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# Developing plant fibre composites for structural applications by optimising composite parameters: a critical review

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# Abstract

Plant fibres, perceived as environmentally sustainable substitutes to E-glass, are increasingly being employed as reinforcements in polymer matrix composites. However, despite the promising technical properties of cellulose-based fibres and the historic use of plant fibre reinforced plastics (PFRPs) in load-bearing components, the industrial uptake of PFRPs in structural applications has been limited. Through an up-to-date critical review of literature, this manuscript presents an overview on key aspects that need consideration when developing PFRPs for structural applications, including the selection of *I*) the fibre type, fibre extraction process, and fibre surface modification technique, *II*) fibre volume fraction, *III*) reinforcement geometry and interfacial properties, *IV*) reinforcement packing arrangement and orientation, and *V*) matrix type and composite manufacturing technique. A comprehensive materials selection chart (Ashby plot) is also produced to facilitate the design of a PFRP component, based on (absolute and specific) tensile properties.

Keywords: Polymer-matrix composites; Natural fibres; Structural composites; Mechanical properties; Composite manufacture; Ashby plot

## 1 Introduction

While the total global production of fibre reinforced plastics (FRPs) amounted to 5.9 million tonnes in 1999 [1], this figure increased to 8.7 million tonnes in 2011 [1]. With the increasing consumption of FRPs, environmental concerns relating not only to the energy-intensive unsustainable production processes of the reinforcing synthetic fibres and plastics [2, 3], but also to the limited recyclability and end-of-life disposal options of the FRPs have been highlighted [4, 5]. The perceived scale of the problem has even led to stringent government legislations, such as the EU Directive on Landfill of Waste (Directive 99/31/EC) and the End-of-life Vehicle Directive (Directive 2000/53/EC), which are seen as barriers to the development or even continued use of FRPs in some markets [4]. To alleviate some of the environmental issues associated with using synthetics in FRPs, there has been a resurgent interest in biocomposites. Materials from renewable resources are being developed to replace not only the reinforcing fibres but also the polymer matrix of composites [6-13]. This review article is concerned with the development of plant fibres as reinforcements for FRPs. While it is acknowledged that the use of synthetic thermoset matrices, for instance, will produce bio-based composites that are not biodegradable or strictly recyclable, the presented research will nonetheless play a valuable role in the future increasing use of eco-materials.

## 1.1 Survey of current applications of plant fibre composites

Composites originated as biomaterials employing plant fibres as reinforcements. References have been made to the use of linen and hemp textiles as reinforcements of ceramics as early as 6500 BC [11]. The Egyptians have also been known to use grass and straw as reinforcing fibres in mud and clay bricks for the building of walls over 3000 years ago [14]. While synthetic fibres, specifically E-glass, dominate today's FRP market [1], awareness of the scarcity of non-renewable resources and a demand for environmental sustainability have led to a renewed and ever-increasing interest in biocomposites. This is reflected by the increasing number of publications on biocomposites during recent years, including books [9, 11, 14-17] and review articles [6-8, 18-28].

Plant fibres, such as flax, sisal and bamboo, offer several economical, technical and ecological advantages over synthetic fibres in reinforcing polymer composites (Table 1) [29]. The wide availability, low cost, low density, high specific properties and eco-friendly image of plant fibres has portrayed them as prospective substitutes to traditional composite reinforcements, specifically E-glass [2, 18, 30-33]. As 87% of the 8.7 million tonne global FRP market is based on E-glass composites (GFRPs) [1], plant fibres and their composites have a great opportunity for market capture.

	Properties	<b>Plant Fibres</b> <sup>a</sup>	Glass Fibres <sup>b</sup>	Carbon Fibres <sup>c</sup>
ny	Annual global production of fibres [tonnes] <sup>d</sup>	31,000,000	4,000,000	55,000
ono	Distribution of fibres for FRPs in EU [tonnes] <sup>d</sup>	Moderate (~60,000)	Wide (600,000)	Low (15,000)
Ec	Cost of raw fibre [£/kg]	Low (~0.5-1.5)	Low (~1.3-20.0)	High (>12.0)
	Density [gcm <sup>-3</sup> ]	Low (~1.35-1.55)	High (2.50-2.70)	Low (1.70-2.20)
	Tensile stiffness [GPa]	Moderate (~30-80)	Moderate (70-85)	High (150-500)
cal	Tensile strength [GPa]	Low (~0.4-1.5)	Moderate (2.0-3.7)	High (1.3-6.3)
chni	Tensile failure strain [%]	Low (~1.4-3.2)	High (2.5-5.3)	Low (0.3-2.2)
Te	Specific tensile stiffness [GPa/gcm <sup>-3</sup> ]	Moderate (~20-60)	Low (27-34)	High (68-290)
	Specific tensile strength [GPa/gcm <sup>-3</sup> ]	Moderate (~0.3-1.1)	Moderate (0.7-1.5)	High (0.6-3.7)
	Abrasive to machines	No	Yes	Yes
	Energy consumption/kg of raw fibre [MJ]	Low (4-15) <sup>e</sup>	Moderate (30-50)	High (>130)
cal	Renewable source	Yes	No	No <sup>f</sup>
igolo	Recyclable	Yes	Partly	Partly
Ecc	Biodegradable	Yes	No	No
	Hazardous/toxic (upon inhalation)	No	Yes	Yes

Table 1. Comparison between plant and synthetic fibres [29] (and references therein).

<sup>a</sup> Includes bast, leaf and seed fibres, but does not include wood and grass/reed fibres.

<sup>b</sup> Includes E- and S-glass fibres.

<sup>c</sup> Includes PAN- and pitch-based carbon fibres.

<sup>d</sup> Estimated values for the year 2010, from [34] for global fibre production values and from [32, 33, 35] for values on the distribution of fibres for FRPs in EU.

<sup>e</sup> While the energy required in the cultivation of plant fibres is low (4-15 MJ/kg), further processing steps (*e.g.* retting and spinning) can significantly increase the cumulative energy demand, for instance, to up to 146 MJ/kg for flax yarn [20]. This is discussed further in *Section 2.3.1 Plant growth and fibre extraction*.

<sup>f</sup> Carbon fibres based on cellulosic precursors currently account for only 1-2% of the total carbon fibre market [36].

Although the use of plant fibres (non-wood and non-cotton) in reinforced plastics has tripled to 45,000 tonnes over the last decade [11, 21, 33], plant fibre composites (PFRPs) make up only ~1.9% of the 2.4 million tonne EU FRP market (Fig. 1) [33]. Notably, the use of carbon fibre composites (CFRPs), globally and in the EU, is lower than the use of biocomposites and on the same level as the use of PFRPs (Fig. 1) [1, 33]. It is of interest to note that while PFRPs were developed and are viewed as alternatives to GFRPs [2, 18], they have mainly replaced wood fibre reinforced thermosets in the EU automotive industry [35, 37].



Fig. 1. PFRPs, primarily manufactured via compression moulding, account for only ~1.9% of the 2.4 million tonne EU FRP market in 2010 [33, 35].

By commercial application, over 95% of PFRPs produced in the EU are used for nonstructural automotive components, which are manufactured primarily via compression moulding [20, 33, 35]. Up to 30% of these PFRPs are based on thermoset matrices, while the rest are based on thermoplastics (Fig. 1) [35]. Other than automotive applications (for interior components such as door and instrumental panels) [11, 33, 38, 39], PFRPs are being considered for applications in:

i) construction and infrastructure (such as beams, roof panels, bridges) [10, 11, 27, 28, 39-44],

- *ii)* sports and leisure (for boat hulls, canoes, bicycle frames, tennis rackets) [10, 11, 28, 35, 39, 41],
- *iii)* furniture and consumer goods (such as packaging, cases, urns, chairs, tables, helmets, ironing boards) [10, 11, 28, 33, 35, 39, 41-44],
- iv) pipes and tanks (for water drainage/transportation) [7, 10, 11, 39, 40, 44, 45], and
- v) small-scale wind energy (as rotor blade materials) [46-51].

In many of these applications, plant fibres are being employed primarily as light, cheap and 'green' reinforcements, playing little or no structural role. Interestingly, this is different to what was envisaged in the mid-twentieth century, when the potential of plant fibres as structural reinforcing agents was acknowledged by pioneers like Ford to manufacture the first 'green car' with an all-plastic-body using 70 wt% lignocellulosic fibres [52]. Ford was even able to demonstrate the strength and impact resistance of the material by famously taking a sledgehammer onto the car's deck lid [52]. At the same time, Aero Research Ltd developed Gordon Aerolite, a flax/phenolic composite, to replace light-alloy sheets for building the structural members of Spitfire fuselages for British military aircrafts during the Second Great War [53]. With Britain facing potential shortages of aluminium, Gordon Aerolite was then the most promising material for aircraft [53]. Furthermore, the structural potential of plant fibres is revealed by the fact that bast fibres (like flax, hemp and jute) are high in cellulose content (~60-80% of the dry chemical composition [30]) and native cellulose has remarkable tensile stiffness (138 GPa) and strength (>2 GPa) [11, 54, 55]. Therefore, investigating and eventually promoting the potential use of plant fibres in load-bearing composite components, as a possible replacement to E-glass, is a natural step ahead.

There are several excellent review articles available in literature, including [6-8, 20, 56], which provide a wide-ranging critical survey on plant fibres and their composites. Specific topics, such as *i*) fibre-surface modification techniques [24, 57, 58], *ii*) plant fibre based hybrid composites [59], *iii*) modelling and predicting the mechanical properties of PFRPs [20, 60-62], *iv*) biocomposites based on specific fibres (*e.g.* jute, hemp, coir) [21-23, 63-66], and *v*) wholly bio-based composites (*i.e.* both fibres and matrix are from renewable sources) [6, 10, <u>19</u>, <u>67</u>, <u>68</u>], have also been the subject of

review articles. In contrast to these, the review presented here aims to address the key issue of *designing PFRPs specifically for structural applications*. While some researchers, for instance [25, 27, 40, 44], have presented overviews and critical assessments on the current status of PFRPs for structural applications, this timely article highlights some of the key requirements and developments in the utilisation of plant fibres for structural composites. This article also provides a comprehensive Ashby plot to facilitate the material design of a PFRP component.

## **1.2** Designing composite materials for structural applications

While structural composites are required to sustain external loads in addition to selfsupport (like the shear web of a wind turbine blade) or play a principal role in supporting the structure of the final component (like the airframe of an aircraft), non-structural composites are primarily for aesthetic purposes enduring minimal loads (like the interior panels of a car). Hence, the design and construction of structural and non-structural composites is different.

During product development, materials selection is a process where a range of material properties are taken into consideration. Asbhy [69] describes a method to compare the relative performance of a variety of materials for a specific constructive element by using material *performance indices* – defined by the component function, objective and constraint – as design criteria. Generally, minimising material weight (density  $\rho$ ) and/or cost are key objectives for industrial products. The key mechanical parameters, defined by the component function and constraint, are typically stiffness *E* and strength  $\sigma$ . Following Asbhy [69], the critical material performance indices that need to be maximised for a beam/plate loaded in pure tension are specific tensile stiffness  $E/\rho$  and specific tensile strength  $\sigma/\rho$ . For a beam/plate loaded in bending mode, specific flexural stiffness  $E^{1/3}/\rho$  and strength  $\sigma^{1/2}/\rho$  need to be maximised. Material selection, on the basis of these performance indices, is best achieved by plotting the performance indices (which are typically a mathematical combination of material properties) on each axis of a *materials selection chart*, also known as an *Ashby plot*. Individual materials or material sub-classes appear as balloons, which define the range of their properties.

