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Abstract

Over the past decades, the use of plant fibre reinforced composites has increased significantly due to their many attractive attributes such as high specific strength and modulus, wide availability, low cost and high environmental credibility compared to their synthetic counterparts. These attributes are especially attractive for lightweight applications in automotive, marine, aerospace and sporting goods sectors. This growth is expected to continue in the future. To improve the design and performance of bio-based composites, an improved understanding of processing-structure-property relations in such bio-based composites is required, the fibres

being the key component of the composite to obtain performing properties. This is due to the sensitivity of the constituent plant fibres to mechanical stress (pressure), temperature, water and other parameters. The purpose of this review is to critically synthesise literature on the impact of composites processing steps on plant fibre cell wall structure and properties. The impact of plant fibre composites processing steps from the polymer impregnation stage right through to the end-of-life recycling stage is reviewed. Additionally, mechanical, morphological and hygroscopic properties of plant fibres are considered in conjunction with process times, temperature and shear rate. This review will aid process and product designers to develop new performing plant fibre composite products, taking into account the process parameters to select the most optimised process and (their effects on) plant fibres. Considering how fibre properties change with biocomposites processing steps is indeed essential to understanding the links between the micro and macro scales, and to be able to design optimised plant fibre composite materials.

Keywords: Plant fibres; Processing; Time; Temperature; Water sorption; Mechanical properties

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58 **5. Concluding discussion**

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61

62 **1. Introduction**

63 Plant fibres have been an integral part of the 'fabric' of human history. Their use in ropes or
64 clothing dates back several tens of thousands of years (Kvavadze et al., 2009). Among the
65 latter, flax (*Linum Usitatissimum L.)* and hemp (*Cannabis Sativa*) fibres were originally cultivated
66 by the advanced civilizations of the Fertile Crescent; they are now the two most produced plant
67 fibres in Europe (Bourmaud et al., 2018). Over time, their uses have widened and for the past
68 twenty years, they have been of particular interest in the field of technical materials, in the form
69 of plant fibre composites.

70 Plant fibre reinforced composites are becoming an important area of development for a large
71 number of industries. This is particularly the case in the transport, marine and construction
72 sectors. These fibres have important environmental advantages, good specific mechanical
73 properties and are often available at a viable cost (Bourmaud et al., 2018). They offer a credible
74 alternative to synthetic fibres such as glass fibres for semi-structural applications. In the context
75 of impoverishment and the decline of fossil-based resources available to our civilization, their
76 use addresses a real societal and industrial challenge, driven by a regulatory framework that
77 promotes the use of bio-based, recyclable, biodegradable or lighter materials, and therefore
78 creating a less negative environmental impact (Mohanty et al., 2018).

79 To produce these bio-based composites, the plant fibres are processed with thermoplastic or
80 thermoset matrices by extrusion, injection, compression moulding, and also by the emerging
81 process of fused deposition moulding (3D printing), amongst various other techniques. During
82 these material transformation processes, the plant fibres are subject to thermomechanical
83 stresses that are not benign to the integrity of plant fibre cell walls (Bourmaud and Baley, 2010).
84 While the mechanical properties of synthetic fibres (such as glass or carbon fibres) may only be
85 slightly impacted by process stages (Shah and Clifford, 2015), this is not the case for plant cell
86 walls made of cellulose but also of non-cellulosic polymers, many of which have low thermal

stability (Siniscalco et al., 2018). In addition, they have a very specific architecture and structure that can be significantly altered by the mechanical stresses involved in conventional composites tools (Berzin et al., 2014). These structural modifications have a significant impact on the dimensions of the fibres, and therefore on their aspect ratio (length/diameter) which strongly conditions their reinforcement ability (G Ausias et al., 2013). Similarly, lengths of synthetic fibres are drastically affected by shear rate and process parameters (Bourmaud and Baley, 2007). In addition, structural modifications of the constituent polymers, induced by thermomechanical stresses, will have an impact on their architecture, degree of crystallinity, chain length, but also on inter-polymer bonds and on the overall parietal structure of plant fibres (Placet, 2009). These different impacts, whether morphological or structural, will modify the mechanical behaviour of the reinforcing fibres (Gourier et al., 2014). They can also have a major influence on the water absorption capacity of the principally hydrophilic non-cellulosic cell wall polymers, which will have consequences on the quality of multi-scale interfaces (polymer-fibre, fibre-fibre or between layers of a fibre) and therefore on the performance and durability of the resulting composites (Hill et al., 2009).

Thus, at both the fibre and composite scale, it is desirable to take into account all the relevant parameters that can alter the integrity of the plant fibres during all the processing stages. Whether during the drying, extrusion, compression, injection phase or even during a resin infusion process, the plant fibre must be considered as an evolutionary object whose structure and performance changes according to the process parameters used. This data is also important for mechanical modelling purposes as the properties of the fibres used as inputs into such models should be those following mechanical and thermal exposures, and not those of virgin fibres as the latter may no longer be valid model inputs. For example, the input permeability during flow process modelling should not be unsaturated permeability (alone), but rather saturated permeability that is a function of time, and incorporates effects of changing pressure and temperature during impregnation and curing, changing fibre structural properties (such as stiffness) as they get ‘wet’ and are being compacted, as well as any dimensional changes (e.g. swelling) of fibres due to resin absorption. Similarly, any micro-mechanical or finite element modelling should employ inputs of fibre geometry (vis. length, diameter, aspect

ratio, orientation) or structural properties (e.g. stiffness, strength, failure strain) based not on the virgin reinforcement form (e.g. pellets for injection/extrusion moulding) alone, but rather, make an effort to account for property changes due to the various processing steps.

In this critical review, an overview of these phenomena is provided. Firstly, the main thermal and mechanical parameters involved during a plant fibre composite processing transformation stage will be detailed. This will bear in mind the choice of the family and characteristics of the polymer matrix and the specific processing method. Secondly, the impacts of the plant fibre composite processing methods on the morphology of plant fibres are analysed. Following a detailed description of the architecture and composition of the fibres, the damage and rupture mechanisms occurring at the fibre scale, alongside the dimensional consequences on the fibre object is discussed. In addition, the effect of multiple re-processing cycles (injection recycling) on fibre property changes will also be elucidated. Thirdly, the impact of the plant fibre composite processing method and temperature on the mechanical performance of plant cell walls, in relation to the changes in their structure, as well as the hygroscopic behaviour of the plant fibres is discussed. The final section of this review will be a concluding analysis that will identify and account the key parameters to be defined when producing biocomposites and the compromise to be found between time, pressure, shear rate, and exposure temperature. Consideration will also be given to the choice of the nature of the fibres and the need (or not) to develop specific processing tools. We clarify here that the scope of this review does not include exploring the changes in properties of composites; rather the (changes in properties of) plant fibre reinforcements themselves are the subject of this review.

2. Choice of temperature and shear rate for plant fibre composite materials processing

Depending on the processing method chosen, which is largely determined by the nature of the polymer, its viscosity and its processing temperature, the thermal and mechanical stresses on the materials will vary greatly. They will impact both the polymer and the fibres, plant fibres being particularly sensitive to temperature but also to the exposure time. In this section, the

145 temperature and time ranges to which materials are subjected to during processing will be
146 discussed. In addition, a parallel will be established with the shear rates, which are also very
147 varied depending on the nature of the polymer used but especially on the processing technique.

148 When using a plant fibre composite, the temperature is highly dependent on the process used
149 and the nature of the matrix. In general, the constituents of plant fibres (particularly amorphous
150 polysaccharides) begin to degrade and lose their native properties above 200°C (Velde and
151 Baetens, 2001). But for some, such as pectins, the glass transition occurs at much lower values
152 (around 50°C), leading to a change in behaviour of the constituent cell wall polymer and
153 premature change in fibre performance. For more details on the impact of temperature on the
154 performance and structure of plant fibres, the reader is invited to refer to section 4.a of this
155 review.

156 The first parameter to consider is the choice of the polymer matrix, which will in turn determine
157 the temperature undergone by the plant walls during the transformation process. The use of
158 matrices with low process temperatures is then of major interest (Liang et al., 2010). This is of
159 course possible with thermosetting resins, but also with thermoplastic matrices, which also offer
160 a wide processing temperature window. In addition, many low melting temperature
161 thermoplastics, such as poly-(butylene succinate) (PBS) (Teramoto et al., 2004), poly-
162 (caprolactone) (PCL) (Berzin et al., 2014) or poly-(lactid) (PLA) (Duc et al., 2014) are also
163 compostable, which is an additional advantage when the reinforcement is also compostable
164 (Pantaloni et al., 2020). Bourmaud et al. (A. Bourmaud et al., 2015) found for composites
165 injected and reinforced with flax fibre that the choice of matrix had an impact on the mechanical
166 performance of the reinforcements, with fibres impregnated at 140°C with PBS being
167 significantly less impacted than those embedded in PLA at 190°C. Thus, the process
168 temperature is important because of its impact on the performance of the fibres as well as its
169 influence on the rheological flow properties of the polymer. A rise in temperature generally leads
170 (except in special cases) to a decrease in the viscosity of the resin, which will be important for
171 the quality of the final impregnation. Indeed, due to the complexity of the plant fibre
172 reinforcements, the presence of fibre bundles (Coroller et al., 2013), preforms made of twisted
173 (Baets et al., 2014) or sometimes treated yarns, impregnation can be impeded and high fluidity

is preferable to obtain quality impregnation and thereby minimize porosity in the composite materials.

With regards to plant fibres, the choice of a suitable temperature that allows for better fibre impregnation is a key consideration. A compromise must therefore be found between possible fibre degradation and resin impregnation quality. This link between temperature, structural defects and viscosity has been studied in depth on PP/flax composites by Ramakrishnan et al. (Ramakrishnan et al., 2019). They have shown that the increase in pressure during compression moulding, temperature, and forming time allows for better impregnation but also a reduced porosity content. However, if the temperature and exposure time are too high, fibre degradation occurs, causing the release of volatile organic compounds and consequently an increase in porosity. The authors showed that although the composite stiffness is only slightly affected by these variations, tensile strength is substantially diminished. A low temperature is recommended as this allows for satisfactory porosity values to be obtained while also protecting the plant fibres. This relationship between time and temperature has also been studied at the composite scale by Liang et al. (Liang et al., 2015) on poly-(amide 11) composites reinforced with woven flax preforms. A decrease in composite performance with an increase in temperature and process time was observed; a 64% decrease in tensile strength of the material was measured with an increase in processing temperature from 230°C to 250°C and process time from 2 to 5 minutes. These conclusions are in line with those of Ramakrishnan et al. (Ramakrishnan et al., 2019). In addition to temperature, time also has a major impact and these various works show that a compromise is necessary to obtain parts of suitable quality. This is true for a compression moulding process, as well as for extrusion and injection moulding. By varying the nature of the extrusion tools and therefore the residence times of the flax fibres in a composite extruded and then injected with a poly(propylene) matrix (PP), Doumbia et al. (Doumbia et al., 2015) showed a decrease in the performance of the mechanical properties of the materials, which can be explained by a decrease in the rigidity of the flax fibres measured by nano-indentation. The choice of a process is often dictated by economic production constraints, but when several variables require consideration, it is appealing to select them

202 according to the residence time, which can vary from a few hundredths of a second to a few
203 minutes depending on the process (Fig. 1).

