0 laminate present significant lower properties except for the strain at break than others despite the dispersion.

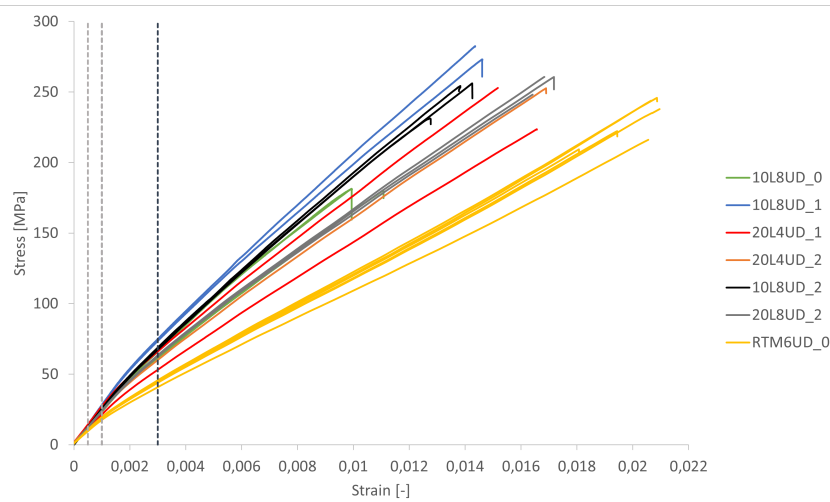


Figure 5.1: Tensile stress-strain curves of UD flax/InfuGreen 810 in the fiber direction. The strain range defined by the standard ASTM D3039 is represented by the black dotted lines $-1000\mu\epsilon$ to $3000\mu\epsilon$ while the new strain range is represented by the grey dotted lines $-500\mu\epsilon$ to $1000\mu\epsilon$.

Table 5.2: Tensile properties of UD flax/InfuGreen 810 laminates in fiber direction.

Specimen	E_T [GPa] ASTM D3039	E_T [GPa] New range	σ_{break} [MPa]	ϵ_{break} [%]
10L8UD_0	20.43 ± 1.13	23.40 ± 0.90	180.89 ± 0.74	1.03 ± 0.06
10L8UD_1	23.02 ± 0.43	27.80 ± 0.02	277.86 ± 6.63	1.45 ± 0.018
10L8UD_2	21.58 ± 0.36	25.06 ± 0.43	247.25 ± 13.87	1.36 ± 0.08
20L4UD_1	17.62 ± 2.51	22.97 ± 4.07	238.27 ± 20.69	1.59 ± 0.1
20L4UD_2	17.84	22.91	252.7	1.69
20L8UD_2	19.47 ± 0.15	23.21 ± 0.19	256.52 ± 7.22	1.68 ± 0.04
RTM6UD_0	12.54 ± 0.49	17.29 ± 0.72	229.25 ± 15.36	2.01 ± 0.11

As shown in Fig. 5.2, the UD specimens present different failures between each other.



Figure 5.2: Tensile specimens after failure in the fiber direction (a) specimens with a failure perpendicular to fibers. On the top, specimens was manufactured with the new UD fabric and on the bottom, the specimen was manufactured with the old one (b) Two cracks perpendicular to fibers located at the two extremities of the sample joined by a longitudinal one.

According to Eq. 2.23, the estimated fiber volume fraction are represented in Fig.5.3. It shows a significant variation between the different tested laminates.

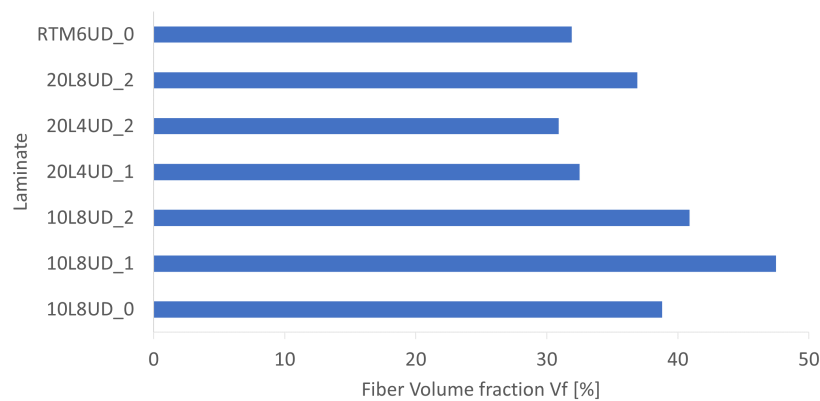


Figure 5.3: Approximation of fiber volume fraction of UD laminates according to Eq. 2.23.

In the transverse direction

For this test, stress-strain curve according to ASTM D3039 for the only specimen is shown in Fig. 5.4. Other samples were not usable because of their destruction during the cutting step. This sample has a linear curve until failure. Main properties of this specimen are available in Table 5.3.

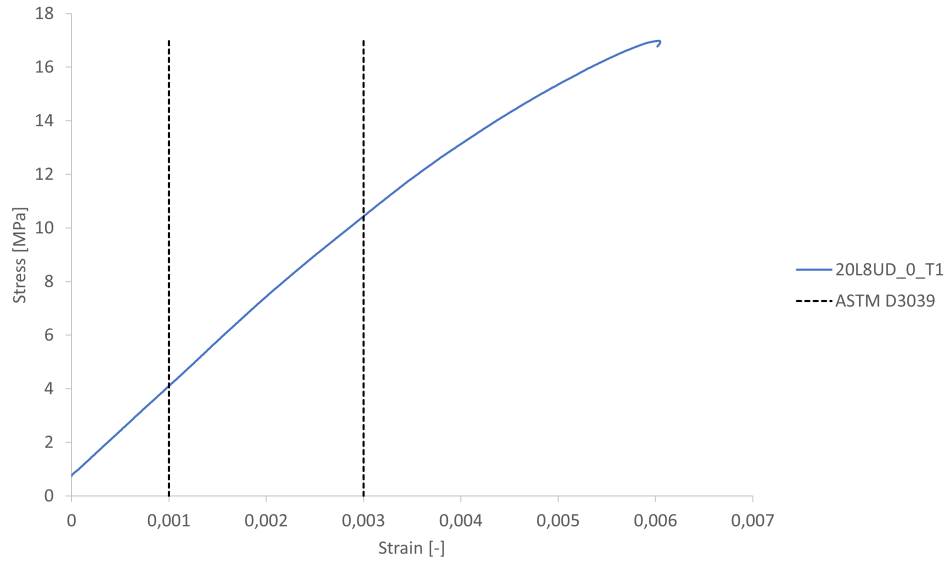


Figure 5.4: Tensile stress-strain curve in the transverse direction for UD flax/InfuGreen 810. Black dotted lines represent the strain range defined by the standard ASTM D3039.

Table 5.3: Tensile properties of UD flax/InfuGreen 810 sample in transverse direction. The black dotted lines represent the strain range defined by ASTM D3039 for the modulus computation $-1000\mu\epsilon$ to $3000\mu\epsilon$.

Specimen	E_T [GPa]	ASTM 3039	σ_{break} [MPa]	ϵ_{break} [%]
20L8UD_0		3.21	16.99	0.6

As shown in Fig. 5.5, the failure of the tested specimen is parallel to the fibers and perpendicular to the load direction.



Figure 5.5: Tensile specimen failure after the tensile test in the transverse direction.

5.1.2 Flax fabric

For all the fabrics specimens, stress-strain curves computed according to the standard ASTM D3039 are available in Fig. 5.6. All specimens have a bilinear curve but the strain value corresponding to the knee-joint is not obvious and lies between 0.4-0.6% strain. Because the strain range defined by the standard is close to the knee joint, to

avoid underestimation of stiffness property, a new strain range for the tensile modulus was also defined. The resulting properties are available in Table 5.4. All specimens show reproducible behavior in the first part of the curve but after the knee-joint, the samples from the 20L12S_0 laminate show lower properties than others until failure.

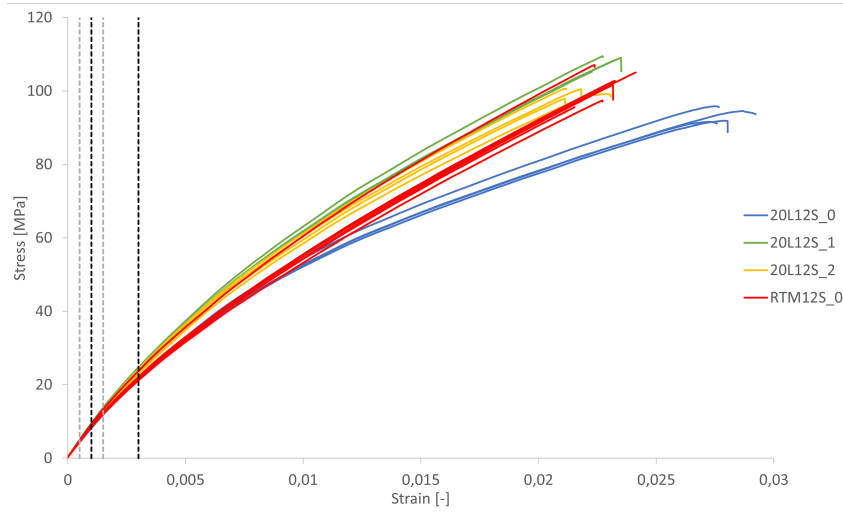


Figure 5.6: Tensile stress-strain curves for flax twill/InfuGreen 810. The strain range defined by the standard ASTM D3039 is represented by the black dotted lines $-1000\mu\epsilon$ to $3000\mu\epsilon$ while the new strain range is represented by the grey dotted lines $-500\mu\epsilon$ to $1500\mu\epsilon$.

Table 5.4: Tensile properties of flax twill/InfuGreen 810 laminates.

Specimen	E_T [GPa] ASTM 3039	E_T [GPa] New range	σ_{break} [MPa]	ϵ_{break} [%]
20L12S_0	6.56 ± 0.08	7.55 ± 0.14	93.48 ± 2.04	2.78 ± 0.06
20L12S_1	7.51 ± 0.08	8.64 ± 0.92	108.09 ± 1.96	2.3 ± 0.06
20L12S_2	7.32 ± 0.19	8.23 ± 0.24	99.55 ± 1.31	2.17 ± 0.08
RTM12S_0	6.67 ± 0.28	7.82 ± 0.33	101.6 ± 4.4	2.28 ± 0.09

Regarding the estimation of the fiber volume fraction, results are shown in Fig. 5.7. Laminates have low volume variation between each other.

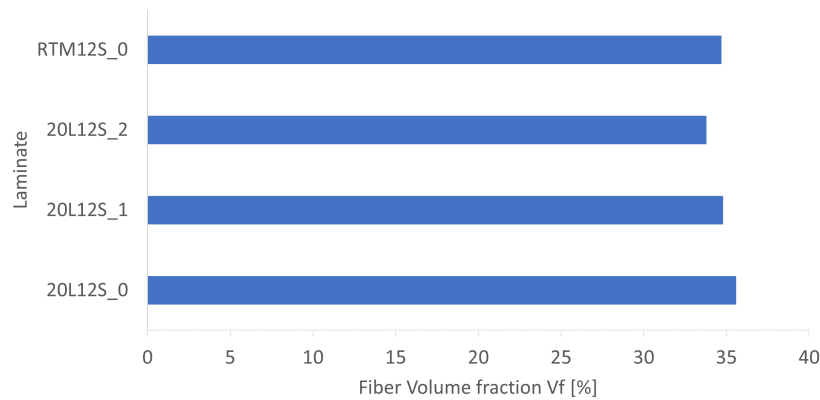


Figure 5.7: Approximation of fiber volume fraction of flax twill laminates according to Eq. 2.23.

5.1.3 Carbon

For carbon reinforced specimens, tensile stress-strain curves according to ASTM D3039 are all available in Fig. 5.8. Carbon samples show linear behavior until failure with a high reproducibility. All specimens have similar properties except for the RTM8G_0 which has higher properties at failure, as shown in Table 5.5 as well. Fiber volume fraction values are all given in Fig. 5.9.

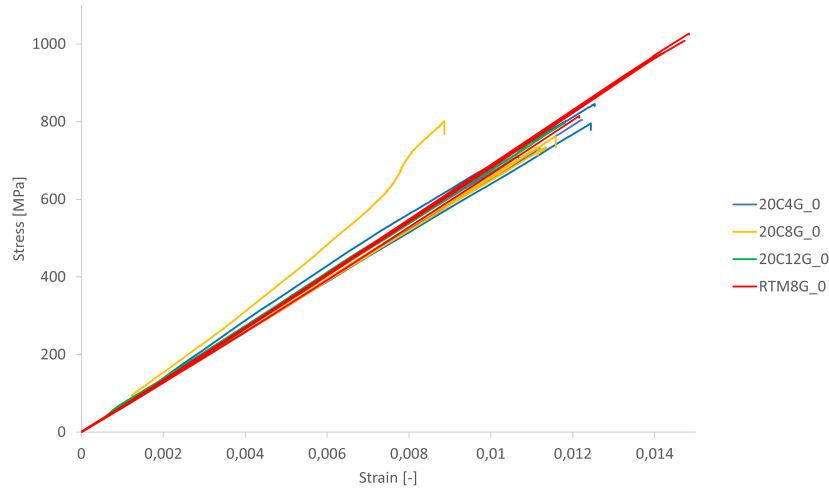


Figure 5.8: Tensile stress-strain curve of Carbon G0926/InfuGreen 810

Table 5.5: Tensile properties of Carbon G0926/InfuGreen 810 samples.