The Asbhy plot in Fig. 2 compares the specific tensile performance of various natural fibres (from animals and plants) with synthetic fibres. It is observed that several plant fibres, including flax, hemp and jute, have better specific tensile stiffness than E-glass. On the other hand, specific tensile strength of plant fibres is consistently lower than that of synthetic fibres.



Fig. 2. Ashby plot comparing the position of natural fibres against synthetic fibres with respect to specific tensile properties (from [70]).

As the density of plant fibres (~1.30-1.55 gcm<sup>-3</sup>) is approximately half of E-glass (2.60 gcm<sup>-3</sup>), at the same fibre content PFRPs are significantly (30-40%) lighter than GFRPs [71, 72]. A lower density gives PFRPs a good chance to compete against GFRPs in terms of specific stiffness and strength. Nonetheless, to construct a comprehensive materials selection chart for PFRPs, the influence of various composite parameters on PFRP mechanical properties needs to be well-characterised and well-understood.

#### 1.2.1 Composite material parameters

FRPs are heterogeneous materials, consisting of reinforcing fibres embedded in a continuous matrix. While the fibres provide strength and stiffness to the composite, the matrix transmits externally applied loads, via shear stresses at the interface, to the

reinforcing fibres and protects the fibres from external damage. The advantage of coupling the two distinct constituents is that the high strength and stiffness of the fibres, which in practical situations would be difficult to realise, may be exploited.

One of the many advantages of composite materials, in general, is the possibility of tailoring material properties to meet different requirements. It is well-known that the macro-mechanical behaviour of FRPs depends on the following parameters: *I*) the fibre properties, *II*) the volumetric composition (where the sum of the volume fraction of the fibres  $v_f$ , matrix  $v_m$  and voids  $v_p$  is unity, *i.e.*  $v_f + v_m + v_p = 1$ ), *III*) the geometry of the fibres and the fibre/matrix interface properties, *IV*) the packing arrangement, orientation and stacking sequence of the fibre reinforcements, and *V*) the matrix properties. The effect of all these parameters is elegantly demonstrated by the fundamental equations in composites engineering: the generalised rule-of-mixtures (ROM) model for the tensile modulus  $E_c$  (Eq. 1) and strength  $\sigma_c$  (Eq. 2) of discontinuous fibre composites.

$$E_c = E_f v_f \eta_{lE} \eta_o + v_m E_m$$
 Eq. 1

$$\sigma_c = \sigma_f v_f \eta_{lS} \eta_o + v_m \sigma'_m \qquad \text{Eq. 2}$$

where, *I*)  $E_f$  and  $\sigma_f$  are the fibre modulus and fibre strength, *II*)  $v_f$  and  $v_m$  are the fibre and matrix volume fraction, *III*)  $\eta_{IE}$  and  $\eta_{IS}$  are the reinforcement length efficiency factors for stiffness and strength (incorporating the effect of fibre geometry and interfacial properties), *IV*)  $\eta_o$  is the reinforcement orientation distribution factor (incorporating the effect of packing arrangement and orientation of the fibre reinforcements), and *V*)  $E_m$  and  $\sigma'_m$  are the matrix modulus and matrix tensile stress at the fibre failure strain. Note that the order of the parameters of Eq. 1 and 2 is homologous to the order of the composite parameters defined previously.

The basic assumptions of the above micromechanical models include: *i*) all fibres have identical geometry and properties, *ii*) homogenous and uniform distribution of fibres in the matrix, *iii*) iso-strain conditions within the composite, *iv*) ideal fibre/matrix interface, *v*) elastic deformation of the fibre and matrix, *vi*) no transverse deformations (*i.e.* ignore Poisson's contractions), *vii*) zero and maximum tensile stress at the fibre ends and centre, respectively, and *viii*) no effect of porosity content  $v_p$  on composite properties (other than

reducing  $v_f$  and  $v_m$ ). Although many of these simplifications and assumptions do not hold true for FRPs in general, the ROM model has proved to be adequate for the prediction/estimation of the properties of synthetic fibre composites and for the determination of the reinforcing potential of the fibres (by 'back-calculation').

The simplicity of the generalised ROM model implies that it has become a widely used model for PFRPs as well. Interestingly, as plant fibres are inherently discontinuous, the ROM model can be used for PFRPs even if plant yarns/rovings (i.e. 'continuous' reinforcements) are employed. Nonetheless, as plant fibres require specific considerations, recent pioneering work has led to a modified ROM model that has been shown to be more suitable for PFRPs [60, 61]. The modified ROM model, presented in Eq. 3 and Eq. 4, includes i) a factor of  $(1 - v_p)^2$  to simulate the detrimental effect of porosity on the tensile properties of PFRPs [73-75], ii) a fibre diameter distribution factor  $\eta_d$  to incorporate the effect of approximately linear  $(E_f = E_{f0} - m \cdot d_f)$  decline in fibre tensile modulus with increasing fibre diameter  $d_f$  [20, 60, 61, 76, 77], *iii*) a fibre area correction factor  $\kappa$  to address the discrepancy between the true (non-circular, irregular and variable) cross-sectional area of the fibre and the apparent circular cross-sectional area calculated by the measurement of the apparent fibre diameter [20, 60, 61, 78], and *iv)* a modified reinforcement orientation distribution factor of  $\eta_o = \cos^2(2\alpha)$ , where  $\alpha$  is the surface twist angle of the reinforcing yarn in yarn reinforced PFRPs, to elucidate the detrimental effect of yarn twist on yarn reinforced PFRP tensile strength [60, 79]. While these modifications to the general ROM model have been validated with experimental results on PFRPs in the relevant studies, they have been validated only for limited data sets. Therefore, the applicability of the modified ROM model to PFRPs needs to be investigated further.

$$E_c = \left(E_f v_f \eta_{lE} \eta_o \eta_d \kappa + v_m E_m\right) (1 - v_p)^2$$
 Eq. 3

$$\sigma_c = (\sigma_f v_f \eta_{lS} \eta_o \kappa + v_m \sigma'_m)(1 - v_p)^2$$
 Eq. 4

In Eq. 1 and Eq. 3, the length efficiency factor for stiffness  $\eta_{lE}$  can be estimated by the Cox's shear lag model (Eq. 5) [80], where  $l_f$  is the fibre length,  $d_f$  is the fibre diameter,  $G_m$  is the matrix shear stiffness, and  $v_{f,max,FRP}$  is the maximum achievable fibre volume

fraction (dependent on fibre packing geometry; e.g.  $v_{f,max,FRP} = \pi/4$  for square-packing arrangement). In Eq. 2 and Eq. 4, the length efficiency factor for strength  $\eta_{lS}$  is given by the Kelly-Tyson's model (Eq. 6) [81], where  $l_c$  is the critical or ineffective fibre length. Sub-critical length fibres ( $l_f < l_c$ ) will not carry the maximum load. If a composite has both sub-critical length ( $l_f < l_c$ ) and super-critical length ( $l_f > l_c$ ) fibres, Eq. 6 can be expressed as a summation of the contribution from different fibre lengths. It is useful to note that the critical fibre length is a function of the fibre tensile strength  $\sigma_{f}$ , the fibre diameter  $d_f$ , and the interfacial shear strength  $\tau$  (Eq. 6). The length efficiency factors for stiffness and strength range between 0 (for  $l_f \ll d_f$  or  $l_f \ll l_c$ ) and 1 (for  $l_f \gg d_f$  or  $l_f \gg$  $l_c$ ). This is graphically demonstrated in Fig. 3 using typical values for PFRPs. It can be inferred from the graphs that as  $l_f \rightarrow 0.5 \text{ mm} (l_f/d_f \rightarrow 25)$ , the length efficiency factors increase rapidly towards a value of about 0.80. Thereafter, the length efficiency factors asymptotically approach unity as fibre length (or fibre aspect ratio) increases. Fig. 3b also demonstrates the effect of interfacial properties on the length efficiency factor for strength  $\eta_{IS}$ ; an increase in the fibre/matrix interfacial shear strength  $\tau$  (and a subsequent decrease in the critical fibre length  $l_c$ ) has a noticeable effect on  $\eta_{lS}$  for short fibres ( $l_f < 3$ mm), but a negligible effect on  $\eta_{lS}$  if the fibre length is over 10 times the critical length (*i.e.*  $l_f = 10l_c$ ) [82]. These observations are critical to selecting the reinforcement form.



Fig. 3. Predictions of the fibre length efficiency factors for a) stiffness  $\eta_{lE}$  and b) strength  $\eta_{lS}$ , based on Cox's shear lag model (Eq. 5) and Kelly-Tyson's model (Eq. 6), respectively. Typical values for flax reinforced PFRPs are used in the calculations:  $d_f = 20 \ \mu m$ ,  $G_m = 1 \ GPa$ ,  $E_f = 50 \ GPa$ ,  $v_{f,max,FRP} = \pi/4$ ,  $v_f = 0.30$ ,  $\sigma_f = 1000 \ MPa$  and  $\tau = 30 \ MPa$  or 15 MPa ( $l_c = 0.333 \ mm$  or 0.667 mm, respectively).

$$\eta_{lE} = 1 - \frac{\tanh\left(\frac{\beta l_f}{2}\right)}{\frac{\beta l_f}{2}}, \quad \frac{\beta l_f}{2} = \frac{l_f}{d_f} \sqrt{\frac{2G_m}{E_f \ln\left(\sqrt{v_{f,\max,FRP}} / v_f\right)}}, \quad \eta_{lE} \in [0,1]$$
 Eq. 5

$$\eta_{lS} = \begin{cases} 1 - l_c / 2l_f & \text{for } l_f \ge l_c \\ l_f / 2l_c & \text{for } l_f \le l_c \end{cases}, \ l_c = \frac{\sigma_f d_f}{2\tau}, \ \eta_{lS} \in [0,1] \end{cases}$$
Eq. 6

The reinforcement orientation distribution factor  $\eta_o$  in Eq. 1-4 can be estimated by the Krenchel orientation distribution factor (Eq. 7) [83], where  $a_n$  is the fraction of fibre with orientation angle  $\theta_n$  with respect to the axis of loading. The reinforcement orientation distribution factor ranges between 0 (fibres aligned transverse to the stress direction) and 1 (fibres aligned parallel to the stress direction).

$$\eta_o = \sum_n a_n \cos^4 \theta_n, \ \sum_n a_n = 1, \ \eta_o \in [0,1]$$
 Eq. 7

Although the fibre diameter distribution factor  $\eta_d$  in Eq. 3 has not been formally defined [20, 77], it may be a complex function of the fibre structure [77] or be correlated to the probability density function of the fibre diameter [20, 76].  $\eta_d$  ranges between 0 and 1.

Coming to estimating the fibre area correction factor  $\kappa$ , while it is well known that the cross-section of plant fibres is variable, irregular and non-circular, only recently have researchers quantitatively estimated the deviation of the fibre cross-section shape from circularity [61, 84, 85]. The studies suggest that calculating the cross-section area  $A_C$ , assuming a circular cross-section with an average fibre diameter  $d_f$ , overestimates the true cross-section area  $A_T$  by a fibre area correction factor  $\kappa$  ( $A_C/A_T$ ) of 1.42–2.55 [84-86]. Virk *et al.* [61] have shown that a fibre area correction factor of  $\kappa = 1.42$  for jute fibres offers a better prediction for the composite mechanical properties (than assuming circular fibre cross-section, *i.e.*  $\kappa = 1$ ).

Other than the fibre area correction factor  $\kappa$ , which is used to account for fibre area measurement discrepancies, all parameters in Eq. 1-4 can be maximised to achieve improvements in the mechanical properties of PFRPs, and FRPs in general. If  $\eta_{IE}$ ,  $\eta_{IS}$ ,  $\eta_o$ and  $\eta_d$  are taken to be unity, the generalised and modified ROM models (in Eq. 1-4) are equivalent to the Voigt 'upper bound' for continuous fibre composites.

