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Variability of mechanical properties of flax fibres for composite reinforcement. A review

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Abstract :

Flax fibres are a promising reinforcement in the development of biocomposites and are finding new applications in transport structures. However, there is a perceived problem with plant fibres related to the variability of the properties of these natural materials. This paper describes the factors which affect variability, from plant growth conditions to fibre sampling and testing. A large number of test results are presented (characterization of elementary fibres, bundles, assemblies of bundles, and unidirectional composites), and it is shown that provided fibre supply is carefully controlled, characterization procedures are appropriate, and manufacturing processes are optimal then excellent composite properties can be achieved with low variability.

Highlights

► This paper provides a unique review of a large quantity of data from hundreds of tests on flax fibers ► Data are discussed in terms of testing procedures and sample geometry ► Variability of composite properties is discussed in terms of fibre variability

Keywords : Flax fibre, Composite, Variability, Mechanical properties

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1 / Introduction to the use of flax fibres as a composite material reinforcement

The reinforcement of polymers by natural fibres makes it possible to propose a new class of composite materials (natural fibre composites, NFCs) that are distinguished from synthetic fibre composites (SFCs) by their lightness and a lower environmental impact. Numerous reasons explain the adoption of plant fibres in place of mineral or animal ones. For example, plants such as flax provide relatively long fibres (> 20 mm), with good tensile properties and low density (Bledzki and Gassan, 1999)(Müssig, 2010)(Bourmaud et al., 2018). They originate from renewable resources, are available in large quantities (significant annual crop yields) and allow, through appropriate use, reduced environmental impacts (Le Duigou and Baley, 2014). Concerning mineral fibres, asbestos is present in the earth's crust. Asbestos fibres are natural fibres, and were used for many years as a reinforcement for both organic and cement matrix materials (Gordon, 1994). They are no longer used in Europe today due to their toxicity. In the animal world, silk fibres show remarkable properties, in particular those of certain spiders, with very long elongation at failure and a large energy absorption capacity (Eisoldt et al., 2011)(Ko and Wan, 2018). To make a web a spider will produce different threads, whose properties depend on their function such as suspension, capture…

(Agnarsson et al., 2010). While these fibres are of great scientific interest they are not available in sufficient amounts for industrial use.

Of the plant fibres, bast fibres, particularly flax, hemp and ramie, are the most efficient in terms of Young's modulus and tensile strength relative to density, due to small microfibrillar angles and high cellulose content. Concerning matrix polymers, given that the performance of fully biobased polymers is not yet sufficient (especially with respect to moisture), partially biobased polymers are used but those from the petroleum route are still the most common. Today, the development of these biocomposites using the processes developed for SFCs is being considered for semi-structural parts in the fields of transport, leisure and furniture (Baley et al., 2018). Among the plant fibres used for reinforcing polymers, flax shows a variety of benefits and this paper will focus on this fibre.

The mechanical properties of plant fibres are often considered to show high variability. This large scatter is believed to depend, among other factors, on meteorological conditions during growth, and it is often cited as one of the limitations to the use of these fibres in structural applications. However, it must be remembered that the mechanical properties of synthetic fibres also show variations, even though they are produced by controlled industrial processes. The tensile strength of freshly drawn E glass fibres of diameter of around 10 microns is 3200 - 3400 MPa , but that of fibres taken from a sized yarn is in the range 2000 – 2400 MPa, and 1200 – 1550 MPa for a yarn with a large number of fibres (Berthereau and Dallies, 2008). This loss of mechanical properties (Coroller et al., 2013)(Lefeuvre et al., 2014a) may be explained by several effects operating during the production of the yarns: abrasion by contact between fibres or with tools, chemical effects due to moisture. Moreover, fibre sizing may be modified during storage, which can result in changes in fibre/matrix adhesion. Significant changes have been reported after glass fibre storage for 15 days (Peters, 2018). The variability in the mechanical properties of plant fibers does not limit their use in the textile field for two reasons: First, these fibres are used in the form of twisted yarns. And second, tolerable variations in the textile industry are much higher than those required in the design of structural parts.

The specific terms used to designate the different fibre assemblies are defined below. Figures 1.A and 1.B show the cross-section of a flax stem and a sector of this section, respectively. Within a unidirectional composite, the reinforcement is in the form of unit fibres (Fig 1.E), bundles (Fig 1.D) or bundle assemblies (Fig 1.C).

Fig. 1: Location of fibres in the stem (A, B and C) and representation of bundles (D) and unit fibres (E).

Flax is a member of the genus *Linum* in the Linaceae family. Among the 298 species of flax (Montaigne, 1997), the one which is most widely cultivated is *linum usitatissimum L.* : an annual plant that provides both fibres (Figure 2) and seeds rich in oil, reputed for their nutritional qualities. If the purpose of the crop is to produce textile fibres, a variety of textile flax is sown. On the other hand, if seed production for oil is preferred, oilseed flax will be sown. According to the main aim of the crop, textile fibres or oil, the choice of the cultivation soil and the desired growth conditions (number of plants per square metre, maturity at harvest, machines required ...) will be different. It may be noted that the oilseed crop also provides good quality fibres (Pillin et al., 2011) which are used to produce paper, including the paper used for the manufacture of banknotes (US dollars for example) which represents a very significant market.

Figure 2: Scanning electron microscope image of raw flax fibres

The flax fibres are found in the periphery of the stem (figure 1) (Morvan et al., 2003) (C Baley et al., 2018a) and are the mechanical support of the plant which is very slender (Goudenhooft et al., 2019a). Their remarkable properties (length of the unit fibres and good mechanical properties) can be mainly explained by their morphology and the nature of the constituents. The formation of the fibre, from the cell divisions in the apical meristem and their differentiation, includes elongation and growth in apparent diameter. At the beginning of the elongation, cells with different tissue destinations extend simultaneously (Gorshkova et al., 2003) (Snegirevaet al, 2006) to reach 200 microns with a diameter of 4 to 7 microns. After this stage of slow elongation (50 microns/hour) an intrusive type elongation follows, which is characterized by an extension of several orders of magnitude without increasing the wall thickness (Snegireva et al., 2010). Intrusive elongation is accompanied by overlapping neighboring cell walls (A) and increases the number of fibres in the cross-section of the bundles. According to (Esau, 1950) and (Rihouey et al., 2017), but contrary to a former opinion (Esau, 1950), it is after elongation that thickening of the walls occurs. The intensive thickening of the cell walls lasts about 60 days and takes place mainly below the snap point (Gorshkova et al., 2003). More precisely, the thicker cell wall of the fibres, called either S2 or G-layer, with a gelatinous appearance due to its gel-like matrix similar to tension wood fibres (Gorshkova et al., 2015), is progressively formed from the conversion of an inner sub-layer known as Gn-layer (Mikshina et al., 2013) (Rihouey et al., 2017). Later on, the G-layer increases in thickness until complete conversion of Gn-layer, which leads to a more homogeneous and compacted layer over cell maturation (Mikshina et al., 2013). The two-step development of the S2 layer was mechanically visualized by AFM (Method: Peak-Force Quantitative Nano-Mechanical property mapping (PF-QNM)) (Arnould et al., 2017). During the fibre development, the presence of two layers (layer G and layer Gn) with different indentation moduli illustrates their progressive development during thickening of the secondary cell wall. Finally, measurements were carried out on mature flax fibre cell wall samples; in this case, no significant stiffness gradient could be identified in the secondary S2 layer. At a low scale, this result highlights the homogeneity of the cell wall stiffness, independent of the area considered.

There would therefore be a temporal separation of the stages of elongation and thickening. However, there is also a spatial separation of these two stages, the transition being at the level of the "snap point".

The snap point on the stem corresponds to a point at which there is a variation in the flexural rigidity, visible on the plant. Above this point the stem is flexible, while below it is stiff (Gorshkova et al., 2003)(Goudenhooft et al., 2019b).

The cells of the bundles below this point no longer have intrusive elongation; the thickening of the walls begins in the cell layer located at the periphery of the bundle. This seems to be confirmed by the nanoindentation measurements of Bourmaud & Baley (Bourmaud & Baley, 2012), who report a greater apparent longitudinal Young's modulus for these peripheral fibres. However, the thickening of the walls is identical for all the fibres of the bundles located more than 200 mm below the snap point. The walls thicken over time, until the plants are uprooted. Thickening operates by cyclic deposits of cellulose on the walls, the number of cycles being greater at the foot than at the top of the stem. These deposits are in the form of nano-sized cellulose fibrils (almost completely crystalline) embedded in a complex organic compound consisting of other components of hemicelluloses, pectins and lignin. The cellulose fibrils are oriented in a helix at an angle of around 10° with respect to the stem axis. The large amount of these fibrils (> 65 wt.%) governs the mechanical properties of the flax fibre.

Figure 3 shows a 3D reconstruction of a fibre bundle including the fibre ends. The approach used to trace a portion of the bundle is described in Baley et al. (Baley, Goudenhooft, et al., 2018).

Figure 3. 3D reconstruction of a bundle in an area with a fibre extremity.

This figure was obtained by microscopy, reconstructed using observations of several sections of a short fibre bundle (Baley, Goudenhooft, et al., 2018).
The flax fibres have limited lengths and polygon cross-sections, and they show tapered ends. The figure indicates that the fibre ends are randomly distributed. Their distribution and geometry represent two aspects which limit strain concentrations within the bundles. This type of architecture and geometry results in little misalignment of the fibres. Within the bundles load transfer is through middle lamella.

To summarize, there is a longitudinal variability of the properties of the fibres due to a greater or lesser elongation of varying duration, between the fibres of the foot, the middle or the top of the stem. In addition, there is a transverse variability of the properties of the fibres due to the thickening of the walls and thus a decrease in the size of the lumen depending on the internal or external position of the fibre in the bundle. It is assumed that despite transverse variations in properties between the fibres of the same bundle, two bundles of the same stem and located at the same altitude will be more similar than two bundles taken at different altitudes in the stem.

In addition, a flax stem can also be considered globally: mechanical characterization shows that flax fibres contribute 71% on average to the flexural stiffness of the stem (Réquilé et al., 2018). The flax stem can be considered as a composite structure (Baley, Goudenhooft, et al., 2018) with an outer protection (equivalent to a top coat), a unidirectional ply on the periphery (supporting tissues assembled in bundles and absorbing the tensile or compressive loads) and a porous core (conduction tissues contributing to the flexural strength of the stem by limiting ovalization of the cross section and local buckling of the bundles) (Baley, Goudenhooft, et al., 2018). The good mechanical properties of stems and fibres (both are composite structures) can be explained by the nature of the compounds, their specific morphology and the mechanical characteristics of their components. In addition, the slenderness and layer morphology of these structures gives them exceptional resistance to buckling (Goudenhooft et al., 2019a)

2 / Control of the different stages of flax fibre production

2.1. Crop rotation

For farmers, good practices have been well documented (Bert, 2013). Flax is one of the crops farmed according to a rotation cycle so that the same plot of land will typically produce flax once every 6 -7 years. This is intended to avoid certain diseases or parasites that remain in the soil and it can also break the growth cycle of some weeds.

2.2. Selection of a species for a given location

Plant variety selection is a continuous occupation and is made with respect to the particular location and conditions. For example in France, each new variety is registered in the official French catalogue of species and varieties (and then in the European catalogue). Strict rules are applied; the new variety must be quite different from existing ones, homogeneous and stable, i.e. keeping its characteristics from generation to generation. It must also have a sufficiently high added value, with respect to agronomy, technology and the environment. For the selectors, the main criteria are the fibre yield, tolerance to diseases, and stability of the plant with respect to bending. The intellectual property (and thus the protection) of a new variety put on the market is guaranteed for 20 years.

It is important to note that flax plants are self-fertilizing. This guarantees the production of plants of the same grade from one year to the next. It takes between 9 and 11 years for a new variety to become genetically stable. To illustrate how new varieties can improve yield it should be noted that the production yield of flax fibre per hectare has increased by 2.3% (35 kg of fibres per hectare and by year) between 2003 and 2013, and is still increasing (Bert, 2013).

2.3. The number of plants per m²

The use of certified seeds provides a number of guarantees (germination power above 92%, cleanliness, sanitary quality), so losses are limited. Farmers also now have precision sowing equipment. It is recommended to sow between 1500 and 1900 seeds/m² in order to obtain between 1500 and 1600 viable plants per m² (Bert, 2013) (Figure 4). This parameter influences the morphology of the stem (height and diameter), and the resistance to lodging (bending of the plant near ground level), but also the mechanical properties of unit fibres (Bourmaud, Gibaud, & Baley, 2016). Above around 2000 plants per m², both mechanical properties and resistance to plant lodging drop.

Figure 4: Flax plants before flowering

2.4. Tracking the growth of the plant

In general, the life of a plant goes through five stages: germination, growth, flowering, seed formation and aging. Its development time is well defined (around 100 days for flax) despite variable weather conditions in terms of temperature (flax can grow provided the temperature is above 5°C), rain, sunshine and daylight time.

In France, textile flax is harvested when the fibres are mature. In order to define this time, the temperature is measured daily. Above 5°C, the rate of development of flax is directly proportional to the cumulative effective temperature (CT) seen by the plants from sowing (Bert, 2013): , defined as :

$$CT = \sum_{i=1}^{n} \left(\left(\frac{Tmax + Tmin}{2} \right) - 5 \right)$$

with *n* the number of days, Tmax the maximum daily temperature and Tmin the minimum temperature. Fibre maturity is obtained for a value of CT between 950 and 1100°C (Bert, 2013). The value of CT is known for each stage of the plant development: CT = 50°C for seedling emergence, CT= 550°C for flowering, CT= 650°C for capsules formation, CT= 950 – 1 100°C for fibres maturity. Above 950°C, the fibre richness no longer increases whatever the growing conditions or the variety (Bert, 2013).