Specimen	E_T [GPa] ASTM 3039	σ_{break} [MPa]	ϵ_{break} [%]
20C4G_0	67.74 ± 3.81	789.79 ± 55.74	1.20 ± 0.09
20C8G_0	64.64 ± 0.77	743.46 ± 16.06	1.14 ± 0.02
20C12G_0	66.32 ± 1.8	770.53 ± 29.38	1.13 ± 0.04
RTM8G_0	66.53 ± 1.06	974.28 ± 83.84	1.42 ± 0.11

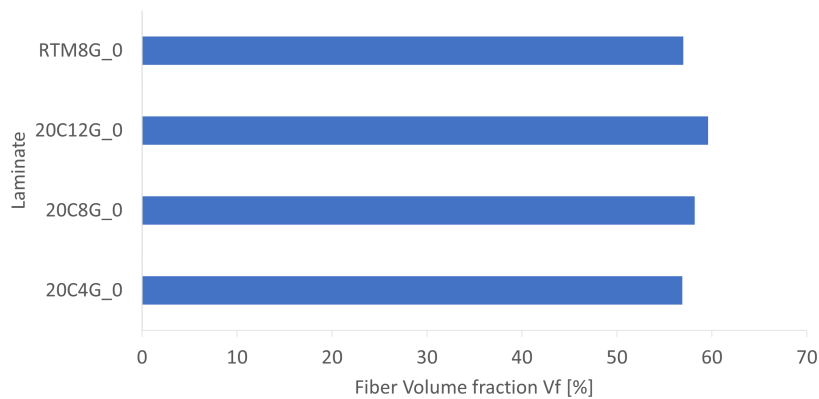


Figure 5.9: Approximation of fiber volume fraction of carbon laminates according to Eq. 2.23.

5.2 Compressive properties

5.2.1 UD laminates

As for tension, results for specimens tested in the fiber direction are first presented and results for specimens tested in the transverse direction are shown next.

In the fiber direction

For the UD samples, only two laminates were tested because of the minimal thickness and the dimensions imposed by the standard ASTM D6641.

For all specimens, stress-strain curves shown in Fig. 5.10 are computed according the given standard. For both laminates, the curves are bilinear and with a low scatter, the knee-joint is approximately at 0.25% and 0.5% strain for RTM6UD_0 and 20L8UD_2, respectively. As for tensile tests, the strain range given by ASTM D6641 for the chord modulus is not consistent anymore and a new one had to be defined –Fig. 5.10. As shown in Table 5.6, RTM6UD_0 samples have lower properties for both modulus and stress at break. Because of gauges saturation, the strain at break is not available.

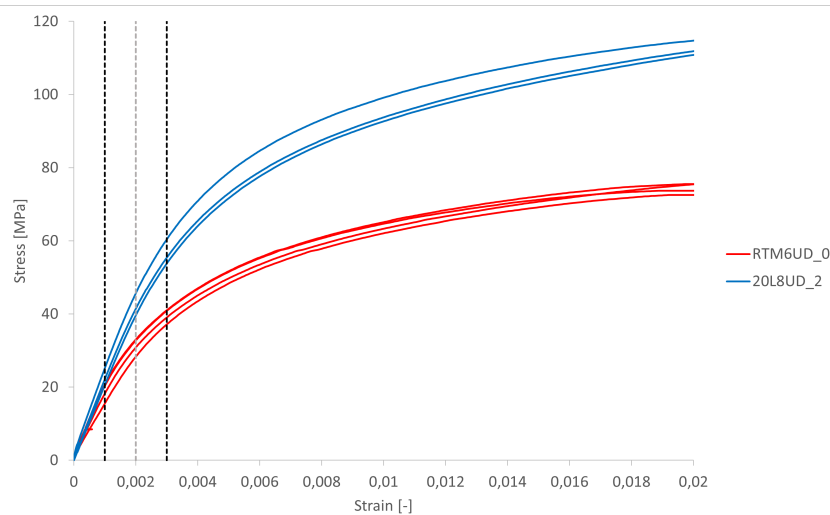


Figure 5.10: Compression stress-strain curves in the fiber direction of UD flax/Infugreen 810. Because of the gauges saturation during the test, only the first part of the curves is published. The strain range defined by the standard ASTM D6641 is represented by the black dotted lines –1000 $\mu\epsilon$ to 3000 $\mu\epsilon$ – while the new strain range is represented by the grey dotted lines –1000 $\mu\epsilon$ to 2000 $\mu\epsilon$.

Table 5.6: Compression properties in the fiber direction of UD flax/Infugreen 810 samples.

Specimen	E_C [GPa] ASTM D6641	E_C [GPa] new range	σ_{break} [MPa]	ϵ_{break} [%]
20L8UD_2	16.94 ± 0.58	19.58 ± 0.77	114.93 ± 3.1	N.A
RTM6UD_0	10.43 ± 0.28	12.56 ± 0.19	74.98 ± 1.81	N.A

In the transverse direction

For the UD specimens, stress-strain curves computed according ASTM D6641 are available in Fig. 5.11. For the transverse direction, curves show linear behavior between 0-1.5% strain and have a slope drop just before the saturation. Even with this linearity at the beginning of the curve, a new range for the modulus had to be defined because of a significant mounting effect –Fig 5.11. One of the specimen shows lower modulus. At failure, specimens show reliable stress at break while the strain at break is not available because of the gauges saturation. Properties for this set are available in Table 5.7.

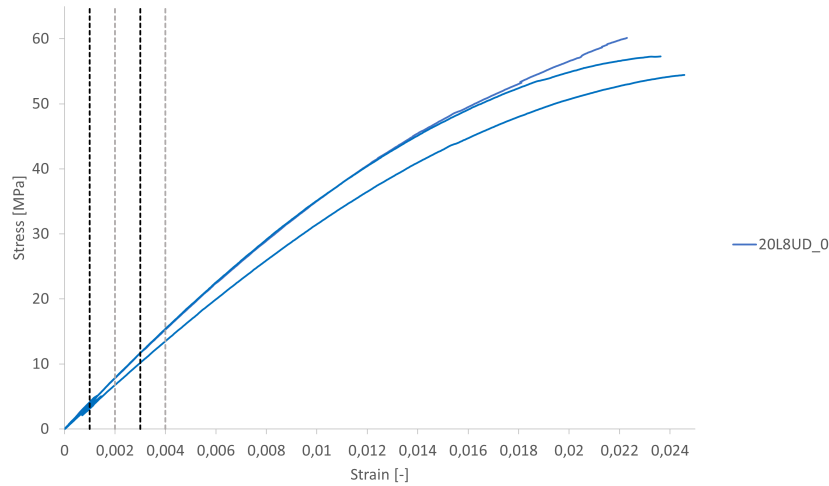


Figure 5.11: Compression stress-strain curves in the transverse direction of UD flax/Infugreen 810. Because of the gauges saturation during the test, only the first part of the curves is published. The strain range defined by the standard ASTM D6641 is represented by the black dotted lines $-1000\mu\epsilon$ to $3000\mu\epsilon$ – while the new strain range is represented by the grey dotted lines $-2000\mu\epsilon$ to $4000\mu\epsilon$.

Table 5.7: Compression properties in the fiber direction of UD flax/Infugreen 810

Specimen	E_C [GPa] ASTM D6641	E_C [GPa] new range	σ_{break} [MPa]	ϵ_{break} [%]
20L8UD_0	3.84 ± 0.28	3.63 ± 0.22	58.1 ± 3.88	N.A

5.2.2 Fabrics

For fabrics in compression, stress-strain curves computed with the standard ASTM D6641 in Fig. 5.12. All specimens have bilinear behavior with the knee-joint at approximately 0.6% strain. This transition being after 0.5% strain, the strain range defined by the standard is still consistent. All laminates present a good reproducibility except for RTM12S_0 for which the curves are more scattered. Despite this, all samples have similar elastic properties and stress at break as shown in Table 5.8. As for UD samples, gauges saturation occurred before the end of the test and strain at break is therefore unavailable.

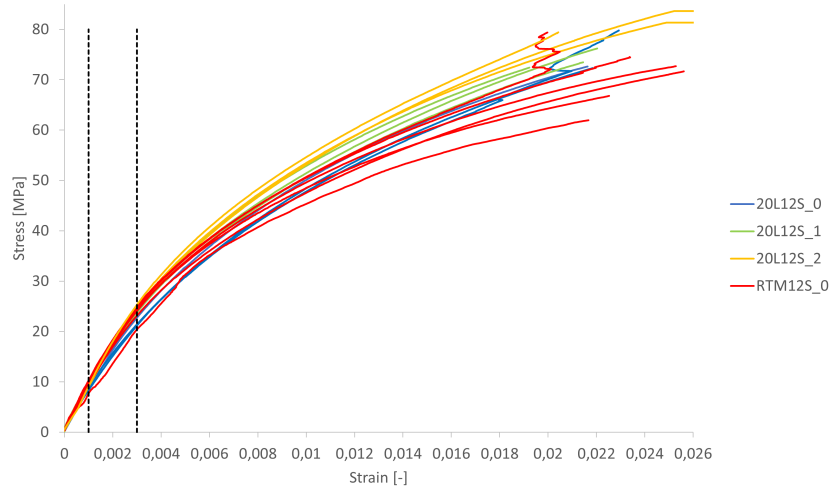


Figure 5.12: Compression stress-strain curves in the transverse direction of UD flax/Infugreen 810. Because of the gauges saturation during the test, only the first part of the curves is published. The strain range defined by the standard ASTM D6641 is represented by the black dotted lines $-1000\mu\epsilon$ to $3000\mu\epsilon$.

Table 5.8: Compression properties of flax twill/Infugreen 810 samples.

Specimen	E_C [GPa]	ASTM D6641	σ_{break} [MPa]	ϵ_{break} [%]
20L12S_0	6.6 ± 0.47		84.47 ± 4.14	N.A
20L12S_1	7.26 ± 0.04		87.05 ± 6.98	N.A
20L12S_2	7.61 ± 0.34		94.46 ± 2.71	N.A
RTM12S_0	7.48 ± 0.18		80.04 ± 4.77	N.A

5.2.3 Carbon

As for previous compressive tests, stress-strain curves for carbon laminates were computed according to ASTM D6641 and are available in Fig. 5.13. First of all, all specimen curves are linear. Although all samples have similar modulus with high reproducibility, the stress at break values are more scattered. For carbon laminates, any gauges saturated before the failure of the samples which allows to determine strain at failure for this experiment. All properties are available in Table 5.9.

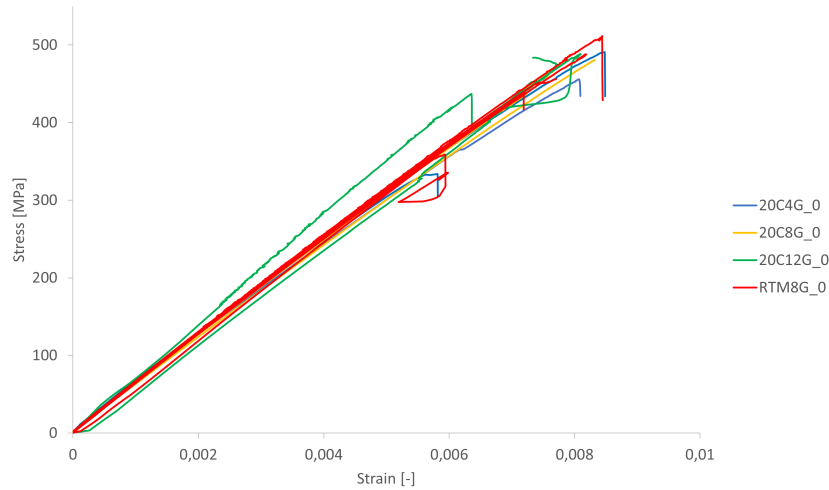


Figure 5.13: Compression stress-strain curves of Carbon G0926/InfuGreen 810 samples.

Table 5.9: Compression properties of Carbon G0926/Infugreen 810 samples

Specimen	E_C [GPa] ASTM D6641	σ_{break} [MPa]	ϵ_{break} [%]
20C4G_0	60.79 ± 1.44	426.86 ± 82.59	0.75 ± 0.14
20C8G_0	60.96 ± 1.24	476.47 ± 6.16	0.8 ± 0.04
20C12G_0	67.23 ± 3.65	443.92 ± 36.9	0.68 ± 0.05
RTM8G_0	64.20 ± 1.94	450.56 ± 58.59	0.75 ± 0.10

5.3 Flexural properties

5.3.1 UD laminates

In the fiber direction

For UD specimens in bending, stress-strain curves are computed according to the standard ISO 178 and are available in Fig. 5.14. UD flax samples show stress-strain curves in two regimes: the first one is linear and the second one has a progressive modulus drop until failure. The knee-joint between the two stages is at approximately 1% strain. Despite a similar global behavior for 20L4UD_0, 20L4UD_1 and 20L8UD_2, the curves are highly scattered at the end. Furthermore, RTM samples, despite a similar bilinear behavior, have all the same plateau at the beginning of the curve. Therefore a new strain range for the modulus had to be defined for this set of specimens –Fig. 5.14. Those also present lower stress at break but higher strain at break compared to other samples. Properties for all samples are summarized in Table 5.10.