# 2 Plant fibres as structural reinforcements

This section focuses on selecting plant fibre reinforcements for structural composites applications.

# 2.1 Plant fibre type

There are five basic types of plant fibres, classified as follows: *i*) bast fibres, from the inner bark of the plant stems, *iii*) leaf fibres, *iv*) seed fibres, *v*) grass and reed fibres, and *vi*) all other fibres (including wood fibres). Examples of the different fibre types and their estimated annual global production values are shown in Table 2.

Table 2. Classification of plant fibres. If data was available from the FAO database [34], the global production estimates  $(10^3 \text{ tonnes})$  for 2010 are given in brackets. The bottommost row gives the estimated total global production quantity  $(10^3 \text{ tonnes}; \text{ from } [11, 34])$  for each category.

Bast	Leaf	Seed				Grass/ Reed	Other	
		Fibres	Pod	Husk	Fruit	Hulls		
Flax	Sisal	Cotton					Wheat	Wood
(622)	(361)	(23295)						
Jute	Pineapple		Kapok				Corn	Roots
(3056)			(99)					
Hemp	Agave			Coir			Rice	
(214)	(34)			(1058)				
Kenaf	Banana				Oil		Bamboo	
(500)					palm		(30000)	
Ramie	Abaca					Rice	Bagasse	
(118)	(95)						(75000)	
(5000)	(600)			(25000)			(>100000)	(>100000)

In terms of utilization, plant fibres can be classed as being from primary or secondary plants. Primary plants (like flax, sisal, cotton, bamboo, hardwood/softwood trees) are cultivated specifically for their fibre content, while fibres from secondary plants (like pineapple leaf, coir, oil palm (empty fruit bunch), bagasse, rice straw) are a by-product from some other primary utilization. Hence, although plant straws and stalks (secondary source) are a potentially larger source of fibre than even wood fibres (primary source) (Table 2), the former are predominantly used as livestock feed or bio-fuel [87-89].

Other than wood fibres (including flour and pulp), commercially useful fibres come mainly from the bast, leaf, and seed coverings of specific plants, whose principal application lies in textiles. Notably, while the total global production of wood fibres and cotton exceeded 100 million tonnes and 20 million tonnes in 2010 (Table 2), respectively, the total global production of all bast, leaf and other seed fibres amounted to only  $\sim$ 5 million tonnes in the same year [30, 34]. Therefore, the significant consumption of wood and cotton fibres in FRP manufacture is not surprising. In fact, biocomposites accounted for  $\sim$ 13% of the 2.4 million tonne EU FRP market in 2010 (Fig. 1), of which 170,000 tonnes is attributable to wood fibre composites and 100,000 tonnes is attributable to cotton fibres [33, 35]. Only 45,000 tonnes of the biocomposites manufactured employed non-wood, non-cotton fibres [33, 35], primarily flax (64% of the market share), jute (11%), hemp (10%) and sisal (7%) [25].

It should be noted, however, that wood and cotton fibres are used as 'fillers' in the plastics, with no reinforcing role, due to the short length (*i.e.* low aspect ratio) of the fibres (or particles, in the case of wood flour) [11, 90]. Nonetheless, the use of wood and cotton fibres for non-structural PFRPs has been attractive due to, *i*) the abundance of these low-cost fibres, *ii*) the weight savings that the resulting PFRPs provide, and *iii*) the improved green credentials of the material due to lower polymer use [90]. The latter is attributable to the fact that raw plant fibre production requires <10-20% of the energy used in the production of the polymer matrix (*e.g.* 15 MJ/kg for hemp and 70-90 MJ/kg for polypropylene) [20, 33]. In fact, noting the regional availability of certain fibre types, there are an increasing number of studies which demonstrate that for such non-structural applications even fibres from secondary sources with poor mechanical properties (due to a lack of biological and evolutionary incentive) like rice straw [91, 92], coir [65], banana leaf [93], oil palm (empty fruit bunch) [66], and pineapple leaf [64], may be suitable.

Table 3 presents the physio-mechanical properties of different plant fibres. With some exceptions, it is observed that the tensile properties (absolute and specific) are in the following order: bast fibres > leaf fibres > seed fibres. In fact, only bast fibres have tensile stiffness and specific tensile properties comparable to E-glass (Table 3 and Fig. 2). Notably, the tensile strength of even bast fibres is considerably lower than that of E-glass.

	Fibre	Density [gcm <sup>-3</sup> ]	Tensile modulus [GPa]	Specific tensile modulus [GPa/gcm <sup>-3</sup> ]	Tensile strength [MPa]	Specific tensile strength [MPa/gcm <sup>-3</sup> ]	Failure strain [%]	Source
	Flax	1.45-1.55	28-100	19-65	343-1035	237-668	2.7-3.2	[56]
Bast	Hemp	1.45-1.55	32-60	22-39	310-900	214-581	1.3-2.1	[21]
	Jute	1.35-1.45	25-55	19-38	393-773	291-533	1.4-3.1	[56]
	Sisal	1.40-1.45	9-28	6-19	347-700	248-483	2.0-2.9	[23]
Leaf	Pineapple	1.44-1.56	6-42	4-27	170-727	118-466	0.8-1.6	[64, 93]
	Banana	1.30-1.35	8-32	6-24	503-790	387-585	3.0-10.0	[93]
	Cotton	1.50-1.60	5-13	3-8	287-597	191-373	6.0-8.0	[94]
Seed	Coir	1.10-1.20	4-6	3-5	131-175	119-146	15.0-30.0	[19]
•1	Oil palm	0.70-1.55	3-4	2-4	248	160-354	25.0	[8]
ner	Bamboo	0.60-1.10	11-30	18-27	140-230	210-233	1.3	[8]
Oth	Wood pulp <sup>a</sup>	1.30-1.50	40	26-31	1000	667-769	4.4	[19]
	E-glass	2.55	78.5	31	1956	767	2.5	[95]

Table 3. Comparison of the mechanical properties of various plant fibres and E-glass.Sources include those listed and [7, 30].

<sup>a</sup> Particulate form of softwood pulp (produced using Kraft separation method)

To observe the reinforcing effect of the different plant fibres in a composite, Table 4 presents typically reported mechanical properties of compression moulded polypropylene (PP) composites reinforced with randomly-oriented short-fibre mats. Expectedly, it is observed that PP reinforced with bast fibres exhibit significantly superior mechanical properties in comparison to leaf and seed fibre reinforced PP. In fact, the tensile properties (absolute and specific) of leaf, seed and wood fibre reinforced PP is barely comparable to unreinforced PP. On the other hand, bast fibre reinforcements not only improve the tensile properties of the matrix considerably, but the resulting composites can compete against even GFRPs in terms of (absolute and specific) tensile stiffness and strength. Therefore, if certain structural requirements need to be met, it is essential that bast fibres (or other selective fibres like sisal and bamboo) are used as reinforcements (not fillers) in FRPs. Perhaps, this is why composites reinforced with bast fibres are now replacing under-performing wood fibre composites and even GFRPs in automotive applications [35].

Table 4. Typically reported mechanical properties of compression moulded PP composites reinforced with various nonwoven (randomly-oriented short-fibre) plant fibre mats. For comparison, the mechanical properties of neat PP and chopped strand E-glass mat reinforced PP are also given.

	Fibre reinforcement	Fibre content <sup>a</sup> [wt%]	Tensile modulus [GPa]	Specific tensile modulus <sup>b</sup> [GPa/gcm <sup>-3</sup> ]	Tensile strength [MPa]	Specific tensile strength <sup>b</sup> [MPa/gcm <sup>-3</sup> ]	Source
	PP	0	0.7-1.7	1.1-1.9	19-35	21-39	[21]
<u>ц</u>	Flax	40	8.8	8.0	57	52	[96]
Bast	Hemp	40	6.9	6.3	52	47	[18]
	Jute	40	3.7	3.5	27	25	[18]
Leaf	Sisal	40	5.3	4.9	34	31	[18]
	Pineapple	20	0.6	0.6	32	32	[97]
	Banana	50	1.5	1.4	31	29	[98]
_	Cotton	30	1.9	1.8	27	26	[99]
Seed	Coir	40	1.2	1.2	10	10	[18]
•1	Oil palm	40	0.7	0.7	8	8	[100]
her	Bamboo	50	3.6	3.7	30	30	[101]
0f]	Wood fibre	35	1.4	1.3	21	19	[90]
_	E-glass	50	7.0	4.8	33	68	[102]
	E-glass	42	6.2	4.9	89	23	[18]

<sup>a</sup> Fibre content is approximate.

<sup>b</sup> Estimated values. Composite density  $\rho_c$  is estimated ( $\rho_c = (\rho_f \rho_m)/(\rho_f - w_f(\rho_f - \rho_m))$ ) assuming no porosity and using fibre weight fractions  $w_f$  from Table 4, fibre densities  $\rho_f$  in Table 3 and a density for PP of  $\rho_m = 0.91$  gcm<sup>-3</sup>.

## 2.2 Plant fibre structure

While it is clear from the previous sub-section that bast fibres have superior mechanical properties in comparison to leaf and seed fibres, understanding the reasons behind this may prove useful in developing structural PFRPs.

One approach is to consider the role of the fibre in the living plant [11]. Bast fibres (and some grass fibres like bamboo) provide rigidity and strength to the plant stems, so they would be ideal in stiffening/strengthening composites. Leaf fibres experience repetitive flexing from the wind, so they would be useful for toughening composites. As seed fibres have no structural role, they would not reinforce a plastic effectively.

A more fundamental and quantitative approach involves understanding the influence of the chemical and physical structure of plant fibres on their mechanical properties. Each elementary plant fibre is a single cell with an elongated thick cell wall surrounding a central luminal cavity (Fig. 4). While the cell wall is responsible for the structural integrity of the living plant, the luminal cavity facilitates transportation of nutrients. Although having a high aspect ratio, the cross-sectional shape and dimensions of the cells are highly variable [30]. Typically, elementary plant fibres are found in bundles (in the form of a technical fibre), where the middle lamella (a pectin layer) cements the cell walls of two adjoining cells together (Fig. 4).



Fig. 4. The structure of an elementary fibre (i.e. a unit cell) in a technical fibre bundle, where the middle lamella (M) glues adjacent cells together, and each unit cell composes of primary (P) and secondary (S) cell walls and a central lumen (L).

As depicted in Fig. 4, the cell wall has a hierarchical structure, including a thin primary (P) cell wall, and a thick secondary (S) cell wall which exists in three sub-layers (S1, S2, S3). Typically, the primary cell wall accounts for less than 2% of the total cell wall thickness, while the secondary cell wall accounts for up to 90% of the total cell wall thickness [103]. Notably, the S2 cell wall is the main sub-layer, accounting for more than 80% of the total cell wall thickness [103]. The luminal cavity is typically up to 25% of the total cross-sectional area for non-wood plant fibres [30, 104], and usually between 2-16% for bast fibres [103].

Plant fibres themselves can be referred to as composites as the cell wall composes of reinforcing oriented semi-crystalline cellulose microfibrils which are embedded in a twophase (lignin-hemicellulose) amorphous matrix. The content of the three main polymers (*i.e.* cellulose, hemicellulose and lignin) is known to vary between plant fibre types [30]. The typical chemical composition of flax is given in Table 5.