204

205 **Figure 1**

206

207 Depending on the processes and materials used, despite set temperatures appropriate for the
208 use of plant fibres, self-heating can occur; the latter can be caused by the exothermic nature of
209 the resin in the case of thermosetting matrices but also by energy dissipation during major
210 shearing operations. This is particularly true in extrusion where conjugates and single threads
211 are used. This phenomenon also occurs commonly in injection at the mould thresholds which
212 are intended to fluidize the material under the effect of pressure but which also causes
213 significant increases in its temperature. On PCL-extruded hemp fibre PCL blends, Beaugrand et
214 al. (Beaugrand and Berzin, 2013) found self-heating between 3°C and 67°C depending on the
215 screw rotation speed, for a barrel temperature imposed at 100°C. This strong difference shows
216 how difficult it is to control the real process conditions and above all the need to adapt the
217 machine parameters to the plant fibres in order to limit both the self-heating of the parts and the
218 shear rate, although the latter is necessary to homogenize the compounds and reinforcement
219 homogeneity.

220 Depending on the processes considered, shear rates vary considerably; Figure 1 summarizes
221 the stress ranges that can be encountered in the various tools used to transform plant fibre
222 composites.

223 Numerical tools exist to model the energy supplied to plant fibres during a process stage (with
224 particular attention to the extrusion process). These design assistance tools make it possible to
225 control the parameters during a process stage and thereby maintain plant cell wall integrity as
226 much as possible to generate materials with optimized performance.

227 To limit temperature and shear rate, one way is to use softer moulding stages such as
228 thermocompression (Liang et al., 2015), thermoforming (Bhattacharyya et al., 2003),

compression with induction heating (Ramakrishnan et al., 2019), 3D printing (Duigou et al., 2016) or automated fibre deposition (Baley et al., 2016; McGregor et al., 2017). As can be seen in Figure 1, they generally reduce pressure or time significantly; however, in the case of powder impregnation, 3D printing or automated deposition (Badouard et al., 2019; Baley et al., 2016), initial porosity rates are often high and require the implementation of an additional thermal consolidation stage. Infusion or thermocompression processes also have their advantages in terms of moderate shear rate but they are often carried out with thermosetting resins that offer good impregnation but also require longer processing times. For example, a complete curing stage of a flax epoxy composite can take between 2 and 8 hours, while it will only take 7 to 8 minutes in the case of PLA-flax (Liang et al., 2015). Moreover, thermosetting matrices are not easily recyclable, which is an environmental contradiction when it comes to plant fibre composites.

3. Impact of process on plant fibre morphology

3.1. Initial morphology of plant fibres

This section discusses the initial morphology of natural and extracted plant fibres, prior to being subjected to any biocomposites processing step. Fibres are ubiquitous in vascular plants and are found in various organs including stems, leaves, seeds, and roots. Indeed, the structure and properties of a fibre are governed by their growth and development process ('ontogenesis' in biology = 'processing' in technology) as well as their function in the plant (Shah, 2013).

For the materials community, fibres are loosely referred to as long cells or bundles of cells. They include i) sclerenchyma fibres in dicotyledon stems, such as phloem fibres of flax and hemp, ii) fibres around vascular bundles in monocotyledon stems and leaves, such as bamboo and sisal, respectively, iii) trichomes and mesocarp of seeds, such as cotton and coir, respectively, iv) and xylem fibres, such as tracheids (Bourmaud et al., 2018; Leuwin, 2007). Of course, in botanical terms, fibres have a tighter definition: a fibre is an individual cell that belongs to the sclerenchyma, and has characteristics including elongated shape (high aspect ratio), thick secondary cell wall, tapered ends, and structure-supporting role in the plant (Esau,

1953; Gorshkova et al., 2012; Mokshina et al., 2018; Van Dam and Gorshkova, 2003). Many other classifications of fibres exist, such as primary and secondary fibres, but no classification is sufficiently all-encompassing to exclusively capture the diversity (Van Dam and Gorshkova, 2003). For the purposes of this wide review, we will use the looser definition frequently adopted in the plant fibre composite materials community. Figure 2 illustrates structural properties (length, diameter, aspect ratio) and mechanical properties (stiffness, strength) of a range plant fibres, commonly used for composites reinforcement.

In terms of ontogenesis, fibres typically develop through three key stages (Bourmaud et al., 2018; Gorshkova et al., 2012): i) initiation, to up to around 30µm length and 3-5 µm thickness, ii) elongation, by up to more than 1000-fold in length and 10-fold in thickness, and iii) further specialisation (vis. cell wall thickening). Of course, environmental and agronomical factors would have compounded effects on plant - and therefore fibre - growth, however such effects are not the focus of this section.

Figure 2

During the elongation growth phase, fibres increase substantially in length, but also, albeit to a lesser degree, in width. The resulting aspect ratio (l/d) of a fibre is a governing geometric parameter of mechanical properties (Legland and Beaugrand, 2013), with high aspect ratios correlating to high stiffness and strength (Fig. 2) (Bourmaud et al., 2018; Mukherjee and Satyanarayana, 1986; Pickering and Rowell, 2008; Shah, 2013). However, the aspect ratio of fibre elements is also dependent on its form: if it exists as an elementary fibre (idioblast) or in bundles. Although bundles may have comparable lengths, they also have much higher diameters leading to substantially reduced aspect ratios. Most non-bast fibres have a shorter elongation phase and exist in large bundles (Leuwin, 2007; Van Dam and Gorshkova, 2003) leading to smaller aspect ratios (Fig.2). Indeed, even secondary phloem bast fibres, which grow in the secondary (cambial) meristem, have (up to an order of magnitude) shorter lengths, aspect ratios and mechanical properties in comparison to primary phloem bast fibres that grow in the

primary (apex) meristem (Gorshkova et al., 2012). This is because secondary phloem fibres grow in already formed tissue, while primary phloem fibre cells undergo coordinated growth, thereby developing at the same rate as the surrounding soft tissues (Bourmaud et al., 2018). Moreover, the embedding of fibres in the surrounding tissues to form bundles affects the ease of extraction of these fibres (Van Dam and Gorshkova, 2003) through retting, decortication, scutching and hackling. These processes further influence fibre aspect ratio, as well as impart defects (Hughes, 2012), leading to a further deterioration in fibre mechanical properties.

Secondary cell wall thickening is also an important growth phase, particularly in phloem and xylem fibres (Bourmaud et al., 2018; Gorshkova et al., 2012). Indeed, ratio of cell wall thickness to lumen diameter, referred to as luminal porosity, is also an important geometric parameter influencing fibre mechanical properties; larger cell wall thickness and lower luminal porosity correlate to higher strength and stiffness (Pickering and Rowell, 2008; Shah, 2013). As there is significant heterogeneity in dimensional characteristics of natural fibres, the luminal porosity may also vary along the length of the fibres (Beaugrand et al., 2017) and therefore may influence the fracture mechanics.

Figure 3

It is not just the cell wall thickening process in this third growth phase that is important, but also the emerging biochemical composition and the structural organisation of the fibre (Fig. 3). Secondary cell wall thickening involves the formation of thick, sometimes lignified, secondary cell wall layers with properties substantially better than the primary cell wall. The principal cell wall polysaccharides are cellulose, hemicellulose, with pectin-rich middle lamella adjoining the fibre cells. In addition, the lignin, a polyphenolic polymer, is non homogenously distributed over the cell wall structure with a reported enrichment in the middle lamella and primary cell wall. To a lesser extent, proteins are also part of the cell wall as some of them are playing a structural role, and are well known to act as the cytoskeleton of the cell, which impact on the plant cell remodelling (Wasteneys and Galway, 2003). The cellulose content, and in particular, cellulose

crystallinity are the key biochemical parameters influencing mechanical properties (Bledzki and Gassan, 1999; Pickering and Rowell, 2008; Shah, 2013). Cellulose crystallinity is higher for structure-supporting bast fibres and decreases with reducing structural requirements, as in leaf and seed fibres. The S2 secondary cell wall, the thickest of the cell wall layers (Fig.3), dominates the structural behaviour of a plant fibre cell (Bledzki and Gassan, 1999; Leuwin, 2007). The cell wall layer is comprised of cellulose microfibrils that helically orient at specific microfibril angle (MFA) to the fibre axis, and are held together by amorphous hemicellulose and lignin polymers. The MFA of the S2 cell wall is the key structural organisation parameter influence fibre mechanical properties. Bast fibres have low MFA (<10°), leaf fibres moderate MFA (10-25°) and seed fibres have high MFA (30-50°) leading to their corresponding mechanical properties of high, moderate and low stiffness (Figure 2), but low, moderate and high elongation at failure (Gorshkova et al., 2012; Shah, 2013).

The initial morphology and properties of plant fibres evolve through their extraction phases, and subsequent processing into polymer reinforcing fibre composites. Henceforth, we explore the main premise of this review paper, the evolution in fibre morphology and properties from an initial stage through the various processing methods employed to produce plant fibre composite.

3.2. Breakage mechanism during a process stage

As an introduction, we must recall that it seems unrealistic to demand for a description of one sole prevalent breakage mechanism, due to the large panel of stress modes generated by the multiple processes and stages in use. One can expect that according to the process used, the dominant mechanical stress will vary between shear, impact, compression, tension and wear. Hence, it would be a very challenging task to generalise ‘the’ breakage mechanism that is sufficiently malleable to adequately describe the effects of all the plant fibre composites process stages used for plant fibres.

Figure 4

Along this line of thought, it is relevant to compare the different processes. To conduct an empirical comparison of the severity of dedicated processes is not a trivial task. Some have already embraced this challenge with noticeable success when comparing effects of vacuum-assisted resin transfer moulding (VARTM), pultrusion and extrusion-injection moulding (Haag et al., 2017).

Establishing a relationship between process intensity and breakage is fundamental for natural fibres. Due to their natural biochemical anisotropy and composite poly-laminate structure (Barber and Meylan, 1964), a dynamic succession of predominant breakages mechanisms is observed, which take place in a more or less coordinated fashion according to the process scale and duration (Fig. 4, illustration of the decohesion-delamination of individual fibre in a bundle).