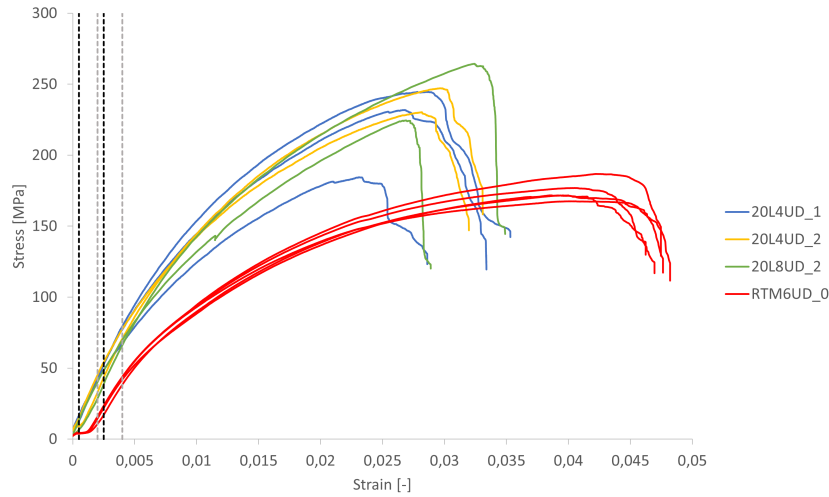


Figure 5.14: Flexion stress-strain curves in the fiber direction of UD flax/InfuGreen 810. The strain range defined by the standard ISO 178 is represented by the black dotted lines $-500\mu\epsilon$ to $2500\mu\epsilon$ while the new strain range for RTM6UD_0 specimens is represented by the grey dotted lines $-2000\mu\epsilon$ to $4000\mu\epsilon$.

Table 5.10: Flexural properties of UD flax/InfuGreen810 samples in fiber direction.

Specimen	E_C [GPa] ISO 178	E_F [GPa] new range	σ_{break} [MPa]	ϵ_{break} [%]
20L4UD_1	18.32 ± 2	/	220.33 ± 31.7	2.62 ± 0.28
20L4UD_2	18.34 ± 1.66	/	238.73 ± 11.91	2.89 ± 0.11
20L8UD_2	16.69 ± 1.02	/	244.44 ± 28.2	2.96 ± 0.39
RTM6UD_0	8.68 ± 1.35	14.04 ± 0.36	174.92 ± 7.42	4.02 ± 0.14

In the transverse direction

As for test in the fiber direction, all stress-strain curves for samples in transverse direction are computed with ISO 178 as well and are available in Fig 5.15. Specimens are highly scattered between laminates but all present linear behavior until the failure. For the flexural modulus, 20L4UD_0, 20L4UD_1 and 20L8UD_2 laminates show similar values while the RTM6UD_0 samples have lower elastic property. Furthermore, those present a similar plateau at the beginning as well. At the end of the curves, samples show a more progressive failure in contrast to samples loaded in the fiber direction. Some of them even have a small plateau before breaking. Regarding the stress at break, 20L8UD_2 and RTM6UD_0 laminates have lower properties. Main properties are available in Table 5.11.

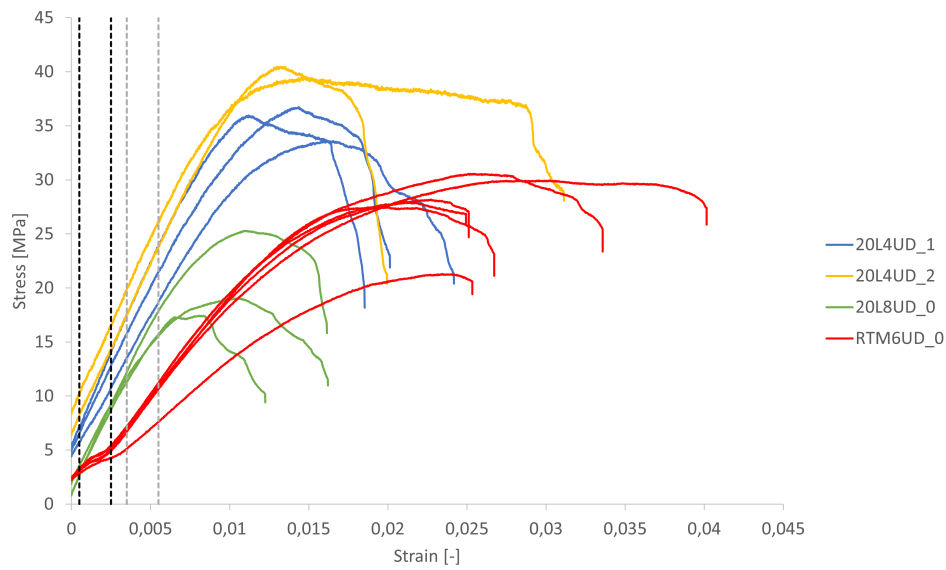


Figure 5.15: Flexion stress-strain curves in the transverse direction of UD flax/InfuGreen 810. The strain range defined by the standard ISO 178 is represented by the black dotted lines $-500\mu\epsilon$ to $2500\mu\epsilon$ while the new strain range for RTM6UD_0 specimens is represented by the grey dotted lines $-3500\mu\epsilon$ to $5500\mu\epsilon$.

Table 5.11: Flexural properties of UD flax/InfuGreen810 samples in transverse direction.

Specimen	E_F [GPa] ISO 178	E_F [GPa] new range	σ_{break} [MPa]	ϵ_{break} [%]
20L4UD_1	3.05 ± 0.5	/	35.44 ± 1.6	1.39 ± 0.25
20L4UD_2	3.12 ± 0.13	/	40.01 ± 0.66	1.39 ± 0.12
20L8UD_0	3.05 ± 0.22	/	20.59 ± 4.15	0.98 ± 0.15
RTM6UD_0	1.08 ± 0.37	1.84 ± 0.30	27.56 ± 3.29	2.37 ± 0.35

5.3.2 Fabrics

For fabrics, stress-strain curves computed according to ISO 178 are shown in Fig. 5.16. Those present the usual bilinear behavior with a knee-joint at approximately 1% strain. Before the regime transition, curves show close properties but in the second part, the scattering increases. This dispersion is reflected in the strain at break. However, the stress at break is similar for all sets of specimens except for the RTM12S_0_T samples which show lower failure property as noted in Table 5.12.

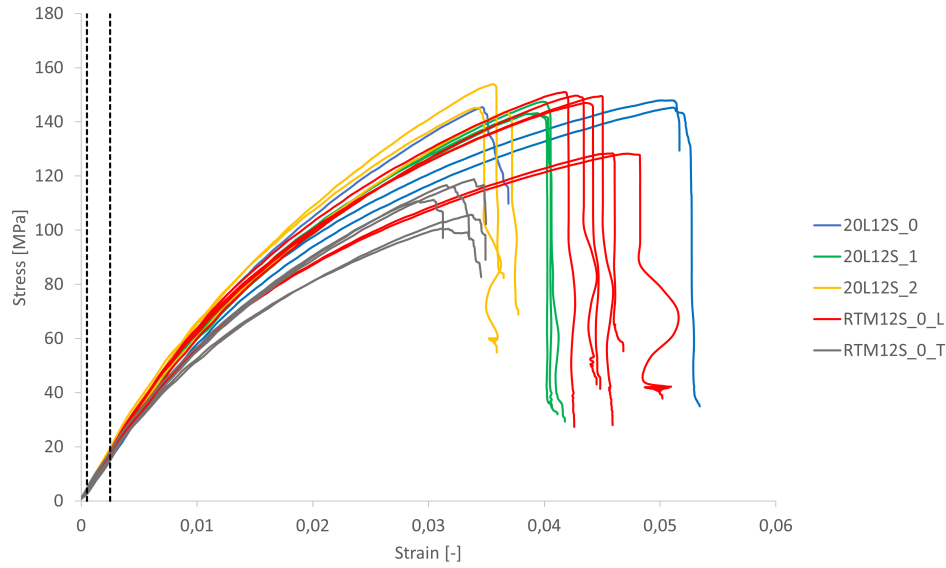


Figure 5.16: Bending stress-strain curves of twill flax/InfuGreen 810. The strain range defined by the standard ISO 178 is represented by the black dotted lines $-500\mu\epsilon$ to $2500\mu\epsilon$.

Table 5.12: Flexural properties of flax twill/InfuGreen810 samples.

Specimen	E_F [GPa] ISO 178	σ_{break} [MPa]	ϵ_{break} [%]
20L12S_0	6.7 ± 0.51	146.24 ± 1.49	4.56 ± 0.95
20L12S_1	6.79 ± 0.37	144.58 ± 2.48	3.93 ± 0.06
20L12S_2	7.34 ± 0.37	147.69 ± 5.47	3.55 ± 0.12
RTM12S_0 long	7.01 ± 0.28	142.33 ± 10.93	4.43 ± 0.2
RTM12S_0 trans	6.4 ± 0.28	110.58 ± 7.57	3.21 ± 0.16

5.3.3 Carbon

For all carbon specimens, stress-strain curves according to ISO 178 are available in Fig. 5.17. Carbon samples show a linear behavior until failure but highly variable in bending. However, because of a large toe region for all samples, a new strain range had to be defined for the modulus computation –Fig. 5.17. Even with the dispersion, a more significant drop in properties can be seen for all samples with 12 plies and for some of the samples with 8 plies. These latter were arbitrarily rejected for the computation of the average modulus. Main properties are available in Table 5.13.

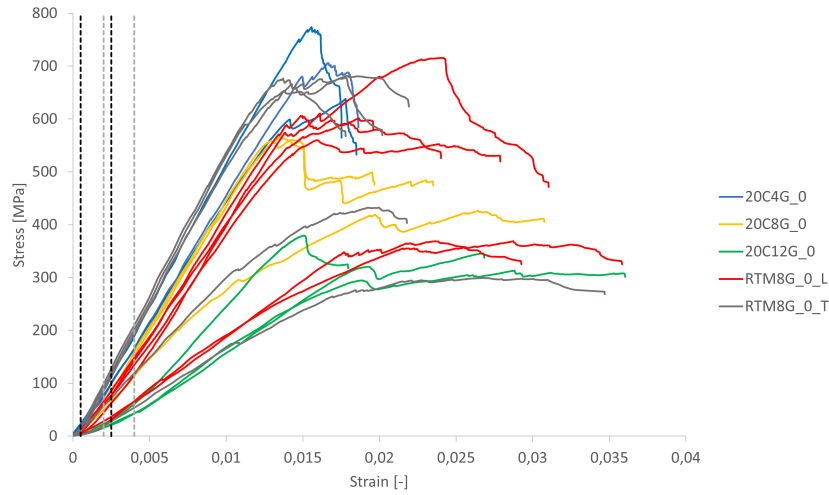


Figure 5.17: Bending stress-strain curves of Carbon/InfuGreen 810. The strain range defined by the standard ISO 178 is represented by the black dotted lines $-500\mu\epsilon$ to $2500\mu\epsilon$ while the new strain range for RTM6UD_0 specimens is represented by the grey dotted lines $-2000\mu\epsilon$ to $4000\mu\epsilon$.

Table 5.13: Flexural properties of Carbon G0926/InfuGreen810 samples.

Specimen	E_F [GPa] ISO 178	E_F [GPa] new range	σ_{break} [MPa]	ϵ_{break} [%]
20C4G_0	42.65 ± 2.64	47.01 ± 3.55	705.92 ± 67.94	1.67 ± 0.11
20C8G_0	39.72 ± 1.06	45.21 ± 4.10	563.63 ± 4.71	13.9 ± 0.06
20C12G_0	10.94 ± 2.00	15.28 ± 2.96	346.05 ± 33.09	2.35 ± 0.74
RTM8G_0 long	31.34 ± 3.18	40.25 ± 3.06	627.2 ± 79.94	1.83 ± 0.5
RTM18G_0 trans	52.73 ± 3	55.06 ± 0.58	670.84 ± 15.37	1.65 ± 0.24

5.4 Interlaminar shear strength

For this part of the work, stress-strain curves are not available. But for a better comprehension of the behavior of samples during loading, shear stress-displacements curves are available in Appendix A. These curves may help for the identification of failure mechanisms as defined in the standard EN 2563.

5.4.1 UD laminates

As shown in Table 5.14, all computed apparent shear stresses are in the same range of magnitude except for the 20L8UD_2 laminate which shows higher properties. For this test, RTM laminate does not show lower properties as for previous tests.

Table 5.14: Interlaminar shear strength results for UD flax/InfuGreen 810 samples

Specimen	Apparent shear strength [MPa]	Mode of fracture according to EN 2563
10L8UD_0	18.87	Plastic deformation
10L8UD_1	18.65 ± 1.03	Plastic deformation
10L8UD_2	19.61 ± 0.71	Plastic deformation
20L4UD_1	19.82 ± 3.82	Plastic deformation and bending
20L4UD_2	17.54 ± 0.93	Bending
20L8UD_2	30.16 ± 0.02	Bending or simple shear
RTM6UD_0	17.43 ± 0.7	Plastic deformation

5.4.2 Fabrics

For twill fabrics, small laminates present similar apparent shear stress with lower dispersion than RTM fabrics which show lower properties especially in the transverse direction.

Table 5.15: Interlaminar shear strength results for fabrics flax/InfuGreen 810 samples

Specimen	Apparent shear strength [MPa]	Mode of fracture according to EN 2563
20L12S_0	17.23 ± 0.94	Bending or simple shear
20L12S_1	17.44 ± 0.67	Bending or simple shear
20L12S_2	16.57 ± 0.55	Bending or simple shear
RTM12S_0 long	15.9 ± 1.14	Bending or simple shear
RTM12S_0 trans	13.07 ± 1.02	Bending or multiple shear

5.4.3 Carbon

For carbon samples, results are scattered between and within the different sets. 20C4G_0 laminate shows higher properties than other carbon and the RTM8G_0 in the longitudinal direction presents the lowest average value.