Cellulose, a non-branched macromolecule (Fig. 5), is usually the major component of plant fibres (Table 5). Molecular chains of cellulose, comprising of about 10,000 pairs of covalent-bonded glucose units, are oriented in the fibre direction. Each repeating glucose unit contains three hydroxyl groups, which enables cellulose to form strong hydrogen bonds with its own chains to form fibrils, and with neighbouring chains to form microfibrils [105]. It is well known that cellulose has both crystalline and amorphous regions, depending on whether the cellulose chains are held in a highly ordered (crystalline) structure due to intermolecular hydrogen bonding. Notably, crystalline and amorphous cellulose have very different mechanical properties; for instance, the tensile stiffness of crystalline cellulose (in the chain direction) is up to 15 times more than that of amorphous cellulose (Table 5). Furthermore, while amorphous cellulose is isotropic, the molecular linearity of crystalline cellulose makes it very anisotropic [103]. Flax fibres, for instance, comprise of 55-75 wt% cellulose, of which 53-70 % is crystalline (Table 5).



Fig. 5. Molecular structure of cellulose [75].

Cellulose microfibrils are helically wound around the cell wall, and thus are at an angle with respect to the fibre axis (Fig. 4). The cell walls also consist of heterogeneous, nonlinear and highly-branched hemicellulose and lignin molecules. It is agreed that the hemicellulose molecules are hydrogen bonded to the cellulose microfibrils and act as a cementing matrix between adjacent microfibrils. These structural cellulose/hemicellulose units are then encapsulated by a lignin matrix.

	Crystalline cellulose	Amorphous cellulose	Hemicellulose	Lignin	Pectin
Content in dry flax [ <i>wt%</i> ]	30-50	20-30	14-18	2-3	2-3
Density [gcm <sup>-3</sup> ]	1.6	1.42	1.4	1.4	-
Tensile modulus [ <i>GPa</i> ]	74-168	8-11	7-8	2-4	-

Table 5. Typical chemical composition of flax fibre, alongside the density and tensilestiffness of the various constituents. From [30, 103].

Importantly, both the chemical composition and the orientation of the cellulose microfibrils with respect to the fibre axis, vary between cell wall layers [30, <u>103</u>]. As the S2 cell wall layer is the thickest, it is the microfibril angle (MFA) of the S2 cell wall that is of particular interest.

It is obvious that the chemical composition of a plant fibre would strongly affect its properties. For instance, the hydrophilic nature of cellulose and hemicellulose implies that plant fibres have a high moisture content (typically 5-15 wt% [9]) and the resulting composites have poor moisture resistance. Furthermore, it is known that fibre chemical composition, cellulose crystallinity and density are well-correlated [104].

Four critical micro-structural parameters that affect the mechanical properties of plant fibres include: *i*) cellulose content, *ii*) cellulose crystallinity, *iii*) microfibril angle, and *iv*) fibre aspect ratio. Studies performed by McLaughlin and Tait [106] and Satyanarayana *et al.* [107, 108] conclude that these four parameters are strongly correlated to the tensile properties of plant fibres. Several studies on the prediction of plant fibre tensile properties also incorporate these four key parameters [103, 109, 110].

Table 5 presents the typical tensile modulus of the different chemical constituents of a plant fibre. Crystalline cellulose has significantly better stiffness than all other constituents. In fact, even the transverse stiffness of crystalline cellulose (about 27 GPa [103]) is over 3 times higher than the stiffness of amorphous cellulose, hemicellulose and lignin. Hence, it is clear that not only high cellulose content but high cellulose crystallinity is also desirable when selecting plant fibres for use as reinforcements in structural applications. Furthermore, due to the highly anisotropic nature of crystalline

cellulose, a low MFA is desirable so that the cellulose microfibrils are oriented in the fibre direction. Finally, several studies [109, 111] confirm that for a constant test gauge length, the tensile modulus of a plant fibre increases with decreasing fibre diameter (*i.e.* increasing fibre aspect ratio). A high fibre aspect ratio is also desirable for improved load transfer capability in a fibre reinforced composite (as demonstrated in Section 1.2.1).

Table 6 presents typical values of the four critical structural parameters for various plant fibres. It is found that bast fibres exhibit a high cellulose content (60-70 wt%) and crystallinity (50-90 %), low microfibril angle ( $<10^{\circ}$ ) and high aspect ratio. With some exceptions, leaf and seed fibres, exhibit lower cellulose content and crystallinity, higher microfibril angles (10-50°) and lower aspect ratios. Hence, the superior mechanical properties of bast fibres and their composites, observed in Table 3 and Table 4, is logical.

	Fibre	Cellulose content [wt%]	Cellulose crystallinity [%]	Microfibril angle MFA [°]	Aspect ratio l <sub>f</sub> /d <sub>f</sub> [-]	Luminal porosity [%]	Source
	Flax	64-71	50-90	5-10	1750	2-11	[103]
Bast	Hemp	70-74	50-90	2-6	900	2-11	[103, 112]
	Jute	61-72	50-80	8	100	10-16	
e	Sisal	66-78	50-70	10-25	100	10-22	[23]
Leaf	Pineapple	70-82	44-60	10-15	450	10-22	[64]
	Banana	44-64	45-55	10-12	150	35-53	[93]
_	Cotton	85-93	50-90	46	1000	5	[113]
Seed	Coir	32-43	27-33	30-49	35	30-50	[93]
•1	Oil palm	40-50	20-30	42-46	100	5-10	[66]
ner	Bamboo	26-60	40-60	8-11	100	-	[101]
Oth	Wood fibre <sup>a</sup>	40-60	60-70	10-25	50	20-70	[114]

*Table 6. Typical values of critical structural parameters for various plant fibres. Sources include those listed and [7, 27, 30, 107].* 

<sup>a</sup> Including softwoods and hardwoods.

It is noteworthy, that apart from the four micro-structural parameters identified previously, fibre cross-sectional shape and dimensions (particularly, fibre diameter and luminal porosity) are also thought to be important parameters in determining fibre mechanical properties [103, 107, 109, 110]. Table 6 presents typical values of surface

area proportion of the lumen in different plant fibres; lower luminal porosity would lead to better tensile properties. Several authors, for instance [20, 76, 77, 103, 109, 115], have reported that lower fibre diameter also leads to improved fibre tensile stiffness. While there is no ready explanation in literature to explain this phenomenon [103], Baley *et al.* [76, 109, 115] and Summerscales *et al.* [77] have hypothesised that this may be due to the lumen size increasing with fibre diameter. Placet *et al.* [103] and Gassan *et al.* [110] have also demonstrated through their models on the elastic properties of bast fibres that an only an increase in surface area proportion of the lumen (*i.e.* a reduction in the loadbearing area of the fibre), as a function of fibre diameter, could justify a decrease in fibre stiffness. However, both Placet *et al.* [103] and Summerscales *et al.* [77] acknowledge that this assumed relationship of increasing lumen size with increasing fibre diameter is not currently supported by morphological studies on hemp and jute fibres. Structural effects, such as the microfibril angle being a function of the fibre diameter, have been deemed unlikely to explain the diameter dependence of fibre modulus [77, 103].

Of interest is a recent analysis by Porter et al. [116] which shows that the fibre diameter plays a key role in determining fibre properties for both natural and synthetic polymer fibres. Applying Griffith observations, which combines fracture mechanics and inelastic deformations, to a variety of fibres, Porter et al. [116] find that the fibre fracture strength is directly proportional ( $R^2 = 0.90$ ) to the square root of the ratio of the fibre stiffness to the fibre diameter, *i.e.*  $\sigma_f = \sqrt{(G \cdot E_f/d_f)}$ , where G is the strain energy release rate (determined to be 1000 Jm<sup>-2</sup>), for a large range of polymer fibres. As is suggested by the results of Porter et al. [116], for a given fibre (with a given characteristic fibre strength), the fibre stiffness would thus be characteristically inversely proportional to the fibre diameter. The latter is observed by Virk et al. [77], inspiring them to define a fibre diameter distribution factor  $\eta_d$  for the modified ROM model (discussed in Section 1.2.1). Other than the diameter dependence of fibre tensile properties, Gassan et al. [110] have shown that the cross-sectional shape of the fibre may affect the fibre tensile properties. In fact, the tensile modulus is lower for circular cross-section shaped fibres than for elliptical cross-sectional shaped fibres [110]. This is possibly due to higher transverse fibre aspect ratio for elliptical cross-sectional shaped fibres.

## 2.3 Plant fibre processing

### 2.3.1 Plant growth and fibre extraction

Plant fibres, even of the same type, have highly variable properties. The variability in properties can be ascribed to the variability in the previously described fibre micro-structural parameters. Indeed, even for a given plant fibre type, the fibre micro-structural parameters, which dictate the fibre quality, are themselves influenced by *i*) plant growth conditions (including, plant species, geographic location, climate, soil characteristics, crop cultivation), *ii*) fibre extraction and preparation (including, age of plant, fibre location in plant, type of retting method, decortification and carding processes), and *iii*) fibre processing (including, spinning to produce rovings from slivers and yarns from rovings, and production of mats and textile preforms from slivers/rovings/yarns). Several review articles and studies (for instance, [25, 27, 56, 117-123]) have discussed the influence of these factors on the fibre and composite properties. To ensure that the quality of their products is consistent (*i.e.* the variability in properties is within acceptable limits) and independent of plant growth conditions, suppliers of plant fibres/yarns typically use 'batch-mixing', across several crops/harvests/years.

Regarding optimising fibre extraction and processing, the resounding message of scientific studies is that an increasing number of mechanical processing steps leads to an increase in defect count (in the form of kink bands, for instance), a reduction in degree of polymerization of the cellulose chains, and a subsequent reduction in fibre mechanical properties [118, 121]. Minimally-processed fibres that have undergone retting and hackling produce high quality fibres and good quality composites [25, 119, 120]. However, to ensure full utilisation of fibre properties in a composite, a continuous and aligned reinforcement product is required [79]. Once fibres have been carded or cottonised to produce a (typically coarse *i.e.* high linear density) sliver, rovings can be produced through a wet-spinning process, and yarns can produced through a dry-spinning process. Notably, the level of twist imparted to the product increases at each stage [120]. Increasing twist levels have various detrimental effects on composite properties, including hindered resin impregnation, reduced wettability, increased intra-yarn void formation and a significant quantifiable drop in tensile properties, similar to an off-axis

composite, due to increased fibre misorientation [71, 72, 79, 120, 124]. Interestingly, structure-property relations of twisted yarns imply that for the same twist level, yarns of fine count (low linear density) have a smaller diameter than heavier yarns [79]. The result is that the twist angle in fine count yarns, and the induced reinforcement misorientation and subsequent reduction in composite properties, is smaller [79, 120]. Therefore, to achieve a compromise between *i*) minimal fibre processing, *ii*) employing aligned/continuous reinforcements, and *iii*) limiting the detrimental effects of yarn twist, the order of preference for a reinforcement product is: slivers, followed by rovings, followed by fine-count yarns [25, 79, 120].

Complementary to the studies on the effect of fibre processing on fibre and composite mechanical properties are life cycle assessment studies by Joshi et al. [2], Dissanayake et al. [20, 125-127], Steger [3] and Le Duigou et al. [128]. Dissanayake et al. [20, 125-127] quantified the energy required in the production of UK flax fibres, and found that while the energy required for cultivating plant fibres is low (4-15 MJ/kg of processed fibre), the use of agrochemicals and retting processes increases the energy consumption significantly (by 38-110 MJ/kg of processed fibre). An independent analysis by Le Duigou et al. [128] on French flax fibres, based on a different set of assumptions, provides a similar conclusion. Water retting is found to be least energy intensive, followed by dew retting and bio-retting [20, 125, 127]. Conversion from fibres to semiproducts through textile processes increases the energy consumption further by 2-15 and 26-40 MJ/kg of processed fibre, for slivers and yarns respectively [20, 125]. The total energy required is 54-118 MJ/kg for flax sliver and 81-146 MJ/kg for flax yarn [20]. This compares to 55 MJ/kg for E-glass reinforcement mats and 90 MJ/kg for polypropylene fibres [20]. Hence, even in terms of minimising the environmental impact of plant fibre reinforcements, minimal processing is attractive.