Understanding and predicting process-induced damage mechanisms will enable better control of fibre length. Indeed, longer fibres with high aspect ratio (Pickering et al., 2016) may improve composite mechanical performance (G Ausias et al., 2013; Beaugrand and Berzin, 2013). Many works on natural fibre are inspired from previous studies on synthetic glass fibres (Shon and White, 1999). Process-induced damage affects the structural geometry of the fibre, the latter being characterized by its aspect ratio (Grishanov et al., 2006; Nyström et al., 2007). Moreover, improvement in thermoplastic composite strength is reported with increasing fibre aspect ratio (Beaugrand and Berzin, 2013). Notably, the more individualised the fibres are (see Fig.4), the higher the aspect ratio. The resulting decohesion from process stresses may expose various surface chemicals, but also substantially increases the specific surface area on offer for interfacial adhesion.

This hypothesis is not new and is supported by research in the field of pulp and paper. Indeed with wood fibres, which are relatively short in length and rather homogeneous in dimension compared to plant fibres, this community has understood that the breakage mechanisms of the bundles and individual fibres of wood are governed by the refining intensity (e.g. torque), which

also leads to specific desirable technical characteristic of the pulps and products thereof (Padovani et al., 2019).

Among processes that induce damage, an arguably aggressive process is milling (Mayer-Laigle et al., 2020). The fibre milling process may lead to plant fibre particles less than a few µm in length (see Fig 5, d), and of non-elongated shape (aspect ratio lower than 10). Extrusion with a co-malaxor system, single- or twin-screw, is yet another harsh process (Doumbia et al., 2015). Many reports are available on the impact of single-screw profiles on a batch of natural fibres (El-Sabbagh, 2014). The rich literature on this topic is explained by the economic implication of this research for the current plant fibre composite sector, given that the twin-screw process is the main industrial process for incorporating lignocellulosic fibres into polymers (Antoine Gallos et al., 2017) and it may offer a numerical damage descriptor for a common scale unit. Indeed, using this approach, one is able to scale the fibre breakage observed, depict it in terms of damage mechanisms (Castellani et al., 2017), and even model the progressing mechanisms for twin screw (Berzin et al., 2017) or extrusion-injection moulding (Albrecht et al., 2018). Technically, mechanistic models of evolution-simulation for lignocellulosic fibre breakage are based on observations, such as through static (Le Moigne et al., 2013) and dynamic (Le Duc et al., 2011) methods, quantification by morphometric measurements from 2D image analysis (Hamdi et al., 2015) (Hamdi et al. 2015 CPA), and even 3D particle volumes from X-ray tomography (Hamdi et al., 2018).

Figure 5

One recent breakthrough in plant fibre breakage understanding is a breakage classification based on fatigue, fragility or where peeling behaviour is present (Castellani et al., 2017). The study used a setup of four batches of fibres of botanical origin and proposed correlations between breakages, intrinsic fibre properties, and biochemical composition. Lignin, a recalcitrant polyphenolic polymer, seems to be strongly involved in the breakage behaviour. Regarding the morphometry of the fibre elements, it seems to be a first order criteria for

breakage mechanisms. A breakage index of lignocellulosic fibres was defined by Berzin et al. (Berzin et al., 2017). The authors report a strong influence of the initial dimensions of the fibre elements: longer fibres are more susceptible to length breakage (i.e. more susceptible to a decrease in length) rather than decohesion of bundles (which result in decrease of diameter). Note, for instance, that there is much variation between miscanthus and hemp. Figure 5 gives a schematic representation of the plant fibre-breakage mechanisms during a process stage along with a kinetic of reduction of both the length and the diameter of the fibre elements. At the beginning of the process stage, both the rate of decohesion (mostly longitudinal break) and fragmentation (due to transverse break), are high but not identical. Several types of damages are often visible at the surface of the fibre elements (Fig 5) like surface flaws (vestiges of residual middle lamella, primary cell wall). And along with the process, there is an enrichment of fine elements according to the duration or severity (Fig 5). The kinetics of breakages have been measured for fibres of various origin, but the influence of central cavity (vis. lumen) (Fig.3), which is often considered a defect, is not usually considered in breakage predictions due to technical considerations. More generally, the ratio between respective area of cell wall and lumen is known to be important in the rupture properties of fibre elements (Lucas et al., 1997). The filling of these lumen by polymer can occur during the hot melt processing for thermoplastic based composites. This filling can significantly increase the strength of the composites (Mohanty et al.). The lumen may be partially or totally filled with the polymer depending on parameters such as the lumen/cell wall size ratio, the viscosity of the matrix and the process parameters. Reported lumen filling values range from 5 to 50% for wood fibers (Mohanty et al., 2005), whereas such data is unavailable for bast fibres.

For annual plant fibres, the heterogeneity of the morphometries (Legland and Beaugrand, 2013), biochemical compositions and macromolecular cell wall organisations have discouraged many researchers to propose breakage mechanisms during a process stage. Indeed, these fibre elements are often organised as bundles, rarely as individual fibres, in an industrial lot. These fibre elements, comprising fibrils, macrostructural defects such as kink bands, and a myriad of broken parts, are often called ‘fines’ (see Fig 5). The overall challenge is in i) the technical description and the labelling of the morphometric features, and ii) relating the

functional properties (mechanical/thermal) of each of those micron features and in explaining an homogenised property at the composite scale.

Fines are defined as particles of length smaller than 200µm (Mayer-Laigle et al., 2020), though these fines can be underestimated due to technical detection limits. Fines are known to have an impact on rheological and even mechanical properties of plant fibre composites, particularly when they are highly concentrated (Bourmaud et al., 2019).

The damage mechanism during a process stage does not necessarily mean 'breakage' that leads to division of the fibre elements in at least two bodies. Indeed, there are also damage-induced defects, like compression zone, kink band, and fibrillation (Fig 5), that are even more difficult to assess in comparison to decohesion and rupture. Indeed, they require more focused instruments for visualisation, such as microscopy techniques (optical or electronic). Arguably, these are defects or weakness points that could initiate further breakage mechanisms. In fibrillation, often part of the fibre sub-layer remains, i.e. there is partial delamination. They could enhance paper properties, whereas their positive contribution in plant fibre composites is more questionable and rather difficult to quantify by direct observation (Padovani et al., 2019). The kink bands are submicronic damages present in natural fibres and could drive failure, as observed in hemp fibres (Beaugrand et al., 2017). They are naturally present in stems (Thygesen et al., 2011) but their content increases drastically with the decortication process (hackling/scutching) where the fibre bundles are extracted from the plant. Kink bands are zones of disorganisation of crystalline cellulose in the cell wall, and span across the full diameter of the fibre elements. Recent work investigated by (Beaugrand et al., 2017) reported some crack bridging between two adjacent kink bands, thanks to the propagation of the main crack via the fibre lumen (Beaugrand et al., 2017). Lumen porosity and empty space between fibres are often viewed as defects (Madsen and Lilholt, 2003; Shah et al., 2016) (Madsen CST 2003, Shah et al Cellulose 2016); the smaller they are, the higher the mechanical properties of the technical fibre (Fuentes et al., 2017; Müssig and Stevens, 2010). In addition, the surface defects (crack, pith, notch) are often forgotten, however it has been shown using finite element simulation and high-resolution X-ray microtomography that they contribute significantly to the development of severe damage evolution (Guessasma and Beaugrand, 2019), as well as the fibre ends. Finally, it

seems useful to highlight that in terms of damage rate, the development of damage growth is often related to the presence of localized or diffused damage sites (Andersons and Joffe, 2011; Gourier et al., 2017; Guessasma and Beaugrand, 2019).

3.3. Impact of process on plant fibre diameter and aspect ratio: focus on recycling.

Processing of thermoplastic composites generally cause a drastic reduction in the length of the plant fibres, especially after an extrusion stage (Albrecht et al., 2018; Beaugrand and Berzin, 2013; Berzin et al., 2014; Subasinghe et al., 2015). The diameters of the fibres are also reduced during this transformation phase. The design of the processing screws in an injection moulding machine or single- or twin-screw extruder influences the final dimensions, morphology and properties of the fibre, and consequently affects the properties of the resulting fibre reinforced composite. Indeed, different screw designs may generate very different shearing and mixing conditions during the process. Generally, to attain homogeneity and good dispersion of fibres in the composites, twin-screw extruders are favored. Buss co-mixers can also be an efficient alternative (Shon and White, 1999). Doumbia et al. (Doumbia et al., 2015) compare the transformation of flax-PP following processing through a Buss co-malaxor and by twin screw extrusion; they show that individualisation was more efficient with the co-malaxor but the latter induces shorter fibre lengths. It is, nevertheless, recommended that fibre individualization and dispersion are promoted. This is due to their positive effect on the resulting composite material's tensile strength and modulus, as demonstrated on flax (Doumbia et al., 2015) and kenaf (Sallih et al., 2014) fibre composites; better dispersion leads to homogenization and also possible penetration of resin into hollow cores of plant fibres especially for high lumen-size fibres such as kenaf. A 2D simulation comparison between single-screw and twin-screw extruders (Connelly and Kokini, 2007) has shown better spreading of fibres/particles through mixing by twin-screw extruders.

The screw profile is also an important factor to be considered. The screws can be tailored with multiple block elements of various shapes for specific functions such as conveying and mixing (A Gallos et al., 2017). Based on the shaped elements used for the screw profile, fibres will be

subject to different residence times by using conveyors or reverse elements. The evolution in bast fibre lengths during processing through various screw elements was recently studied by X-ray microtomography (Hamdi et al., 2018), revealing that the shaped profile of the screw has a strong influence on fibre length. The use of reverse screw elements increases the shear rate and the severity of the process (Gogoi et al., 1996). Shearing increases with mixing or kneader elements (Berzin et al., 2017), as reported for bast fibre, and also short wood fibre (Dickson et al., 2020); inducing a drastic reduction in the length of the fibres (Teuber et al., 2016). The screw profile also influences the final properties of composites. For instance, it has been demonstrated that the screw profile can be used to substantially improve the exfoliation of organoclay in PP (Lertwimolnun and Vergnes, 2007), and the mechanical properties of short glass fibre reinforced polypropylene composites (Lekube et al., 2019). Aside from the screw configuration itself, several other extruder parameters can be optimised, such as the feed rate or screw speed.

Depending on the nature of the fibres, their behaviour may vary during an extrusion or injection stage. Originally, fibres are assembled into bundles in plants (Bourmaud et al., 2015) and these bundles may still be present after one or more transformation cycles in a composite material. However, their ability to divide depends on the cohesion of these bundles and the nature of the middle lamellae, and in particular their lignin content. Thus, jute bundles, made of very cohesive short fibres, will be difficult to individualise (Tanguy et al., 2018), which will be detrimental to maintaining a good aspect ratio. In contrast, flax fibres, already highly individualised during scutching, can be almost elementarised at the end of an extrusion or injection stage (Oksman et al., 2009). This has also been demonstrated on hemp by Peltola et al. (Peltola et al., 2011).