Beyond a CT of 1100°C, flax plants are over-matured, i.e. they produce lignin components. The development of lignin depends on the local conditions and corresponds to plant aging. For flax, the presence of lignin in large amounts complicates retting and mechanical extraction of the fibres. It is therefore important to follow the maturity of the plant closely to avoid it.

2.5. Harvesting

The quality of the fibres depends not only on growing conditions but also on harvesting. The main steps are uprooting the plants, retting, turning and rolling. These are all organized in order to protect the fibres. The stems are not cut but pulled out of the ground using special machines, the bottoms of the stems break in the ground. The harvested stems are then carefully spread evenly on the ground. They then undergo dew retting (figure 5). During this step they are attacked by the combined effects of bacteria and fungi under the combined actions of sun and rain (Djemiel et al., 2017). These microorganisms attack the libero-ligneous bonds, both within and between bundles, and separate the wood and the fibres. In order to achieve homogeneous retting, the stems have to be turned over. The degree of retting is checked by periodically examining the samples. The retting process has been employed for centuries for textile applications and it is also essential for composite reinforcements. An increase in the mechanical properties of fibres during retting has been reported (Martin et al, 2013), but there is a retting limit not to be exceeded, otherwise the fibres will be degraded.

Figure 5: Flax plants during retting in the field (dew retting)

The stem layers are then carefully rolled together, keeping the stems parallel and with the tops of the plants together. In order to store them they must not have a humidity level greater than 16% during rolling. The flax bales are bound together by flax twine to avoid any pollution by synthetic materials. The bales are then stored in hangars with concrete floors, to limit humidity.

2.6. Mechanical extraction of fibres

Fibres are extracted from the plant stalks by scutching operations. These involve beating the stalks to remove the outer woody part, the inner shives, seeds and dust. Special machines are used, with parameters adjusted to each batch. These mechanical operations also produce short fibres. The long fibre tows correspond to the fibre bundles and may be of plant length. These are sorted into homogeneous batches of tows, by visual observation of the color of the fibres. The flax fibres are then conditioned in bales weighing around 100 kg. An additional combing operation may be performed to refine and further separate the fibre bundles. Fibre batches are evaluated according to organoleptic parameters (colour, strength, fineness and unit fibre length) (Bert, 2013). These criteria come from the textile industry.

The environmental conditions during scutching (in particular the humidity) affect not only the tow yield but also the average length of fibre bundles and the propensity for bundle splitting (Barbulée, 2015). The mechanical settings of the scutching machines (advancing speed of the straw, beating rate, grinding intensity) control the quality of the tows (structure, cleanliness and mechanical properties). A detailed study of the influence of these parameters reveals that the role of scutching could be limited to elimination of short fibres, while an improvement in fibre cleanliness is related to the final steps of the extraction line (Barbulée, 2015).

2.7. Consequences of the different steps on the morphology of the fibres

After mechanical extraction the flax fibres are in the form of single fibres, bundles and assemblies of bundles. The individual fibres are rarely completely detached, as this would make them difficult to recover in an industrial context.

In order to understand the mechanics of plant fibres and to take into account the scatter in morphology and mechanical properties the fibres can be studied at three different scales: the unit fibre, the fibre bundle, and a group of fibre bundles.
During mechanical loading, these different morphological variations will lead to a stress-strain response similar to the ones associated with structural defects in brittle materials. As a result, the physical and mechanical properties of plant fibre reinforcements will be strongly dependent on the

variety, growth conditions, cultural practices and the defects introduced during the various stages from retting and fibre extraction through the assembly and preparation of the reinforcements.
In the following paragraphs results from characterization studies at the three scales will be presented.

3. Parameters that influence the mechanical properties of single fibres.

3.1 Difficulties in characterizing a unit flax fibre under tensile loading

Single flax fibres have characteristics which affect their tensile behaviour and the way the test results are analysed. In particular, their cross section is not circular and the mean diameter varies between 10 and 25 µm, for a mean length between 15 and 36 mm, and a section which varies along the fibre length and tapers at the ends (Baley, Goudenhooft, et al., 2018). In the absence of a full volumetric 3-D characterization it is necessary to estimate the effective cross-sectional area.

The apparent diameter of a single fibre is defined as the width of the fibre as observed with a microscope. An area is calculated from the apparent mean diameter.
By representing the fibre as a collection of n elements of different cross-sectional areas and moduli, S_i and E_i respectively, connected in series, the mean apparent area, S_m, is the arithmetical average of the apparent cross-section areas along the fibre: $S_m = \sum_1^n \frac{S_i}{n}$. The effective cross-section area is that of a hypothetical fibre with a constant area, S_u, along its gauge length, which shows the same stiffness as the real fibre: $S_u = \frac{n}{\sum_1^n \frac{1}{S_i}}$ (Barbulée, 2015).
The average apparent (effective) Young's modulus is calculated from the mean apparent (effective) cross-section area and the fibre stiffness.

The variations of the apparent longitudinal Young's modulus as a function of the effective cross-section area for different fibre gauge lengths (Fig. 6) show some interesting features. For all gauge lengths examined (5, 10, 20 and 30 mm), the modulus decreases as the fibre cross-section area increases. The rate of decrease and the scatter decrease as the gauge length increases. This can be explained when assuming that each fibre is made up of elements connected in series, with different Young's modulus and different cross-section areas. In fibres with long gauge lengths, the variations in cross-section area are more likely, and there is an averaging effect on fibre modulus, which reduces the scatter. On the contrary, for short lengths there is less effect of averaging.

Figure 6: Interrelations between the apparent Young's moduli and the effective cross-section areas of unit fibres (fibre free length 20 mm). The average trend is represented by the blue dashed line (Barbulée, 2015).

In order to investigate the decreases in modulus and in scattering when the gauge length is increased, the experimental results shown in figure 6 were used to set up a model. Taking a gauge length of 5 mm as the basic element, gauge lengths of 10, 20 and 30 mm can be obtained by associating 2, 4 or 6 of these elements. For example, the modulus associated with a gauge length of 10 mm can be found by combining two elements of length 5 mm according to the following expression: $E_{series} = \frac{n}{S_m * \sum_1^n \frac{1}{S_i * E_i}}$

The modulus values generated are shown versus gauge length in Figure 7a. They are somewhat higher than the values calculated by using average cross-section areas. This indicates that taking account of cross-section area variations along the fibre is sufficient to determine an effective modulus.
The strengths and ultimate strains calculated using a Weibull approach are shown in figures 7b and 7c, respectively, as a function of gauge length. The low failure strains relate to particular test conditions (30 to 40% RH instead of 50%RH as usual).

Fig. 7: Variations of the mechanical parameters of unit flax fibres as a function of the gauge length:(A) Modulus, (B) Strength, (C) Ultimate strain (Barbulée, 2015)

3.2. Fibre location along the stem

The different stages in the growth of flax fibres were presented in Section 2. Given the strong bonding within fibre bundles, removal of unit fibres from a stem during growth is not a simple task. The zone within the plant from which samples are taken is also an important parameter; the central lumen reduces in size and within a bundle at a given height the fibres do not all develop at the same rate (Gorshkova et al., 2003).

At different heights and development times, the indentation modulus of the S2 layer of the fibre was characterized by atomic force microscopy (AFM) using peak-force quantitative nano-mechanical property mapping (PF-QNM) (Goudenhooft et al., 2018). Changes in the modulus with the cell wall thickening were highlighted. For growing plants, fibres from top and middle heights show a not very consistent inner layer Gn with a lower indentation modulus (13-14 GPa) than mature fibres (about 18 GPa), which exhibit thickened homogeneous cell walls made only of G layer. The evolution of mechanical properties over fibre development was confirmed by tensile tests on unit fibres (fibres extracted at mid-stem height from plants of age 60 days and 120 days after sowing). For growing fibres with a filling rate of 63±8 %, average tangent modulus of 32±12 GPa and strength of 680±337 MPa were estimated, whereas much higher performance was observed for mature fibres (namely a tangent modulus of 45±10 GPa and a strength of 965±302 MPa). These values correspond to the whole cell wall average longitudinal Young's modulus and strength, as the presence of the lumen has been taken into account. However, both batches exhibit similar ultimate strain, namely 2.1±0.5% for growing fibres and 2.2±0.5% for mature ones. It is important to keep in mind here that the indentation moduli of the S2 layer are lower than the longitudinal tangent modulus due to the anisotropic behaviour of the cell wall layers (Jäger et al, 2011). In addition, the relationship between the indentation modulus and the tensile modulus of both growing fibres and mature fibres is in accordance with published values (Tanguy et al., 2016).

This very interesting work will need to be repeated with a larger number of samples in order to generate reliable average values of the mechanical properties of fibres. The next section will compare the tensile mechanical properties of fibres from a large number of batches tested using the same test protocol and under the same conditions. The influence of the location in the stem will be discussed further in section 3.8.

3.3. <u>Fibres extracted from plants harvested at maturity and dew retted: comparison between fibres from different batches tested with the same protocol</u>

In order to allow comparisons to be made it is essential that the same test conditions be used. There are standard test methods for unit flax fibres (AFNOR NF T 25-501-2, 2015) and for fibre bundles sometimes known as technical fibres (AFNOR NF T 25-501-3, 2015). In this section, results have been obtained using these standards. While standards can sometimes be criticized, they do allow comparisons to be made on the same basis.

Comparisons of average mechanical properties gathered from 65 different batches are shown in figures 8 and 9 (storage and characterization conditions: 23°C and 50% HR). This represents 4298 fibre tests, on 17 varieties of flax (textile and oilseed) grown in Normandy (France) between 1993 and 2017. The average tangent modulus is 52.3±8.3 GPa, the average value for the strength is 945 ± 200 MPa and 2.4 ±0.4% for the failure strain. The mean diameter of these fibres was 16.64 ±2.65 µm.

Analysis of these results does not reveal a clear influence of the fibre variety nor of the year of culture. It may also be noted that no batch shows very low performance, which is reassuring. Thus, despite very different climatic conditions, the fibres' mechanical properties remain stable, which is a positive message for industrial developments.

Figure 8: Average strength vs tangent modulus for all the batches of flax under analysis. (Lefeuvre et al, 2013)(Lefeuvre et al, 2014a)(Bourmaud et al, 2013)(Coroller et al., 2013)(Charlet et al., 2009)(Charlet et al., 2007)(Baley and Bourmaud, 2014) (Alix et al., 2012)(Le Duigou et al, 2012)(Baley, 2002)(Bourmaud et al, 2010)(Roussière et al, 2012)(Martin et al, 2013)(Pillin et al., 2011)(Bourmaud et al, 2016)(Tanguy et al, 2018)(Goudenhooft et al, 2017)(Bourmaud, et al., 2016)(Gibaud et al, 2015) 2015)(Martin et al, 2014)(Bensadoun et al., 2017)

Figure 9: Average ultimate strain vs tangent modulus for all the batches of flax under analysis. (Lefeuvre et al., 2013) (Bourmaud et al., 2013)(Coroller et al., 2013)(Charlet et al., 2009)(Lefeuvre et al., 2014b)(Charlet et al., 2007)(Baley and Bourmaud, 2014) (Alix et al., 2012)(Le Duigou et al., 2012)(Baley, 2002)(Bourmaud et al., 2010)(Roussière et al., 2012)(Martin et al., 2013)(Pillin et al., 2011)(Bourmaud, et al., 2016)(Tanguy et al., 2018)(Goudenhooft et al., 2017)(Bourmaud et al., 2016)(Gibaud et al., 2015)(Lefeuvre et al., 2015)(Martin et al., 2014)(Bensadoun et al., 2017)(Lefeuvre et al., 2014a)

3.4. <u>Variations in fibre density</u>

In addition to the average mechanical properties of the unit fibres described above, it is important to know the properties at the composite ply level. In order to estimate the properties of composites it is essential to know the fibre content. This requires knowledge of the fibre density, ρ. For glass fibres this value is well known and shows low variability. Plant fibres show more scatter and recent papers have quantified this (Le Gallet al, 2018). It was shown that weighing in air and in fluid provides density values that allow predictions with a better correlation with measured composite properties than other density measurement methods.

Material comparisons are usually made on an equivalent weight basis. For a structure loaded in tension the specific stiffness is defined as the elastic modulus E divided by the density (E/ρ) and the specific strength is the strength divided by the density (σ/ρ). For a beam under flexural loading the specific properties are respectively ($E^{1/2}/\rho$) and ($\sigma^{2/3}/\rho$) and ($E^{1/3}/\rho$) or ($\sigma^{1/2}/\rho$) for a plate (Ashby and Jones, 2013). The ability to evaluate the composite density accurately is therefore important.

3.5. The same flax variety grown at the same place for several years

For one variety (Marylin), the reproducibility of the fibre mechanical properties was studied over four consecutive years (from 2009 to 2012), for one region (the plateau du Neubourg in France) and one fibre extraction centre (CTLN) (Lefeuvre et al., 2014a). As crop rotation is unavoidable it was necessary to select fields which were close to each other (the same field is only used once every seven years). Many parameters were identical over the four years: the same seed batch, same density, harvesting at the same degree of maturity (according to ITL recommendations, (Bert, 2013). The weather conditions were different (two normal years, one very wet year and one dry), which affected the yields (Lefeuvre et al., 2014a). Thirteen batches of fibres were selected from the four years; they were taken from the mid-height of the plants. The test results (Lefeuvre et al., 2014a) indicate that the average Young's modulus varies from 53.3 to 58.9 GPa, the strength from 970 to 1109 MPa and the failure strain from 2 to 2.2%. The variations are similar to those reported in the study mentioned above (Baley and Bourmaud, 2014) . In an industrial context, fibres are supplied from the production of the year (homogenization effect), not just from one plot of land.