## 2.3.2 Fibre surface modification

The hydrophilic nature of plant fibres has led to the popular view, particularly amongst researchers of PFRPs, regarding the vulnerability of plant fibres and their composites to moisture absorption and the poor compatibility of highly polar plant fibres with typically non-polar polymer matrices [18]. While the former is a concern for the long-term

durability of PFRPs, the latter is a concern for the general mechanical performance of PFRPs. Not surprisingly, a significant amount of work has been undertaken, reviewed by several authors in [8, 24, 27, 57, 58], to explore various avenues in improving the fibre/matrix interfacial properties. The two fundamental routes are fibre surface physical/chemical modification and matrix modification. The former is usually preferred over the latter. The aim of physical modification techniques, such as plasma treatment or mercerisation, is to roughen the fibre surface topography and/or remove surface impurities (such as oils, waxes, pectin), enabling improved mechanical adhesion between the fibre and the matrix. In chemical modification techniques, a third material is introduced, as a compatibiliser or coupling agent, between the fibre and the matrix.

The question is: Is fibre surface modification necessary to achieve good mechanical properties in all PFRPs? In Section 1.2.1 it has been described that there is an ineffective fibre length below which the fibre does not carry the maximum load. The contribution of the fibre in reinforcing the composite (*i.e.* the length efficiency factor) is determined by the ratio of the critical fibre length to the reinforcing fibre length (Eq. 6). Notably, the critical fibre length is directly proportional to the ratio of the fibre tensile strength and fibre/matrix interfacial shear strength (Eq. 6). An interesting inference of these relationships is the following: assuming that i) a given plant fibre has the same diameter as E-glass (which is true in the case of flax [30]), and *ii*) a PFRP and GFRP are to be manufactured with reinforcing fibres of the same length, then for the critical fibre length (and thus length efficiency factor) to be the same in the PFRP and the GFRP, the ratio of the fibre strength to the interfacial shear strength needs to be the same in PFRP and the GFRP. In essence, as plant fibres have a lower tensile strength than E-glass, PFRPs require a proportionally lower interfacial shear strength than GFRPs. Therefore, the common notion that PFRPs have poor interfacial shear strength in comparison to GFRPs, is rather trivial.

$$\eta_{lS,plant} = \eta_{lS,Eglas} \Leftrightarrow l_{c,plant} = l_{c,Eglas} \Leftrightarrow \frac{\sigma_{plant}d_{plant}}{2\tau_{plant}} = \frac{\sigma_{Eglass}d_{Eglass}}{2\tau_{Eglass}} \Leftrightarrow \frac{\sigma_{plant}}{\tau_{plant}} = \frac{\sigma_{Eglass}}{\tau_{Eglass}}$$

The case for improving the interfacial shear strength of PFRPs (and thus employing fibre surface pre-treatments) becomes important when the reinforcing fibres are 'short', that is

short in comparison to the critical fibre length. As was demonstrated in Section 1.2.1 (see Fig. 3), an increase in the interfacial strength from 15 MPa to 30 MPa (and a consequent reduction in the critical fibre length from 0.667 mm to 0.333 mm) leads to a significant increase in the length efficiency factor (from 0.667 to 0.833 for a constant fibre length of 1 mm). (Please refer to the typical values used for these calculations listed in the caption of Fig. 3). As a significant amount of research on PFRPs has focussed on short-fibre randomly oriented composites, based on the compression moulding of nonwovens (typically  $l_f \approx 3-30$  mm [8, 11, 129]) or the injection/extrusion moulding of pellets/granules (typically  $l_f \approx 0.2-3$  mm [8, 11, 129-131]), it is appreciable why some researchers report significant improvements in the mechanical properties of the resulting PFRPs if the fibres are subjected to pre-treatments.

On the other hand, if the reinforcing fibres are 'long', that is more than 10 times the critical fibre length [82], improvements to the interfacial shear strength (through fibre surface pre-treatment) have negligible effect on the length efficiency factor (see Fig. 3). Essentially, as the fibres are carrying the maximum load over a majority of the fibre length, a reduction in the ineffective fibre length does not have a significant effect on the contribution of the fibre in reinforcing the composite. Therefore, it can be argued, that when considering PFRPs for structural applications, as long fibre reinforcements (typically  $l_f > 30$  mm) are used, the use of fibre surface modification is unnecessary. Indeed, PFRPs with impressive mechanical properties can be produced, without any active fibre surface treatment, by using an optimised reinforcement form (*i.e.* slivers or rovings) and high fibre volume fractions [71, 120, 132]. In fact, considering that i) fibre surface treatment techniques may employ expensive (e.g. silanes) and/or toxic (e.g. isocyanates) chemical reagents which tarnish the low-cost eco-friendly image of plant fibres [11], *ii*) unoptimised fibre surface treatments have been known to slash the raw fibre tensile strength by up to 50% [133], *iii*) there is a lack of consensus in literature on the surface treatment parameters to use (e.g. concentration of reagent, treatment time, temperature) to achieve improvements in PFRP mechanical properties [133], and iv) improvements on interfacial properties often lead to a reduction in impact performance [18], the use of fibre surface modification to potentially improve the mechanical properties of PFRPs that are to be used for structural applications is discouraged.

#### 3 Fibre volume fraction

As already intimated in Section 1.2.1, the mechanical properties of a composite are dependent not only on the properties of the constituents, but more so on the volumetric composition of the composite. In fact, the fibre volume fraction  $v_f$  is the single-most important factor in the ROM model (Eq. 1-4).

Amongst other researchers (including [75, 76, 117, 134]), Shah *et al.* [135] have shown that the tensile modulus of yarn reinforced unidirectional PFRPs is linearly related ( $R^2 = 0.99$ ) to the fibre volume fraction (Fig. 6). This is in agreement with the generalised and modified ROM model (Eq. 1 and Eq. 3), assuming no porosity (so that  $v_m = 1 - v_f$ ). Fig. 6 presents data on the tensile modulus of unidirectional flax/polyester composites (from [135]); absorbing all the efficiency and correction factors into an effective fibre property (*e.g.*  $E_f \eta_{IE} \eta_o \eta_d \kappa = E_{f,eff}$  in Eq. 3), it is found that the modified ROM model for PFRPs is a good fit to the experimental data for an effective fibre modulus of  $E_{f,eff} = 44.3$  GPa, which is an acceptable estimate of the modulus of flax [135]. Madsen *et al.* [136] have also validated the modified ROM model (in Eq. 3), incorporating the effect of porosity, for hemp yarn reinforced PET composites, to determine an effective fibre modulus of  $E_{f,eff} = 63.1$  GPa.



Fig. 6. Influence of fibre volume fraction on the tensile properties of a unidirectional flax/polyester composite. From [135].

In composite theory [82], for brittle-fibre ductile-matrix systems, such as PFRPs (Fig. 7), the strength-fibre content relationship is well-understood (Fig. 8). The minimum fibre volume fraction  $v_{f,min}$ , which is the volume fraction that results in the lowest composite strength, determines the transition point from one failure regime/mechanism to another. If

there are very few fibres present ( $0 < v_f < v_{f,min}$ ), the stress on a composite may be high enough to break the fibres. However, the matrix, due to its high failure strain, would be able to support the applied load on the composite until its tensile strength  $\sigma_m$  is reached. The broken fibres, which carry no load, can be regarded as an array of aligned holes. The net effect is that the composite tensile strength  $\sigma_c$  is even below that of the matrix  $\sigma_m$  (Eq. 8). This defines a minimum fibre volume fraction  $v_{f,min}$  below which the fibres weaken the material rather than strengthen it and composite failure is controlled by the matrix [82]. When  $v_f > v_{f,min}$ , there are a sufficient number of fibres to bear some of the load when the matrix reaches it failure strength  $\sigma_m$ . An increase in composite tensile strength beyond the matrix strength only occurs once the fibre volume fraction exceeds the critical fibre volume fraction ( $v_f > v_{f,crit}$ ). This is mathematically presented in Eq. 8 and diagrammatically illustrated in Fig. 8. Note that  $\sigma'_m$  is the matrix stress at the fibre failure strain  $\varepsilon_f$  (schematic in Fig. 7). Eq. 9 and Eq. 10 mathematically define the minimum  $v_{f,min}$ and critical  $v_{f,crit}$  fibre volume fractions, respectively [82].



Fig. 7. PFRPs are a brittle-fibre ductile-matrix system. From [135].

$$\sigma_c = \begin{cases} \sigma_m (1 - v_f) & \text{for } 0 < v_f < v_{f,\min} \\ \sigma_f v_f + \sigma'_m (1 - v_f) & \text{for } v_f > v_{f,\min} \end{cases}$$
Eq. 8

$$v_{f,\min} = \frac{\sigma_m - \sigma'_m}{\sigma_f + (\sigma_m - \sigma'_m)}$$
 Eq. 9

$$v_{f,crit} = \frac{\sigma_m - \sigma'_m}{\sigma_f - \sigma'_m}$$
Eq. 10



Fig. 8. Schematic illustration of the variation of the strength of a unidirectional (brittlefibre ductile-matrix) composite with fibre volume fraction.

For structural composites,  $v_f > v_{f,crit}$  is required. An aligned carbon/polyester composite would have  $v_{f,min} = 2.3\%$  and  $v_{f,crit} = 2.4\%$  [82]. PFRPs have much higher minimum and critical fibre volume fractions. Reproduced from [135] in Fig. 6, Shah *et al.* [135] report that yarn-reinforced unidirectional PFRPs have a  $v_{f,min} \approx 11\%$  and  $v_{f,crit} \approx 13\%$ . For their analysis, Shah *et al.* [135] use a value of  $\sigma'_m = 6.2$  MPa and  $\sigma_f = 502.7$  MPa, which they determine through curve fitting of tensile strength with fibre volume fraction (for  $v_f >$ 15%). Eq. 9 and Eq. 10 are then used to determine  $v_{f,min} \approx 11\%$  and  $v_{f,crit}$ , respectively. Note that determination of the true  $\sigma'_m$  is difficult, noting the non-linear stress-strain curve of the resin and the stochastic variation in the failure strain of the fibre.

For FRPs in general, improvements in most mechanical properties, including stiffness (tensile, flexural, compressive, shear) and strength (tensile, flexural, compressive, shear, impact), can be made by simply increasing the fibre volume fraction [82]. Indeed, several studies, particularly those employing aligned reinforcements, have shown this to be the case for PFRPs (such as [132, 134-136]), as illustrated for the tensile properties in Fig. 6.

Although increasing the fibre content is attractive, there is a theoretical maximum fibre volume fraction  $v_{f,max,theo}$ , dependent on the fibre packing arrangement, which cannot be surpassed (Fig. 8) [82]. Quadratic arrangement of the fibres leads to a  $v_{f,max,theo}$  of  $\pi/4$  (= 78.5%) while hexagonally-packed fibres generate a higher  $v_{f,max,theo}$  of  $\pi/2\sqrt{3}$  (= 90.7%) [137]. If twisted yarns are used as reinforcements,  $v_{f,max,theo}$  would be lower [135, 136].

The maximum packing fraction of a yarn  $\emptyset_{max}$  is 75.0% assuming an open-packed structure [138] or 90.7% assuming hexagonal close-packed fibre arrangement [139, 140]. The maximum achievable composite fibre volume fraction can be estimated to be the product of the maximum packing fraction (accounting for the packing arrangement of fibres within the yarn) and the maximum packing-ability of yarns in the composite. Hence, the absolute limit of the maximum fibre volume fraction of a PFRP reinforced with twisted yarns ranges from 58.9% (= 78.5%.75.0%) to 82.2% (= 90.7%.90.7%), depending on the combination of packing assumed in the composite and the yarn [135].