The concentration of fibres can affect fibre morphology but also viscosity and overall rheology of the melt blend during moulding or extrusion stage; and several models have tried to describe and quantify this phenomenon. These models use numerical simulation software (e.g. Cadmould® and MoldFlow®) and determine fibre interaction coefficients (Albrecht et al., 2018) that reflect the adhesion forces between the fibres within fibre bundles. When the volume fraction (and thereby concentration) of plant fibre increases, the fibre lengths and diameters decrease, as shown for wood (Teuber et al., 2016) and flax (Puglia et al., 2008). These trends were confirmed experimentally by Ausias et al. (G. Ausias et al., 2013). They observed a

510 substantial decrease in flax fibre length with increasing fibre content following extrusion or
511 injection molding. They also noted a slight decrease in the aspect ratio at low fibre contents. A
512 threshold fibre content of 30%-vol. was highlighted, indicating the estimated critical aspect ratio.
513 This critical aspect ratio depends on fibre intrinsic characteristics, matrix properties and quality
514 of fibre/matrix adhesion. These changes in fibre morphology induced by changes in fibre
515 volume fraction significantly impact the viscosity and rheology of polymers and consequently the
516 flow behaviour in processing tools; this decrease in compound viscosity may be an advantage
517 for industrial users, especially considering the high initial mixing viscosity resulting from the
518 presence of the notable plant fibre components (Bourmaud et al., 2016; Doumbia et al., 2015).

519 By applying successive injection cycles after this compounding, these evolutions will naturally
520 continue with particular characteristics related to the nature and the structure of the
521 reinforcements. A distinction can be made here between i) fibres originating from the supporting
522 tissues which have initial lengths of a few tens of mm and high aspect ratio (case of flax and
523 hemp), and ii) shorter fibres, such as cell walls of wood, which are naturally short and are
524 generally incorporated in the form of powder or flour because of their extraction method or their
525 origin (sawmill waste, for example). Figure 6 shows the evolution in length distribution of flax
526 fibres from an initial length of ca. 2 mm after successive cycles of compounding and injection
527 within a PLLA-PBS matrix (Bourmaud et al., 2016). We observe a steady decrease in fibre
528 length with recycling steps; as the injection cycles continue, the decrease in length continues to
529 reach a threshold value from the fourth cycle. Interestingly, we have been able to demonstrate a
530 correlation, on PA11-flax composites (Gourier et al., 2017), between the inter-defect distances
531 and the final lengths of the same fibres after recycling; Indeed, as shown by Le Duc et al. (Le
532 Duc et al., 2011), the break of fibres during compounding preferentially occurs in the kink band
533 zones.

534 This strong decrease in length after compounding has already been observed on flax, hemp
535 and even sisal and kenaf fibres (Bourmaud and Baley, 2009, 2007; Dickson et al., 2014; Le
536 Duigou et al., 2008; Subasinghe et al., 2015). This has also been observed on synthetic fibres
537 such as glass. Due to the different structure of the fibre plants, the observations are different for
538 changes in diameters and therefore in aspect ratio (L/D). Regarding glass fibres, one can note a

539 significant decrease in this form factor (L/D) with the number of cycles due to the shear rate of
540 the injection process.

541

542 **Figure 6**

543

544 This decrease is caused by the constant reduction in the length of the fibres associated with the
545 fact that their diameter remains constant throughout the injection cycles. Indeed, glass fibres
546 are initially individualized within the compound. The behaviour of plant fibres is different. These
547 fibres are initially present in the plant in the form of bundles which can contain several tens of
548 fibres. After recycling, these bundles will divide, which will cause a reduction in the diameters of
549 the reinforcements. In parallel, fibre lengths decrease but this decrease is less marked than for
550 glass fibres (Bourmaud and Baley, 2007). The combination of these two decreases (length and
551 diameter) leads to a relative stabilization of the aspect ratios as can be seen in Figure 6, for flax
552 fibre elements.

553 One can notice that the aspect ratio obtained on plant fibres are relatively low compared to
554 those of glass fibres - 13.0 and 10.5 for hemp and sisal against 54.0 for the glass after an
555 injection cycle (Bourmaud and Baley, 2007). This can lead to a lower reinforcement quality. This
556 factor must be taken into account in the realization of future compounds where the emphasis
557 will be on the dispersion of the elementary fibres in order to favour performing aspect ratios.

558 Works have also been carried out on composites reinforced with wood flour (Beg and Pickering,
559 2008; Dickson et al., 2014; Soccalingame et al., 2015). Given the specific morphology of these
560 reinforcements, it is sometimes difficult to define a length and a diameter, these are therefore
561 mostly defined by a single dimension. Even if the geometry of these reinforcements is
562 noticeable, it remains modest and the wood particles very quickly reach a floor value given their
563 initial geometrical characteristics which are already small. By working with wood fibres extracted
564 by methods that better preserve the lengths, it is possible to approach the trends observed for
565 flax and hemp fibres, the fibres then maintaining a significant aspect ratio over the cycles

566 (Dickson et al., 2014). However, these extraction processes require the use of heavy
567 mechanical and chemical equipment and the environmental impacts must be considered in
568 parallel.

569 After compounding and injection, the plant reinforcements are embedded into a polymer matrix.
570 If it is possible to extract them from the latter chemically in order to study their morphologies, it
571 is not possible to carry out tensile tests because of their short lengths and especially the impact
572 of the solvents on the integrity of the plant walls. In addition, the nanoindentation allows to
573 obtain an estimate of the stiffness and hardness of the plant cell walls by working on cross-
574 sections of injected parts. Figure 6 illustrates the evolution in mechanical properties measured
575 by nanoindentation (Bourmaud et al., 2016) of flax fibres after injection cycles in a PLLA-PBS
576 matrix. These images show that in this specific case the surface morphologies and the
577 geometries of the indents are similar, evidencing no major structural differences, contrary to
578 what can be observed, for example, between two species of plant cell walls (Tanguy et al.,
579 2016). We can note an important difference between the performances of the virgin fibres and
580 after a process cycle, this being particularly marked for the hardness which exhibits a significant
581 drop after compounding.

582 As was pointed out by the wood community (Eder et al., 2012), the nanoindentation modulus is
583 representative of the behaviour of cellulose microfibrils (especially when the microfibrillary
584 angles are weak as it is the case for flax or reaction wood) whereas the hardness is much more
585 sensitive to changes linked to the matrix polysaccharide. This is mainly due to its method of
586 calculation, based on a theoretical contact area which can be modified in the event of alteration
587 of this matrix. Thus, the large drop in hardness after a process cycle reveals a probable
588 alteration of the pectins and hemicellulose constituting the matrix of the S2 wall. This was also
589 demonstrated by tensile tests on heated fibres (Gourier et al., 2014) or by nanoindentation by
590 studying the effect of an injection cycle on the mechanical performance of flax cell walls
591 (Bourmaud and Baley, 2010).

592 Similar tests were carried out on PP-wood compounds (Soccalingame et al., 2015; Stanzl-
593 Tschegg et al., 2009). The observations are different, especially for the hardness whose value

tends to increase with the recycling cycles. This contradictory phenomenon can be explained by the nature of the wood cell walls which, contrary to flax, constitute a large part of lignin and xylan. It has been demonstrated (Yin et al., 2011) that in the case of wood, partial crosslinking of the cellulose-xylan-lignin system could occur after a thermal stage, this phenomenon has been exploited to develop stabilization treatments for the walls such as wood refining.

In spite of these morphological and mechanical changes of the reinforcing plant fibres, bio-based composites have stable mechanical performances after recycling, even if caveats are made based on the nature of matrix; some such as PLA may be more sensitive to shear rate and hydrolysis. In this case, a significant decrease in the stress at break of the composite appears after a few cycles (Le Duigou et al., 2008). In case of use of a polyolefin or PA11 matrix (Gourier et al., 2017), the values of Young's modulus and strength at break are mostly stable after several process cycles. This is mainly due to the good stability of the fibres' aspect ratio but also to a relatively stable fibre-matrix adhesion compared to the one between PP and glass fibres. The latter benefits from optimized sizing but this sizing loses efficiency with subsequent process cycles. Moreover, one can notice that recycled PP offers also good recycling behaviour and enables to manufacture reliable plant fibre composites, even after several process cycles (Bourmaud et al., 2011).

At the same time, due to the smaller size of the fibrous elements but also the alteration of the chain length of the matrix, the plant fibre composite viscosity is generally considerably reduced after recycling. This point does not constitute a disadvantage, in particular for applications requiring high fluidity such as injection moulding. Thus, all these elements ensure that the plant fibre composites generally present useful properties after recycling, the main limit being their collection and the waste separation, which is not yet really implemented mainly because of the low volumes available. The increase in market shares in the automotive sector must be a trigger for taking their end-of-life into account.

4. Impact of process on plant fibre properties

4.1. Impact of the temperature at the single fibre scale

As discussed previously, important morphological or mechanical degradations of the plant cell walls may occur during a process stage, with significant decreases in length but also in terms of mechanical performance. Shear rate and pressure are responsible for a large part of this damage but temperature, by modifying structure of fibre constitutive polymers must also be considered. As evidenced by Subasinghe et al. (Subasinghe et al., 2015), extrusion temperature has an impact on fibre length and also on fibre ultrastructure and properties. The temperature regime undergone by the cell walls induces a significant decrease in the properties of these walls which is accentuated with the rise in temperature, either due to the choice of the matrix or self-heating (often uncontrolled) (Beaugrand and Berzin, 2013) induced by the shear rate within the process tools and especially the extrusion screws. This last point still requires work to be done: the temperatures really experienced by the fibres are poorly known and often underestimated, even when approximated using modelling software (Berzin et al., 2014).

Sometimes, thermal treatment is used; this is especially the case for wood fibres with regards to the torrefaction process (Pushkin et al., 2015; Volfson et al., 2015). The purpose of torrefaction is to modify the chemical composition of wood filler in order to improve its compatibility with polymers, particularly polyolefin matrices. In these two works, the effect of torrefaction (soft pyrolysis (200–300°C) in inert atmosphere) on the samples of hardwood (birch) and softwood (pine) has been compared. The authors observed strong differences between the torrefied samples between 225 and 250°C, mainly due to the specific behaviour of hemicellulose constituents. A significant decrease in the xylose content was observed at a temperature of 250°C whereas mananes were more resistant to degradation. Thus, an important modification in hemicelluloses was reported. Interestingly, the content of cellulose remained stable up to 250°C and the crystallinity rate increased in torrefied samples. Moreover, the lignin/carbohydrate ratio gradually increased with the temperature of treatment, probably due to both carbohydrate decomposition and the formation of new aromatic clusters due to thermochemical reactions. This last point is considered to be very promising for the forthcoming plant fibre composite blends, lignin being an interesting promoter of adherence, especially with polyolefin matrices (Graupner et al., 2014).

