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Title:

Variability of flax fibre morphology and mechanical properties in injection moulded short straw flax fibre reinforced PP composites

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Abstract

Abstract

The influence of compounding and injection moulding on the initial variability and morphology of short straw flax fibres is determined and the mechanical properties for the injection moulded fibre reinforced composites are measured. It is found that the composition of the straw flax, flax fibre bundles and woody parts, together with the cutting process strongly affects the initial fibre morphology and its variability. In the initial fibres, small particles as well as long fibres with large width were found. A filter was used to reject the fibres with an aspect ratio below 15 before calculating statistics because these fibres have a negligible contribution to the composite reinforcement. After processing, the initial fibre length and width decrease strongly (-38% to -66% for length and -22% to -72% for width). Also the variability is affected resulting in a standard deviation shifted towards lower fibre lengths and widths (-55% for length and -71% for width). The improvement of mechanical properties of the flax compound compared to the pure matrix material for the injection moulded samples is found to be similar to the results for compounds with further processed flax fibres like scutched and hackled fibres. An increase of tensile strength by 20% was found, for stiffness the increase is in the order of 50-70%. This indicates that despite the very large variability of the initial straw flax fibres and the strong changes of the variability in each processing step, a compound is obtained with improved mechanical properties.

Keywords

Short flax fibre composite, Variability, Mechanical properties, Fibre morphology

Introduction

The use of natural fibres in composites is gaining interest because of the different advantages they have compared to man-made fibres. Their carbon footprint is significantly lower compared to glass and carbon fibres. Not only during the production the environmental impact is lower but also during their lifetime[1,2]. At the end-of-life, in the recycling phase by reprocessing, the reduction in mechanical properties of natural fibres is less severe compared to man-made fibres. This is mainly due to the higher ductility which causes less fibre breakage resulting in maintaining higher length over diameter ratios or aspect ratios. The natural fibre also has the possibility to be spliced compared to glass fibres, resulting in more fibres with higher aspect ratio[1,3-6].

This study focusses on flax fibre reinforced thermoplastic polypropylene composites (FFRTPC) for high volume polymer processing techniques. Because of the low density of flax fibres (1.40-1.50 g/cm³) and the high stiffness (55-75 GPa), the specific stiffness of flax fibre is good compared to other natural fibres and similar to or even better than some man-made fibres like glass fibres. Natural fibres also have some inherent disadvantages. Due to the hydrophilic behaviour of flax fibres, the compatibility with the hydrophobic polypropylene is low. Arbelaiz et al.[7] showed that the use of maleic anhydride grafted polypropylene (MAPP) is more efficient than fibre pre-treatment to improve fibre matrix bonding. For this reason, MAPP is used in this study as a coupling agent. The inherent variability of the natural fibre geometry and properties is much higher than for, for example, glass fibres, which creates uncertainties in the design parameters.

During processing, the morphological properties of the short flax fibre change. Studies in literature focus on the effect of processing conditions on the morphology of the fibres after processing for initially scutched or hackled flax fibres[8-11]. It was found that the average fibre length decreases after compounding and injection moulding.

In this study straw flax fibres are used. Compared to the scutched and hackled flax fibres used in literature, less processing steps are needed. The more processing steps the plant has been subjected to, the purer and the finer the fibre (see Figure 1). After pulling the plant from the field, first a retting phase is necessary to reduce the amount of pectin's and lignin's in the plant. By leaving the plant on the field for several weeks, the presence of natural moist conditions like dew initiates a natural retting process. This process reduces the pectin and lignin percentage in the plant and facilitates the extraction of the fibre from the stem and improves fibre quality. After collecting the plants from the field, first the roots and the seeds are removed. Removing the seeds is also called rippling. In the breaking step, the plant is crushed between several breaking rolls which break the straw in smaller parts and loosens the interaction between the straw parts and the fibres . In the subsequent scutching phase, the broken straw parts of the stem, which did not fell off during the breaking phase, are scraped off the fibres by several rotating plates. At the end of this phase, most of the straw parts and other materials like dust coming from the field are separated from the fibres. The fibres are now still bundled together and defined as bast fibre bundles. To remove all remaining smaller impurities and separate the fibre bundles into more refined fibres and remove the shorter fibres, the scutched flax fibres are hackled by pulling the fibres through “hackles” or combs. Depending on the number of hackling cycles and the distance between the pins of the hackle, the fibre bundles are more and more separated into finer fibres. The result is the so-called technical fibre or hackled fibre.