3.6. Influence of the variety of flax

Since the varieties grown change with time it is logical to consider the influence of variety selection on the mechanical properties of fibres. Four different varieties were grown on the same plot the same year with the same plant density (Goudenhooft et al., 2017). The varieties were Liral Prince (marketed since 1946), Arianne (since 1978), Eden (since 2009) and Aramis (since 2011). The proportion of fibres in the stems was shown to increase with time because of the constant work of improving varieties, (from 7.8% to 13.4% of the section of the stem including the cells); this is one of the aims of improving fibre variety, but the mechanical properties of the unit fibres (taken at mid-height of the stem) were all quite similar.

3.7. Differences between long fibres and tows

During extraction both long fibres (assemblies of fibre bundles nearly as long as the plant height) and tows (small bundles pulled out during extraction, of random length) are obtained. It is interesting to compare these two types of fibre. For one batch of stems (variety Alizé), the tensile properties measured on unit fibres taken from long fibre bundles and tows were found to be identical (Martin et al., 2014) as was their ability to reinforce an epoxy matrix. This means that mixing long fibres and tows will not necessarily lead to a property reduction.

3.8. Influence of the location of fibre sampling

Growing conditions can change during the 100 days of plant growth. However, the quantity of fibres and their properties (mechanical and physical) will also change as a function of their position in the stem. The fibres are more numerous and have better properties at mid-height (Charlet et al., 2007) (Lefeuvre et al., 2015). On average the diameter of the fibre is larger near the ground than at the top of the plant (A. Bourmaud et al., 2016a). It is therefore interesting to separate fibres according to their location in the plant, in order to optimize the properties of composite reinforcements. A study for unidirectional plies (Lefeuvre et al., 2015) showed that it is also necessary to account for the weight distribution of fibres versus height, so in order to profit from the benefits a complex selection organisation would be required.

4. Mechanical properties of fibre bundles

4.1. Tensile characterization of fibre bundles (volume effect)

Work on the effect of fibre volume on mechanical behaviour of fibres started with glass fibres (Griffith and Taylor, 1921). Those authors indicated that the tensile strength of glass fibres decreased when the diameter was increased, and this was attributed to the larger number of defects in a larger volume of material. The statistical approach to brittle failure (Weibull, 1951) is the main basis of these studies. In the fibre bundle model (FBM), the bundles are a group of parallel fibres, each showing a Weibull failure stress distribution (Coleman, 1958) (D. G. Harlow and Phoenix, 1978)(D. Harlow and Phoenix, 1978)(D. Harlow and Phoenix, 1978). More recently, Chudoba et al. (Chudoba et al., 2006) and Vořechovský et Chudoba (Vořechovský and Chudoba, 2006) have studied the influence of structural imperfections in the fibre bundles (including those of the mechanical testing equipment) on the failure behaviour. Using a stochastic approach, based on previous work of (Phoenix, 1974) and (Phoenix and Taylor, 1973), they analysed the effects of structural disorder in the bundles (variations in diameter and length of unit fibres and their delayed response to loading) on mechanical performance. For plant fibres, many studies have investigated the mechanical properties of unit fibres in a similar way to studies of synthetic fibres (carbon, glass), which show brittle behaviour at room temperature. In particular, the influence of gauge length has often been examined using the Weibull statistical approach, considering fibres to have the same cross-section area (Xia et al., 2009) or variable cross-section area (Zhang et al., 2002)(Aslan et al., 2011). Curiously, the effect of fibre diameter on mechanical properties has not been studied from the point of view of Weibull's statistical approach. However, work on fibre bundle variability is rarer and often limited to the observation that mechanical properties decrease when the gauge length is increased.

In addition, this type of study does not reveal the links between the observed variations and the structural characteristics of the reinforcement.

The fibres described in the following sections were taken from the central part of the plant stems, where the best mechanical properties are found (Charlet et al., 2007).

4.2. Tensile behaviour of flax fibre bundles

For convenience, the influence of gauge length and bundle section on the tensile mechanical properties is usually studied on unidirectional specimens. Bundles of fibres are bonded to cardboard supports, in a similar way to the tests on unit fibres.
The variations in bundle cross-section area are related to the location in the plant or to local stress concentrations. Figure 10 shows how measured average cross-section areas of flax fibre bundles vary for three varieties (Charlet, 2008). An additional parameter which could be included in such a study is the position of the leaves, as the fibre bundle is modified locally by leaf attachment.

Fig. 10 Variations of bundles cross-section areas along the stem (Charlet, 2008)

The strength and ultimate strain of fibres and fibre bundles for different gauge lengths are plotted in Figure 11. The slower reductions for the bundles can be explained by the fact that the failure of a fibre may transfer the load to neighbouring fibres, which avoids a catastrophic failure of the entire bundle. The bundles, with more regular cross-sections, are less sensitive to stress concentration and thus can reach higher failure stresses for long gauge lengths.

Fig 11: Variations of the strength (A) and the ultimate strain (B) of flax bundles and fibres as a function of the gauge length (Barbulée, 2015)

4.3. Tensile characterization of an assembly of fibre bundles

A clamping device especially designed for gripping an assembly of fibre bundles (made up of bundles from the same stem) was developed for mechanical tensile tests on plant fibres; it is described in detail in (Barbulée et al, 2014). For testing plant fibres it is essential that all fibre bundles are equally tensioned, and that the bundles are perfectly aligned along the tension axis, in order to avoid shear in the gripping zone. The end grips here were made of tubular metal shells, and the gauge length was defined as the distance between these. The bundles are solidly held after the curing of an epoxy adhesive cast in the shells. The moulding of the bundle ends allows them to be held without gripping them directly in the machine grips. The specimen is placed in the testing machine without touching the fibres. After testing it is possible to section the metallic shells (containing the fibre bundles in the epoxy) in order to measure the actual cross-sectional area using image analysis software. The repeatability of sample preparation was checked by statistical analysis of fibre bundles: an Anova test showed no differences between several specimens taken from the same industrial bundle assembly. Mechanical tests were performed on an Instron 5800R machine equipped with a 2N capacity load cell and strained at a rate of 1 %/ minute.
The dependency of the mechanical properties on the cross-section area is shown in figure 12. The secant modulus is the value which is the most sensitive to size effects and shows a large scatter and decreasing trend when the cross-section area of the bundle assemblies is increased (Fig. 12a), even though compliance corrections were made. The values at failure are almost independent of the cross-section area (Fig. 12b and Fig. 12c), and the values are similar to single fibre values.

Fig 12: Variations of the mechanical parameters of fibre bundles assemblies as a function of their cross-section area: (A) Modulus, (B) Strength, (C) Ultimate strain (Barbulée, 2015)

5. Longitudinal tensile tests on unidirectional composites to assess variability in fibre mechanical properties.

In this section we present results from tests on biocomposites reinforced with unidirectional flax fibres. The aim is to check whether such tests are sensitive to variations in fibre mechanical properties. It should be noted that for the materials studied here the fibre length is greater than the critical length necessary to ensure load transfer (Gibson, 2012) (C Baley et al., 2018a).

5.1. Correlation between mechanical properties of fibres and those of unidirectional composites for man-made fibres

Correlating the tensile response of unit fibres with that of a unidirectional composite ply reinforced by continuous fibres loaded in the fibre direction is not simple. Nevertheless, it is often assumed that, at small strains, the ply modulus is simply related to the elastic properties of the fibres and the matrix and the fibre volume fraction by the law of mixtures. This approach is not directly applicable to estimate strength, as various other parameters must be considered such as:

- The volume of material under load (Soutis, 2000; Lavoie et al, 2000) and thus the thermal and hygrothermal history of the material following manufacture
- The variability of the fibre strength, which depends on the presence of defects (Rosen, 1964), requires a probabilistic approach. Ply failure occurs after accumulation of damage.
- The initial fibre lengths (Rosen, 1964). As fibre length increases the probability of large defects increases and therefore the strength decreases.
- The meso-structure of the ply (Piggott, 1996), i.e. the fibre distribution, the presence of voids and inclusions, local or global misalignment of fibres (waviness), local fibre failures...
- Residual stresses may be present in the composite upon manufacture. They may originate from differences in thermal expansion coefficients between the fibres and the matrix, stacking sequence, cooling conditions or cure shrinkage
- The structure of the matrix (degree of cross-linking, crystallization…) and its mechanical behaviour
- The fibre volume fraction (local and global in the ply)
- Fibre/matrix adhesion. This characteristic is usually studied using micro-mechanics tests (pull-out, microbond test, single fibre fragmentation) and measuring an apparent interfacial shear strength. This value strongly depends on the test method. Some values have been published for flax fibres, with both thermoplastic (Graupner et al., 2014) and thermoset matrix polymers (Le Duigou et al., 2016). The concept of a fibre/matrix interface with flax reinforcements is more complex than for conventional composites due to both the presence of fibre bundles, with internal interfaces, and the multilayer nature of elementary fibres (Baley et al., 2014).
- Loading conditions (loading rate, temperature, humidity…) and the test protocol (presence of end tabs, for example)…

5.2. Some specific considerations about flax fibres, which can affect their mechanical behaviour

Plant fibres show some specific characteristics which must be taken into account when analysing tensile behaviour of biocomposites. For example:

• The consequences of the multi-scale organization of flax fibres. When glass fibre composites fail there are three main types of damage: fibre failure, matrix cracking, and fibre/matrix interfacial failure. Failure of plant fibre composites involves additional mechanisms, as the fibres themselves have a composite structure (in flax fibres stacking of different layers reinforced by cellulose fibrils). Damage can therefore also appear both within the fibres and at shared layers between fibre bundles (Baley et al, 2012a) (Baley, 2002). There is thus a hierarchy of interfaces within the biocomposite which must be considered (Baley et al., 2014).

• A non-linear behaviour in tension, which can be explained by the composition and organization of cell walls (Baley et al., 2018)

• The unit fibres are not continuous and are assembled in bundles within the plant. After extraction, these bundles will be more or less separated. This organization is visible on a cross-section of the composite. In order to benefit from the potential of reinforcement of plant fibres, it is usual to work with unidirectional layers whose fibres are held together by either a bonding agent or a thread (Baley et al, 2018). It is also possible to overlay and stitch together the layers to produce multidirectional stacks. Unidirectional bundles without torsion can also be produced in the form of rovings by bonding them together (Khalfallah et al., 2014) (Baley et al., 2018). In this case, care is necessary to ensure that the matrix and the bonding agent are compatible. In a similar way, glass fibre rovings are coated with a sizing, whose role is to ensure cohesion between the fibres. It is also possible to twist the fibres together, as in the textile industry. Plant fibres are discontinuous and in order to achieve strength (or weaving and use as a textile) twisting is required. In this case load transfer occurs by friction and the strength increases, at least initially, with the number of turns per length) (Goutianos et al, 2006)(Baets et al, 2014). In a composite, load transfer onto neighbouring fibres occurs by interfacial bonds between fibre and matrix. Reinforcing a ply with twisted plant fibres results in a loss in mechanical properties (Shah et al, 2013)(Baets et al., 2014) due to the off-axis orientation. Twisted fibres also result in a less homogeneous distribution within the composite.

• Flax fibres under usual storage conditions will contain water. The amount absorbed at equilibrium at 65% RH and 21°C is around 7% by weight (Faruk et al, 2012)(Rowell, 2008). Placing the fibres in an oven before impregnation with a thermoset matrix will influence the mechanical properties of the composite (Baley et al., 2012). Furthermore, composites (flax/polyester) made of fibres which have not been dried show considerably lower moisture sorption and desorption behaviour, and degree of swelling and shrinkage, than composites reinforced with dried fibres (Lu and Vuure, 2019).

5.3. Analysis of the tensile behaviour of unidirectional composites reinforced by flax fibres

5.3.1. Analysis of published data

As noted above, any comparison among published data on mechanical properties is delicate as the fibre properties can vary significantly (Lefeuvre et al., 2015) and are not always measured.

Figures 13 and 14 report published values of the tensile properties of unidirectional flax/thermoset matrix (mainly epoxy or polyester) as a function of the fibre content. Although not always stated, it is assumed that the materials were made according to best practice, the reported tensile modulus values (E1) were measured in the low strain range of the stress-strain curves, and the void content is low.

Figure 13. Longitudinal tensile modulus (E1) of unidirectional composites versus fibre volume fraction (Coroller et al., 2013) (Charlet et al., 2007) (Poilâne et al., 2014) (Liang et al, 2014) (Martin et al., 2014) (Van de Weyenberg et al., 2003) (Oksman, 2001))(Cherif et al, 2011))(Hughes et al, 2007) (Marrot et al, 2014)(Khalfallah et al., 2014)(Lefeuvre et al., 2015)(Lebrun et al, 2013)(Habibi et al, 2017)(Goutianos et al., 2006)

In spite of the uncertainties about some parameters (fibres origins, possible treatments, fibre content determination method non-specified, orientation control, manufacturing cycles, void content …) there is a reasonable correlation between the Young's modulus and the fibre volume fraction (figure 13). Using the law of mixtures, it is possible to determine an average fibre modulus to be 50.48 GPa. This value is close to the one measured on single fibres from different batches (Baley and Bourmaud, 2014) i.e. 52.5 GPa (measurements made according to French standard (AFNOR NF T 25-501-2, 2015) in the high strain domain of the stress-strain curve.