In addition, there is a practical maximum fibre volume fraction  $v_{f,max,prac}$ , typically lower than the theoretical maximum fibre volume fraction  $v_{f,max,theo}$  (Fig. 8), dependent on manufacturing process limitations, above which composite properties deteriorate [134, 141], often due to increased porosity [74, 136, 142] (resulting from impregnation and wettability issues) or increased fibre-fibre interactions [134, 143, 144] (causing inefficient stress-transfer between fibre and matrix). For instance, Madsen et al. [136] found that when aligned hemp/polypropylene laminates were fabricated at a nominal fibre volume content of 61%, the actual measured fibre volume content was only 51% with a larger porosity content of 17%. The experimentally determined optimal (or practical) maximum fibre volume fractions  $v_{f,max,prac}$  for PFRPs range from about 60% for aligned jute roving reinforced polyester [134], 46-54% for aligned hemp yarn reinforced polyethyleneterephthalate (PET) [74, 136], and between 33% and 46% for short random flax and jute reinforced polypropylene [74]. Notably, some authors, for instance Hepworth et al. [145], have manufactured aligned PFRPs with fibre volume fractions of up to 85%; the composites have good mechanical properties, although the authors do not state i) how the fibre volume fraction was measured, and ii) the porosity content in the composites. For synthetic fibre composites,  $v_{f,max,prac}$  is much higher at up to 80% [82].

Not only does the high minimum and critical fibre volume fraction and low maximum fibre volume fraction of PFRPs, in comparison to synthetic fibre composites, limit the maximum exploitation of the mechanical properties of plant fibres in FRPs due to a small processing window, but it is also found that PFRPs manufactured in an identical manner to GFRPs consistently produce lower fibre volume fractions [71, <u>72</u>, 75]. This is due to the poor packing ability of plant fibre assemblies in comparison to synthetic fibre

assemblies. Madsen [75] has studied the compaction of PFRPs in comparison to GFRPs. They report that for a constant compaction pressure, unidirectional flax yarn and E-glass composites have a fibre volume fraction of 56% and 71%, while random flax fibre and E-glass composites have a fibre volume fraction of 38% and 52%, respectively. It is interesting to see that the ratio in achievable volume fractions of PFRPs to GFRPs is ~0.75. Shah *et al.* [71] and Goutianos *et al.* [72] also find that when employing liquid moulding processes, GFRPs produce higher fibre volume fractions than PFRPs.

To produce PFRPs with high fibre content, it is generally suggested that due to the low compactability of plant fibre assemblies [11, 136], the plant fibre preforms need to be compacted using external force. As the literature survey in Table 7 reveals, compression moulding (including hot- and cold-pressing) has been the most popular method so far. It is being used for the manufacture of both thermoplastic- and thermoset-based PFRPs [8, 11]. Indeed, current commercial applications of PFRPs are primarily based on compression moulded components (Fig. 1.1) [33, 35]. In the case of liquid thermoset resins, a 'leaky mould' is typically used, where the excess resin is forced out during mould compaction [11]. From Table 7 it is clearly observed that amongst thermosetbased PFRPs, hand lay-up and vacuum infusion produce lower fibre content than compression moulding. Comparing hand layup and compression moulding techniques in the manufacture of flax/epoxy composites, Charlet et al. [146] find that the maximum achievable fibre volume fractions were  $\sim 15\%$  and  $\sim 40\%$ , respectively. Other researchers have been able to achieve fibre volume fractions typically up to 60% [73, 134, 147], but even up to 85% [145], using very high compaction pressures in compression moulding. While compression moulding is suitable for high-volume part production at low-cycle times, the limitation with compression moulding is the component size that can be manufactured. Large structural components produced at a much lower rate, such as wind turbine blades, are typically manufactured through vacuum infusion, resin transfer moulding (RTM) or prepregging technology (with autoclave consolidation). The literature survey in Table 7 finds that fibre volume fractions achievable through RTM and prepregging technology (up to 50%) are comparable to compression moulding (up to 60%), noting that the latter is more prone to porosity-related issues [82].

# 4 Ashby plot for PFRPs

The construction of a materials selection chart (*i.e.* Ashby plot) relies heavily on a large database that captures, and is representative of the variability in (*i.e.* range of), typical properties. To generate such a database, an extensive literature survey was conducted on the (absolute and specific) tensile properties of bast fibre reinforced PFRPs. The literature survey is partly presented in the form of Table 7. The wide-ranging database looks to particularly elucidate the effects of *i*) reinforcement geometry and orientation (pellets, short-random nonwovens, and long-aligned fibres for unidirectional and multiaxials), *ii*) matrix type (thermoplastic *vs.* thermoset), and *iii*) manufacturing technique (injection moulding, compression moulding, hand lay-up, vacuum infusion, resin transfer moulding and prepregging), on the tensile properties of bast fibre reinforced PFRPs. While the specific effects of each will be discussed in some detail in the following sections, here Ashby plots are presented for the PFRP materials (Fig. 9), showing the absolute and specific tensile stiffness, respectively. Note that the fibre volume fraction of the PFRPs may be dissimilar.

Ashby plots, such as the ones presented in Fig. 9, are very useful for four key reasons [69, 70]: *i*) they allow quick retrieval of the typical properties of a particular material, *ii*) they allow quick comparison of the properties of different materials, revealing their comparative efficiencies, *iii*) they facilitate the selection of the materials/manufacturing processes during the product design stage, and *iv*) they enable substitution studies exploring the potential of one material to replace another.

It is quite clear from the Ashby plots in Fig. 9 that PFRPs can be categorised into four distinct sub-groups, with increasing tensile properties in the following order: *i*) Injection-moulded PFRPs, whose mechanical properties are low and comparable to the matrix material, *ii*) PFRPs based on nonwoven reinforcements (randomly-oriented short fibres), *iii*) PFRPs based on textile reinforcements (woven and stitched biaxials, for instance) and *iv*) unidirectional PFRPs. It is also observed that tensile strength and stiffness tend to increase linearly with each other. Observing the variation in properties within each sub-group, it is found that thermoset-based PFRPs have better mechanical properties than thermoplastic-based PFRPs. Furthermore, the manufacturing technique can have a

noticeable effect on PFRP mechanical properties, particularly in the case of unidirectional PFRPs.

The Asbhy plot in Fig. 9 can be expanded to include typical tensile properties of various GFRPs to enable a comparison between properties achievable with GFRPs and PFRPs. In fact, although the data has not been graphically shown in Fig. 9, the literature survey of Table 7 includes example tensile properties of GFRPs. The comparison reveals that when comparing short-fibre reinforced composites (*i.e.* injection moulded and nonwoven composites), PFRPs have better tensile modulus (specific and absolute) and comparable specific tensile strength than GFRPs. On the other hand, when comparing long-fibre reinforced composites (*i.e.* textile and unidirectional composites), PFRPs have better specific tensile modulus than GFRPs; the specific tensile strength of PFRPs is only up to half that of GFRPs.

Although the Asbhy plot in Fig. 9 suggests that unidirectional PFRPs, for instance provide 2 to 20 times better tensile properties than nonwoven PFRPs and up to 5 times better tensile properties than multiaxial PFRPs, this does not necessarily mean that unidirectional PFRPs would be preferred over the other materials for all structural applications. To truly enable substitution studies exploring the potential of one material to replace another, other material properties may need to be taken into other, depending on the specific component function, objectives and constraint for a given application. For instance, minimising cost cost may be an important objective. Due to the high cost of plant fibre intermediate products (*i.e.* yarns/rovings) in comparison to the raw fibres [51, 75], nonwoven plant fibre reinforcements (up to 36.7 f/kg) and up to 30 times cheaper than optimised textile (biaxial) reinforcements (up to 45.9 f/kg) [51]. Other mechanical properties, such as fatigue properties and fracture toughness, may also need to be considered. It would be beneficial to the industrial uptake of PFRPs in wider applications, if other researchers were to build on the Asbhy plot that has been presented here.



Fig. 9. Typical tensile properties (absolute and specific) for PFRPs manufactured with thermoplastic/thermoset resins, short-random/long-aligned fibre reinforcements, and various manufacturing routes. Refer to the main text and Table 7 for more information, comparison with GFRPs, and the primary literature sources used in the production of this chart.

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Manufacturing technique	Reinforcement form	Matrix type	Composite	Fibre content <sup>a</sup> [%]	Tensile modulus [GPa]	Specific tensile modulus <sup>a</sup> [GPa/gcm <sup>-3</sup> ]	Tensile strength [MPa]	Specific tensile strength <sup>a</sup> [MPa/gcm <sup>-3</sup> ]	Source
_	<u> </u>	Thermoplastic	PP <sup>b</sup>	0	0.7-1.7	0.8-1.9	20-35	22-39	
-	-	Thermoset	UP <sup>b</sup>	0	1.4-4.7	1.2-3.9	12-75	10-63	
-	-	Thermoset	Ероху	0	3.1-3.7	2.7-3.2	60-75	50-63	
0	ູ້ເ	Thermoplastic	Flax/PP	30 wt	1.7	1.6	27	26	[148]
Injection noulding	short fibre <sup>°</sup> 3D-randon		Hemp/PP	30 wt	1.5	1.5	30	29	[149]
			Flax/PP 5 wt% MAPP <sup>b</sup>	30 wt	2.1	2.0	38	36	[148]
			E-glass/PP	30 wt	2.2	1.5	49	35	[148]
u r	Compression moulding <sup>d</sup> short fibre <sup>e</sup> , 2D-random	Thermoplastic	Flax/PP	40 v	8.8	8.0	57	52	[96]
sessic ing			Jute/PP	30 v	3.7	3.6	27	26	[18]
ompro			Flax/PP 3.5 wt% MAPP	40 v	8.6	7.8	68	62	[96]
- 0			E-glass/PP	22 v	6.2	4.8	89	69	[18]
u -	ູ້		Flax/UP	21 wt	11.0	8.7	80	63	[150]
essic ing'	lbre	loset	Hemp/UP	47 wt	5.6	4.3	36	28	[151]
ompre	hort fi 2D-rar	Therm	Hemp/UP 5% NaOH treated	46 wt	7.5	5.8	46	35	[151]
- 0	S ( )		E-glass/UP	20 wt	8.5	6.1	95	68	[150]
	» <sup>^</sup> ۲		Flax/UP	30 v	6.3	5.2	61	50	[152]
ion	short fibre <sup>°</sup> 2D-random	loset	Jute/UP	30 v	8.0	6.3	50	39	[152]
Vacu infus		Therm	Flax/epoxy 1% NaOH treated	22 v	9.2	6.5	60	42	[153]
			E-glass/UP	30 v	14.9	9.1	190	116	[152]

Table 7. Literature survey of typically reported mechanical properties of various PFRPs, specifically focussing on the effect of *i*) matrix type, *ii*) reinforcement form, *iii*) manufacturing technique, and *iv*) interface engineering, on PFRP mechanical properties. For comparison, the mechanical properties of the neat matrix and similarly manufactured GFRPs are also given.