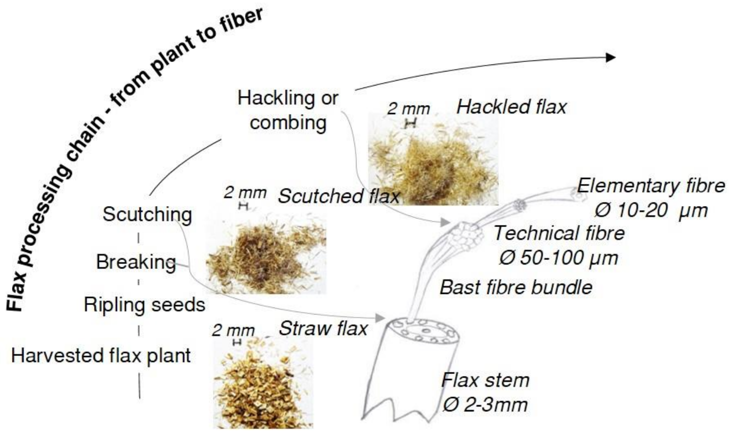

Figure 1. Composition of straw, scutched and hackled flax fibres following Bos9 and flax processing chain.

Straw flax fibres are used in this study. For straw flax, the complete stem of the flax plant is used, except the seeds (12% of the plant total[12]) and the root which are removed. The rest of the stem is cut at a length of 2 mm. After scutching almost 60% of the weight of the remaining plant (after rippling and removing the roots) is removed and forms the fraction of the shives[12,13]. In these shives some fibres are still present. It is estimated that the cut straw flax fibres are composed of about 50% shives (woody parts) and 50% flax fibres, mainly still present in fibre bundles inside the stem.

Straw flax has several advantages compared to hackled or scutched flax, not only on the aspect of cost. As measured in the present study scutched and hackled flax fibres have lower bulk density than the straw flax fibres, respectively 0.120 g/cm³, 0.072 g/cm³ and 0.144 g/cm³ for 2 mm cut scutched, hackled and straw flax. Next to the cutting also the feeding of the fibres into the compounder is easier for straw flax fibres. For the scutched and hackled flax fibres or low bulk density fibres in general, an extra processing step like premixing or pelletizing is needed to get the fibres into the matrix[11,14]. For the cutting, the stem keeps all fibres together and its stiffness makes the feeding of the fibres or plant to the cutting machine easier than for the flexible fibres. The result is a reduced production cost, not only for the production of the cut flax fibres themselves but also for the compound.

Straw flax fibres are composed of both shives and fibres. The effect on the morphological and mechanical properties is unknown. In the interest of product quality it is useful to quantify scatter on fibre morphology.

Suijano-Solis et al.[15] found (1) that the initial fibre morphology of wood fibres has an influence on the final width (diameter or the cross-sectional area) of the fibres after processing, (2) that it does not have a significant influence on the final length of the fibres, (3) while the processing conditions do affect both the final width and length of the fibres after processing. The objective of this study is to investigate the evolution in fibre morphology, including scatter in dimensions, before and after processing to quantify the effect of processing on the morphology. A proper quantification is useful for an economically interesting compound. The mechanical performance of these compounds, with their specific morphology, must be similar to the ones with the more expensive scutched and hackled fibres.

The processing parameters are set to generate the lowest possible fibre degradation. The fibre length and aspect ratio (a ratio between the fibre length and the characteristic dimension of its cross section) are crucial to have a reinforcing effect in the composite. The higher the fibre length and aspect ratio, the higher the reinforcing effect, thus improving the mechanical properties of the technical material. Not only the fibre lengths but also the strength of the fibres and the interfacial bonding between fibres and matrix play an important role in the reinforcing effect. To use the full potential of a reinforcing fibre, the minimum fibre length for fibres or critical fibre length can be determined by the model of Kelly and Tyson[16] as follows:

$$l_c = \frac{\sigma_f \, . d_t}{2 \, . \, \tau}$$

The fibre strength σ_f and its variation are strongly related to the fibre width d_t (diameter or the cross-sectional area)[17,18], obviously affecting the overall mechanical properties of the final product. MAPP is used as a coupling agent to meet the matrix-fibre bonding and thus the interfacial shear strength τ of the polymer matrix. As a rule of Thumb, an aspect ratio of 15 is accepted as a minimum required to have a reinforcing effect for a flax/MAPP composite[19]. Literature[7,14,19] shows that for similar fibre-matrix system as used here, the addition of the 3wt% MAPP coupling agent is enough to create better stress transfer from the matrix to the fibre bundles and indicating that for the aspect ratio larger than 15, determined based on analysis of interfacial strength and corresponding critical length, a considerable reinforcing effect can be found for similar fibre-matrix systems.

Besides the fibre aspect ratio, fibre orientation is one of the major aspects that affect the mechanical behaviour of fibre reinforced composites. When the load is applied in an orientation parallel to the fibre direction, mechanical properties are best. However, for short fibre filled composites produced by injection moulding, fibre orientation depends on matrix viscosity, processing parameters and mould design[20]. Fibres near the surfaces of the product show a high alignment in flow direction because of high shear forces[11] while the fibres in the core of the product are more randomly oriented.

For the compounding phase, process parameters are chosen according to Puch et. al.[14], who studied the effect of the compounding process on the mechanical properties for scutched fibres on a twin screw extruder with a premixing step. It was found that a low rotational speed, high throughputs and moderate shear energy inputs by the screw configuration lead to an optimum set of mechanical properties. For injection moulding screw rotation speed, injection flow rate and back pressure were chosen to limit further fibre deterioration[11].