Figure 14. Strength versus fibre volume fraction for unidirectional flax/thermoset composites (Coroller et al., 2013) (Poilâne et al., 2014) (Liang et al., 2014) (Martin et al., 2014) (Van de Weyenberg et al., 2003) (Oksman, 2001) (Baets et al., 2014) (Cherif et al., 2011) (Hughes et al., 2007) (Marrot et al., 2014)(Khalfallah et al., 2014)(Lefeuvre et al., 2015)(Lebrun et al., 2013)(Habibi et al., 2017)(Goutianos et al., 2006)

An analysis of the correlation between the strength and fibre content shows more scatter than for the modulus, (figure 14). A simple law of mixtures estimation of fibre strength indicates a value of 486 MPa, significantly lower than the unit flax fibre average strength value (945 MPa) (Baley and Bourmaud, 2014). There are several explanations for this discrepancy: all fibres do not span the gauge length, variability in fibre properties, influence of the manufacturing process, quality of fibre/matrix interface and homogeneity of the reinforcement (mixture of unit fibres and fibre bundles). Various authors have studied the mechanical properties of plant fibres and their unidirectional composites, for example: (Andersons et al, 2015)(Coroller et al., 2013)(Shah et al, 2016)(Charlet et al., 2007)(Bensadoun et al., 2017).

5.3.2. Intercomparison of results issued from different laboratories using the same batch of fibres (Round Robin program)

In order to define a fibre bundle test similar to the one used on impregnated continuous carbon fibre tows (ASTM D 4018-99 and Afnor NF EN ISO 10618), round robin tests on impregnated fibre bundle (IFBT) were performed in 5 European laboratories on the same batch of flax fibres (Aramis variety, combed flax) (Bensadoun et al., 2017) and epoxy or vinylester matrix. Each laboratory manufactured its own specimens. Unidirectional fibres taken from a combed roving were placed in a mould, impregnated with the polymer and transverse compression was applied. After curing the specimens were tested in tension. In addition to composite properties the unit fibre properties were also investigated, in order to evaluate the load transfer ability in the composites (Bensadoun et al., 2017).

Figure 15 and figure 16 show the variations of the stiffness and the strength versus fibre content. In order to facilitate the analysis, literature data discussed previously are reported on the same plot (Bensadoun et al., 2017).

It should be noted that the literature data are published as average values, while each IFBT value (Bensadoun et al., 2017) corresponds to an individual sample.

Figure 15. Initial longitudinal tensile modulus (E1) of unidirectional composites versus fibre volume fraction. Filled circles: published data (Coroller et al., 2013) (Charlet et al., 2007) (Poilâne et al., 2014) (Liang et al., 2014) (Martin et al., 2014) (Van de Weyenberg et al., 2003) (Oksman, 2001) (Baets et al., 2014))(Cherif et al., 2011))(Hughes et al., 2007) (Marrot et al., 2014)(Khalfallah et al.,

2014)(Lefeuvre et al., 2015)(Lebrun et al., 2013)(Habibi et al., 2017)(Goutianos et al., 2006). Open circles: Values from a single batch of flax fibres: (Bensadoun et al., 2017)

The plots of modulus E1 as a function of fibre content (Fig. 15) show higher values on average for IFBT. This can be explained by the uniqueness of the fibre source in this case whereas the results associated with fibres of different varieties and different origins are amalgamated in the data issued from the literature. However, the scatter is similar for both sets of values.

Figure 16. Strength versus fibre volume fraction for unidirectional flax/thermoset composites

Filled circles: published data (Coroller et al, 2013) (Charlet et al., 2007) (Poilâne et al., 2014) (Liang et al., 2014) (Martin et al., 2014) (Van de Weyenberg et al., 2003) (Oksman, 2001) (Baley et al., 2012) (Baets et al., 2014))(Cherif et al., 2011))(Hughes et al., 2007) (Marrot et al., 2014)(Khalfallah et al., 2014)(Lefeuvre et al., 2015)(Lebrun et al., 2013)(Habibi et al., 2017)(Goutianos et al., 2006). Open circles: Values from a single batch of flax fibres: (Bensadoun et al., 2017)

In contrast to the apparent modulus E1, the two sets of tensile strength values seem to form a homogeneous population. It is concluded that the manufacture further impacts the structure-sensitive property (longitudinal Young's modulus) via fibre misalignments, for example. Remember that each of the laboratories participating in the round robin had manufactured its own materials. These results thus demonstrate the importance of the stage of specimen preparation of these biocomposites, even when using the same reinforcement.

Figure 17 shows the comparison between the initial modulus E1 measured in the strain range 0 – 0.1%) and the slope determined in the linear region with strain between 0.3% and 0.5% (tangent modulus, E2). (Bensadoun et al., 2017). The change of slope on the stress-strain curve around 0.2% deformation complicates the comparison between the behaviours of the fibre and that of the composite as long as the mechanisms at the origin of this change are not clearly identified.

Figure 17 : Initial (E1) and tangent (E2) moduli measured for longitudinal tensile loading of samples made from the same flax fibre batch (Bensadoun et al., 2017)

5.3.3. Flax/PA11 produced under closely controlled manufacturing conditions

Figure 18 shows a typical tensile stress-strain plot for a unidirectional composite manufactured by a film stacking process (alternating polymer films and layers of untwisted unidirectional fibres held together by pectins, 70.3% fibre volume content), in a heated press at 210°C (Khalfallah et al., 2014). All batches were produced by the same operator following the same protocol. By calibrating the polymer films and the applied pressure, the fibre volume fraction could be controlled and the presence of porosity is almost avoided. As noted above the curve is non-linear (Andersons et al., 2015). Similar behaviour has been noted for composites with thermoset (Baets et al., 2014) (Coroller et al., 2013) and thermoplastic matrix polymers, such as PP (Baley et al., 2016) and PA11 (Bourmaud, 2016). The behaviour is generally considered in two strain ranges and thus low strain and high strain tangent modulus values can be determined (Bensadoun et al, 2015) (Shah, 2016) (Bensadoun et al., 2017).

Figure 18 also shows the influence of fibre volume fraction on the axial tensile behaviour of flax/polyamide 11 composites (A. Bourmaud et al., 2016b). For all fibre contents, the loading curve shows the same trend, but the non-linearity is more important when the fibre ratio is increased.

Figure 18 : Influence of fibre volume fraction on longitudinal tensile behaviour of a unidirectional flax/PA11 composite (A. Bourmaud et al., 2016b)

The analysis of these curves shows a linear growth of the modulus E1 (in the strain range <0.1%) and of the strength as a function of the fibre content (Fig. 18). These results show that it is possible to obtain increasing stiffness (E1 at low strain) and strength with low scatter as fibre content is increased for these unidirectional flax/PA11 composites (Bourmaud et al., 2016). From Fig. 19a the Young's modulus of the fibres can be evaluated at 60 GPa (see also figure 7) using the law of mixtures and neglecting the contribution of the matrix to the stiffness of the composite. This estimated value is among the highest of the experimental values reported in Figure 12a. Nevertheless, one can notice that with the use of PA11 matrix, a significant thermal heating (210°C) is mandatory, the latter has a significant impact on fibre stiffness with a possible cell wall stiffening with temperature increase and associated water content decrease. Plant fibres are mainly composed of cellulose but also include non-cellulosic polymers, which for flax are mainly pectins and hemicelluloses. Research work has shown that the first polymers to be affected by a thermal cycle are these non-cellulosic polymers and in particular pectin; cellulose does not start to degrade until about 200°C (Gourier et al., 2014). At the fibre scale, Gassan and Bledzki (Gassan and Bledzki, 2001)have shown that the mechanical properties of jute and flax fibres start to be affected by the temperature at around 170°C but time at temperature must also be considered. For the results presented above, the cycle time was 8 minutes, which is relatively low compared to compression cycles with thermosetting resins (which can also be very exothermic). During the work mentioned (Bourmaud et al., 2016b), the authors have checked the performance of the elementary flax fibres after an 8-min heating cycle at 210°C. It appears that their mechanical performance decreases by about 10% for the modulus and 30% for the strength; this decrease is mainly attributed to the degradation of the non-cellulosic matrix and not to the cellulose itself. This assessment is confirmed by a range of literature results: for example, Paris et al. (Paris et al., 2005) have studied spruce and pine heating between 35 °C and 250 °C, these authors highlight the evaporation of water and dehydration with slight depolymerization, but no change of cellulose microfibrils. In addition, Zolfrank and Fromm (Zollfrank and Fromm, 2009) have studied wood pyrolysis between 200 °C and 300 °C. In the range of 200°C–250 °C, they demonstrate at first a degradation of polyoses but only a disorientation of cellulose micro- fibrils away from the fibre axis; a significant evolution in cellulose structure appears only near 250 °C.

Fig. 19. Initial Young's modulus (A) and strength (B) of flax–PA11 UD composite versus fibre volume fraction (Bourmaud et al., 2016)

5.4. Influence of manufacturing of flax composites

Manufacturing procedures can have a significant influence on the structure of flax fibre reinforced composites, and hence on their mechanical properties and their variability. This can be illustrated by examining the scatter in properties of composites made from a single batch of flax fibres (Bensadoun et al., 2017). Examination of Figures 14 to 17 indicates considerable variations for all tensile properties.

There is an expanding published database on plant fibre-reinforced polymer composites. As noted above, the properties depend on the structure of the fibres, and several studies have focussed on this aspect. However, in many cases the final properties of the biocomposites produced do not fulfil their expectations. Indeed, many published studies report sub-optimal properties, and this may add to a perceived image of large variability. The manufacturing processes employed are generally those developed for synthetic fibre composites (glass and carbon). For these synthetic fibres the high energies deployed during consolidation are not detrimental, but they may be for plant fibres. It is therefore essential that the processing windows (temperature, hold time, environment) suitable for plant fibres be clearly identified.

The understanding and hence the improvement and optimization of the manufacturing processes and the resulting composite properties require a thorough description of the mechanisms controlling the properties of the material. There has been considerable progress in the development of physical and numerical models but the coupling of these models is also necessary. A recent paper by Khanlou et al. (Khanlou et al., 2017) describes models to optimize the process and shows results for flax/PLA composites

Manufacture of composites involves coupled thermal, chemical, hygrometric and mechanical loadings. Current work aims to evaluate these loadings and their impact on the quality of the final material, including parameters such as polymerization, crystallization, porosity, fibre integrity, interfaces, etc. Understanding the behaviour of plant fibre composites is part of a more general research activity on the response of complex fibre reinforcement architectures within a porous medium. The characterization of their internal structure, from the nano-scale to the industrial component is a complex challenge. Predicting changes in this structure with time and environment is also essential if these materials are to realize their potential.

A recent study examined the influence of the variability of the elastic properties of woven flax on their epoxy composites (Scida et al, 2017). An analytical model enabled the parameters describing the fibre reinforcement architecture to be taken into account, and this is another aspect of the manufacturing process which requires more attention.

6. Summary of factors affecting variability

Table 1 summarizes the factors which influence the performance of flax fibres intended for reinforcement of composites. Some of these can be controlled; others such as weather conditions are imposed. While the influence of a parameter on plant growth can be evaluated it is more difficult to establish relationships for fibre development.

Table 1: Factors affecting the performances of flax fibres designed for composites (in red, parameters that cannot be controlled)

7. Conclusion

If flax fibres are to be used in structural applications, it is essential that their mechanical properties, and those of their composites, be fully understood.

The use of fibres from a renewable (agricultural) resource is an advantage, but the manufacture of these fibres in industry requires a broader know-how in order to understand the stages from flax seeds to composite reinforcement. It is often the agricultural aspects which are the most difficult to assimilate for a materials engineer, familiar with synthetic fibres. Common questions concern the influence of growth conditions, and the reproducibility of fibre properties from one year to the next. This paper describes the different steps in the flax growth, harvest and fibre extraction cycles and their influence on properties.

The properties of flax fibres justify their use as reinforcement for structural composite materials. These good properties were not achieved by chance; they are the result of the strengthening role of the fibres in the plant, their genetic history and development. For many years, farmers have worked closely with scutching companies in order to improve fibre quality. This has resulted in growing and harvesting practices which are carefully defined and monitored. The existence of a complete European supply chain is very beneficial today (active selection of varieties, well-developed agricultural know-how, dedicated scutching equipment…) in guaranteeing traceability. Many scientific studies have developed specific characterization tools and understanding of the influence of biological and physical parameters on the mechanical properties of fibres and their composites.

Variability in properties is often cited as a potential disadvantage of plant fibre reinforced composites. In the plant, fibres are assembled in bundles, which are then more or less divided by mechanical and chemical treatments for composite implementation. The behaviour of bundles differs from that of unit fibres as the former are bonded assemblies of discontinuous fibres. There is thus an influence of the volume of material under load. Characterization of unit fibres is time-consuming and it is also possible to use an inverse method to obtain fibre properties from unidirectional ply testing. However, for plant fibre composites there are additional constraints:

- The hierarchical structure of flax fibres involves stacks of layers reinforced by micro-fibrils, so damage can initiate in biocomposites and propagate within the fibres themselves, unlike in glass or carbon fibres.
- Plant fibres loaded in tension show pronounced non-linear behaviour.
- Within a structure there are both single fibres, which are quite short (25 mm on average), and fibre bundles.
- The properties of flax fibres depend on processing conditions; again, this is not the case for man-made fibres.

However, as shown in this paper, composite stiffness, which is the property of interest for most applications, can be predicted accurately from fibre properties. While more work is still required, particularly with respect to long term behaviour, this perceived scatter in properties should not prevent the development of new structural applications using flax fibre composites.