Manufacturing technique	Reinforcement form	Matrix type	Composite	Fibre content <sup>a</sup> [%]	Tensile modulus [GPa]	Specific tensile modulus <sup>a</sup> [GPa/gcm <sup>-3</sup> ]	Tensile strength [MPa]	Specific tensile strength <sup>a</sup> [MPa/gcm <sup>-3</sup> ]	Source
	al	ic	Flax/PP	43 v	26.9	23.6	251	220	[73]
essio essio ding <sup>d</sup>	long fibre <sup>1</sup> unidirection	plast	Hemp/PP	42 v	21.1	18.2	215	185	[75]
Compr		Thermo	Hemp/PP 0.2 wt% MAPP	40 v	20.1	17.5	208	181	[75]
			E-glass/PP	35 v	26.5	17.4	700	461	[154]
dn⁄	.e f, onal	set	Flax/UP	19 v	6.5	5.2	150	120	[146]
ind lay	long fibr unidirecti	ıermo	Flax/UP	28 v	14.0	10.8	140	108	[72]
H		Ē	E-glass/UP	-	-	-	-	-	-
E .	al		Flax/UP	58 v	29.9	23.0	304	233	[155]
essio essio ding <sup>d</sup>	bre <sup>f</sup>	loset	Jute/UP	56 v	35.0	28.2	248	200	[134]
Zompr	long fí nidire	Thern	Jute/epoxy 26% NaOH treated	40 v	24.0	18.8	220	172	[147]
	n		E-glass/UP	42 v	30.6	16.9	695	384	[155]
n) ng	e <sup>f</sup> , onal	set	Flax/epoxy	42 v	35.0	28.2	280	226	[132]
RTM neludi 'acuur ifusio	ig fibr	lermo	Flax/VE <sup>b</sup>	37 v	24.0	16.9	248	175	[72]
(jr ir	lor unic	T	E-glass/epoxy	48 v	31.0	18.1	817	478	[132]
ů B	e f, onal	set	Flax/epoxy	42 v	39.9	31.3	378	296	[120]
preggi	ng fibn lirecti	ıermo	Flax/epoxy	48 v	32.0	24.7	268	207	[133]
Prej	lon unid	Th	E-glass/epoxy	55 v	39.0	18.6	1080	514	[156]

Manufacturing technique	Reinforcement form	Matrix type	Composite	Fibre content <sup>a</sup> [%]	Tensile modulus [GPa]	Specific tensile modulus <sup>a</sup> [GPa/gcm <sup>-3</sup> ]	Tensile strength [MPa]	Specific tensile strength <sup>a</sup> [MPa/gcm <sup>-3</sup> ]	Source
uc p	long fibre <sup>f</sup> , stitched balanced biaxial	t	Flax/epoxy [0, 90]	44 v	14.3	11.2	170	133	[157]
essic		nose	E-glass/epoxy [0, 90]	43 v	21.9	12.2	380	212	[157]
nould		Cherr	Flax/epoxy [±45]	44 v	6.5	5.1	79	62	[157]
C			E-glass/epoxy [±45]	43 v	11.1	6.2	103	58	[157]
-0	ibre <sup>f</sup> , veave cial	t	Flax/VE [0, 90]	33 v	7.3	4.8	81	54	[72]
M Iding uut Sion)		loset	Flax/VE [0, 90]	35 v	8.6	5.7	89	59	[72]
RT inclu vacu infus	ng f niai bia	Therr	Jute/VE [0, 90]	41 v	10.0	6.7	111	74	[72]
	ol d		E-glass/UP [0, 90]	51 v	33.0	12.8	483	189	[72]
مہ 10	f ial	t	Flax/epoxy [0, 90]	45 v	11.2	8.2	94	69	[158]
gging	ibre twill biaxi	nose	Flax/epoxy [0, 90]	36 v	10.0	7.5	104	78	[158]
epres	long fi 2x2 t weave l	Them	Flax/epoxy [0, 90]	37 v	11.2	8.5	77	59	[159]
Pr			Flax/epoxy [0, 90]	54 v	9.3	6.8	78	57	[159]

<sup>*a*</sup> Fibre content is approximate – note that some sources have presented fibre content in terms of fibre weight fraction  $w_f$  and not fibre volume fraction  $v_f$ . If the specific properties have not been determined in the referenced source, these have been estimated by either using the composite density measured by the authors in their respective studies, or by estimating the composite density (assuming no porosity) using  $\rho_c = (\rho_f \rho_m)/(\rho_f - w_f(\rho_f - \rho_m))$  if fibre weight fraction is given, or  $\rho_c = \rho_f v_f + \rho_m v_m$  if fibre volume fraction is given.

<sup>b</sup> PP = Polypropylene, MAPP = Maleic Anhydride Polypropylene, UP = Unsaturated Polyester, VE = Vinylester

<sup>c</sup> Injection moulding includes extrusion-injection moulding (*i.e.* pellets/granules obtained from an extruder rather than a melt-blender).

<sup>d</sup> Compression moulding includes press moulding (*i.e.* hand layup or vacuum infusion or filament winding as a pre-cursor to lossy pressing of the mould for compaction of the impregnated preform). Compaction pressures of up to 60 bars, but typically 20-30 bars, are used.

<sup>e</sup> Short fibre = discontinuous reinforcement with fibres less than 30 mm in length. Typically, fibre lengths are less than 1 mm for injection moulding and between 3 to 30 mm for compression moulding.

<sup>f</sup> Long fibre = continuous reinforcement, in the form of slivers, rovings and yarns. Single fibres are typically greater than 30 mm in length.

<sup>g</sup> Prepregging with autoclave consolidation and cure. Autoclave pressures of up to 10 bars, but typically 4-6 bars, are used.

# 5 Reinforcement geometry and orientation

## 5.1 Length efficiency factors

To ensure that the full reinforcing potential of plant fibres is realised, it is essential that the highest reinforcement efficiency is utilised. As demonstrated by Fig. 3, the reinforcement geometry (*i.e.* fibre length and aspect ratio) directly affects the length efficiency factors for stiffness  $\eta_{lE}$  and strength  $\eta_{lS}$  (Eq. 5 and Eq. 6).  $\eta_{lE}$  and  $\eta_{lS}$  can be maximised by using high aspect ratio fibres with fibre lengths significantly longer than the critical fibre length. In fact, fibre aspect ratios of  $l_f/d_f > 50$  (*i.e.* fibre lengths of  $l_f > 1$ mm) would yield  $\eta_{lE} > 0.93$  [74] and provided that the fibre length is about 10 times the critical length ( $l_f/l_c > 10$ ),  $\eta_{lS} > 0.95$  can be achieved [82]. This is confirmed by the plots in Fig. 3.

Critical fibre lengths for bast fibre reinforced PFRPs have been measured to be in the range of 0.2-3 mm [11, 37, 129-131, 160-162]. While a majority of bast fibres are typically >30 mm in length [27] and have high aspect ratios (between 100-2000; Table 6), depending on the composite manufacturing route, the utilised fibre length and aspect ratio can be much lower. For instance, injection moulding employs fibres with lengths of 1.2-0.1 mm and aspect ratios <20 [130, 149, 163-165]; the resulting length efficiency factors are thus <0.30 [130, 149, 163]. Bos *et al.* [130] have determined the length efficiency factors to be in the range of 0.17-0.20 for injection moulded flax composites. On the other hand, Sawpan *et al.* [160] determine  $\eta_{1S}$  to be up to 0.9 for compression moulded hemp/polyester composites based on nonwoven reinforcements (fibre length of  $l \approx 2-3$  mm). Finally, yarns/rovings compose of fibres that are >30 mm in length [15, 37, 119], hence composites utilising textile or unidirectional reinforcements yield length efficiency factors of approximately unity [60, 61, 162]. These results are summarised in Table 8.

## 5.2 Orientation distribution factors

Due to the anisotropic nature of many fibres, reinforcement orientation has a significant effect on composite properties. The anisotropy of fibre reinforcements may result from the natural structure of the fibre (as is the case of cellulose-based fibres) [166] and/or

from the larger aspect ratio along the axis of the fibre in comparison to the cross-sectional aspect ratio [82].

PFRP subgroup (see Fig. 9)	Typical fibre length [mm]	$\eta_{lE}$ or $\eta_{lS}$	$\eta_o$	$\eta_l \cdot \eta_o$
Injection moulded	<1	< 0.3	~0.20-0.37	< 0.11
Nonwovens	3-30	0.5-0.9	~0.38-0.40	0.19-0.36
Multiaxials	>30	~1.0	0.25-0.50	~0.25-0.5
Unidirectionals	>30	~1.0	~1.00	~1.0

 Table 8. Typical fibre length efficiency factors and fibre orientation distribution factors for various PFRP categories.

Once again, the composite manufacturing route can dictate the orientation distribution that is likely in the resulting composite. For a 3D-random orientation of the fibres, it can be shown that  $\eta_o = 1/5$  (= 0.2). In injection moulded PFRPs, fibre orientation is nominally 3D-random, but typically show a preferred orientation [164]. While Garkhail *et al.* [129] and Bos *et al.* [130] have found  $\eta_o$  to be 0.21-0.31, Vallejos *et al.* [163] and Serrano *et al.* [164] have determined  $\eta_o$  to be in the range of 0.28-0.37, for injection moulded PFRPs. For a 2D-random orientation of the fibres, it can be shown that  $\eta_o = 3/8$  (= 0.375). Conventional nonwoven mat reinforced PFRPs have a nominally 2D-random orientation, but may show a preferred orientation. Bos *et al.* [130] have determined  $\eta_o$  to be ~0.40 for nonwoven PFRPs. Composites reinforced with multiaxial fabrics may have a range of orientation distribution factors, depending on the ply orientation. For composites with balanced biaxial reinforcements in a [0,90] and [±45] stacking sequence, it can be shown that  $\eta_o = 1/2$  (= 0.5) and  $\eta_o = 1/4$  (= 0.25), respectively. Finally, to ensure the orientation distribution factor  $\eta_o$  is close to unity, unidirectional fibres are required. These results are summarised in Table 8.

It is noteworthy that using plant fibre reinforcements in the form of yarns can induce fibre obliquity, waviness and misalignment (to the composite loading axis) which results in a drastic drop in mechanical properties of the composite [79, 124]. Shah *et al.* [79] have developed a mathematical model, based on *i*) the modified ROM for PFRPs (Eq. 4), *ii*) the Krenchel orientation distribution factor (Eq. 7) and *iii*) structure-property relations in an idealised staple fibre yarn, to predict the influence of yarn twist (and other structural parameters such as linear density and packing fraction) on PFRP tensile strength. The

model is found to be a near-perfect fit (with  $R^2 = 0.950$ ) for the extensive experimental data from Goutinos *et al.* [124] and in good correlation with another study on aligned PFRPs by Baets *et al.* [120]. An interesting inference of the model is that employing yarns with surface twist angle  $\alpha > 26^\circ$  or  $\alpha > 32^\circ$  as composite reinforcements will reduce the reinforcement orientation distribution factor as in a 2D-random and 3D-random composite, respectively [79, 162]. Baets *et al.* [120] have also observed the evolution of the tensile modulus of unidirectional flax/epoxy composites with changing yarn twist levels, and find good agreement between their experimental data and the predictive models on the elastic properties of impregnated yarns by Rao *et al.* [167] and Naik *et al.* [168]. The use of minimally-twisted yarns (*i.e.* rovings or even slivers) [71, 72, 79, 120] and even wrap-spun yarns [25, 169] is attractive to ensure minimal losses through fibre misalignment.

Table 8 presents the typical length efficiency factors  $\eta_i$ , orientation distribution factors  $\eta_o$ and their product (*i.e.*  $\eta_i \eta_o$ ) for the four PFRP subgroups identified in the Asbhy plot in Fig. 9. The product  $\eta_i \eta_o$  is a good estimate of the reinforcing contribution of the fibre to the composite (Eq. 1-4). The difference in the product of the efficiency factors between the subgroups (Table 8) clearly demonstrates the difference in properties of the materials (Fig. 9). Sub-critical length fibre reinforced 3D-random composites have tensile stiffness and strength in the range of 1.0-2.5 GPa and 20-50 MPa, respectively. This is comparable to the tensile properties of the polymer matrix. Short-fibre 2D-random composites have higher tensile stiffness and strength in the range of 2.5-11.0 GPa and 25-80 MPa. Textile reinforcement based PFRPs have tensile stiffness and strength in the range of 5-15 GPa and 75-175 MPa. Barring the performance of aligned hand-layup PFRPs (with inherently low fibre content), which is still better than that of 2D-random composites, unidirectional PFRPs reinforced with slivers/yarns/rovings exhibit 3-5 times better tensile stiffness and strength than 2D-random composites.