Much research and development have already been done on the characterisation of different types of natural fibres (flax, hemp, jute, sisal, bamboo …), with remarkably high levels for fibre stiffness and strength. Specific strength is in the same range as E-glass and specific stiffness is even better. However, when compared to the commonly used man-made fibres like glass and carbon, mechanical properties exhibit a very high degree of scatter, with coefficients of variation in the order of 10% to even beyond 20%. This is a significant problem for the quality of the final product, as variability may become unacceptably high.

The overall objective of this research is to quantify variability at different levels of the full cycle of product design, starting with the extraction of the fibre, over the composite production chain, to the macroscopic properties of the product, using experimental identification and numerical analysis[21]. Vanaerschot et al. developed a methodology to quantify variability in the geometrical characteristics of a fibre reinforced composite: tow paths, tow cross-sections and tow aspect ratio. Although Vanaerschot et al. [21] applies the methodology for carbon fibre reinforced twill weaves the approach is useful for other fibre architectures and raw materials as well. The model which is ultimately envisaged is a quantified random field of the macroscopic component stiffness tensor, based on multi-scale modelling[22,23].

This paper focuses on the first step in this procedure: its objective is to identify variability of straw flax fibre dimensional properties throughout processing and its influence on the mechanical properties of injection moulded samples.

Production

The composition of the flax fibre compound used in this study is 20 wt.-% of straw flax fibres, 77wt.-% PP Daplen EE050AE and 3 wt.-% MAPP. The flax fibre compound is based on retted straw flax fibres, which are cut at 2 mm, provided by ABV (Algemeen Belgisch Vlasverbond[24]). The matrix is a PP copolymer, Daplen EE050AE. The density of the PP is 0.905 g/cm³ and the melt flow rate ($MFR_{230;2.16}$) is 11 g / 10 min. Further, a maleic anhydride grafted polypropylene (MAPP) (ExxonMobil Exxelor PO 1020 (MA content:0.5–1 wt%) with a density of 0.900 g/cm³ and a melt flow rate ($MFR_{230; 2.16}$) of 430 g/10 min was used. This coupling agent increases the adhesion between the hydrophilic cut straw flax fibres and the hydrophobic PP matrix[25]. Using MAPP is found to be even better than pre-treating the fibre before compounding[7]. Characterisation of mechanical properties is done using dog-bone specimens in a tensile test, according to ISO 527.

Samples were taken to determine the fibre morphology in three successive stage of the process (see Figure 2) : before compounding, after compounding and after injection moulding.

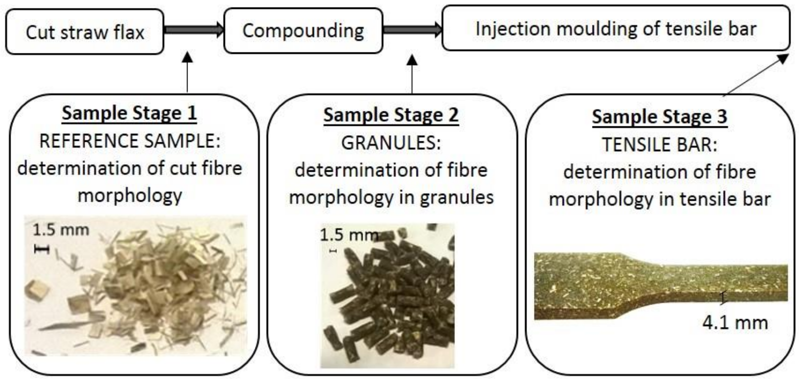

Figure 2. Schematic representation of the sequence and different processing steps and indication of the different phases in the process where samples were gathered to determine fibre morphology.

The short flax fibre reinforced composites are compounded using a co-rotating twin screw Leistritz ZSE18MAXX lab extruder. Prior to compounding, the fibres were dried for 8 hours in a ventilating hot air oven Venticell 222 at 80°C. The compound is then dried for 6 hours at 105°C and used for injection moulding of the dog-bone specimens. Dog-bone specimens are produced using an Arburg 320S injection moulding machine (processing parameters are listed in Table 1), as specified by ISO 527.

Table 1. Processing parameters for the injection moulding of the 20 wt.-% flax fibre – 77wt.-% PP Daplen EE050AE – 3wt.-% MAPP compound.

Processing parameter		
Melt temperature	[°C]	195
Injection flow rate	[cc/s]	40
Screw rotation speed	[m/min]	40
Cooling water temperature	[°C]	40
Mould temperature	[°C]	40
Back pressure	[bar]	0
Holding pressure	[bar]	500
Holding pressure time	[s]	20

Fibre morphology is quantified by fibre length and fibre width (as seen for the fibres dispersed on a flat surface, see details below) which are measured at three different stages throughout the production process. Before compounding, the length and width of the straw flax fibres are measured, with particular attention for their statistical distribution over the batch. For the granules and the tensile bars, the fibres are extracted from the PP-MAPP matrix to be able to measure the fibre length and width again, including the statistics. From the data the influence of the different processing steps on the variability of the flax fibre morphological parameters is determined.