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Table 1: Factors affecting the performance of flax fibers designed for composites (in red, parameters that cannot be controlled)

Stage	
Farming practices	Variety selection / Early variety or not / winter or spring flax / triple resistance variety
	Species, variety, genetic, seed quality
	Work in the field Date, seeding method, seed density Plant organization Fertilization Control of weeds, diseases, pests
Plant growth Growing conditions	Growing practices (number of plants per m²), distance between seed lines
	Physicochemical and biological characteristics of soil Soil structure **Rain, Temperature, wind** daily sunshine duration, shadow **light intensity**
After pulling out in the field	Date of uprooting (plant maturity) Wilting **Retting type and conditions** Moisture level in rolls Storage conditions
Mechanical extraction	Scutching, combing
	Hydrothermal conditions
Manufacture of semi-products for textiles or composites	Hydrothermal conditions Spinning and weaving Bundle production Preform manufacture for composites Carding, needle spinning …

Figure 1: Location of fibers in the stem (A, B and C) and representation of bundles (D) and unit fibers (E)

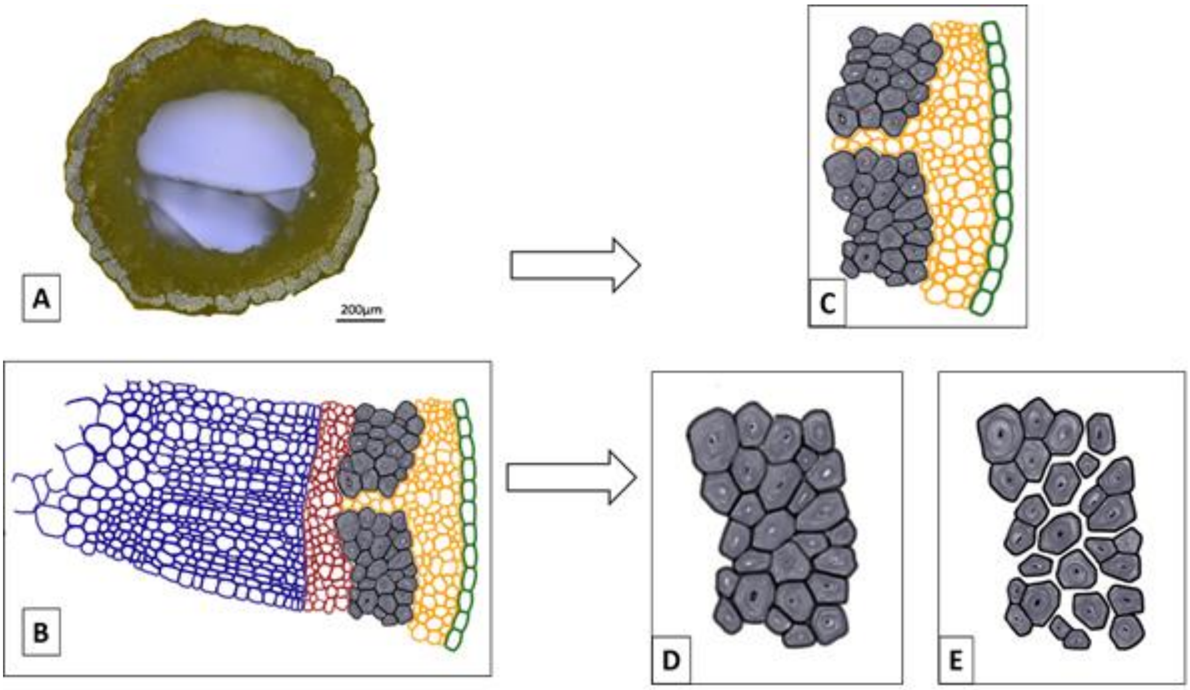


Figure 2: Scanning electron microscope image of raw flax fibers

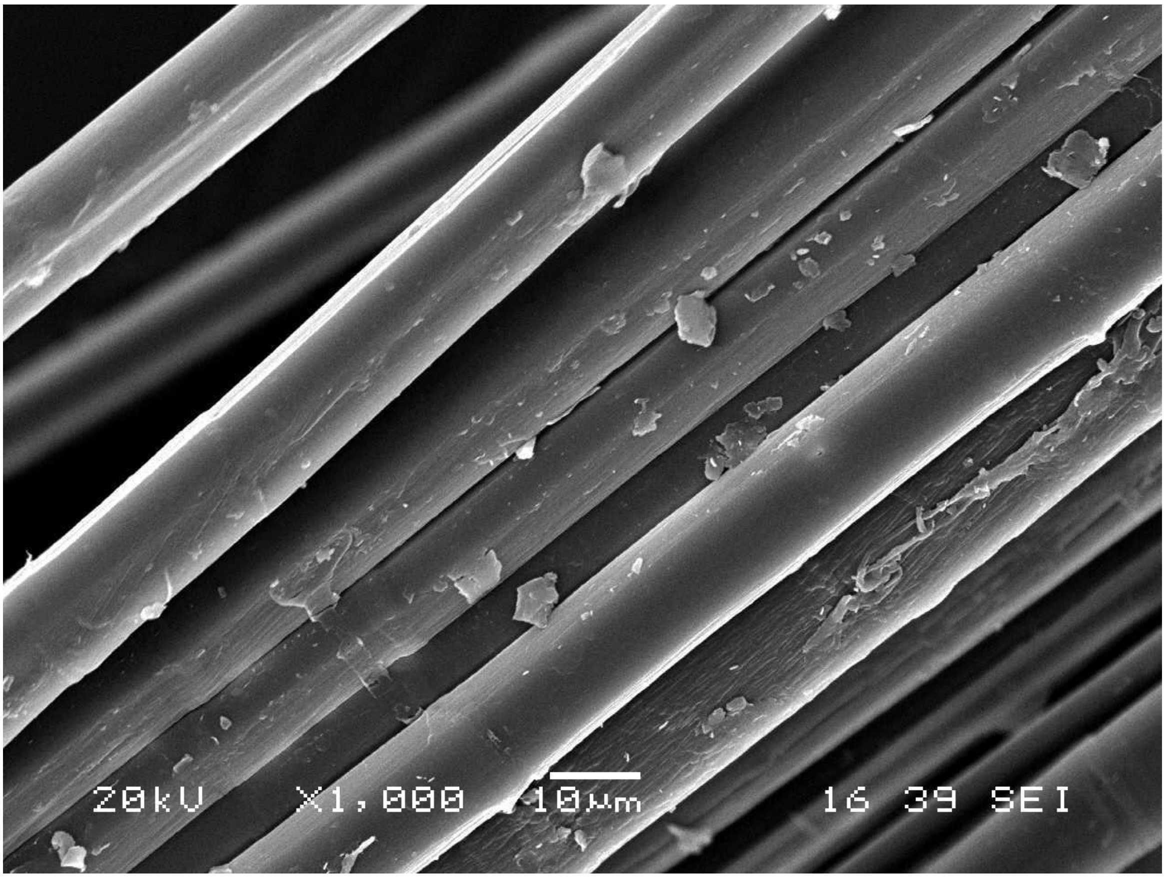


Figure 3. 3D reconstruction of a bundle in an area with a fibre extremity

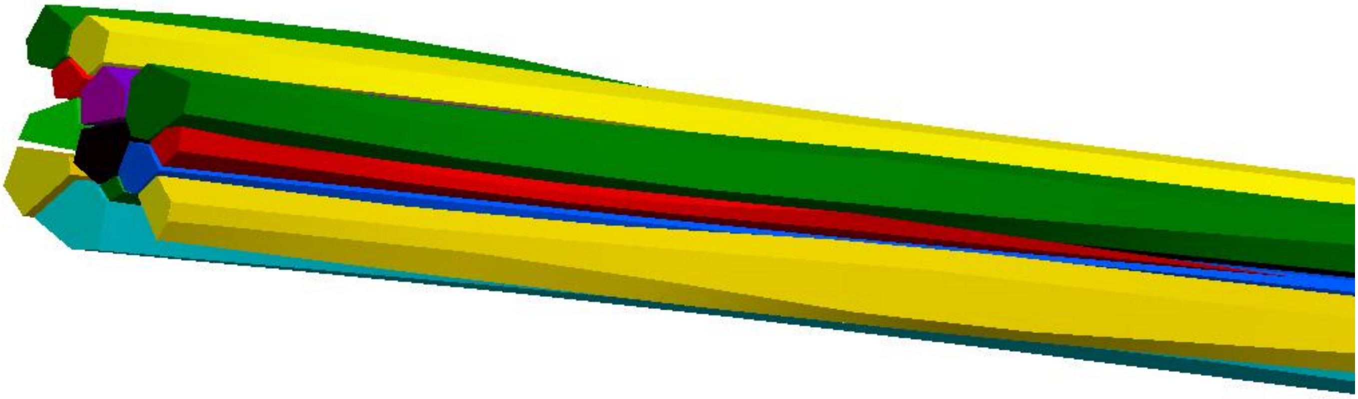


Figure 4: Flax plants before flowering

Figure 5: Flax plants during retting in the field (dew retting)

Figure 6: Interrelations between the apparent Young's moduli and the effective cross-section areas of unit fibers (fiber free length 20 mm). The average trend is represented by the dashed line (Barbulée, 2015).

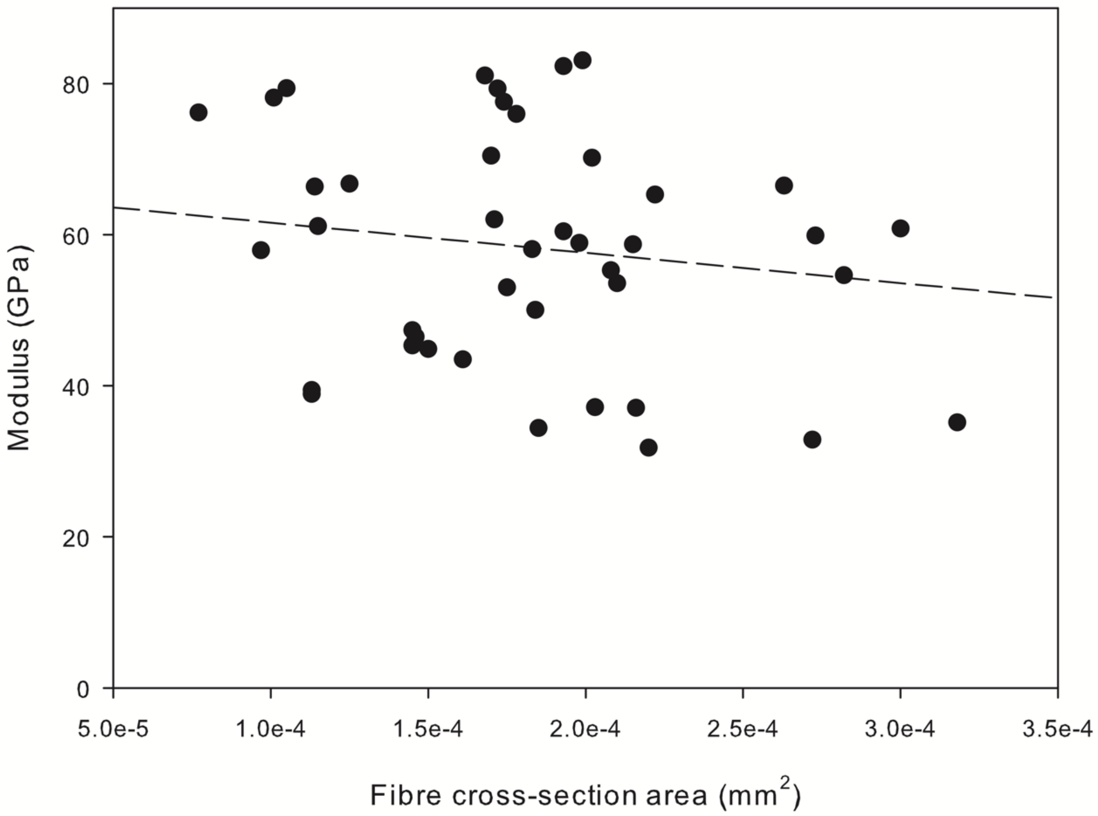


Fig. 7: Variations of the modulus (A), strength (B) and ultimate strain (C) of unit flax fibers as a function of the gauge length [Barbulée 2015]