651 Temperature may also negatively impact the cell walls properties and performances. Plant
652 fibres are mainly composed of cellulose but also of non-cellulosic polymers, which for flax are
653 mainly pectins and hemicelluloses. Research works have shown that the first polymers to be
654 affected by a thermal stage are these non-cellulosic polymers and in particular pectin; cellulose
655 does not start to degrade until about 200°C (Gourier et al., 2014). This assessment is confirmed
656 by several literatures. For example, Paris et al. (Paris et al., 2005) have studied spruce and pine
657 heating between 35 °C and 250 °C, these authors highlight the evaporation of water and
658 dehydration with slight depolymerization, but no change in cellulose microfibrils. In addition,
659 Zolfrank and Fromm (Zollfrank and Fromm, 2009) have studied wood pyrolysis between 200 °C
660 and 300 °C. In the range of 200°C–250 °C, they demonstrate at first a degradation of polyoses
661 but only a disorientation of cellulose micro-fibrils away from the fibre axis; a significant evolution
662 in cellulose structure appears only near 250 °C.

663 At the cell wall scale, nanoindentation or AFM Peak Force technology are powerful tools to
664 study and monitor local mechanical properties of plant cell walls. The review of Eder et al. (Eder
665 et al., 2012) thoroughly synthesis advantages and drawbacks of such investigations, mainly on
666 wood. Other authors have investigated the impact of temperature through this route. For
667 example Li et al. (Li et al., 2015) studied the mechanical properties of bamboo cell walls in order
668 to link the indentation modulus and hardness with an increase in temperature. Zickler et al.
669 (Zickler et al., 2006) studied the behaviour of pyrolysed spruce wood as a function of high
670 temperature up to 2,400°C. They evidenced significant variations depending on the temperature
671 range. Stanszl-Tschegg et al. (Stanzl-Tschegg et al., 2009) highlighted a slight increase of the
672 indentation modulus with temperature but a significant increase of the hardness. This increase
673 can be explained by a cross-linking reaction of lignin and xylan (Yin et al., 2011) and is specific
674 of xylan type fibres (Mikshina et al., 2013). This result was confirmed by (Soccalingame et al.,
675 2015). These authors observed wood cell wall hardening as a function of recycling cycles, the
676 number of cycles resulting in substantial cumulated heating time (Fig.7).

677 Similar work on thermoplastic composites reinforced with flax fibres has shown different results
678 by using nanoindentation (Bourmaud et al., 2015; Doumbia et al., 2015; Gourier et al., 2017; Le
679 Duigou et al., 2016). Flax cell walls, not lignin-rich, are considered as gelatinous cell walls,

680 generally show a decrease in their mechanical performance with temperature and process
681 stages. The nature of its constitutive non-cellulosic polymers make it more heat sensitive than
682 more lignified plant walls such as wood or kenaf (Fig. 7). These results were confirmed by
683 Siniscalco et al. (Siniscalco et al., 2018) with AFM PeakForce measurements in a range of
684 temperatures from room to 250°C.
685
686 **Figure 7**
687
688 Observations at the cell wall scale are confirmed on elementary fibres. Gassan and Bledzki
689 (Gassan and Bledzki, 2001) evidenced that tensile properties of jute and flax fibres start to be
690 affected by temperature at around 170°C but time must also be considered. In their case an
691 exposition time of 8 min was considered. Gourier et al. (Gourier et al., 2014), have checked the
692 performance of the elementary flax fibres after an 8-min heating stage between room
693 temperature and 250°C. A strong decrease in mechanical performance was observed above
694 210°C with a reduction in modulus of 10% and strength of 30%. This decrease is mainly
695 attributed to the degradation of the non-cellulosic matrix and not to the cellulose itself, as
696 previously underlined. This evolution of non-cellulosic parietal polymers induces a decrease into
697 the slipping ability of cellulose macro-fibrils and a more brittle behaviour of single fibres.
698 Consequently a progressive drop in strain and also strength is observed. As explained in
699 section 2 of this review, time needs also to be considered; Baley et al. (Baley et al., 2012) report
700 a significant decrease in tensile properties of flax fibres after 12 hours at 105°C; this
701 temperature is moderate but, conjugated with time, has approximatively the same impact as 8
702 min at 210°C.
703
704
705 **4.2. Impact of process on hygroscopic behaviour of the fibre**
706 The reliability and long-term durability of plant fibres is influenced by their structure (microfibrillar
707 angle, the fibre diameter, fibre surface characteristics) and chemical composition (vis. cellulose,

hemicellulose and lignin content). Similarly, the performance of plant fibres as reinforcements also largely depends on operating environments (temperature and humidity) and the presence of surface defects and the hydrophilic nature of fibres itself (Faruk et al., 2012) (Faruk, et al., 2012). The cell walls of plant fibres are predominantly made up of a number of layers including a primary wall (the first layer deposited during cell development) and the secondary wall (S), which comprises of three sub-layers (S1, S2 and S3) as depicted in Figure 3.

Bast fibres such as hemp, flax, jute and kenaf need to be separated from their barks. In this process, fibrous and non-fibrous materials are separated using different processes such as retting and scutching. These process stages have significant effects on the chemical composition, cellulose structure (Mayer-Laigle et al., 2020) and overall fibre quality. Moreover, the chemical composition is influenced by the weather, growing conditions and processing techniques used, potentially generating variability in plant fibres properties. Consequently, the low maturity of plant cell walls, which manifest with large lumen sizes, as well as defects, such as kink bands, potentially initiated by aggressive extraction stages, are responsible for increase in water sensitivity of plant fibres. Despite several attractive attributes, some of the drawbacks of these fibres include high moisture uptake and moisture gain potential leading to weak fibre-matrix or fiber layer-layer interface and lower mechanical properties compared to their synthetic counterparts such as carbon and glass fibres. Thus, defects on the fibre surfaces, as depicted in Figure 5.a or 5.b, can easily be introduced during process stages hence will significantly influence the overall mechanical properties, as well as act as hot spots for moisture absorption (Le Duc et al., 2011; Placet et al., 2014).

Manufacturing process has a major role to play in moisture absorption behaviour. Void content in composite materials largely depend on the manufacturing methods employed. In addition to their complex structures, the void contents of natural fibre reinforced composites for example are influenced by manufacturing methods and the processing parameters used. For example, if hand lay-up, a commonly used lower-cost composites manufacturing technique, is used to fabricate the composite laminates, there are inherently higher void contents. Moreover, this technique cannot accommodate high fibre volume fraction. Whereas, if resin transfer moulding (RTM) techniques are used, then there will be less void content. Manufacturing process also

737 influences the wettability of reinforcements. When there is not enough resin to wet the fibres,
738 there is chance of having high void content. Therefore, composites manufactured by the hand
739 lay-up method absorb more moisture than parts produced from the RTM method. It is well
740 accepted that high void content will promote higher moisture ingress due to higher diffusion
741 coefficients (Celino et al., 2014; Chilali et al., 2017). Additionally, if composites for example are
742 manufactured using the compression moulding process, then the processing parameters such
743 as compaction pressure and temperature will significantly influence void content. If the applied
744 compaction pressure is high, voids may increase as the individual fibre can be pressed and
745 elongated and defects can be created, and in such a situation overall mechanical properties
746 decrease and moisture ingress can increase.

747 Advanced manufacturing techniques such as autoclave and out-of-autoclave (OoA) play an
748 important role in void formation and represent promising routes for plant fibre composites
749 development. In autoclave manufacturing, voids for example can be brought to an acceptable
750 level. It is suggested that without application of vacuum, by increasing cure temperature, voids
751 can be reduced. However, by increasing cure pressure, voids can form. There are various
752 factors that influence void formation in autoclave processes, including surface roughness,
753 humidity and ply orientation (Mehdikhani et al., 2019). Alternatively, OoA process such as
754 vacuum bag only (VBO) and vacuum assisted resin transfer moulding (VARTM), have been
755 used in the last few decades due to expensive and high energy requirements of the autoclave
756 process. Low capital investment, lower consumable costs, and improved energy efficiency are
757 considered attractive attributes of OoA curing process. Literature suggests that OoA production
758 of large parts has void contents of less than 2% (Mehdikhani et al., 2019) but (Grunenfelder and
759 Nutt, 2010) concludes that the autoclave process can suppress void formation while for the OoA
760 process, void volume fractions (void growth) increased exponentially as a function of moisture
761 and humidity. It is true that void content and moisture content are an interlinked phenomenon.
762 Consequently, the use of these moulding processes can be suitable for plant fibre composite
763 materials processing, provided that the moisture content of the plant cell walls is controlled
764 beforehand.

765 Indeed, plant fibre reinforced polymer composites are affected by humidity. During their service
766 life, plant fibre composites may be exposed to extreme weather conditions. Such conditions
767 could affect the structure of composites by disrupting the bonds between the fibres and the
768 matrix, which in turn can reduce mechanical properties.

769 Here, it is worth noting that fibre volume fraction influences the moisture uptake percentages
770 and diffusion coefficient (rate of moisture uptake). It is well-accepted that void content and
771 service conditions such as humidity and extreme service conditions affect the moisture
772 absorption behaviour of natural fibre composites. Similarly, moisture contents in the fibre
773 (hydrophilic) promotes void contents. Additionally, fibre aspect ratio, thickness of composites
774 and fibre orientation equally play an important role in moisture absorption of plant fibre
775 composites (Gager et al., 2019; Réquilé et al., 2018). Void formation can be attributed to both
776 moisture contents of the matrix and the reinforcements. Void fraction can increase significantly
777 with increased moisture/humid environments during manufacturing. It is well established that
778 voids in composites can severely degrade the mechanical properties, especially in the case of
779 natural fibre composites. These are the key concerns for these reinforcements to be used fully
780 in structural composites as reinforcements.

781 Study of water absorption and its influence on various properties is of great interest. Plant fibre
782 reinforced composites especially have a natural tendency to absorb moisture in extreme
783 hygroscopic conditions. Extensive studies have been carried out by (Akil et al., 2009;
784 Almansour et al., 2017; Dhakal et al., 2007; Errajhi et al., 2005; Hu et al., 2010) on the influence
785 of water on the various mechanical properties of different composite materials. The reported
786 work by (Dhakal et al., 2007) for example on the effects of room and elevated temperature on
787 hemp fibre reinforced unsaturated polyester suggests that the temperature has a significant
788 effect on the tensile and flexural properties. Their findings highlighted that the moisture
789 absorption was directly proportional to fibre volume fraction. Their work further concluded that
790 the water absorption patterns of hemp/UP composites were illustrating Fickian behaviour at
791 room temperature and displaying non-Fickian behaviours at elevated temperature.