Methods

Fibre bundle mechanical testing

Fibre bundle tests are performed on a technical flax fibre, a bundle of elementary fibres (see Figure 1). Scutching and hackling of the straw flax are needed to be able to subtract a technical fibre to conduct the fibre bundle tensile testing. The measurement of the tensile properties of fibre bundles (technical fibres) is based on the French standard XP T 25-501-3-2010: Reinforcement fibres - Flax fibres for plastic composites – Part 3: Determination of tensile properties of technical fibres. Strain is determined by measuring the displacement of the grips. Compliance analysis, according to the standard using glass fibres, is performed to take into account the error of the compliance of the machine for the calculations for strain determination of the fibre. This standard is currently only in an experimental phase.

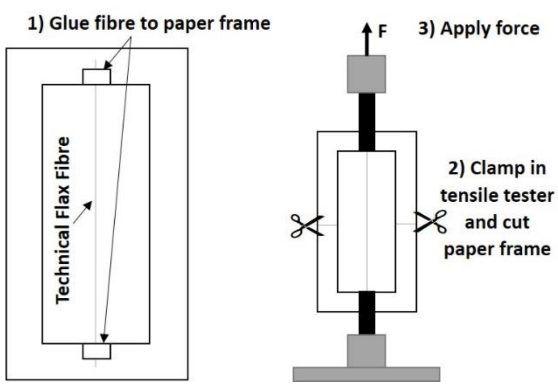

Figure 3. Tensile test on single fibre according to XP T 25-501-3-2010.

Tests are performed at a gauge length of 75 mm. A pre-tension of 1 N is applied by recommendation of the standard. The force at 0.15% and 0.35% elongation is used to calculate the modulus. According to the standard, the coefficients of variation for tensile strength and stiffness should not exceed 40% to have a reliable set of data. These measurements were conducted by Celabor[26], scientific and technical services centre, Verviers, Belgium.

Fibre dimensions

For the unprocessed flax fibres no additional steps are needed to measure the fibre morphology. For the granules and the tensile bar the fibres are extracted from the PP-MAPP matrix before the fibre properties are measured. The extraction of the fibres is done by boiling the granules in perchlorethylene (PCE) for 15 minutes. Then the fibres are filtered and transferred into a new bowl with PCE and boiled again for 15 min, placed in a permanent agitator, and filtered again.

Fibre dimensions are measured at Celabor[26]. The Occhio 500 Nano static particle analyser is used to measure the fibre morphology of the short flax fibres. The measurement system is based on image processing. Fibres are placed on a thin film. Through vacuum explosion, adapted to sticky and entangled fibres, the thin film bursts and the fibres are uniformly dispersed on a 96 mm diameter glass plate. A 6.6 megapixel 2D image is taken by the tele-centric lens which is used to analyse the fibres with the image analysis software. The geodesic length and width of the fibres are measured according to ISO 9276-6. The length and width are calculated by determination of the area A and the perimeter P of the irregularly shaped fibre on the image and by computational transformation to a rectangular bar shaped fibre with the same area and perimeter as the original fibre. The area of the fibres is computed from the visible projected area of the fibre. From the resulting bar shape, the geodesic length X_{LG} and width X_E (referred as “Thickness X_E“ in the standard) of the fibre are determined. Fibres are assumed to have a circular cross-section. The diameter or width of the fibre is defined as the dimension perpendicular to the length dimension of the fibre (see Figure 4).

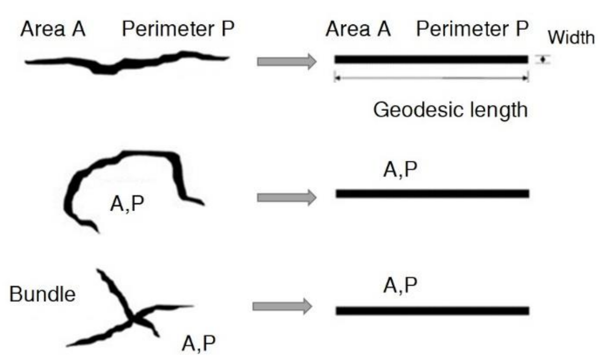

Figure 4. Principle for the identification of fibre morphology according to ISO 9276-6. Measurement of geodesic length and width of the fibres.

About 50 000 fibres are measured for each sampling stage: before the production of the compound, in the compound and in the tensile bar. Studies found in literature on fibre length measurements for short natural fibre filled compounds measure 150[11], 200[6] or 400-700[8] fibres. Using this technique many more fibres are measured and it can be assumed that a sufficiently large batch of data is obtained for reliable statistical processing.

A filter is applied on the data set. All fibres with an aspect ratio below 15 are discarded from the data which are subsequently statistically processed. Short fibres are not relevant for this study because of their negligible contribution to composite reinforcement.