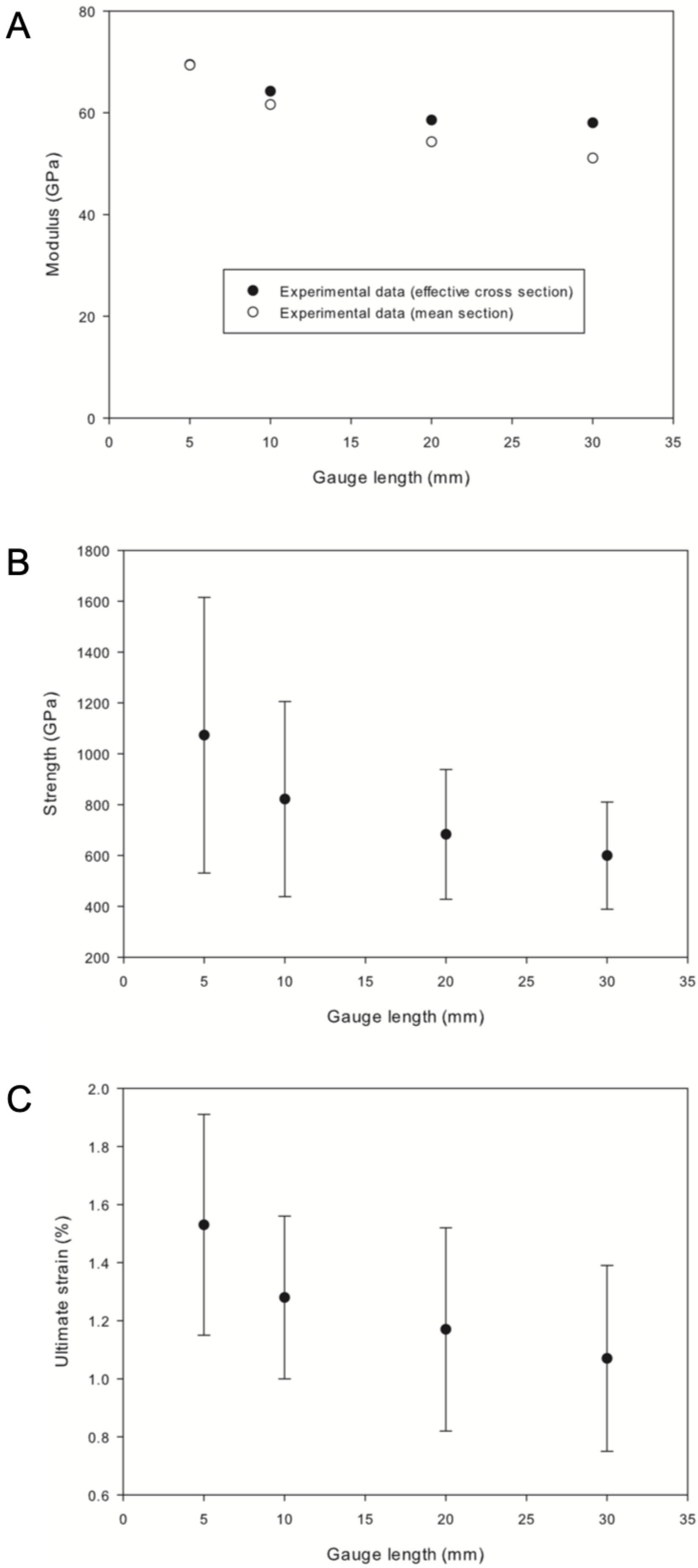


Figure 8: Average strength vs tangent modulus for all the batches of flax under analysis. (Lefeuvre et al, 2013)(Lefeuvre et al, 2014a)(Bourmaud et al, 2013)(Coroller et al., 2013)(Charlet et al., 2009) (Charlet et al., 2007)(Baley & Bourmaud, 2014)(Alix et al., 2012)(Le Duigou et al, 2012)(C. Baley, 2002)(Bourmaud et al, 2010)(Roussière et al, 2012)(Martin et al, 2013)(Pillin et al., 2011)(Bourmaud et al, 2016)(Tanguy et al, 2018)(Goudenhooft et al, 2017)(Bourmaud, et al., 2016)(Gibaud et al, 2015) 2015)(Martin et al, 2014)(Bensadoun et al., 2017)

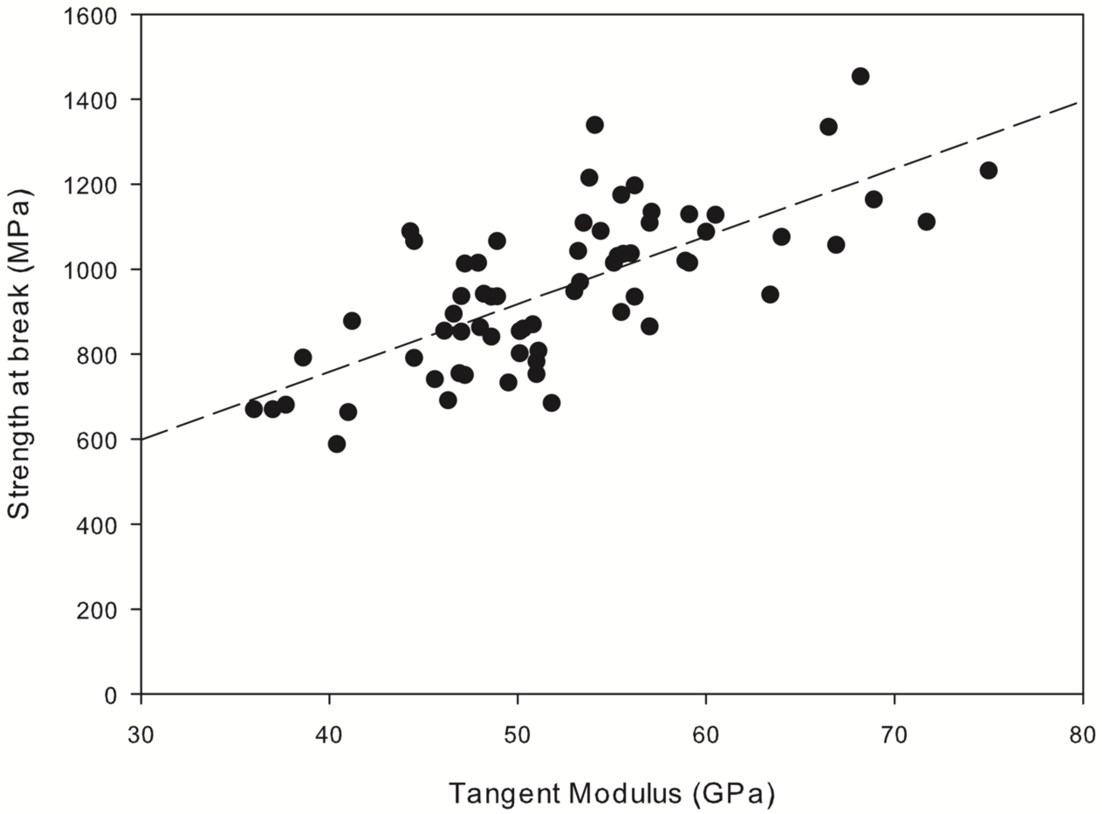


Figure 9: Average ultimate strain vs tangent modulus for all the batches of flax under analysis. (Lefeuvre et al., 2013) (Bourmaud et al., 2013)(Coroller et al., 2013)(Charlet et al., 2009)(Lefeuvre et al., 2014a)(Charlet et al., 2007)(Baley & Bourmaud, 2014)(Alix et al., 2012)(Le Duigou et al., 2012)(Baley, 2002)(Bourmaud et al., 2010)(Roussière et al., 2012)(Martin et al., 2013)(Pillin et al., 2011)(Bourmaud, et al., 2016)(Tanguy et al., 2018)(Goudenhooft et al., 2017)(Bourmaud et al., 2016)(Gibaud et al., 2015)(Lefeuvre et al., 2015)(Martin et al., 2014)(Bensadoun et al., 2017)(Lefeuvre et al., 2014b)

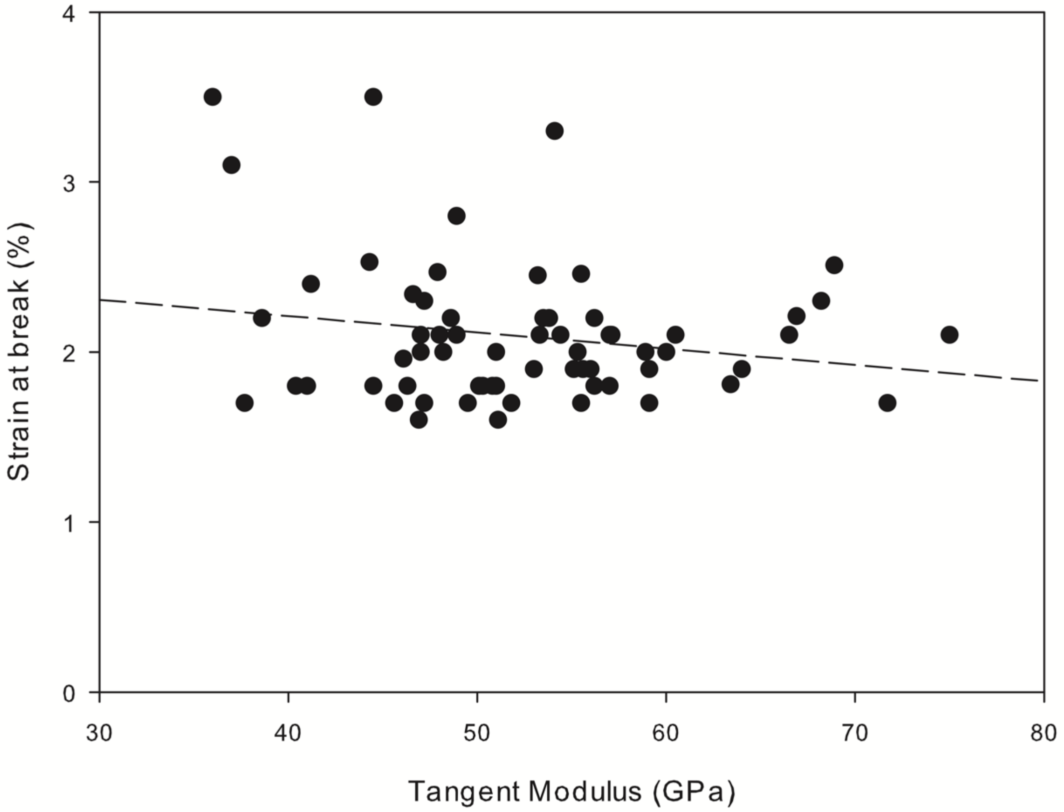


Figure 10 Variations of bundles cross-section areas along the stem (Charlet 2008)

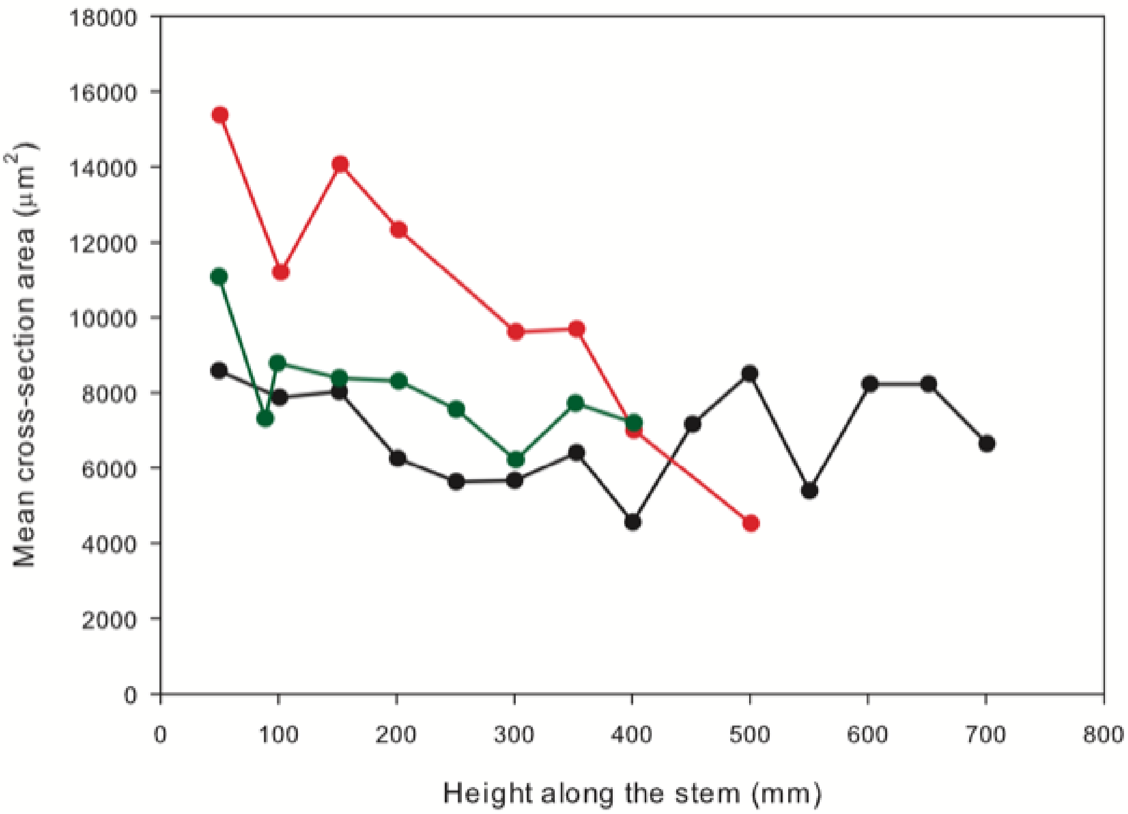


Figure 11: Variations of the strength (A) and ultimate strain (B) of flax bundles and fibers as a function of the gauge length