Table 5.16: Interlaminar shear strength results for Carbon/InfuGreen 810 samples

Specimen	Apparent shear strength [MPa]	Mode of fracture according to EN 2563
20C4G_0	45.5 ± 0.3	Simple shear or bending
20C8G_0	31.64 ± 7.31	Multiple shear
20C12G_0	34.88 ± 1.56	Multiple shear
RTM8G_0 long	24.7 ± 2.7	Multiple shear
RTM8G_0 trans	30.98 ± 9.64	Multiple shear

5.5 In-plane shear

Shear stress-shear strain curves computed according to ASTM 3518 are available in Fig.5.18. For the both composites, curves are bilinear and the inflection point between the two

regimes are at 1% and 2% strain for carbon and flax, respectively. The reproducibility is high between samples from the same laminate. Carbon specimens –RTM8G_0– show significant higher in-plane shear modulus than flax in the first part of the curve. After the transition region, the chord modulus of the second part of the curve is respectively 216 MPa and 167 MPa for carbon and flax. Even if gauges saturated during this test, according to ASTM 3518, only results before 5% strain are relevant. For all specimens, gauges saturated just below this strain value as shown in Table 5.17.

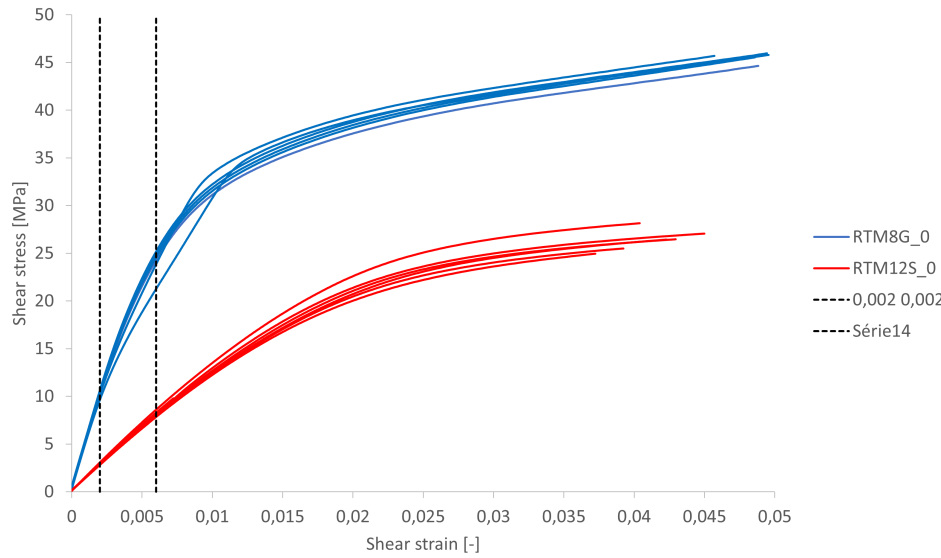


Figure 5.18: In-plane shear stress-strain curve for InfuGreen 810 reinforced with flax fabrics and carbon samples. The strain range defined by the standard ASTM D3518 is represented by the black dotted lines –2000 $\mu\epsilon$ to 6000 $\mu\epsilon$.

To obtain the estimation of the stress at 5%, linear extrapolation was done assuming the last part of the curves was linear.

Table 5.17: In-plane shear results for Infugreen 810 reinforced with carbon and flax twill –*b.s.*: before saturation.

Specimen	G_{xy} [GPa]	τ_{xy} [MPa] b.s	γ_{xy} [%] b.s	τ_{xy} [MPa] at 5%
RTM12S_0	1.3 ± 0.05	26.26 ± 0.86	4.12 ± 0.28	28.07 ± 0.83
RTM8G_0	3.43 ± 0.27	45.53 ± 0.46	4.83 ± 0.14	45.93 ± 0.56

5.6 Micro hardness

The results presented in this part of the work are the Vickers hardness values extracted from the four samples of UD laminates. As reminder samples defined as the "longitudinal" are samples where the indentation direction is the same than fibers axis. "Transversal" are samples where the indentation is perpendicular to fibers axis. As shown in Table 5.18, results are scattered but some difference emerge between the tested samples. Some indentation pictures are shown in Figs. 5.19 and 5.20 and with help for the discussion of the results.

Table 5.18: HV results for tested UD flax/ InfuGreen 810 laminates.

Specimen	Hardness [HV]	Comment
20L8UD_2 - long	17.9 ± 2.1	reject of measures because of presence of adhesive paper
20L4UD_1 - trans	11.4 ± 3.1	reject of measures because of bad quality of print
20L4UD_1 - long	16.3 ± 1.3	reject of measures because of bad quality of print
20L8UD_2 - trans	16.2 ± 1.6	reject of measures because of bad quality of print

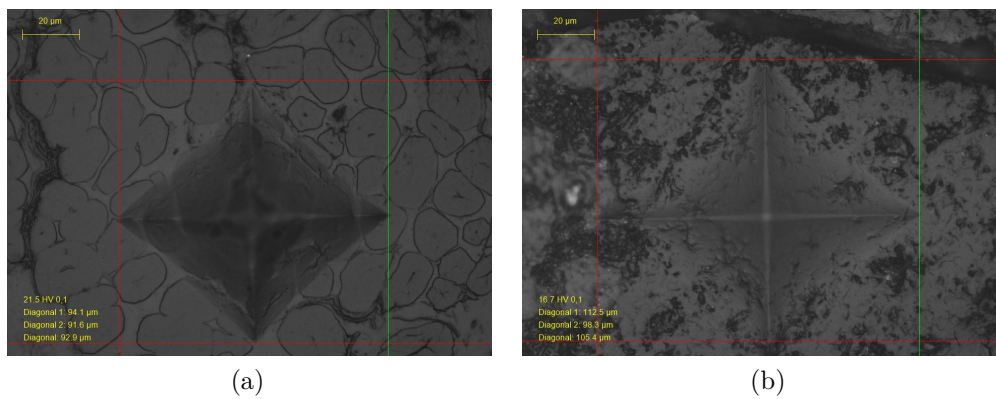


Figure 5.19: Vickers microhardness indentation performed on (a) flax fibre-rich area from 20L8UD_0 sample in the longitudinal direction (b) on Infugreen 810-rich area in the sample 20L4UD_0 in the transverse direction. As shown on both images, the computed HV value is higher for the imprint on fibers than on the matrix.

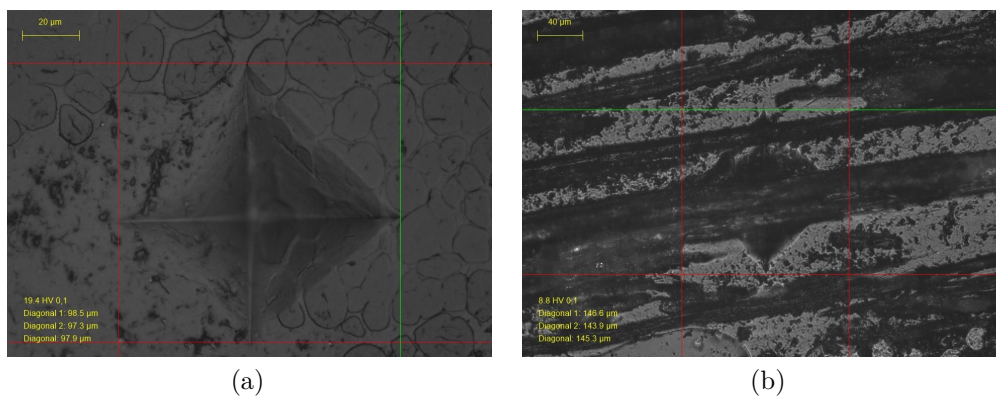


Figure 5.20: Vickers microhardness indentation performed on (a) area containing flax fibres and InfuGreen 810 from sample 20L8UD_0 in the longitudinal direction (b) fiber-rich area from the sample 20L4UD_0 in the transverse direction. In both pictures, the indentation shows a different shape depending on the deformed material. Computed HV values for both are highly different.

5.7 Tensile properties of single flax strand

For the results, typical stress-strain curves of strands from the both UD are shown in Fig. 5.21. First of all, regarding curves, flax strands from old and new UD do not show the same behavior. Strands from new UD roll have a linear curve while a bilinearity can be distinguished for old UD strands. But for both sets, the scatter is high.

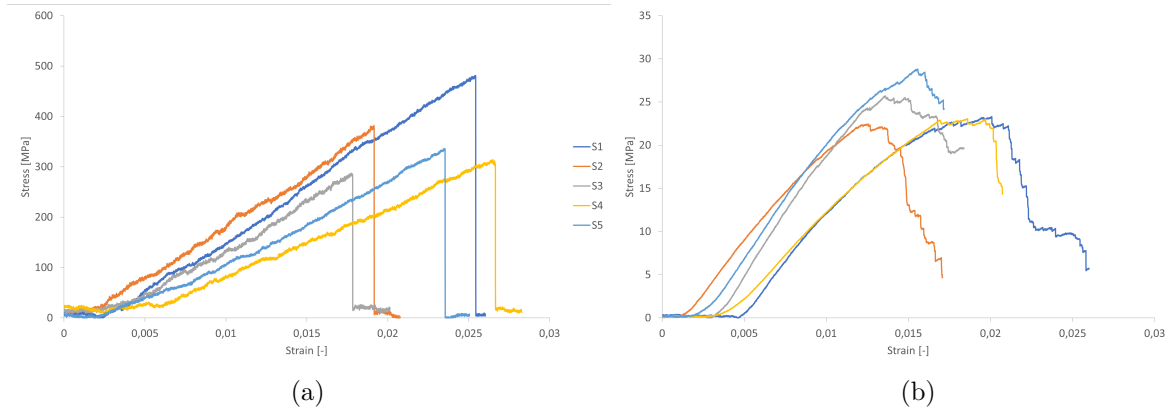


Figure 5.21: Typical tensile stress-strain curves for UD flax with a length of 90 mm (a) new UD (b) old UD.

The modulus and the UTS averages of each flax strands for all tested lengths are available in Fig. 5.22. Regarding the results, the yarns from new UD roll and the fabrics (both directions) have similar properties for tensile modulus and stress at break except for the gauge length of 90 mm. Indeed, fabric yarns have lower properties than new UD yarns. On the other hand, strands from old UD roll have significant lower properties than other strands. However, any significant variation between tested lengths is detected for fibers from the same origin, but on the other hand, the different values are highly scattered.

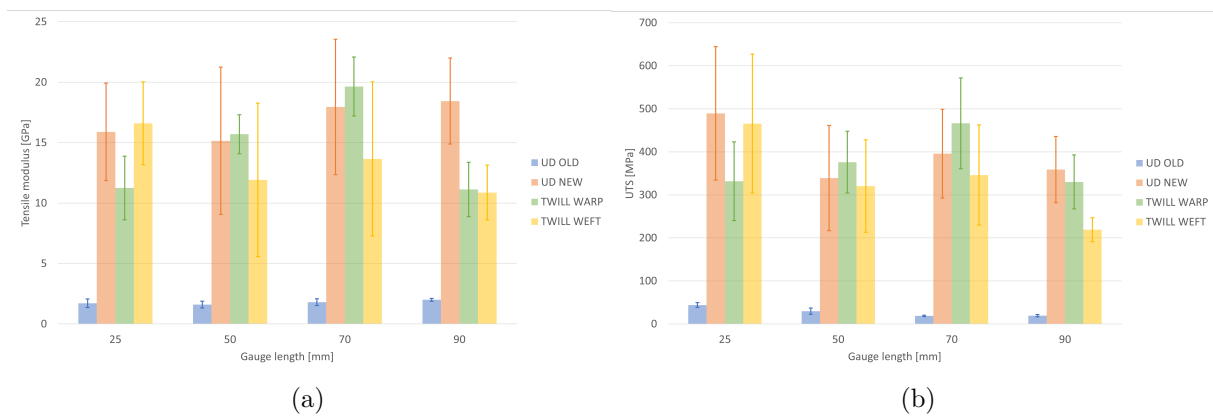


Figure 5.22: Tensile properties for flax strands at different test length (a) Young's modulus (b) stress at failure.

Chapter 6

Discussion

In this chapter, the results available in the previous one are discussed according to the experiment and the reinforcement type. As a reminder, laminates which were manufactured by hand layup are defined as "small laminates" for both used mold sizes. Laminates manufactured with hybrid hand layup-RTM process are named as "RTM laminates". Same shortcut is done when specimens are cited.

Before discussing the results of mechanical tests, it may be relevant to highlight some points about the manufacturing process of laminates. Because the used process was operated for the first time in this work, this one may be not optimized as industrial process and therefore may have caused defects in the plates. These defects are important to discuss because they may have a significant impact on the mechanical properties of the final specimens.

6.1 Samples preparation process

6.1.1 Laminates manufacturing

First, because of the cut of plies was made manually, it was very difficult to obtain square plies with the fibers perfectly aligned in the 0° direction. In addition, as shown in Fig. 6.1 for the UD fabrics, the fibers delaminated at the edges of the ply because of the poor transverse support. To prevent the fibres from separating during cutting, adhesive paper was placed along the edges to hold the fibres together. But this solution could lead to other defects later on, such as poor resin impregnation in these areas. Poor resin impregnation can promote poor interlaminar adhesion at the edges of laminate. To prevent these manufacturing defects from affecting the properties of the samples, they were cut as far away from the edges of the laminate as possible.



Figure 6.1: Delamination of the fibers during the cutting of the first plies.

Another point to notice is the quantity of used resin during impregnation of flax reinforcements. Indeed, the process was quite difficult because flax strands absorbed resin before it can be fully distributed over the entire surface of the ply. The consequence was the preparation of excess resin for the flax/epoxy laminates, this excess being evacuated by the pressure applied during the curing of the plates. Due to lack of time, further analysis could not be carried out to fully understand the causes of this phenomenon but it would be interesting to do so in further research. But this excess of resin had the additional effect of improving the surface of composites. Indeed, for the RTM plate with flax twill fabrics, less resin could be used during impregnation but after the curing, the produced laminate had many non-impregnated areas on its surface. However, this phenomenon was not detected for basalt [52] and carbon fibers.