792 The work carried out by (Hu et al., 2010) explored the effect of moisture absorption on short
793 jute fibre reinforced PLA composites manufactured using a hydrothermal setup. In that study,
794 they reported the changes at a microstructure level due to moisture ingress. They also linked
795 the changes in mechanical properties due to weak fibre matrix interface due to moisture
796 absorption. Similarly, the work undertaken by (Almansour et al., 2017) reported the moisture
797 absorption behaviour of flax fibre reinforced vinyl ester composites and basalt fibre hybridised
798 composites. Their work highlighted that basalt fibre hybridisation improved the water repellence
799 of flax/vinyl ester composites significantly.

800

801 **Figure 8**

802

803 The diffusion mechanisms and the effect of geometric dimensions and the fibre orientation are
804 further illustrated in Figure 8 (Chilali et al., 2017). It can be seen that the degradation of
805 interfacial adhesion due to moisture absorption is significant which will significantly reduce the
806 mechanical properties. This reduction in strength further continues due to the swelling and
807 debonding of the fibres (Dhakal et al., 2013).

808 From the above discussions and review, one can arrive upon three key factors through which
809 water causes changes and degradation to the fibre and composites structure and a significant
810 reduction in overall mechanical performance. The factors are as follows:

811 • Water molecules diffuse through the microscopic gaps (micro-pores) between
812 polymer chains in the fibre and the composites.
813 • The flaws (void and defects) and gaps pave the way for capillary transport
814 between the fibres and the matrix.
815 • Swelling of the reinforced fibres, especially plant fibres, causes expansion of
816 the micro-cracks in the matrix leading to debonding.

817 Additionally, moisture diffusion into the plant fibre composite involves displacement of water
818 molecules from a region of high concentration to a region of lower concentration (Assarar et al.,

819 2011). This diffusion is further exacerbated through poor wetting of the fibre and surface defects
820 leading to mechanical fatigue (Wang et al., 2006).

821 Considering the above-mentioned three key factors, moisture absorption related behaviours
822 could be assigned into one of the following categories:

823 • Linear Fickian behaviour: after an initial increase due to weight gain resulting
824 from moisture absorption, gradual equilibrium is achieved.
825 • Non-Fickian behaviour: after an initial increase due to weight gain resulting from
826 moisture absorption, no equilibrium is achieved.
827 • Two-stage sorption: In this scenario, both Fickian and non-Fickian behaviours
828 are observed which includes both the linear and plateau regions.
829

830

831 **5. Concluding discussion**

832 Through this review, we were able to highlight various major points related to the use and
833 transformation of plant fibres as composites reinforcements. We have studied the impact of
834 transformation processes on the mechanical performance, water absorption and also micro-
835 scale morphology of plant fibre reinforcements. We have mainly focused on extrusion and
836 injection processes involving thermoplastics. These allow, given the nature of the short fibre
837 reinforcements used, a study of the morphology of the fibres. In addition, their thermoplastic
838 nature also makes it possible to carry out recyclability studies, the latter being an essential
839 argument in the development of future plant fibre composite materials. It is not possible to
840 design optimally performing bio-based composite materials without knowledge and
841 understanding of the constituent components (vis. Fibres and matrix), and especially the
842 changes in fibre properties through composite processing steps given the temperature, stress,
843 moisture and time sensitive nature of plant fibres.

844 Given the thermal sensitivity of plant fibres, the choice of matrix is essential. The thermoplastic
845 family offers a wide range of materials with shaping temperatures suitable for plant fibres,

whether or not the polymers are compostable. Process time is also a major factor; from a morphological or mechanical point of view, it plays a key role in the evolution of fibre properties. A temperature indication is only useful if the exposure time is specified. A lower temperature but with a significantly longer exposure time period is often synonymous with damage to plant walls that is at least as severe as in the case of a higher temperature over shorter time period. This thermal exposure time can be modulated by paying attention to the choice of the implementation process. There are processes by induction, fibre placement or electric field that can significantly reduce the thermal exposure times of the fibres. However, they are often expensive and not always compatible with production constraints, and may require a consolidation stage. Another option to reduce exposure time is to choose low-viscosity polymers to facilitate fibre impregnation.

The latter differs according to the nature of the fibres considered; thus, plant fibres are generally classified into gelatinous cell walls (flax, hemp, nettle) or lignified walls (wood, sisal, kenaf, jute). The nature of their parietal constituents differs and so does their behaviour on thermal exposure. Local hardness or stiffness tests generally show an increase in mechanical performances for lignified walls while mechanical performance decreases with thermal exposure for gelatinous fibres. This is explained in the literature by a cross-linking of the xylane and lignin compounds with a heating stage. It is therefore possible, depending on the temperatures considered, to select fibres that will have a more stable mechanical behaviour, or even improved performance with heating.

In addition to mechanical performance, the hygroscopic behaviour and morphology of the fibres are affected by the transformation processes. The nature of the processes and the pressures involved will have a direct impact on the quality of the parts, their porosity rate and therefore the accessibility of water to the fibres. For example, injection moulding, or autoclave moulding for thermosetting materials, will achieve a very low porosity rate and a much higher durability, the action of water being limited in this case. The porosity and quality of the interface between the fibres and the matrix are the main damage source when using materials under variable or severe environmental conditions. From a morphological point of view, plant fibres evolve considerably in length and diameter, especially when high shear injection or extrusion

875 processes are used. The decrease in length can quickly reach 90% for fibrous elements a few
876 mm long but given their assembly in bundles in the plant, their diameter also decreases which
877 allows their shape factor (L/D) to evolve. This is particularly interesting when cycles multiply,
878 compared to glass fibres for example. Thus, plant fibre composite materials generally exhibit
879 stable mechanical performance after recycling, due to the low evolution of the aspect ratio of
880 their reinforcements.

881 Changes in fibre properties observed at the microscopic scale are generally caused by
882 parameters that are difficult to control, such as stress during growth due to weather conditions.
883 However, they have an impact on a macroscopic scale for composite materials; understanding
884 the origin of the properties is thus a valuable aid to avoid damage during the industrial
885 processing of plant fibres. Being the main cause of damage in plant fibres, even before their
886 use, control of the transformation processes into plant fibre composites is a major challenge for
887 their growing industrial development. It is necessary to respect the fibres from the moment the
888 parts are manufactured. The availability of a range of processes and reinforcements allows
889 relevant choices to be made and the best selection of exposure and processing times,
890 temperatures, pressures and shear rates, as well as the nature of the reinforcements, to obtain
891 the material best suited to the specifications. Taking these factors into account is imperative to
892 take full advantage of the extraordinary performance of these natural materials.

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895 **Aknowledgements**

896 The authors thank the INTERREG IV Cross Channel programme for funding this work through
897 the FLOWER project. Justin Mérotte (Coriolis Composites) is also thanked for fruitful
898 discussions on the AFP process.

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Figure Caption

Figure 1. Range of applied pressures and processing times conventionally used for various bio-based composites processing techniques.

Figure 2. Geometric and tensile mechanical properties of various plant fibres. (a) length-diameter ranges of natural fibres is presented, with lines of different slopes representing different fibre aspect ratio (l/d). (b) strength-stiffness ranges of plant fibres is presented, with generic grouping based on fibre origins. Plots illustrated with data from (Bourmaud et al., 2018; Leuwin, 2007; Shah, 2013).

Figure 3. Schematic drawing of an elementary flax fibre; PCW = primary cell wall; SCW = secondary cell wall with three layers: S1, S2 and S3; TZ = Transition zone between S1-S2 and S2-S3 (a). (b) gives details on the transition zones and of the number of structural layers for each cell wall layer (Baley et al., 2018; Bledzki and Gassan, 1999).

Figure 4. A hemp bundle imaged undergoing the decohesion damage mechanism (the 'calamar' shape). Imaged under UV excitation.

Figure 5. Plant fibre-breakage mechanisms during a process cycle. In a) at the beginning of the process cycle, one can observe both bundle decohesion and fragmentation due to transverse break (see arrows decohesion and broken). In b) evidence of several types of damages highlighted by stars. c) evidences surface flaws and d) is a view of an end of an over processing which lead to an enrichment of fine elements.

Figure 6. Evolution in the length, diameter, aspect ratio, indentation modulus and hardness of the fibres as well as the fine particles content with the number of recycling cycles. Example of a PLLA-PBS-flax fibre injected composite (Bourmaud et al., 2016). Axis shows the relative evolution of fibre properties (in percent) compared to raw fibre ones.

Figure 7. Illustration of differences in cell wall composition with the examples of wood and flax and impact on mechanical behaviour after seven injection moulding cycles. Inspired from (Bourmaud et al., 2016; Mikshina et al., 2013; Rihouey et al., 2017; Soccalingame et al., 2015; Yin et al., 2011)

Figure 8. The diffusion mechanisms are illustrated: (a) micro-cracks present in resin; (b) water molecules reaching in the fibre-matrix interface, and (c) filling the hollow part of the flax fibre lumen (Chilali et al 2017). Diffusion phenomenon occurs also through the direction of fibres; (d) water molecules ingress by capillarity through the micro-cracks present at the fibre-matrix interface and through lumen; (e) micro-cracks present in resin and at the fibre matrix interface; (f) fibre swelling and matrix radial cracking (Chilali et al., 2017).