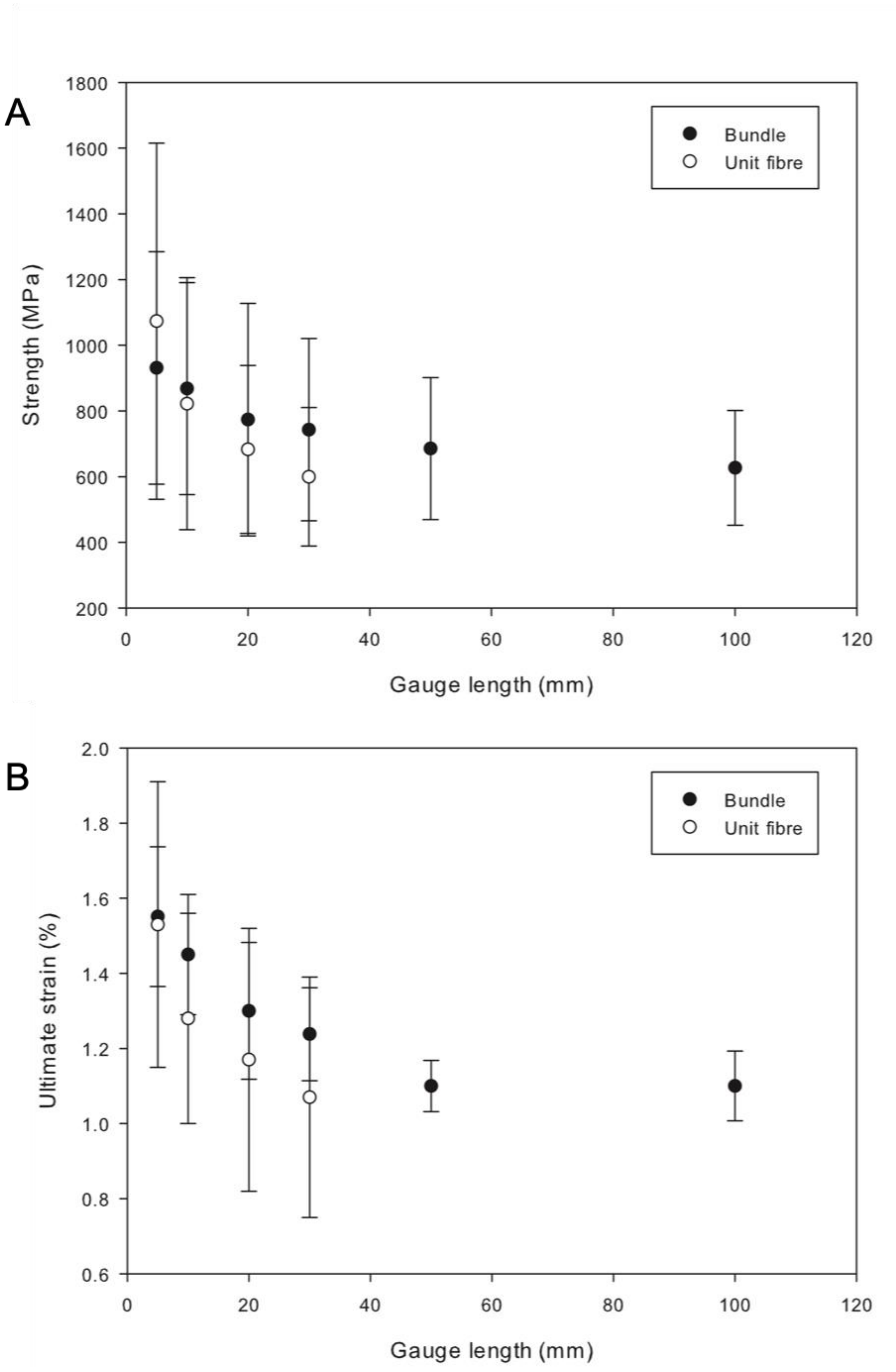


Figure 12: Variations of the modulus (A), strength (B) and ultimate strain (C) of fiber bundles assemblies as a function of their cross-section area (Barbulée, 2015).

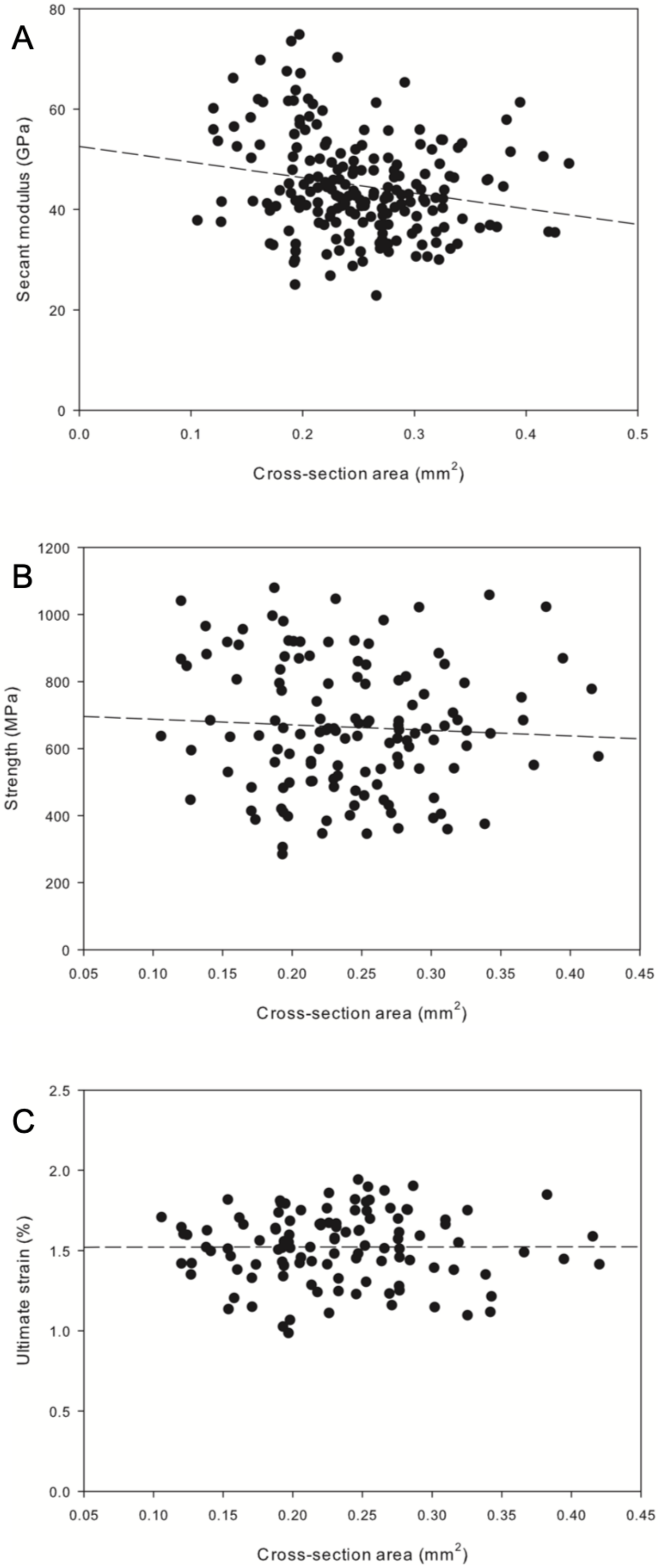


Figure 13. Longitudinal tensile modulus (E1) of unidirectional composites versus fiber volume fraction (Coroller et al., 2013) (Charlet et al., 2007) (Poilâne et al., 2014) (Liang et al, 2014) (Martin et al., 2014) (Van de Weyenberg et al., 2003) (Oksman, 2001))(Cherif et al, 2011))(Hughes et al, 2007) (Marrot et al, 2014)(Khalfallah et al., 2014)(Lefeuvre et al., 2015)(Lebrun et al, 2013)(Habibi et al, 2017)(Goutianos et al., 2006)

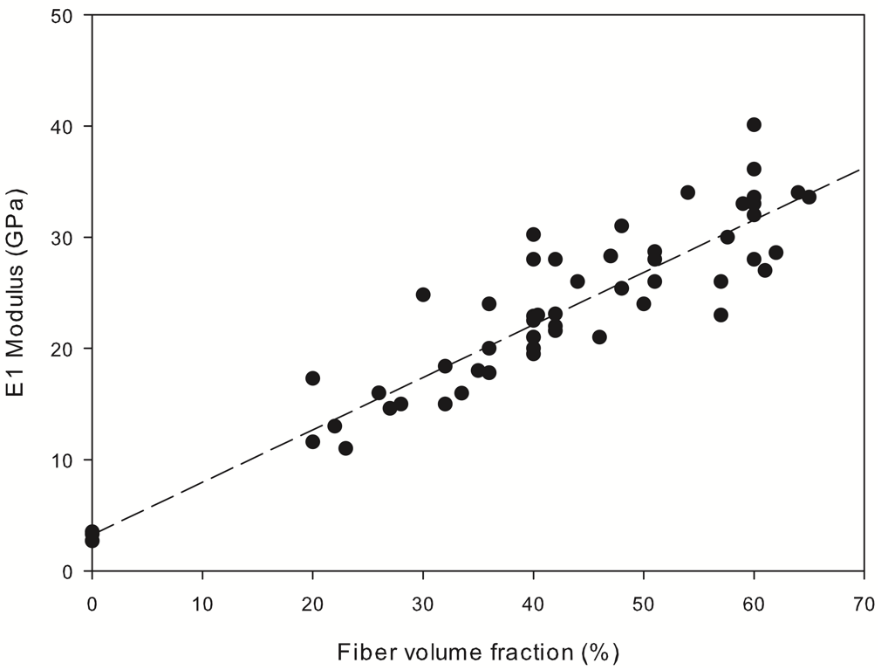


Figure 14. Strength versus fiber volume fraction for unidirectional flax/thermoset composites (Coroller et al., 2013) (Poilâne et al., 2014) (Liang et al., 2014) (Martin et al., 2014) (Van de Weyenberg et al., 2003) (Oksman, 2001) (Baets et al., 2014) (Cherif et al., 2011) (Hughes et al., 2007) (Marrot et al., 2014)(Khalfallah et al., 2014)(Lefeuvre et al., 2015)(Lebrun et al., 2013)(Habibi et al., 2017)(Goutianos et al., 2006)

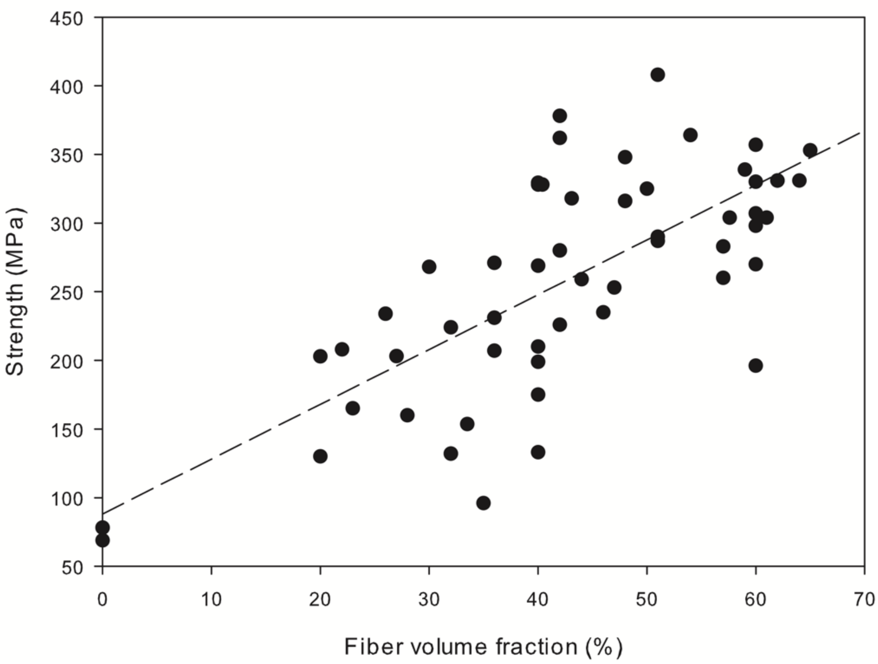


Figure 15. Initial longitudinal tensile modulus (E1) of unidirectional composites versus fiber volume fraction

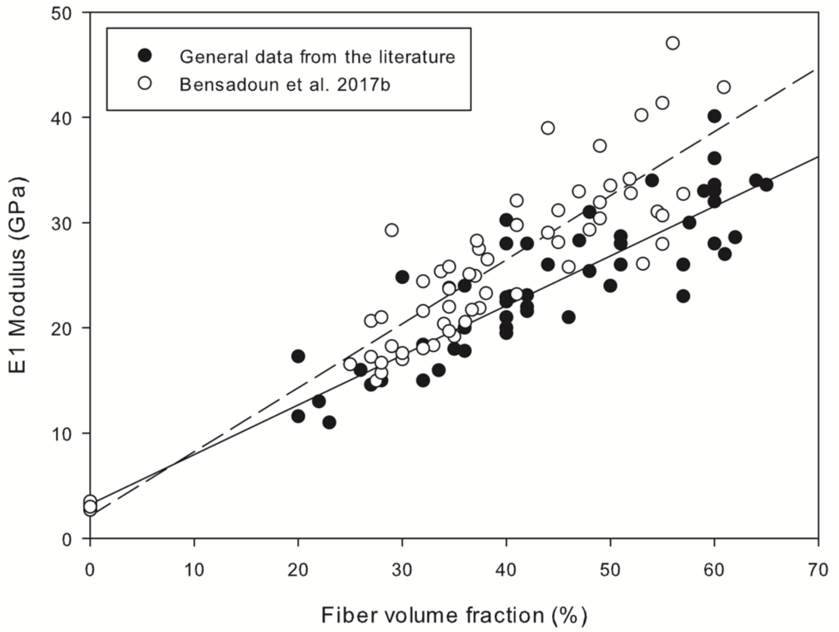


Figure 16. Strength versus fiber volume fraction for unidirectional flax/thermoset composites