After the curing of the samples, it was interesting to qualitatively analyse the porosity of the laminates. As shown in Fig. 6.2, many porous defects are present within the sample despite the applied pressure during the curing. This porosity could be caused partially by the hand layup process, the use of brushes and rolls to distribute the resin could create air bubbles which after stay entrapped in the plate.

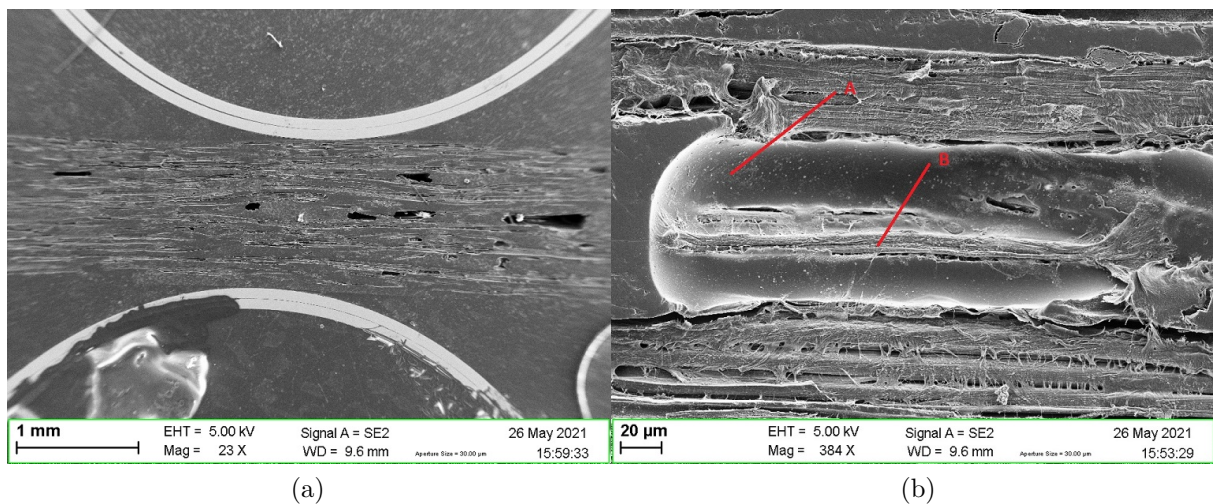


Figure 6.2: SEM micrograph of 20L4UD_1 sample embedded in resin (a) overview of the sample (b) Focus on a porosity defect (A) Bubble imprint (B) exposed fiber.

The porosity is important to notice in this work because these air bubbles can promote crack propagation during loading and impact composite properties. Furthermore if the

defect is in the vicinity of a fiber as shown in Fig 6.2b, it can induce a weak stress transfer and increase the risk of decohesion during loading.

At the microscopic scale, the flax laminates presents a matrix fibre distribution that is interesting to analyse as it could have an impact on the final behaviour of the composite, especially at fracture. As shown in Fig. 6.3a, the elementary fibres are either still assembled in bundles or alone. Concerning the bundles, although the contrast is low, it can be seen that the resin does not impregnate them. This means that the stress transfer between these fibres is not done by the resin but by the pectin. Furthermore, micro-cracks can appear within these bundles which can promote an early failure for loading in transverse direction. Cracks are also present in resin as illustrated in Fig. 6.3a. Even if these cracks are of unknown origin, it can be assumed that they are found in number during the loading of the composite and therefore impact its properties. Furthermore, according to Fig. 6.3a, the volume fraction of the two components seems to be equivalent.

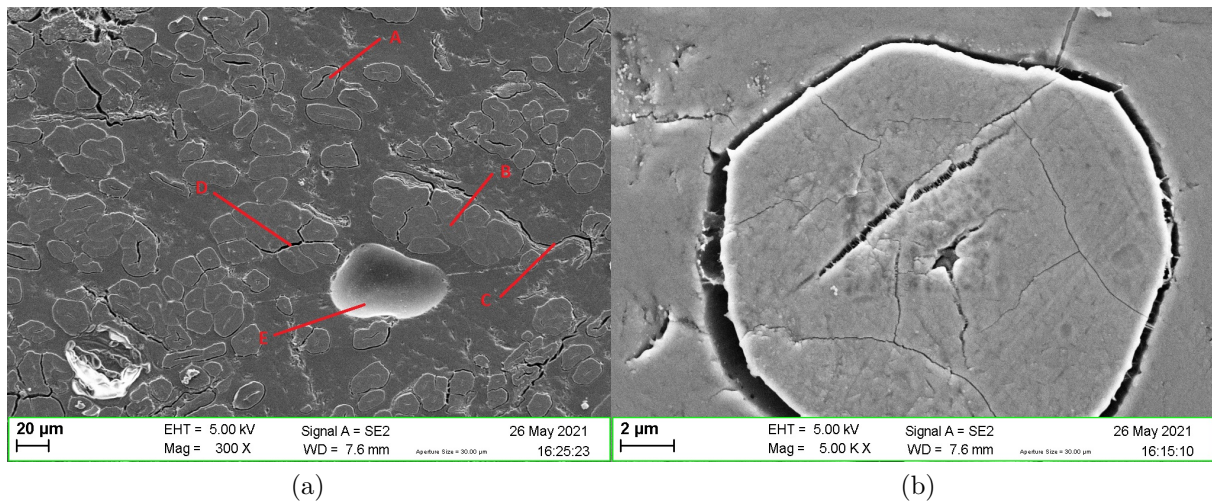


Figure 6.3: (a) SEM micrograph of sample 20L4UD_1 (A) single fiber (B) fibers bundle (C) resin crack (D) crack within bundle (E) air bubble (b) SEM micrograph of single fiber.

At fiber scale, as shown in Fig 6.3b, internal structure is not visible except for the lumen which is also visible on some fibers in Fig. 6.3a. However, it can be seen that fiber does not perfectly adhere to the resin in this case. If this case is isolated, this could be negligible but if a large number of fibres have the same defect, this could considerably reduce the properties of the composite. However, these SEM micrographs were performed on composites reinforced with old UD fabric, the internal appearance of the composite might differ for the samples made with the new reinforcements and not show the same type of defects.

Regarding the curing, the post-curing step was crucial because of the high risk of creep. Indeed, laminates being cured at room temperature, the glass transition temperature T_g was close to the ambient temperature. Moreover, the set temperature for the post-curing was fiber non-destructive because as reminder, the degradation temperature of the main flax components is above 200°C. Therefore, post-curing did not alter mechanical properties of samples.

6.1.2 Specimens cutting

As described in the chapter 4, the prepared laminates had to be cut to obtain the samples with the recommended dimensions for the mechanical tests. The first batch of UD laminates was machined by milling and the second one with fabrics and carbon was cut by water cutting. Although the milling gave samples less sharp edges than water cutting, this technique seemed to be less "aggressive" for samples. Indeed, water cutting damaged samples, in particular those with carbon. Many of them presented delamination defects as shown in Fig. 6.4 which may be prohibitive for some tests as bending and ILSS .



Figure 6.4: Typical delamination defects found on carbon samples after water cutting. Remark: the sample is not loaded.

Those defects could be explained by a poor compatibility between carbon fabrics and InfuGreen 810 or an important increase of stress during water cutting.

6.2 Tensile properties

6.2.1 UD laminates in the fiber direction

In this section, the results obtained for the UD specimens loaded in tension in the fiber direction is discussed. The first point to note is the overall shape of the tensile curves. Indeed, despite the difference between the values such as Young's modulus and stress at break, all the curves present the same shape that was also highlighted in the state of art. As a reminder, *Mahboob et al.* [38] observed a variation of the tensile modulus at approximately 0.2% strain and the same behavior can be noticed in the curves presented in this work. Moreover, as expected, the curves present the same general behavior than the fiber itself. It confirms that in UD composites, when it is loaded in the fiber direction, it is the reinforcements that take up the forces and not the matrix which has the main role to transfer the stress between fibers.

Regarding the tensile modulus, several points can be discussed. First, following the ASTM standard, the approximation of the Young's modulus is wrong because of the inflection region. Indeed, as the latter was within the strain range imposed by the standard, this led to an underestimation of the tensile modulus. The alternative in this case was to modify the range and compute the approximation by avoiding the effects of mounting in small deformations and the inflection point. It is important to take measurements in the first part of the curve because after the transition region, the regime changes for flax and it is no longer the same deformation mechanism which is in place. According to this strain range modification, the results in this work for stiffness –between 17 and 28 GPa– are

similar to the results reported by *Liang et al.* [39] – i.e. 22.8 GPa– but are lower than results presented by *Mahboob et al.* [38] –i.e. 31GPa. It could be explained by the different volumes of fibers reported by the authors, around 43% and 50% respectively. With these information, the current laminates should have a fiber volume approaching 40%. This hypothesis could be confirmed through several approaches. The first one was to use Eq. 2.23 and the second one was to use the rule-of-mixture described in section 2.1.2 and extract the Young's modulus of the composite from Eq. 2.15. According to Eq. 2.23, the fiber volume fraction lies between 30-40 %. Even it only gives an approximation, it is a good approach to investigate if the difference in fiber volume fraction is correlated to the scatter of properties between the laminates. Therefore, if the variation of tensile modulus shown in Table 5.2 is compared to the variation of fiber volume fraction shown in Fig. 5.3, a correlation between both can be noted. But despite the variation, the tensile modulus remains in the same range. The dispersion of the Young's modulus could be explained first by the variation of fibers content between the manufactured laminates as previously described, but also within the laminate itself. The stiffness depending on components volume fraction, if the matrix volume fraction locally increases, it could be sufficient to have variation between specimens from the same laminate. An other hypothesis is the misalignment of the fibers in some regions of the laminate. The variation of fiber volume fraction and misalignment could be caused by the manufacturing technique. In addition to the difficulties described above with the cutting and the use of a brush, it could induce fibers movements during impregnation because of the lack of support in the transverse direction for UD reinforcements.

Regarding the failure of the composite in tension, the stress at break is approximately 250 MPa for specimens tested in the fiber direction with some variations. With these values, the results are lower than the stress results determined by *Mahboob et al.* [38] –287 MPa. This difference remains consistent with the assumption that the fibre volume is lower in this work and it has also an impact on the stress at break. However, it has to be noticed that all specimens break inside the grips area therefore it could be assumed that the stress is underestimated because of the higher stress concentration in the grips. This hypothesis could be confirmed with supplementary tests but this time with additional tabs stuck at the extremities of the specimens to improve load introduction.

Fig. 5.2a shows a fiber dominated failure but some specimens show smoother failure profile. this kind of failure is found for samples manufactured with new UD reinforcements while uneven profile is found on samples reinforced with old UD. For some specimens, as shown in Fig. 5.2b, failure is initiated in two different region of the sample and the final collapse occurs when a crack occurs and joins the two initial damaged areas. This longitudinal crack could be either the failure of the matrix or the decohesion between the fiber and the resin. If the tensile failure for UD specimens is analyzed at micro scale, several points can be highlighted. As shown in Fig. 6.5a, after failure, many fibers are denuded and do not have matrix residues. It could show a weak adhesion between fiber and the matrix. In Fig. 6.5b, close fibers present the same "zigzag" fracture path determined by *Rohmàny et al.* [33] schematized in Fig. 2.25. However, as shown in Fig. 6.6, in addition to the transverse failure of elementary fibers also highlighted by the same author [33], some of these had also longitudinal failure. The roughness of the surface for the transverse failure is due to the failure of all microfibrils constituting the elementary fiber.

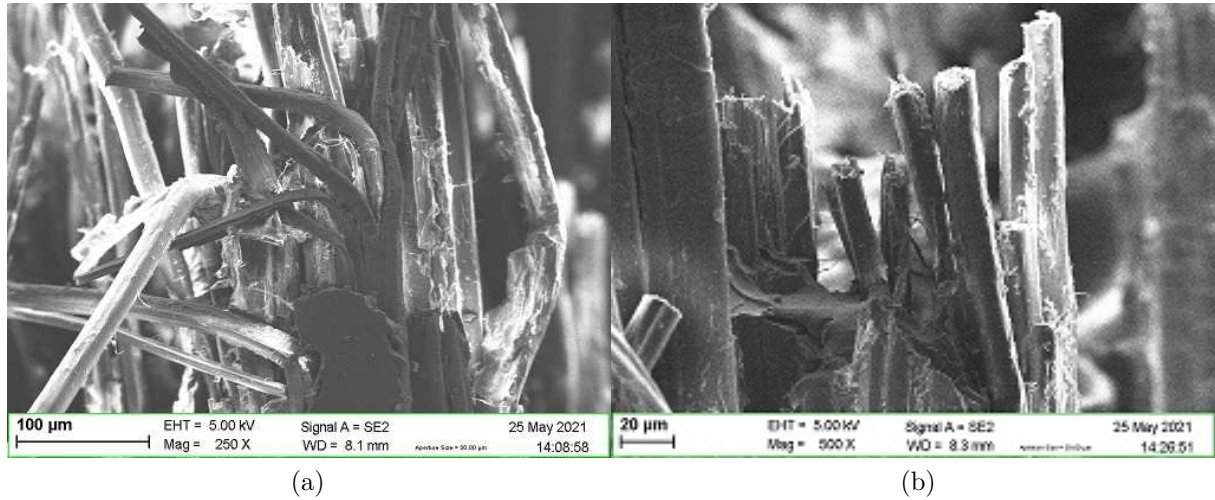


Figure 6.5: SEM micrographs of sample 20L4UD_1 after tensile failure (a) denuded fibers without resin remains (b) broken fibers presented zigzag failure path as reported by [33].