Two different procedures are used to determine average fibre dimensions (width and length): the **number mean** value only takes into account the number of fibres which are identified in the image and the **volume weighted mean** also takes into account the volume of each fibre.

$$\bar{l} = \frac{1}{n}\sum_{i=1}^{n} l_i \qquad (1)$$

$$\overline{l_V} = \frac{\sum_{i=1}^{n} V_i\, l_i}{\sum_{i=1}^{n} V_i} \qquad (2)$$

$$V_i = l_i \frac{\pi {d_i}^2}{4} \qquad (3)$$

The number mean $\bar{l}$ represents the average fibre length $\bar{l}$. In composites the amount of fibres is usually expressed as a certain weight percentage in relation to a specified volume. In this particular material the number of dust particles is large and they do not represent a significant volume in the composite. For this reason, the volume weighted mean $\overline{l_V}$ is calculated. For each fibre the corresponding cylindrical volume V_i is calculated from the length l_i and width of the fibre (which is assumed to possess a circular cross-section with diameter d_i, equal to the measured width). This volume is used to calculate the volume weighted mean.

Mechanical properties of composites

Tensile and flexural tests are performed on the injection moulded tensile bars. Both tensile and flexural properties were measured on an Instron 3345 according to ISO 527-1 for tensile testing and ISO 178 for flexural testing. For both tests, 13 specimens are measured and statistically processed. For flexural testing, the data is collected for an absolute deflection of 1mm.

Results and discussion

Initial fibre statistics

The flax fibres in the compound are straw flax fibres. The fibres have not been processed, the only step done in the flax processing chain was retting of the fibre. The complete plant is used, excluding the seeds and the roots. The flax fibres are composed of woody parts or the shives and the fibres in the form of fibre bundles, technical fibres and elementary fibres[27]. Therefore a large variation in fibre width is already visually apparent in the fibres after cutting. (See Figure 5 (b)).

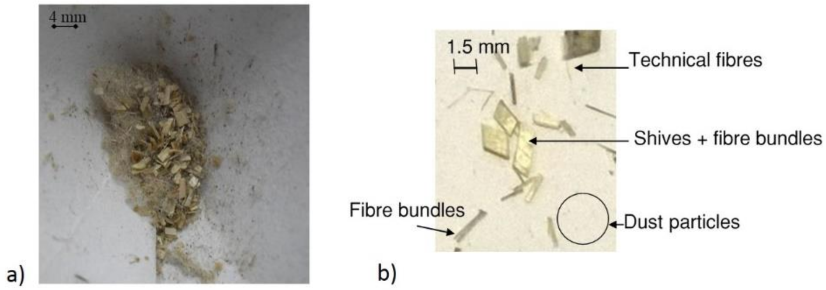

Figure 5. a) Image of fibres extracted from granules 24. b) Image of the cut straw flax fibres with visibly large variation in fibre bundle composition and width. Many smaller particles are present due to the shives because the complete plant is used.

The fibre lengths and widths of the flax fibres cut at a length of 2mm are measured with the Occhio 500 Nanoparticle analyser. Figure 6(a) and (b) show the scatter plot with marginal histograms and the probability density for the fibre length and width, respectively the volume weighted probability density for the length and width of the unprocessed flax fibres after filtering out the fibres with aspect ratio smaller than 15. After the filter, only 899 out 51815 fibres are selected for further processing representing a volume weighted percentage of 15.1% of the total fibre volume (see Table 2). In the category of discarded fibres, only 466 had a fibre width above 100 µm representing the diameter of technical fibres and fibre bundles but still they represent a volume of 80%. The large number of small particles (width < 100 µm) with an aspect ratio below 15 only represent a small volume of fibres in the composite. When looking at the percentages of the different fibre fraction based on number percentage instead of volume weighted percentage, a significant difference is observed. The number of fibres remaining after the filter represents 1.7% of the total number of the fibres which is significantly lower compared to the volume weighted percentage of 15.1%. For the fibres with an aspect ratio below 15 and a diameter below 100 µm, the number percentage is 97.4%. Compared to the volume weighted 4.5% it can be concluded that this fraction of fibres represents only a limited volume in the composite. These numbers show the importance of taking into account the volume of the fibres into statistical calculations and not only the calculations which are based on the number of fibres.

Table 2. Number of fibres in different stages of the composite processing: before filtering with aspect ratio smaller than 15 and the number of fibres remaining and their number and volume weighted percentage after filtering for the unprocessed cut flax fibres, after compounding and after injection moulding of the tensile bars split up in fibres with L/D < 15 and a diameter larger than 100 µm and fibres with L/D<15 and diameter smaller than 100 µm.