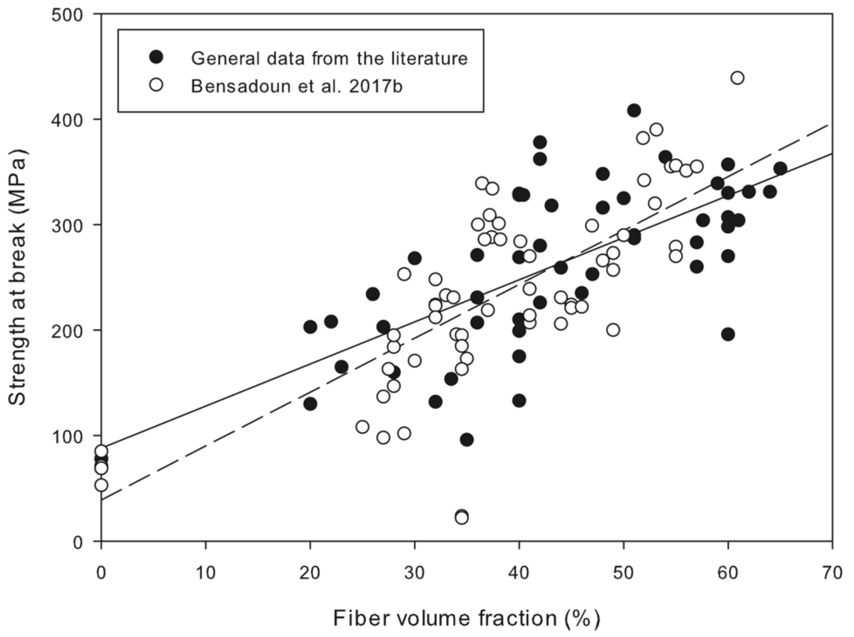


Figure 17 : Initial (E1) and tangent (E2) moduli measured for longitudinal tensile loading of samples made from the same flax fiber batch (Bensadoun et al., 2017a)

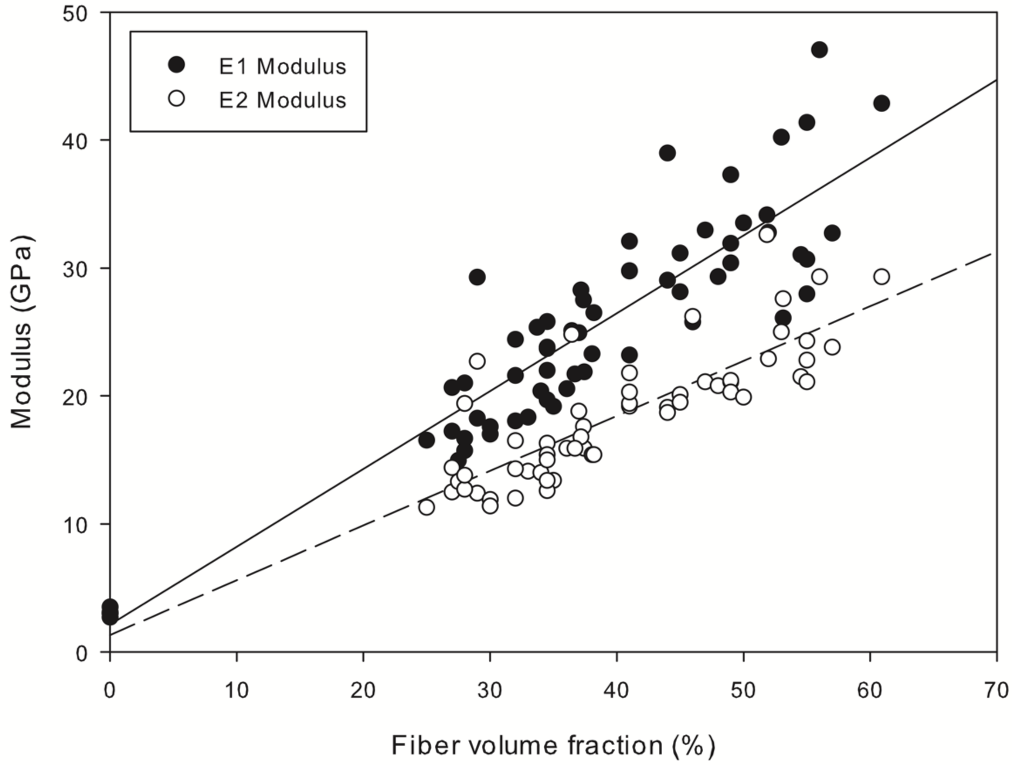


Figure 18 : Influence of fiber volume fraction on longitudinal tensile behaviour of a unidirectional flax/PA11 composite (Bourmaud et al, 2016)

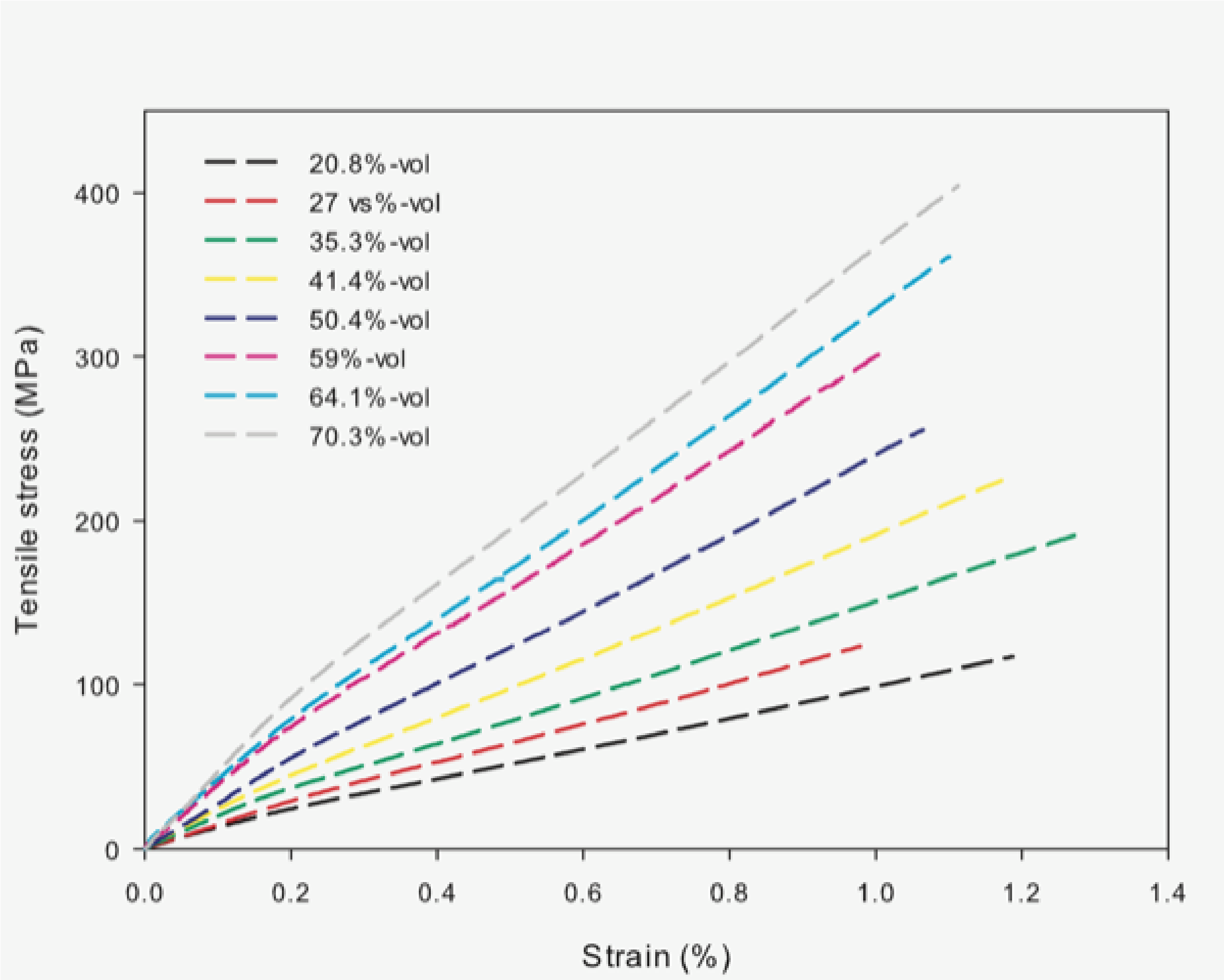
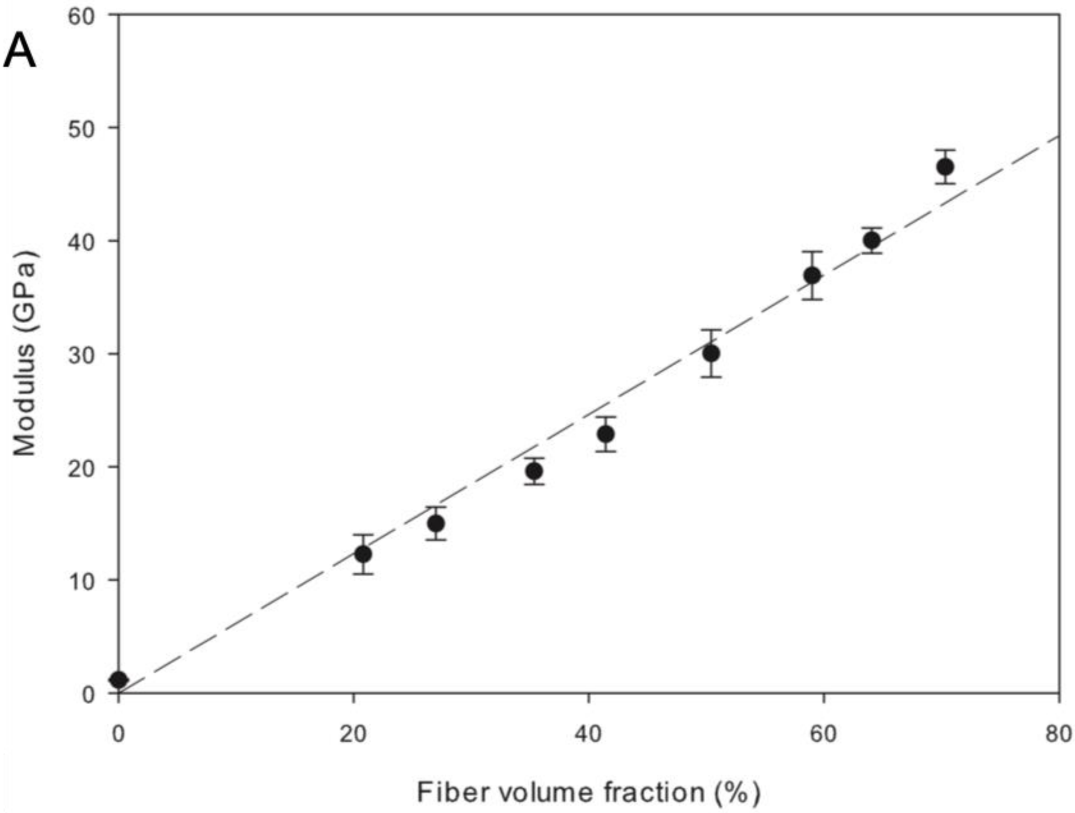


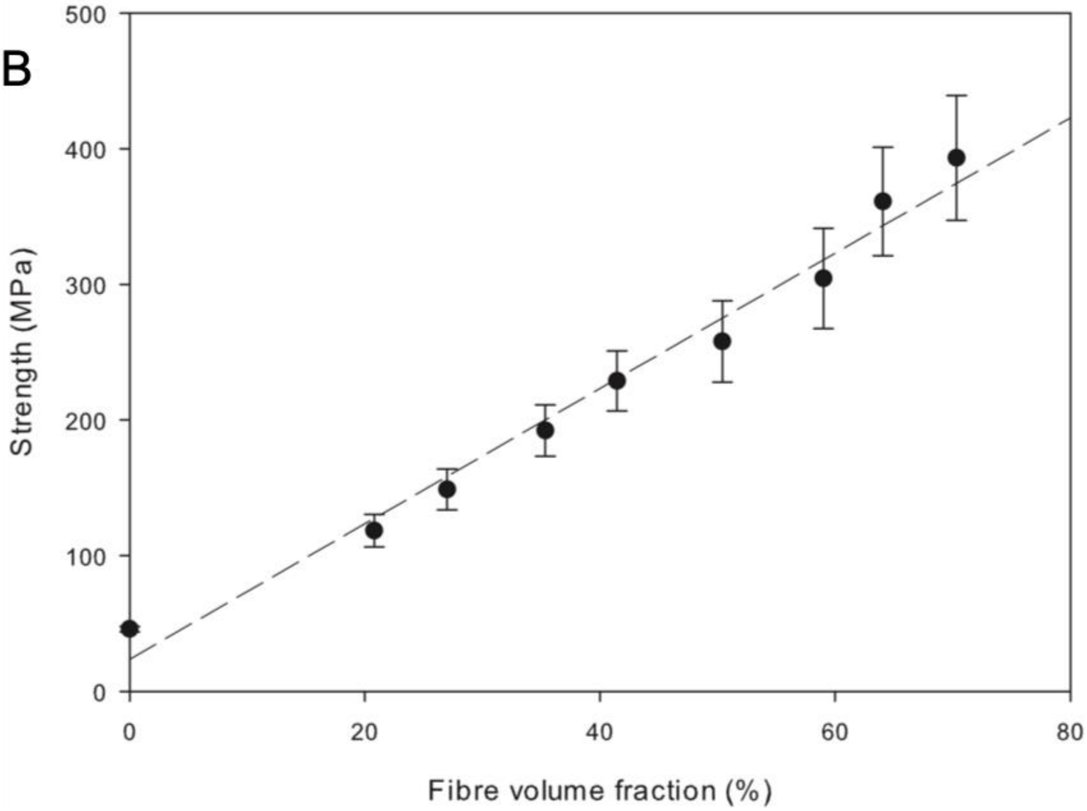
Figure 19. Initial Young's modulus (A) and strength (B) of flax–PA11 UD composite versus fiber volume fraction (Bourmaud et al., 2016)

A



Modulus (GPa)
60
50
40
30
20
10
0
0
20
40
60
80
Fiber volume fraction (%)

B



Strength (MPa)
500
400
300
200
100
0
0
20
40
60
80
Fibre volume fraction (%)