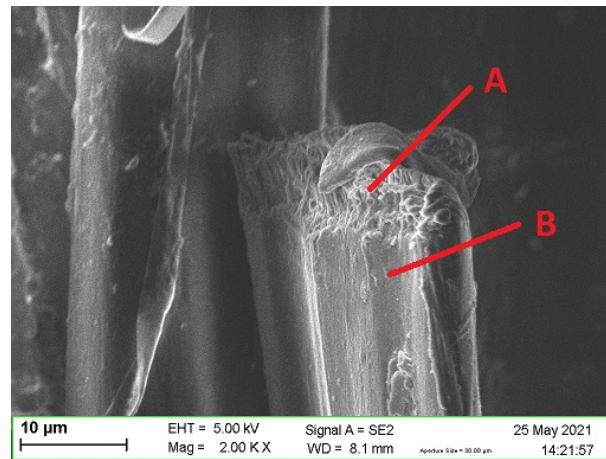


Figure 6.6: SEM micrograph of an elementary fiber from sample 20L4UD_1 after tensile failure (A) transverse failure (B) longitudinal failure.

Although different parameters were changed during the laminates preparation such as the drying and the degassing, any significance variations was reported. Regarding the drying, as reminder, *Baley et al.* [21] determined that drying treatment had a negative impact on the mechanical properties of the fiber and thus the composite. At first sight, the results obtained in this work contradict this idea but a certain nuance must be done. Indeed, for their experiments, *Baley et al.* dried fibers at 105°C during 14h while the fibers in the present work were dried at 70°C during 2 hours. Therefore, it could be assumed that the drying in the present work was less aggressive and perhaps even inefficient to remove enough water from the fibers to induce a drop of tensile mechanical properties. This hypothesis can be reinforced by the comparison between the value of stress at break of 210 MPa determined by *Baley et al.* [21] and the present stress. Regarding the degassing, the results in this work do not present any significant difference between the untreated specimens and the degassed ones. It could be explained by two points, the curing pressure and the degassing step. First, *Li et al.* [43] determined that high pressure –range between

5 and 15 bars– could induce a decrease of the porosity in the laminate. In this case, even if the pressure with clamps is higher than atmospheric pressure, it stays quite low in comparison to the work of *Li et al.* [43]. Furthermore, air bubbles can be trapped inside the yarn and the degassing during 15 minutes could be not efficient enough to remove entrapped air and positively impact the tensile properties.

However, the UD samples extracted from the RTM plates do not show the expected results at all. Indeed, as the technique is normally more efficient in removing air bubbles and excess resin, the properties should be improved. As shown in the results, the tensile properties are lower. Two causes could already partially explain this difference:

1. As described in chapter 2, gauge length has an impact on tensile properties of fibers. With the current results, it could be assumed that this impact is also present for composites. This hypothesis may be rejected according to the results found for tensile properties of flax strands discussed later on.
2. The hybrid process was not optimal and the resulting laminate has no better properties. This hypothesis could be explained by the estimated fiber volume fraction which is lower in this case.

Regarding the change of strands structure between the old UD fabric and the new one, any significant difference in term of properties can be determined except the failure profile. This shows that in this case, the change in the structure of the reinforcements between the two used UD fabrics had no impact on the properties of the composite.

6.2.2 UD laminates in the transverse direction

For the only specimen loaded in tension in the transverse direction, some points could be highlighted as well. First, the look of the curve differs from specimens loaded in the fibers direction because of the absence of transverse fibers. Therefore, the matrix takes up the load. The curve shows the linear behavior of InfuGreen 810 resin and in this case, range defined by ASTM D3039 standard remains consistent. Nevertheless, even if the tensile modulus is similar to the one determined by *Jost N.* [16] –approximately 3GPa–, the current value of the stress at break is lower than the yield stress previously determined. A first hypothesis could be a difference in the failure mechanism. Indeed, when the resin is loaded alone, the failure comes from the breaking of the polymer chains. In this case, other possibilities are the failure occurring at the interface fiber-matrix or a failure within the bundle/strand where the resin could not be able to impregnate. If the adhesion is weak, decohesion can occur before the failure of the matrix itself. The hypothesis of the weak interface adhesion between the two components is reinforced by the information given in Fig. 6.7. This SEM picture shows denuded fibers where any resin remains can be seen. Moreover, the imprint left by removed fibers shows decohesion without resin stripping.

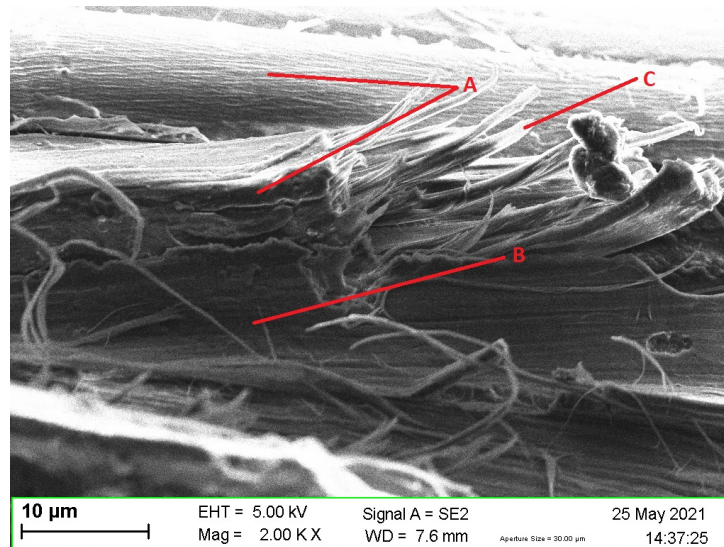


Figure 6.7: SEM micrograph of the sample from 20L8UD_0 laminate broken under tensile load (A) denuded fiber (B) flax fiber imprint (C) broken mesofibril.

6.2.3 Fabric laminates

As expected, the tensile properties of laminates with twill reinforcements are lower than laminates with UD –17-28 GPa for UD against 6 GPa for Twill. These results are in accordance with the literature that shows fabrics are less rigid than UD sheets. Regarding the tensile stress-strain curve, twill laminates show the same behavior than UD with a change in modulus during the loading due to the plastic deformation of the flax fiber. Nevertheless, the knee point in this case is less obvious compared to UD laminates which could be also explained by the architecture difference.

Regarding the failure of fabrics under tensile load, the breaking surface can be interesting to analyse. As shown in Fig. 6.8, broken flax yarns are pulled out of the matrix and some imprints of removed yarns are also visible. These are perfectly circular and there is no resin stripping. Once again, it can support the hypothesis of a weak adhesion between flax yarn and InfuGreen 810. Another event which can confirm this low adhesion is the presence of exposed transverse yarns. Since the fracture surface is exactly at the level of transverse strands, this could be due to the decohesion of the matrix during loading which can be seen as a crack at the fibre-matrix interface and could therefore propagate until the sample breaks.

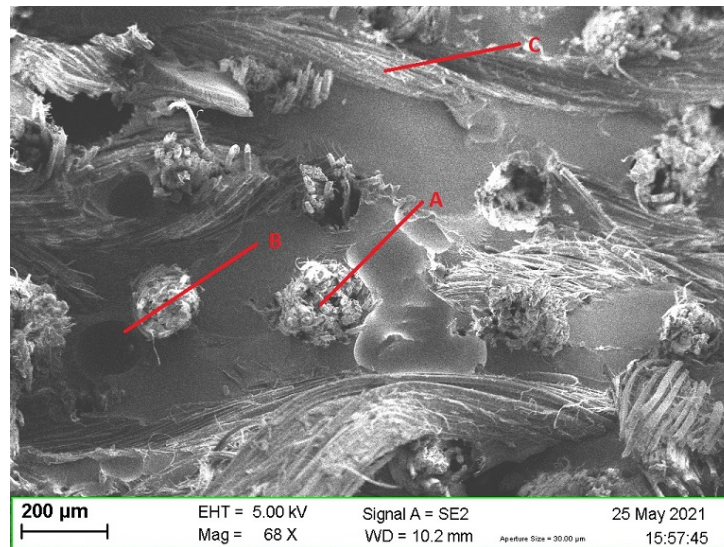


Figure 6.8: SEM micrograph of a sample from 20L12S_0 laminate broken under tensile load (A) pulled-out flax yarn (B) flax yarn imprint (C) denuded transverse yarn.

Regarding the results reproducibility, the lower scatter could also come from the architecture. As already noticed, the UD sheets were difficult to impregnate without induce fiber movements because of the lack of transverse support. But in the case of twill, fibers being equally knitted in both direction, they are less free to move and thus, fibers stay well distributed in the final product despite the manual impregnation.

As for UD, the variation of parameters such as drying and degassing did not impact the samples and show the same general behavior. Furthermore, samples manufactured with the RTM device do not show higher properties which may confirm the hypothesis that the hybrid process is not well optimised for flax fibres in this work.

6.2.4 Carbon laminates

Regarding the carbon laminates, samples show better tensile properties than flax –elastic modulus between 6-30 GPa for flax against 64-67 GPa for carbon– as expected but also higher fiber volume fraction. These results are in accordance with the principle that composites present intermediate properties between those of their components, since carbon fibre has higher performances than flax. The linearity of the curve shows that carbon fibre does not yield like flax fibre before breakage, but loading-unloading tests could be useful to determine whether the matrix also remains in the elastic range. It shows the difference between the inner structure of both fibers. As expected, any significant difference in properties is noted between the laminates possessing varying thicknesses but RTM laminate showed higher stress at break than others. In this case, it could indicate that other parameters than fiber fraction impact stress at break. One of these parameter could be the porosity of the laminate. As reminder, for the hybrid process used with the RTM machine, after fibers impregnation with resin, matrix excess and air bubbles were removed by vacuum process. In this case, the higher stress at break could show the efficiency of the device to remove air bubbles which can be seen as defect and promote crack propagation during loading. Regarding the specimens after tensile failure, as shown in Fig. 6.9, fibers are not perfectly denuded as flax and a lot of residues are still present.

Those could be resin remains and in this case, it could signify that the adhesion between carbon and Infugreen 810 is stronger than flax.

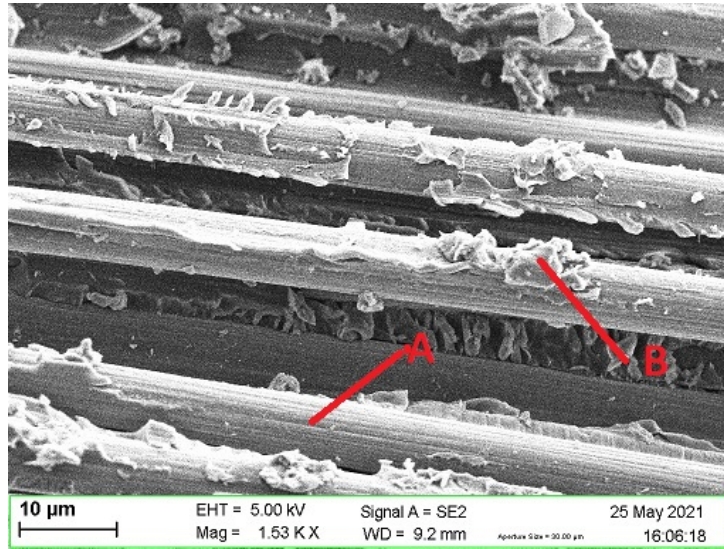


Figure 6.9: SEM Micrograph of a Carbon/Infugreen 810 sample after tensile failure (A) carbon fiber (B) potential resin residue.

For tension, delamination defects in carbon samples described earlier did not have significant impact because all samples broke out of the grips, where defects were.

6.3 Compression properties

6.3.1 UD laminates

In the fiber direction, the biaxial curve described in the work of *Mahboob et al.* [38] is also present for the current specimens. This curve in two parts could be explained by the complex structure of the fiber itself as for tensile test and the second part of the curve could represent a plastic deformation in the fiber. The inflection region is around 0.5% strain, similar result as obtained by [38]. It could show that matrix does not impact the yield resistance of flax. Regarding the compression modulus, according to ASTM D6641 standard, the composite is less stiff in compression than in tension, 10-16 GPa against 17-28 GPa, respectively. This difference could be explained by the presence of irregularities as waviness in the fibres and compression properties are more sensitive to this as mentioned by *Liang et al.* [39]. Regarding the modulus, the difference between the two laminates in this case may depend mainly on the fibre volume but also the reinforcement architecture. Indeed, RTM6UD_0 is manufactured with old UD while 20L8UD_2 is manufactured with the new one. However, over the strain range of the modulus calculation, the percentage of bending remains below the admissible value for all samples, therefore buckling does not impact stiffness property.

Another point that must be highlighted is the difference in term of strength, indeed, the compressive strength is lower than tensile strength. This difference in properties could confirm the presence of kink bands on fibers also described by *Liang et al.* [39]. This defects appear in general during the decortication process and have a very negative

impact on compression properties. In addition, the stress at break could also be influenced by the buckling of the samples. The bending percentage increases more rapidly in the RTM samples, which indicates that buckling occurs earlier in these specimens. It can be explained by the difference in thickness for both sets, small laminates being thicker, they better withstand buckling. However, despite the greater thickness for small laminates, buckling is dominant at the end of the test for both sets of specimens, which distorts the stress at break value.

In the transverse direction, before the gauges saturation, samples show once again more linear behavior. The modulus in compression is also similar to the modulus determined by *Jost N.* [16]. In this case, the strain range for the computing of the modulus was modified because of test assembly misalignment. This issue is not present for other samples for which this defect was compensated. Regarding the stress at failure, this one can not be impacted by kink bands in fibers but buckling can.

6.3.2 Fabrics

For fabrics in compression, even if the specimens have lower properties than UD, the elastic modulus is in the same range either in tension and compression -around 6 GPa. It could show that the presence of weaving in the architecture impact in the same way tensile and compressive elastic properties and defects such as kink bands has less impact compared to weaving. Furthermore, the majority of the samples have admissible bending percent in the given strain range, therefore the average modulus for all samples can be determined as a good estimation of this property. However, all specimens buckled before compressive failure and as for UD laminates, the extracted stress at break values could not well represent the properties at failure.