## 6 Selection of matrix and composite manufacturing technique

#### 6.1 Matrix type

A survey on the applications of PFRPs in the EU in 2010, showed that up to 30% of the PFRPs were based on thermoset matrices, while the rest were based on thermoplastic

matrices (Fig. 1) [35]. There is a general trend, particularly in the automotive industry, of diminishing use of thermoset matrices and increased use of thermoplastic matrices [1, 33, 35]. This is primarily because the latter are faster to process, are fabricated by a cleaner process (dry systems with no toxic by-products), are easier to recycle, and are less expensive (for high volume production). Nonetheless, thermosets may be more suitable for PFRPs in structural applications for three key reasons. Firstly, thermoset matrices have better mechanical properties than thermoplastics, due to the formation of a large cross-linked rigid three-dimensional molecular structure upon curing. Consequently, as highlighted by the literature survey in Table 7 and the graphical analysis in Fig. 9, thermoset-based PFRPs consistently show better tensile properties (absolute and even specific) than thermoplastic-based PFRPs. Secondly, the low processing temperatures (typically below 100 °C) and viscosity (0.1-10 Pas) of thermoset matrices implies that plant fibre mechanical properties are not degraded due to high temperature exposure during composites manufacture, and resin impregnation and preform wettability are easier leading to lower void content and better interfacial properties. The low viscosity of thermoset resins also raises the possibility of using liquid composite moulding techniques, such as vacuum infusion and resin transfer moulding (RTM), which are standard manufacturing procedures in the performance-demanding aerospace, marine and wind energy industries. In contrast, the high processing temperatures (up to 200 °C) and viscosity (100-10000 Pas) of thermoplastics are seen as barriers in the development of optimised thermoplastic PFRPs [11]. Thirdly and finally, thermosets have better shear properties than thermoplastics, and they form a better interface with typically polar plant fibres than thermoplastics (which tend to be non-polar).

It should be noted that in terms of end-of-life disposal, the use of thermosetting matrices, rather than thermoplastic matrices, does not necessarily lower the eco-performance of the PFRP produced. This is because the addition of plant fibres can significantly reduce the recyclability and reusability of a thermoplastic system [8, 20, 170]. All PFRPs can be incinerated for energy recovery or re-used as fillers; the additional option with thermoplastic-based PFRPs is that they can also be granulated and re-processed into extrusion/injection moulded components [20]. Notably, thermoplastic-based PFRPs that are recycled by remoulding into new parts exhibit severely deteriorated mechanical

properties due to repeated thermal exposure [170]. In fact, the 'recyclability' of PFRPs is an altogether different and unresolved issue.

## 6.2 Composite manufacture

Faruk *et al.* [8] and Summerscales *et al.* [20] have discussed the various manufacturing techniques that have been utilised with PFRPs. The literature survey in Table 7 and the graphical analysis in Fig. 9 eloquently present the mechanical properties of PFRPs achievable when produced through a particular manufacturing route. The analysis reveals that to produce PFRPs with optimum mechanical properties, prepregging technology with autoclave consolidation is most suitable. Compression moulding and infusion processes (RTM and vacuum infusion) produce PFRPs with comparable specific tensile properties. Despite the use of aligned reinforcements, hand layup produces composites with only moderate mechanical properties. Injection moulded PFRPs have poorest mechanical properties.

The composite manufacturing technique is interrelated with three key composite parameters, each of which has been discussed separately previously: *i*) volumetric composition (maximum achievable fibre volume fraction and porosity), *ii*) reinforcement form, and *iii*) matrix type. Indeed, the interactive effect of all these parameters can, at least qualitatively, explain the variation in mechanical properties of PFRPs produced through different manufacturing processes.

Firstly, the composite manufacturing technique affects the typical achievable fibre volume fraction and porosity. For high composite mechanical properties, high fibre volume fraction and low porosity are desirable. With increasing consolidation pressure, achievable and typical fibre volume fraction tend to increase. As shown in Table 9, consolidation pressures and thus typically achievable fibre volume fractions increase in the following order: vacuum infusion/RTM, prepregging (with autoclave consolidation), and compression moulding. This was also discussed in previously in Section 3.

Manufacturing	Consolidation	Fibre volume	Porosity volume	Matrix type
tecnnique	pressure	Jraction	jraction	useable
	[bar]	[%]	[%]	
Injection	>1000 bar	Up to 45%	-	Thermoplastic
moulding		(typically 15-30%)		
Compression	Up to 40 bar	Up to 85%	Up to 25%	Thermoplastic
moulding	(typically 20-30 bar)	(typically 25-50%)	(typically 2-8%)	or Thermoset
Prepregging	0-10 bar	Up to 60%	Up to 10%	Thermoset
(with autoclave)	(typically 4-6 bar)	(typically 35-50%)	(typically 0-4%)	
Vacuum	0-4 bar	Up to 60%	Up to 10%	Thermoset
infusion/RTM	(typically 0-2 bar)	(typically 25-50%)	(typically 1-4%)	

Table 9. Manufacturing technique is interrelated with other composite parameters. Here the maximum and typical values of various parameters for PFRPs are quoted. The values are from literatures referenced in Table 7.

Porosity, an almost inevitable phase in a composite material, has significant detrimental effects on composite mechanical performance [82]. As indicated in the modified ROM model (Eq. 3 and 4), Madsen et al. [74] suggest that the influence of porosity on PFRP tensile properties can be modelled by including a factor of  $(1-v_y)^2$  in the generalised ROM model (Eq. 1 and 2). Often, porosity can be managed, if not eliminated, by optimising the manufacturing process [82]. Typically, void contents of <1% are required for aerospace applications, but void contents of up to 5% are acceptable for other less demanding applications (e.g. automotive and marine) [171-173]. In literature [73, 74, 174], PFRPs are often quoted to have high void content. Typically, the void volume fraction is up to 5% for PFRPs with a fibre volume fraction below 40% [136, 141, 142, 152, 169, 174]. However, when the fibre volume fraction exceeds 40%, void content increases drastically and can even approach 25% [136, 141, 142, 174, 175]. Nonetheless, there are some studies [134, 135, 169] which conclude that there is no obvious relationship between fibre volume fraction and void volume fraction for PFRPs. From the literature survey, it is suggested that issues of high porosity in PFRPs are usually related, but not confined, to i) sisal fibre composites due to the large lumen size in sisal fibres which remain unfilled after resin infusion [152, 175], ii) structural porosity in (particularly, high weight fraction) compression-moulded thermoplastic PFRPs due to insufficient amount of matrix to fill the free space between the yarns [174], and iii) randomly-oriented short-fibre PFRPs. It is well known that void content in composites manufactured through different

routes is typically in the following order: Hand lay-up > Compression moulding > Infusion processes (vacuum infusion > RTM > vacuum assisted-RTM) > Prepregging (with autoclave consolidation) [82]. This is in agreement with typical literature values observed for PFRPs (Table 9). Various studies report that vacuum-infused PFRPs have a low void volume fraction of 0.5-4.0% [71, 135, 152] and prepreg-based PFRPs have a typical void volume fraction of 0.0-4.0% [158, 159], although it may be as high as 10% if low autoclave pressures (< 3 bar) are used [158, 159]. It is of interest to note that Madsen *et al.* [75] show that porosity in hemp yarn reinforced thermoplastics increases linearly ( $R^2 = 0.98$ ) with the logarithm of the matrix processing viscosity. As the viscosity of thermosets is several orders of magnitude lower than that of thermoplastics, the significantly lower void content in thermoset-based PFRPs is comprehendible. In addition, while vacuum-infusion and prepregging techniques employ thermoset matrices, compression moulding may employ thermoplastics; therefore, the higher void content in the latter is conceivable.

Secondly, the manufacturing technique may affect the reinforcement form (length and orientation). This is particularly the case of injection moulded compounds, where the process implies that at each stage the fibre length reduces. For instance, initially long fibres (of up to 20 mm in length) are first chopped in a blade mill to a nominal length, of say 10 mm, followed by a melt-blending process where the fibre length reduces to 0.3-0.9 mm, followed by the injection moulding process where the fibre length reduces further to <0.3 mm [149]. Furthermore, the melt-blending process and injection/extrusion moulding process results in mixing of the fibres to produce a nominally 3D random fibre orientation. As discussed in Section 5, this leads to a small product of length efficiency factor and orientation distribution factor for the PFRPs (Table 8).

Thirdly and finally, the manufacturing technique is related to the matrix type that is employed, the effects of which have been analysed previously.

# 7 Conclusions

From the critical review, several recommendations are made in developing PFRPs for structural applications. The recommendations, related to maximising and optimising various composite parameters, are as follows:

- Plant fibre type: Bast fibres are most suitable for reinforcing composites due to their superior mechanical properties which derive from their chemical and structural composition. Typically, fibres with high cellulose content, high cellulose crystallinity, low micro-fibril angles, and high aspect ratios are desirable.
- Plant fibre processing and preparation: Fibres processed specifically for composites applications, rather than textile applications, are desirable. to achieve a compromise between *i*) minimal fibre processing, *ii*) employing aligned/continuous reinforcements, and *iii*) limiting the detrimental effects of yarn twist, the order of preference for a reinforcement product is: slivers, followed by rovings, followed by fine-count yarns. Furthermore, the use of fibre surface modification to improve fibre/matrix adhesion is argued to be unnecessary and possibly detrimental when utilising the previously mentioned long fibre reinforcements.
- Fibre volume fraction: Increasing the fibre content is highly recommended for improving composite properties. The narrow processing window (high minimum and critical fibre volume fractions of the order of 10% for aligned PFRPs, and low maximum fibre volume fractions of the order of 45-60% for aligned PFRPs) of PFRPs and the poor packing ability of plant fibre assemblies, in comparison to GFRPs and synthetic fibre assemblies, limits the possibilities for PFRPs to replace GFRPs in structural applications. Hot-pressing, to increase compaction of plant fibre assemblies, is recommended.
- Reinforcement form: The tensile properties of unidirectional PFRPs are 3-5 times better than short-fibre randomly-oriented composites, due to enhanced reinforcement efficiency. While plant fibres are naturally discontinuous, a continuous product (in the form of slivers, wet-spun rovings and low-count yarns) will ensure that the maximum fibre aspect ratios (or length) and a high degree of alignment are employed. However, aligned plant fibre reinforcements are up to 30 times more expensive than raw and nonwoven plant fibre reinforcements.
- Manufacturing route: Prepregging technology (with autoclave consolidation) is most suitable to produce high quality PFRPs. Compression moulding and RTM/vacuum infusion are follow-up options to produce PFRPs with good mechanical properties.

• Matrix type: Thermosets are more suitable than thermoplastics, due to the formers *i*) capacity in high-performance applications, *ii*) lower viscosity and processing temperatures and, *iii*) better compatibility with plant fibres.

Through a thorough literature survey, a highly useful Ashby plot of (absolute and specific) tensile strength against (absolute and specific) tensile stiffness of PFRPs has also been constructed, which may help in the material selection stage during product design of a natural fibre composite component. It would be beneficial to the industrial uptake of PFRPs in wider applications, if other researchers were to build on the Asbhy plot that has been presented here to build a truly extensive database, by considering other properties of PFRPs (*e.g.* cost of material, cumulative energy demand in the life cycle analysis of the material, other mechanical properties such as fatigue strength at 10<sup>6</sup> cycles and fracture strength), and perhaps including data for synthetic composites (*e.g.* GFRPs and CFRPs).

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