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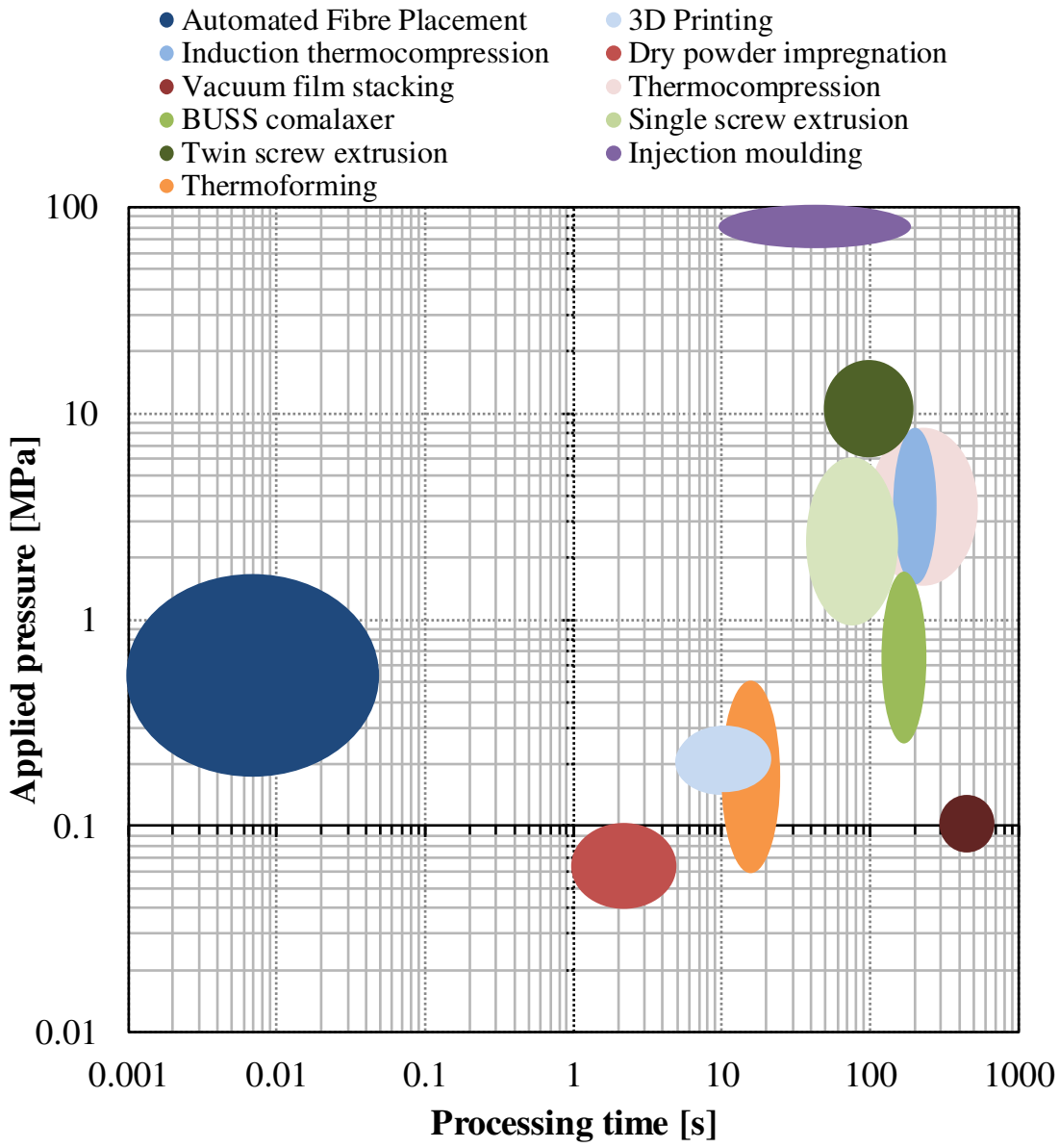

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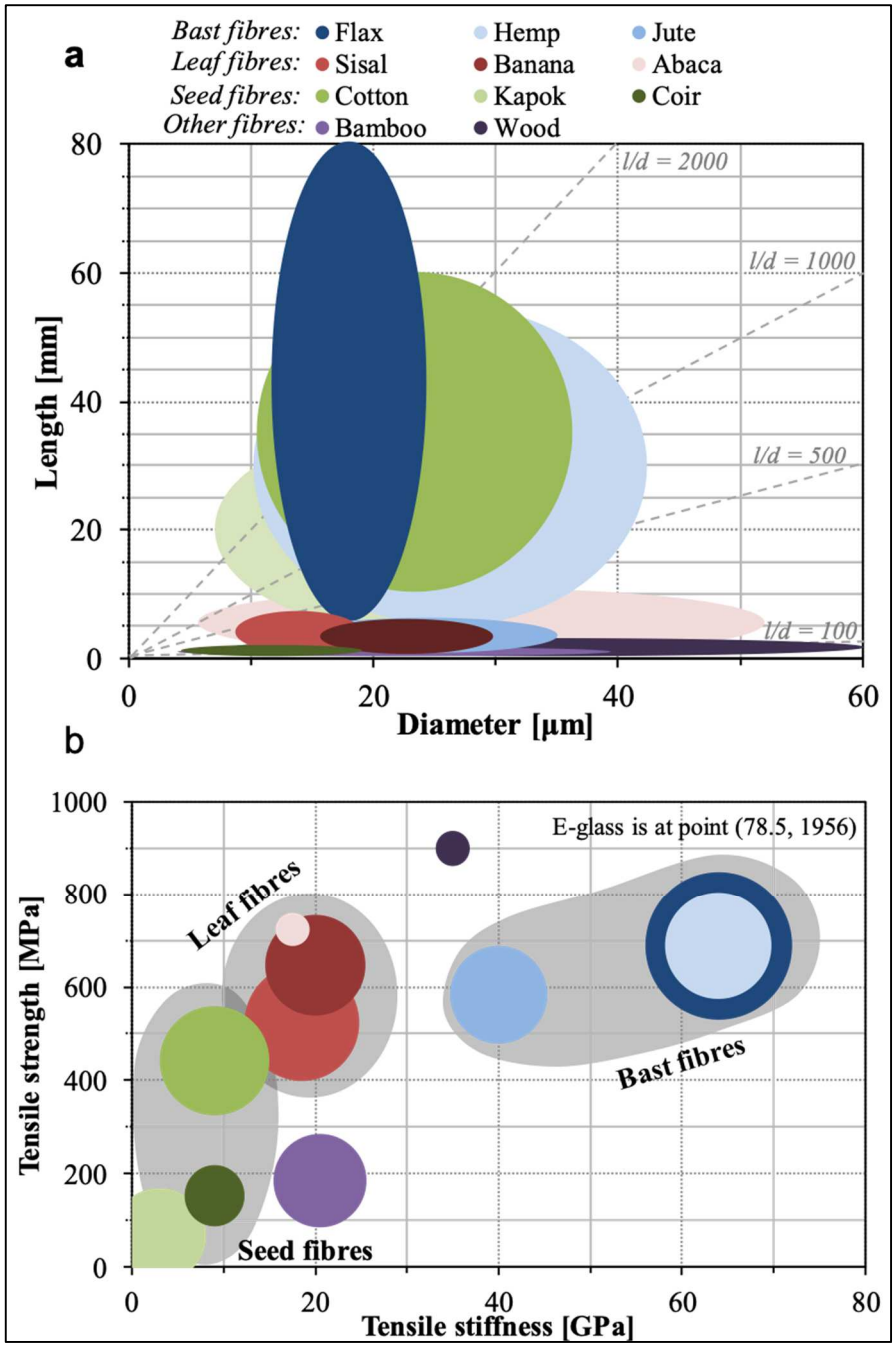

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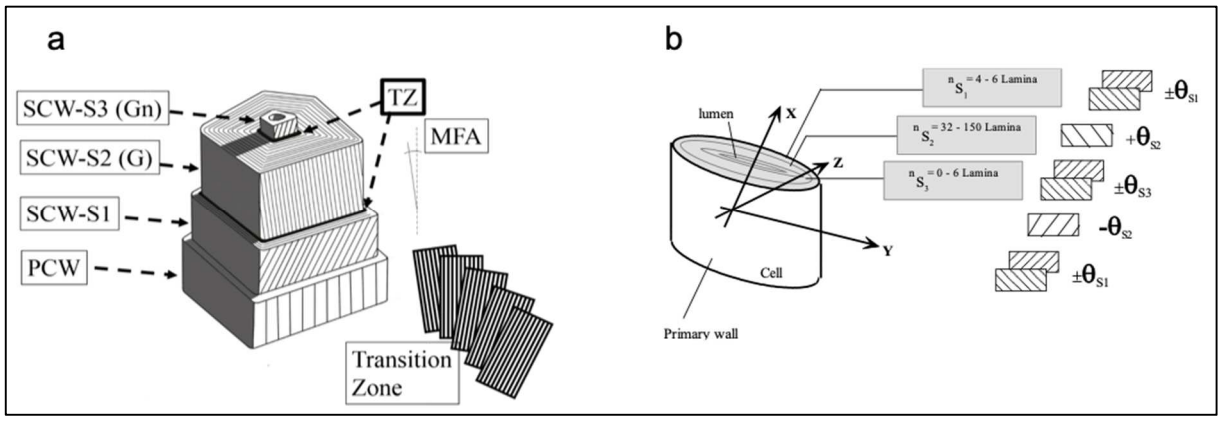

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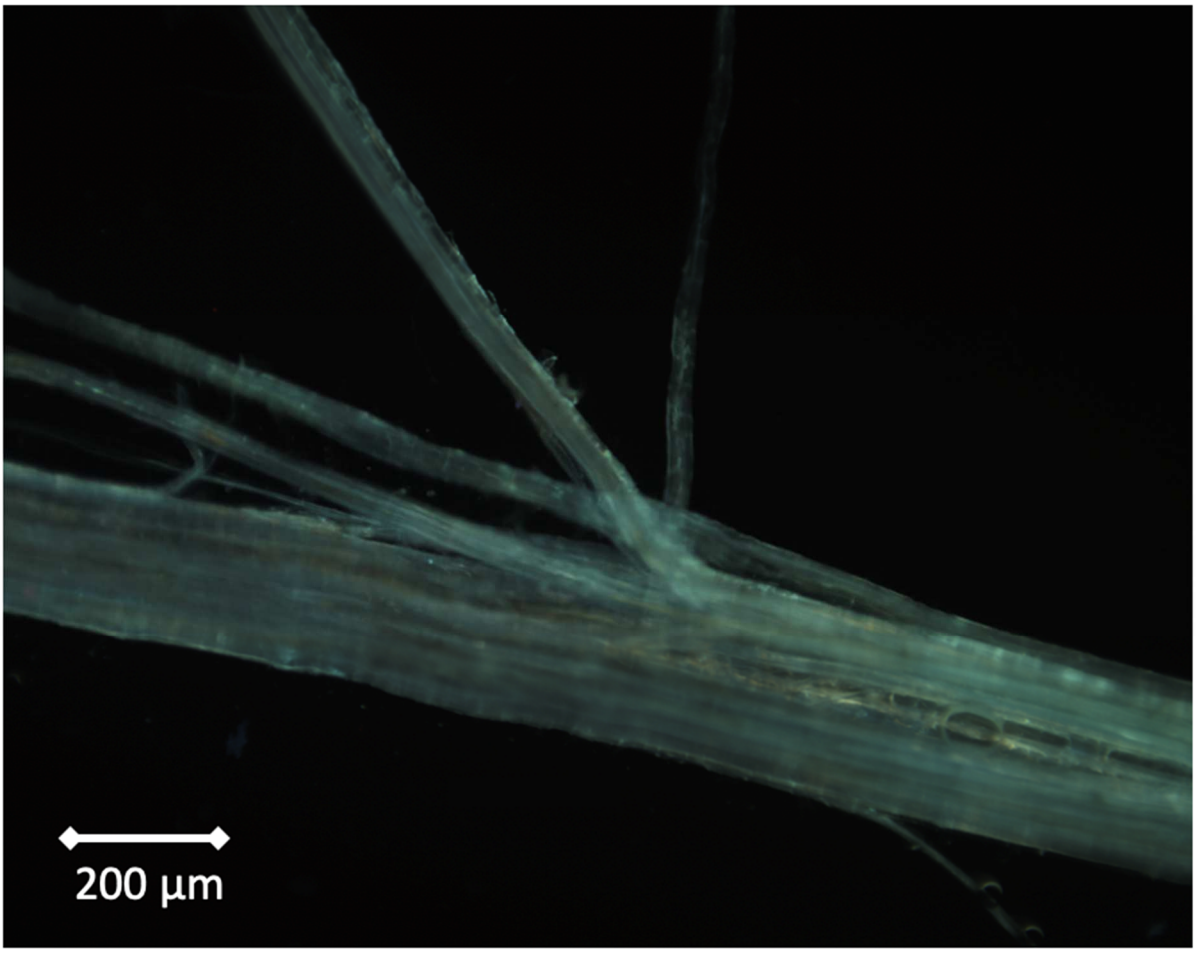