	Cut flax fibres			After compounding			After injection moulding		
Total number of fibres	51815			49641			61167		
	Number of fibres	Number %	Volume weighted %	Number of fibres	Number %	Volume weighted %	Number of fibres	Number %	Volume weighted %
L/D > 15	899	1.7%	15.1%	5570	11.2%	8.8%	2988	4.9%	8.9%
L/D < 15 & D >100 µm	466	0.9%	80.4%	1573	3.2%	87.6%	1419	2.3%	68.7%
L/D < 15 & D < 100 µm	50450	97.4%	4.5%	42498	85.6%	3.6%	56760	92.8%	22.4%

The scatter plot with marginal histograms for the unprocessed flax fibres given in Figure 6(a) shows the number calculated distribution of the fibre width and length on a log scale which is frequently used in particle analysis. Important to notice is that the distributions are not normally distributed. For both length and width, a bimodal distribution is found. An explanation for the presence of these two Gaussian like peaks on the log scale for both length and width can be attributed to the cutting of flax. The length set on the machine for cutting the flax fibres was 2 mm. For the straw flax, the complete stem is cut at once. Because the straw flax is composed of two different materials, shives and fibres, with different properties, this process has an influence on the distribution of the unprocessed fibres. The woody parts or shives are less ductile than the flax fibres themselves. Due to the high forces during cutting some shives split up and break easily into smaller pieces causing the fibre bundles to come loose from the stem. Because of the high friction between the particles themselves, induced by the cutting process, the bundles disentangle and the smaller fibres come loose from the fibre bundles and the shives. Also the remaining smaller particles from the retting process, which are still on and in the plant, together with the smaller fibres, lead to a large number of parts with a low fibre length. The mean length for the first peak of the bimodal distribution is 591 µm with a standard deviation of 242 µm. The mean fibre width for the fibres in this peak is 26 µm with a standard deviation of 9 µm confirming the presence of a large volume of technical and elementary fibres. Because the fibres themselves are more ductile and cutting is more difficult, some fibres tend to be pulled out of the stem instead of being cut, which leads to a large part of fibres with high fibre length and low fibre width, as can be seen in Figure 6(a). The fibres which are pulled out are the technical or elementary fibres which have a lower width. The mean fibre length of the second peak is 2084 µm with a standard deviation of 1020 µm. In this second peak for the fibre length many particles with high width can be detected as well (see Figure 6(a)). These are the parts of the stem with the fibre bundles inside them resulting in a second peak for the fibre width with a mean of 103 µm and a standard deviation of 56 µm.

For the scatter plot with marginal histograms for the unprocessed flax fibres by volume (weighted calculation), a more uniform distribution is found. The volumes of the elementary and technical fibres which are pulled out during cutting do not represent a significant volume in the batch. Most fibres are still bonded to parts of the stem or they form fibre bundles which is indicated by a large number of fibres having a width between 100 and 500 µm and the overall average of the fibre width being 182 µm with a standard distribution of 103 µm, which is significantly larger than the overall average of 42 µm number calculated with a standard deviation of 42 µm. These standard deviations clearly indicate that the distributions are skew rather than Gaussian. Since the cutting process is not fully optimised yet, some fibres lengths exceed 2 mm. Together with taking into account the volume of the fibres, this explains a more uniform nature of the distribution. The result is a high volume weighted mean of 3802 µm with a standard deviation of 1737 µm compared to the lower overall number calculated mean of 1242 µm with standard deviation of 1018 µm which again illustrates the skewness of the distribution.

Variation of fibre length and width after compounding

During compounding, flax fibres and matrix material are mixed. High shear forces on the fibres cause fibre breakage, breaking of the stem and the disentanglement of fibre bundles into smaller particles and more fine fibres like technical and elementary fibres[14]. The effect on the variability and fibre morphology is significant.

After extracting the fibres from the granules fibre morphology is determined (see Figure 5(a)). Again a filter is used to exclude fibres with an aspect ratio below 15 (see Table 2). 5570 fibres are retained from the 49641 measured fibres which represents 11.2% of the total number of fibres or 8.8% of the total volume of the fibres (see Table 2). From the fibres with an aspect ratio below 15, 3.2% of the number of fibres or 87.6% of the volume of fibres have a diameter beyond 100 µm, while 85.6% of the number or 3.6% of the volume of fibres are small particles with a diameter below 100 µm. The number percentage of the smallest particles is lower than for the unprocessed flax fibres (- 11.8% by number and - 1.0% by volume) which may be attributed to the dissolving and filtering of the fibres. This processing step, next to the disentanglement of the fibre bundles that results in more fibres with a higher aspect ratio, may explain why the percentage of fibres by number retained is much higher (11.2%) than for the unprocessed fibres which was only 1.7%. Despite the increase of the retained fibres in number percentage, the volume percentage of the fibres retained decreases from 15.1% for the unprocessed fibres to 8.8% for the fibres from the compound. An explanation may be the fact that the fibre bundles do not only disentangle into elementary and technical fibres but that they (together with the straw parts) also break up into smaller particles or particles with a low aspect ratio, especially the straw parts. Since the largest particles are nearly unaffected during the filtering process the volume percentage for the group of fibres with an aspect ratio below 15 and diameter beyond 100 µm is higher for this batch of fibres. Here they are represented by the straw flax particles. Together with the extraction method, this aspect explains the results found for the volume weighted percentages for the granules.