6.3.3 Carbon

As expected, carbon composite show higher properties than flax –Modulus and stress at break within the ranges of 60-67 GPa and 425-480 MPa, respectively. As shown in results, carbon composites have similar stiffness properties in tension and in compression but samples in compression show lower stress at break. For flax, the presence of weaving in the fiber could impact stiffness, but according to the results in this work, not for carbon. Therefore, weaving in carbon reinforcement could have a low negative impact on stiffness but not significant in contrast to properties at failure.

6.3.4 General remark for compression testing

The use of gauges for the compression strain measure was not straightforward and therefore some bias could appear in results. First for some specimens, only one gauge transmitted signal during testing or one transmitted incoherent signal. For these samples, no bending percent was computed to check specimen buckling. But, these for which the only admissible gauge emitted similar signal than samples with two functional gauges, samples were kept for the properties determination. Furthermore, for some samples, bending percent was above the admissible value of 10%. Most of them were arbitrary kept because their percent was not significantly higher than the limit and their resulting properties were coherent with admissible samples.

A last important point to notice is the difference in fracture mechanisms for the different samples. Indeed the basalt and carbon failed as wanted but many flax samples buckled before the end of the test. This phenomenon is unwanted for this kind of test because, as already said, it does not give the actual properties of the composite. This type of deformation happens in compression when the samples has a too low thickness and it can't handle the load without buckling. Although ASTM D6641 gives formula to estimate the admissible thickness, precise properties need to be known as shear modulus etc, properties that have not been studied in previous works.

6.4 Flexural properties

6.4.1 UD laminates

In the fiber direction

As for tension and compression, the general behavior of the curves in bending in the fiber direction is discussed first. Once again, the biaxial behavior which illustrates two different states is present for flexural curves. In the first part of the curve, the fiber elastically deforms while in the second part, a plastic behavior takes place until the fracture. The inflection point seems to be at higher stress than in tension which could show a better resistance to yield in bending for flax fibers.

Regarding the Flexural modulus, in agreement with literature, samples for each batch have lower flexural modulus than in tension –around 16 GPa for bending against 22 GPa for tension. The similarity between curves shows as for tensile properties that thickness does not impact bending properties. For RTM samples, as for the tensile results, the lower fiber fraction impact properties. In addition, the plateau could be a consequence of non planar sample which leads to a high backlash compensation at the beginning of the test. This deformation could be due to RTM plate creep before the post-curing stage. The deformation may not have been able to influence the other tests as the samples were realigned by the grips.

Regarding the failure of the laminates, all samples present a quite similar failure except for RTM specimens which show a longer plastic region than others. This behavior could be associated to the higher matrix fraction. It could have more impact in the fiber direction for bending than in tension and could increase the ductility of the composites.

In the transverse direction

As expected, properties in transverse direction are lower and modulus is similar to the modulus of the matrix InfuGreen 810 –around 3 GPa. Regarding the stress at break, the value found for the both 4 plies laminate –35 MPa and 4 MPa– are closer than in tension –16 MPa– to the yield stress value determined by *Jost N.* [16]. The plateau after break could also confirm the dominant behaviour of the matrix in this direction. However, as already described, samples with 8 plies have lower stress at break in this direction. As the thickness should normally not affect the properties, it is possible that in this case, other failure mechanisms may have occurred during the loading of the sample such as fiber-matrix decohesion.

6.4.2 Fabrics

First of all, the general remark about the curve behavior for UD in the fiber direction is applicable for fabric samples as well. For fabrics, samples show similar elastic properties in bending than for tension and compression and the dispersion for the flexural modulus is low comparing to UD. For the stress at break, flax fabrics show higher property in bending than in tension –138.3 MPa against 100.5 MPa in average respectively–, it could show a better resistance for this type of loading compared to tension.

Regarding the reproductibility, results are more dispersed compared to the results found in tension. It could be explained by a higher sensitivity to the variation of certain parameters intrinsic to the composite but difficult to identify in this case. Another supposition is the influence of the cutting as described earlier in this chapter. Indeed, even though it was less significant than for the carbon, some of the samples had delamination defects at the entry point of the cutting tool. Depending on the localisation of this defect during the test, this may have more or less influenced the breakdown of the sample. A detail that could confirm this hypothesis is the difference in the stress at break values between the RTM samples in the longitudinal and transverse directions. During the cutting process, as the water jet follows a very precise path, all the "longitudinal" samples have the entry point of the tool along their width. However, the transverse samples having the entry point at the centre of their length, this defect is aligned to the load cylinder of the testing device and could induce an early failure of the sample.

On the other hand, theoretically, when using fabrics as reinforcement, the properties of the composite in the weft direction should be lower than in the warp direction due to the higher weft yarn caking. Although this hypothesis could be raised in view of the results, it must be rejected in this case. Indeed, as the reinforcements were randomly arranged during the manufacturing process in contrast to carbon laminate, the probability that the weft threads are all in the same direction is very low.

6.4.3 Carbon

As expected, the mechanical properties in bending for carbon samples are higher than flax samples. But as noticed in the results, curves are highly dispersed. As for fabrics, this difference could be explained first by the presence of defects and secondly, by the test parameters. For all samples in carbon, delamination defects are often already existing at the edges of the samples. Cracks being not closed by any grips in contrast to tensile test, it can propagate before reaching the maximal bending stress. Two illustrative examples are shown in Fig. 6.10. In Fig. 6.10a, the sample is defined as "RTM8G_0_F1T" and presented large delamination before test. The resulting flexural modulus and stress at break were 31.2 GPa and 432.2 MPa respectively against 55.6 GPa and 681.6 MPa for sample which did not have such a defect. The same behavior was noticed for all samples with 12 plies, Fig 6.10b showing the sample defined as "20C12G_0_F2" –9.4 GPa and 345.8 MPa for modulus and stress at break respectively. Therefore, it can be determined that samples with 12 plies and other samples showing lower values do not show actual flexural properties of the composites.

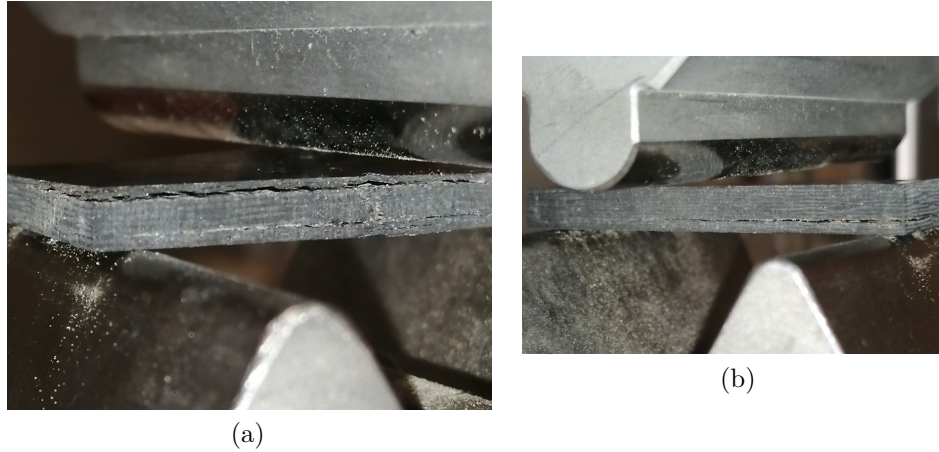


Figure 6.10: Delaminated specimens before bending test (a) RTM8G_0_F1T (b) 20C12G_0_F2.

Another point to highlight, samples for bending test must have varying dimensions depending on the thickness of the sample but it was not taken into account during the cutting design. Samples with 8 and 12 plies should have greater test span than thin samples to prevent deformation mechanisms other than bending. In this case, the span length was smaller than the given length by the standard for thicker samples. Therefore, other mechanisms than bending could take place during the test such as shear and promote early failure.

6.5 Interlaminar shear strength

The ILSS in this case is very difficult to discuss because of the incertitude linked to the experiment. In this case, the failure mechanism was crucial and as specified in the associated standard, if the sample broke in bending or was plastically deformed, the result was not admissible.

Regarding the UD samples, according to the associated curves and their behavior during the test –example shown in Fig. 6.11–, all the samples broke in bending or were plastically deformed. Therefore, the computed apparent shear strength is not admissible. One of the possible reasons is the thickness which was too low compared to the minimal thickness specified in the chosen standard. For ILSS testing, the span between the bottom supports must be tight enough to promote shear mechanism during sample loading. This length being proportional to the thickness of the sample, this parameter must be high enough to obtain a span length within the range allowed by the test setup. Because specimens were too thin, a larger thickness/span ratio had to be used in order to use the assembly and therefore, shear mechanism did not set up and bending took over. The only laminate where the failure could approach a shear failure is the 20L8UD_2 laminate. When the curves of the two tested samples are compared to the other UD specimens, they differ strongly, which shows another type of behaviour than plastic deformation. However, it is difficult to distinguish between bending and shear failure. As reminder, curves are available on Appendix A.

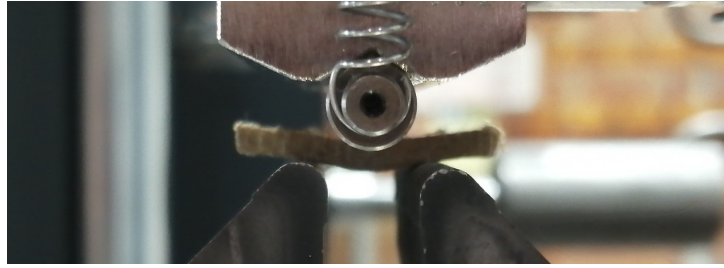


Figure 6.11: Plastic deformation of UD sample during ILSS testing.

For fabrics, the curve plots in Fig. A.2a are really close to bending curves for the same specimens. Therefore, even if the curves are not directly comparable, it could bring a certain incertitude about the failure mechanism. One detail which could show a shear failure for fabrics is the behavior of RTM8G_0 specimens in the transverse directions. Those have successive drop of stress at the end of the test which is defined as multiple shear typical failure in the standard EN 2563. But assuming that the latter curves approach shear failure, the question could arise for the others. Indeed, since the other samples have the same curve except for the failure, it would be logical to assume that they also undergo shear, except that this time, they failed in simple shear.

Regarding the carbon, as already mentioned in the results, the scatter is high. This dispersion could have the same origin than for bending tests, i.e. the presence of delamination defects. Those impact more or less the integrity of each the sample depending on their severity. Therefore, even if the samples failed by shear, the extracted property could be underestimated. However, one laminate can show the actual apparent shear stress which is the laminate 20C4G_0. As shown in Fig A.2b, the samples from this plate have a similar behavior until failure which could show an absence of defect. However, it is still difficult to confirm shear failure instead of bending.

6.6 In-plane shear strength

As noted in the results, the difference for in-plane shear modulus between the two tested fabrics is significant -1.3 GPa for flax against 3.4 GPa for carbon— while the curves scatter remains relatively low. This high gap between the two sets of specimens could show a behavior dominated by fibers in the elastic region. In this case, it is difficult to evaluate the influence of fabric structure —twill for flax and satin 5 for carbon— on results, carbon having much higher properties than flax to begin with. However, after the transition region, the behavior of the two sets are similar, even if the carbon curves show a regime slope a bit higher than flax. The transition part of the curve could therefore show a matrix dominated behaviour and perhaps a supplementary impact of the interface fibre-matrix. The last part of the curve could show a plastic deformation of the matrix but as the tests were stopped when the gauges were saturated, it is difficult to know the level of involvement of the fibres and the matrix in this second part.

Regarding the maximal shear stress, this latter having been computed from an linear extrapolation, an under/over estimation is still possible. Indeed, the last part of the curve is approximated as linear which actually, could be not the case. But since the last valid data were very close to the 5%, the values can still be considered as a good estimation of the properties.

6.7 Micro-hardness

Regarding this last experiment, as noted in the results, all samples presents scatter for the results. First of all, results are scattered because of the localisation of the imprints through the tested surface. Indeed, the samples are highly heterogeneous because of the presence of the two constituents and the results differ depending on whether the indentation is made on fibres or on resin as shown in Fig. 5.19. Therefore, the average hardness is not representative of global properties of the sample. Indeed, 20L8UD_2 appears to be harder than 20L4UD_1 in the longitudinal direction but more indentation were done on fibers in the first one which increases the global hardness of the sample.

The same bias occurs for the samples in the transverse direction but this time, it is the sample with more indentation in the resin which appears harder.

Another point to notice is the difference regarding the elastic springback of the components. In Fig. 5.20a, when the indent touch simultaneously fibers and resin, the indentation remains well defined at the level of the fibres while the resin undergoes an elastic return which distorts the final imprint. In the case illustrated in Fig. 5.20b, the indent pushed on a fiber parallel to the surface. In contrast to the indent shown in Fig. 5.19a, fiber parallel to the surface seems to only elastically deform during the test, the latter shows no trace of an imprint after the punch was removed while it is visible on the surrounding resin. These behaviors could therefore induce bias in the final computed hardness and could increase the results scatter. This also shows the difference in behaviour of the fibre depending on its orientation during the test.

6.8 Tensile properties of single flax strand

The purpose of the experiment was firstly to investigate the variation of mechanical properties according to the strand length during the tensile test. In addition, the theoretical fiber volume fraction could be estimated from the Voigt and Reuss model according to the mechanical properties of tested composites. Regarding the variation of fibers tensile properties, as shown in the results, any significant variation between the tested lengths is detected, in contrast to the results obtained by *Amroune et al.* [35]. But, due to the lack of time and the used material, some nuance must be done between the results found in the literature and the current ones. In this work, only yarns and not technical fibers were tested to obtain properties at the macro scale. The obtained properties and their invariance despite the length change could show that the influence of the testing length disappears at the strand scale. However, only five samples were used for each batch and this quantity was maybe to low to extract a correct tendency curve for the properties.