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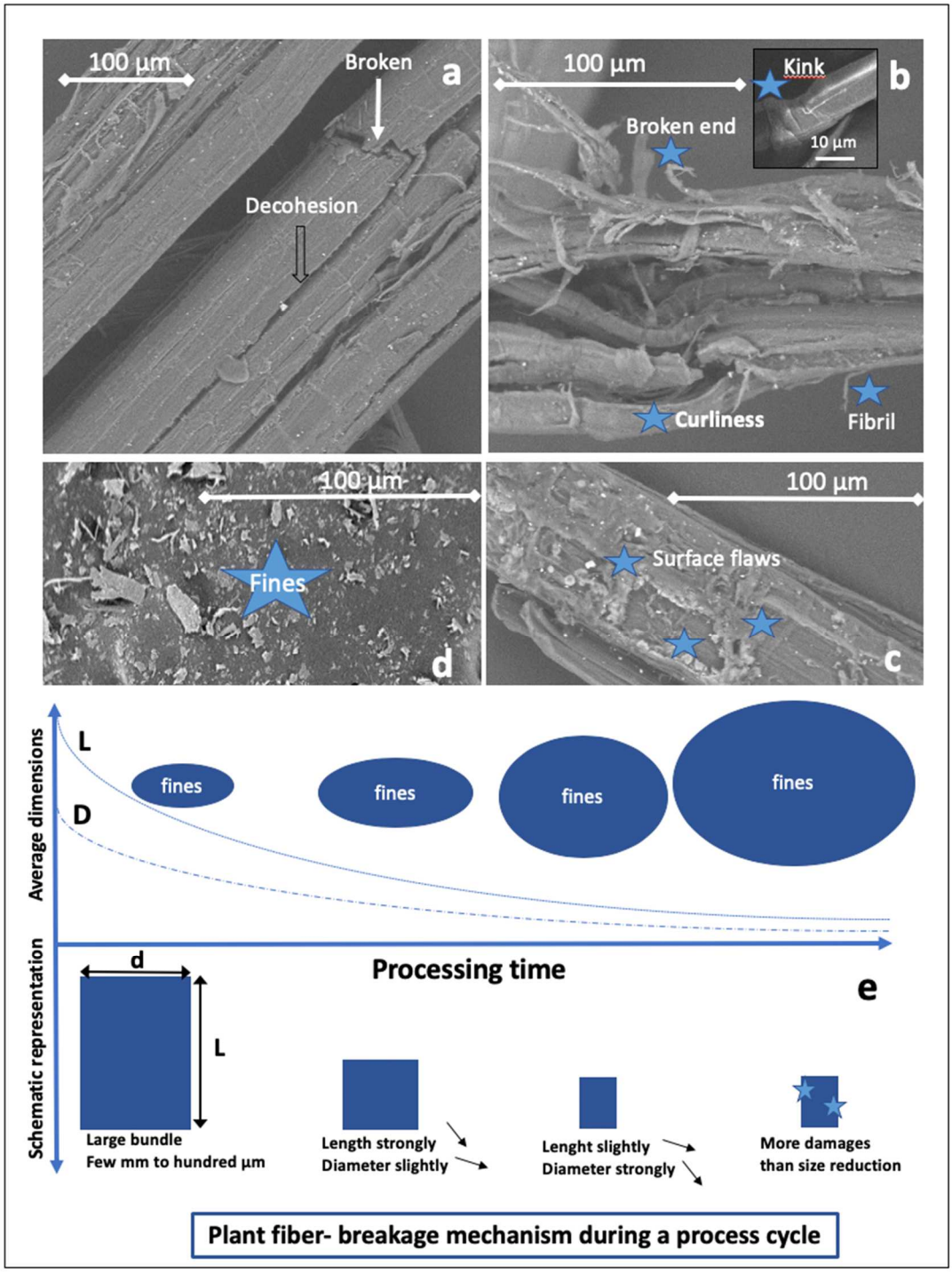

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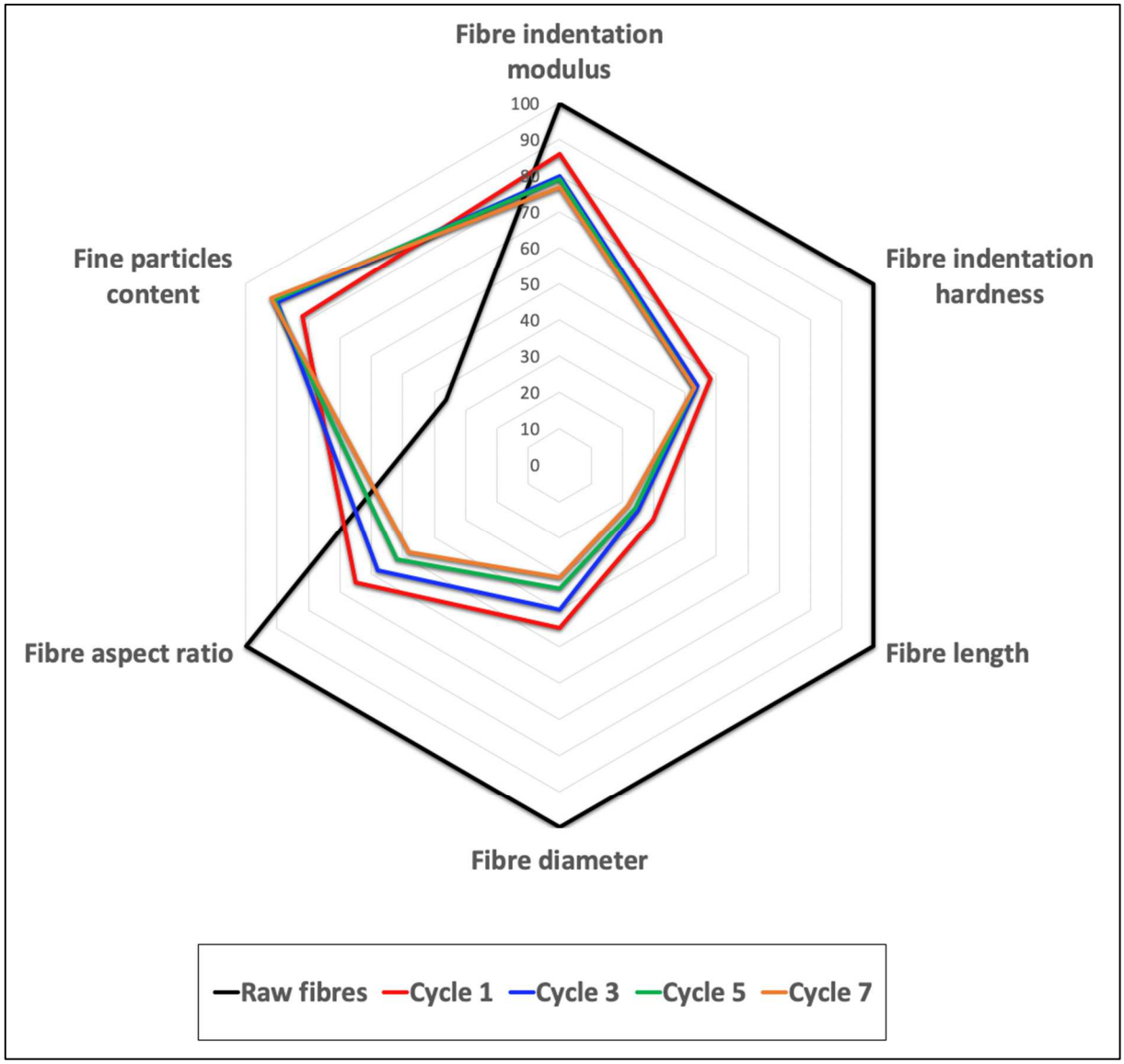

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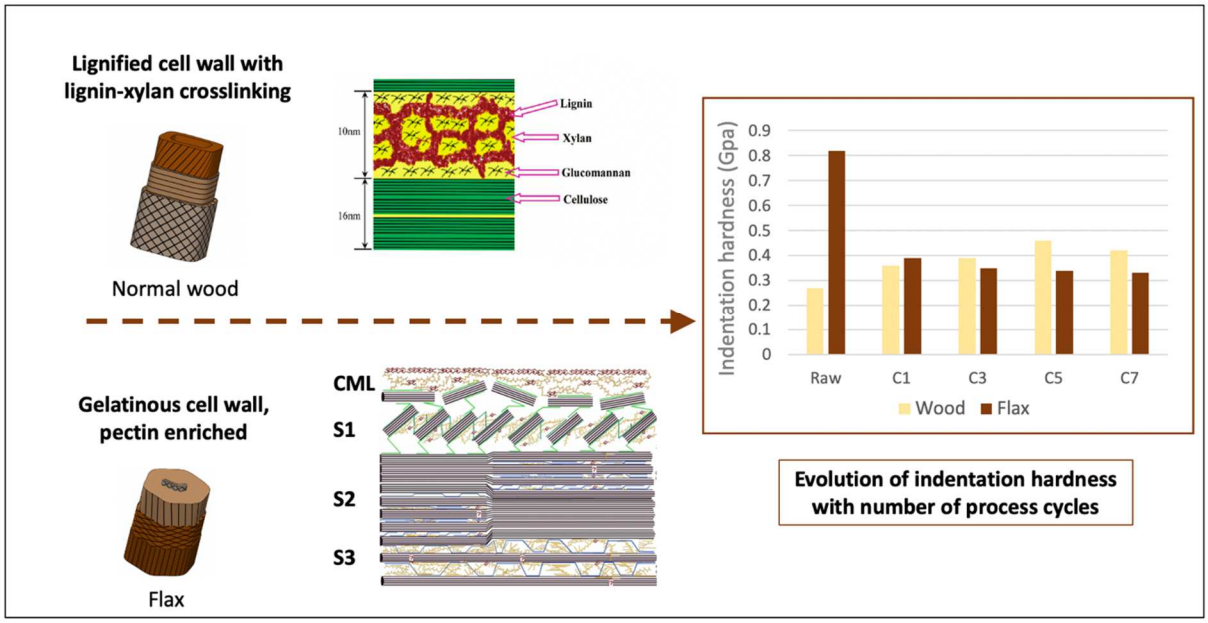

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