Fibre breakage caused by the high shear forces on the fibres results in a slight decrease of 1.4% of number average fibre length but with a smaller standard deviation in the results of 711 µm instead of 1018 µm for the unprocessed fibres (see Table 3). In volume percentage, the decrease is much higher, almost 44% which may be attributed to the higher number of fibres with higher aspect ratio due to disentangling and the filtering of the fibres after dissolving leading to a lower percentage of smallest particles. The disentangling of the fibres causes for both number and volume average percentage a significant decrease of 19% respectively 59% for the fibre width with both a lower scatter in the data (standard deviation of respectively 16 and 64 µm instead of 42 and 103 µm). So overall the compounding step reduces scatter in both fibre length and width properties which is also observed in the scatter plot on Figure 6(c) for the number calculated data and on Figure 6(d) for the volume weighted data. For the number calculated data the probability density shifts from a bimodal to a unimodal distribution for both width and length. Most fibres come near the fibre width of elementary and technical fibres (20-50 µm). For the volume weighted plot also a higher percentage of fibres are elementary or technical fibres but because the volume is taken into account, the few particles with high length and width representing a high volume are grouped in the upper right corner of the plot.

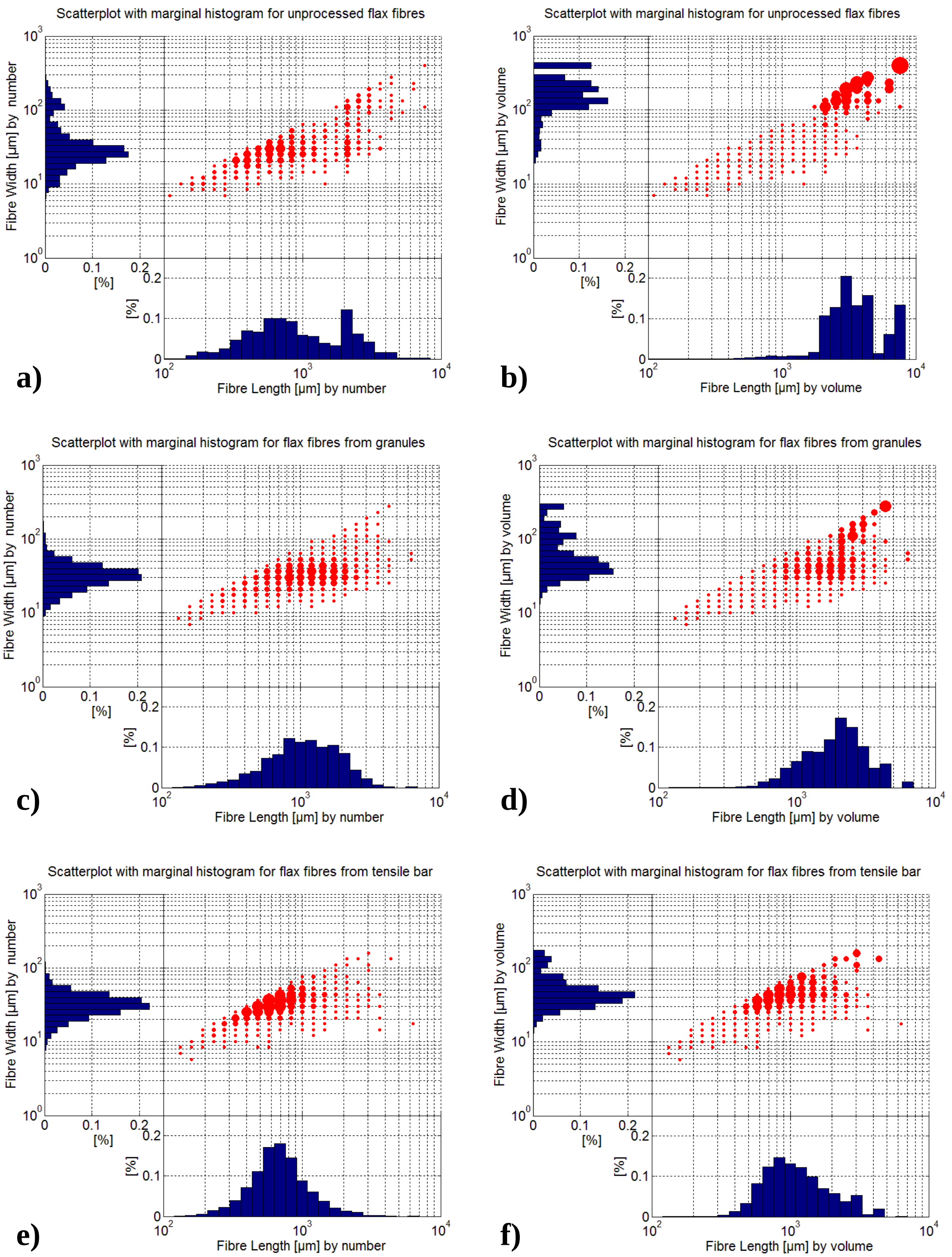

Figure 6. Log scale representations of (a) Scatterplot with marginal histogram for fibre width and length for unprocessed flax fibres based on number of fibres respectively (b) based on the volume of the fibres. (c) Scatterplot with marginal histogram for fibre width and length for flax fibres extracted from the granules based on number of fibres

respectively (d) based on the volume of the fibres. (e) Scatterplot with marginal histogram for fibre width and length for flax fibres extracted from tensile bar based on number of fibres respectively (f) based on the volume of the fibres.