Regarding the curves, the absence of bilinear behavior for new UD yarns could be explained by the used load cell which is not enough sensitive for the actual load range. But for old UD roll, strands being thicker, the range of load better fit with the used cell and the typical curve for flax fibers was visible. In addition, when the current results for new UD yarns are compared with those found by *Yan et al.* [9], they have properties in the same order of magnitude despite the large dispersion.

As indicated in the results, old UD strands have significant lower properties than other tested yarns –2.5 GPa of modulus average for UD OLD against 17.5GPa for UD NEW. It could be explained by the difference of structure between the old roll and the new ones

as shown in Fig. 4.1a. Although strands from old UD roll present lower properties, its structure can result in a larger volume of air between the fibres making up the strand. As this additional volume could not be subtracted when measuring dimensions for the test, the apparent cross-section of the strand is greater than it would be if it did not contain air. But as this air does not provide any resistance in the tensile tests, the final calculated stress is highly underestimated. However, a correlation can be noticed between the Young's modulus and the UTS, both values vary in the same way for each sample.

Finally, strain at failure is not easily extractable because of the absence of extensometer. Only the grips separation is available and because of the mounting, as shown in Fig. 5.21, the loading of each fiber does not begin at the same time and plateau are very variable.

6.8.1 Approximation of fiber volume fraction for UD flax laminates via Voigt and Reuss model

In Fig. 6.12, three Voigt and Reuss models are drawn according to different tensile properties as following, using Eq. 2.15 for the upper limit and Eq. 2.15 for the lower one:

1. The model "UD NEW" is computed from the average of tensile modulus "UD_NEW" found in the previous part for fibers and from tensile modulus reported by *Jost N.* [16] for the InfuGreen810.
2. The model "UD OLD" is computed from the average of tensile modulus "UD_OLD" found in the previous part for fibers and from tensile modulus reported by *Jost N.* [16] for the InfuGreen810.
3. The third model is computed from the tensile modulus reported by *Amaro J.* [45] for fibers and from tensile modulus reported by *Jost N.* [16] for the InfuGreen810.

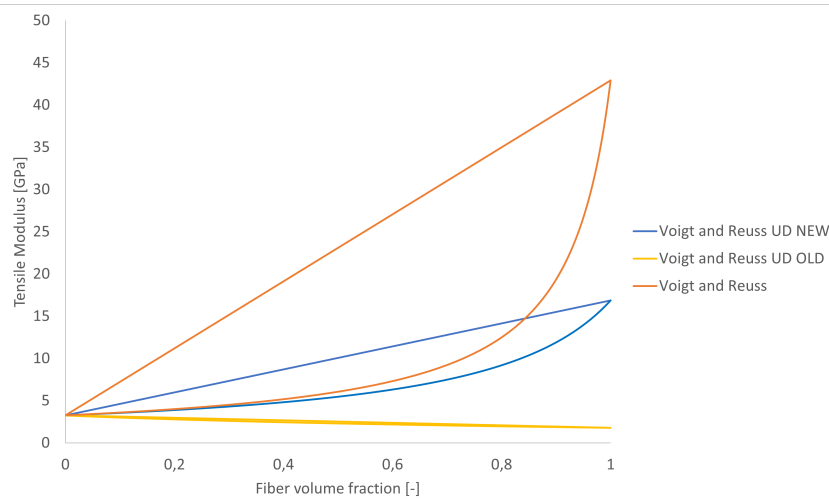


Figure 6.12: Computed upper and lower bounds of UD flax laminates tensile modulus according to the rule-of-mixtures.

As drawn in Fig. 6.12, the three models are very different from each other and the two models based on strands properties from this work are actually not usable for further computations.

For the approximation of the volume fraction, the Voigt and Reuss model is used to interpolate the V_f value according to the determined tensile moduli of laminates available in Table 5.2. Moreover, the current estimated V_f for UD laminates to the values previously computed with Eq. 2.23. As shown in Fig. 6.13, there is a significant gap between the both sets which can indicate significant bias during approximation. Actual values are likely to be found between the two sets of values. However, the volume fraction evolves in the same way for both sets of values between the laminates, which confirms a correlation between the final thickness of a composite, its volume fraction and their impact on the Young's Modulus.

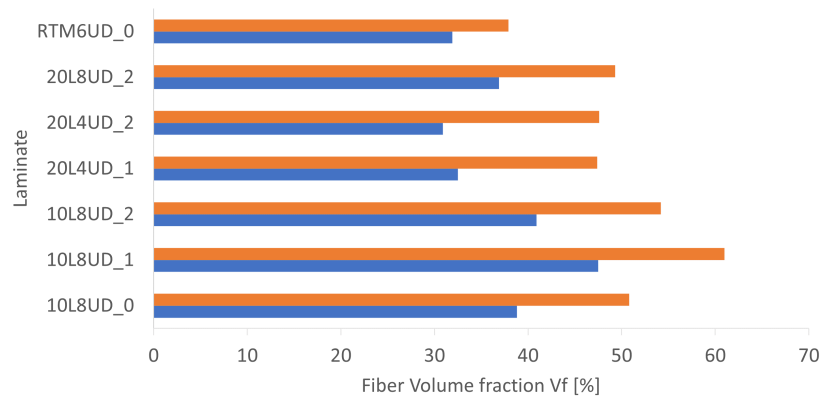


Figure 6.13: Estimated V_f in percent for UD laminates. The values in blue are the values estimated from Eq. 2.23 while values in orange show values estimated from the Voigt and Reuss model.

Chapter 7

Conclusion

The main objective of this work was the mechanical characterization of partially bio-sourced resin InfuGreen 810, mainly reinforced with flax fibers. To reach this goal, composites laminates were first manufactured with hand layup technique and then cured with the curing cycle previously developed for the resin of interest. To assess the influence of manufacturing process, fibers drying and composite degassing were performed on some specimens. Moreover, to investigate the impact of reinforcement architecture, UD and twill fabrics were used to allow a direct comparison between both during testing. Then, mechanical tests were performed on composites specimens to determine quasi-static mechanical properties of manufactured laminates.

First for all, for flax reinforced specimens, the variation of process parameters did not significantly impact the properties of the materials. But some defects such as porosity and delamination associated to the manufacturing may negatively influence the composite behavior. Regarding the general behavior of flax laminates, for twill fabrics and UD in the fiber direction, the bilinear curves for all tests showed a fiber dominated behavior. Linear curves are present for UD flax laminates in the transverse direction, showing a matrix dominated behavior.

For composite laminates reinforced with UD flax, for tension in the fiber direction, the Young's modulus lies between 17-28 GPa, the UTS between 180-280 MPa, with the scatter being mainly caused by the variation of fiber volume fraction. In the transverse direction, the modulus is approximately 3 GPa, a value very close to the modulus previously determined for InfuGreen 810. However, the low stress at break may reflect weak adhesion between fibers yarns and the epoxy resin. In compression, the modulus in the fiber direction was between 12-20 GPa while in the other one, similar property than InfuGreen 810 was noted as well. The dispersion in this case was mainly because of the volume fraction variation but also because of the used UD architecture. For both directions, stress at break may not give actual properties because of the buckling. For bending, the flexural modulus lies between 14-18.4 GPa and the stress at break is between 175-245 MPa in the fiber direction. The scatter was present for the same reason than in tension. In the transverse direction, the modulus was between 1.8-3.1 GPa and stress at break between 20.5-40 MPa. Regarding the micro-hardness, values lie between 11-18 HV, they are highly dependant of the tested region in the composite and the direction of fibers.

Regarding laminates reinforced with twill fabrics, specimens had globally weaker properties

than UD samples in the fiber direction but had lower dispersion both for mechanical properties and estimated fiber volume fraction. For tension, compression and bending, elastic moduli are similar and lie between 6-9 GPa. The stress at break is respectively between 93.5-108 MPa for tension and 110.5-148 MPa for bending. For compression, the stress at break is between 80-94.5 MPa but a bias may be present because of the buckling of the samples.

Regarding the carbon laminates, a comparison with flax allowed to see the high gap between the performance ranges of both resulting composites. First, the linear behavior of carbon in loading can be an advantage over the bilinearity of flax fibers. For tension, carbon specimens have tensile modulus between 64.6-67.8 GPa and an UTS between 743-975 MPa. For compression, the elastic modulus was between 60.5-67.5 GPa and the stress at break between 426-476.5 MPa. For carbon, buckling was not dominant in failure and therefore the computed values at failure can be considered as reliable properties. However, for bending, the scatter is higher because of the presence of cutting defects. The modulus lies between 40-55 GPa and the stress at break between 563-706 MPa.

For ILSS testing, all flax and carbon specimens presented non admissible behavior or uncertain failure mechanism and therefore the apparent shear stress may not be admissible as the actual property. A better thickness control during manufacturing must be done if supplementary tests are performed.

In conclusion, although flax fibers reinforced InfuGreen 810 offer lower performances than CFRP, this material could be a good alternative over other composites such as GFRP which has high carbon footprint as well. But quasi-static mechanical properties are not the only performances that can be determined and it may be interesting to assess further research to investigate other characteristics such as fatigue, thermal or even electrical properties. Another interesting topic related to these materials would be to investigate their recyclability. Indeed, "bio-sourced" does not necessarily mean that it is recyclable. This could therefore become a problem in the long term as single use materials can be considered highly polluting and do not contribute to a sustainable future.

Appendix A

Resulting curves for ILSS test

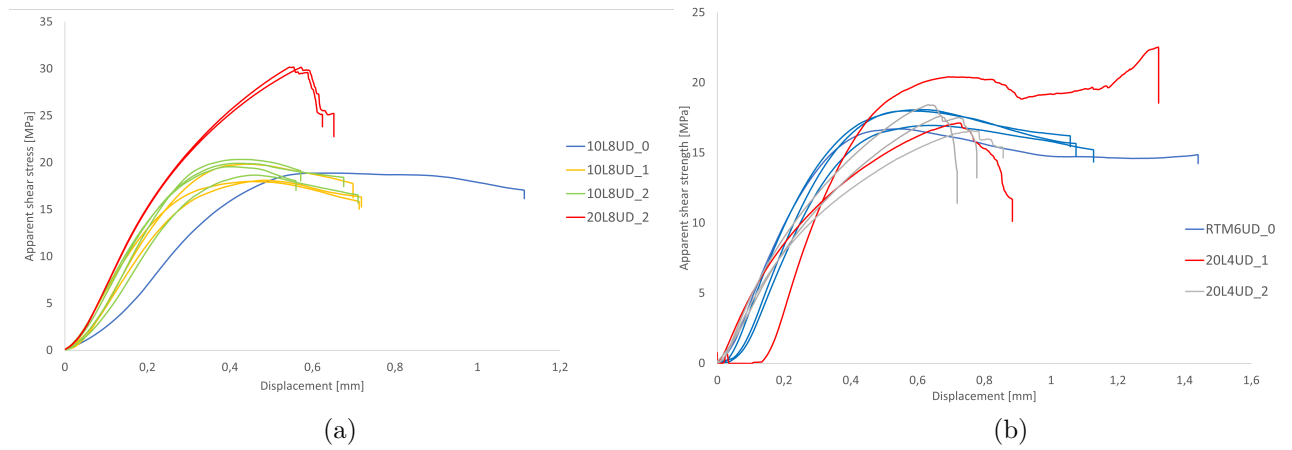


Figure A.1: Apparent shear stress-displacement curves for UD flax/Infugreen 810 laminates manufactured (a) with 8 plies (b) with 4 and 6 plies.

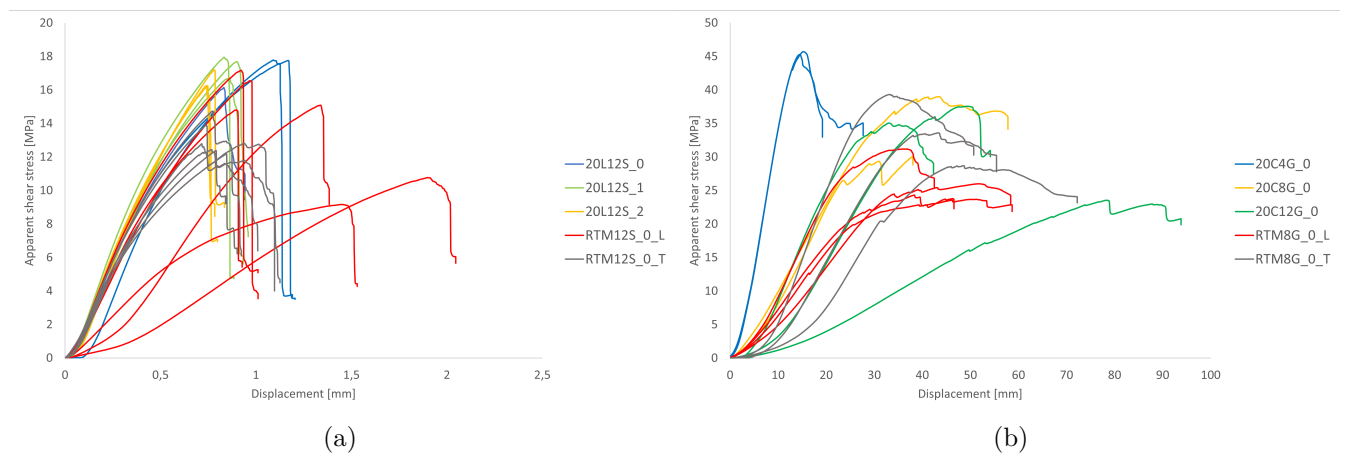


Figure A.2: Apparent shear stress-displacement curves(a) for flax twill/InfuGreen 810 laminates (b) for G0926 carbon/InfuGreen 810 laminates.

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