Table 3. Measured morphological properties of the fibres after cutting, compounding and injection moulding.[a]

		Cut flax fibres				After compounding				After injection moulding			
		Length [µm]		Width [µm]		Length [µm]		Width [µm]		Length [µm]		Width [µm]	
		Numb.	Vol.	Numb.	Vol.	Numb.	Vol.	Numb.	Vol.	Numb.	Vol.	Numb.	Vol.
	μ	1242	3802	42	186	1224	2138	34	76	775	1294	33	52
	σ	1018	1737	42	103	711	1057	16	64	447	782	12	30
Bimodal	μ_1	591		26									
	μ_2	2084		103									
	σ_1	242		9									
	σ_2	1020		56									

[a] Mean values µ and standard deviations σ for the normal distributions and where µ1 and µ2 are the means and σ1 and σ2 the standard deviations of the two normal distributions in the bimodal distribution.

Variation of fibre length and width after injection moulding

During injection moulding the fibre lengths are even further reduced by almost 40% for both number and volume calculated fibre length averages. This result is different when compared to the compounding step (see Table 3). The standard deviations on both averages decrease even further with 37% and 26% respectively for number and volume average length. Both processing steps, compounding and injection moulding, cause high shear forces on the fibres. But during the first processing step, the compounding step, more fibres are disentangled when compared to the injection moulding step. During injection moulding fibres are even further disentangled but since it is the second time that the fibres are being loaded this effect is less pronounced. A significant part of the fibre disentanglement has already occurred during compounding resulting in only a smaller decrease of fibre width after injection moulding of 3% and 32% for respectively the number and volume calculated average fibre width. During injection moulding fibre breakage not only occurs due to plasticising but also due to the mould geometry. The compound is injected into the mould with high pressures (780 bar) causing together with the complexity of the mould (multiple corners, film gate,…) for extra fibre breakage. Together with the lower effect of fibre disentangling and because both batches of fibres had to be dissolved, this explains why the number average fibre length decreases almost 26 times more during injection moulding compared to compounding.

This effect is also reflected in the percentages of the remaining fibre fraction given by number after dissolving and filtering the fibres and applying the selection filter of aspect ratio of 15 on the measured fibres. Compared to the results after compounding which was 11.2%, the retained fibres now represent only 4.9% of the total number of fibres. For the percentage of fibres by volume only a minor difference is observed (see Table 2). Because of the high fibre breakage the percentage of fibres with an aspect ratio lower than 15 and a diameter lower than 100 now increases with 7.2% by number and even 18.9% by volume.

The scatter plots on Figure 6(e) and (f) show the shift to shorter fibres. Again for the volume weighted plot Figure 6(f), some fibres with high fibre length and high width represent a relatively large volume of the fibres but in number they are limited (see Figure 6(e)).

The overall decrease in average fibre length after processing is 38% by number and 66% by volume with a decrease in standard deviation for both around 55%. For the width the decrease is only 22% by number and even 72% by volume (see Table 3). The difference between the overall decrease in fibre length and width by number and by volume is also remarkable in the scatter plots on Figure 6(e) and (f). For the number calculated values, the difference is less distinct than for the volume weighted calculated values.

Overall it can be concluded that the different processing steps strongly affect both mean values and magnitudes of scatter. For the initial fibres a bimodal skew distribution is found for both length and width. By calculating the overall mean and standard variation of the initial fibres, it can be concluded that there is a large variability on the cut fibres for both length and width (see Table 3). This can be attributed to the use of the complete stem of the plant and the cutting process. Both aspects also strongly affect the fibre aspect ratios and the number of fibres retained after filtering. Over 98% of the fibres and particles have an aspect ratio below 15 and were discarded from the analysis. But when taking into account the volume of the fibres only 4.5% were discarded. This justifies the approach of taking into account the volume aspect of the fibres. After processing, the fibre length and width significantly decrease with over 38% by number and 66% by volume for length and 22% by number and even 72% by volume for the width. Coefficients of deviation are around 55% for fibre length and 71% for fibre width. The reduction in COV for the width shows that the initial fibre bundles present in the batch of fibres split up from the shives and disentangle into more fine fibres like elementary and technical fibres. This is also illustrated in Figure 6(e) and (f) showing most fibres with a width between 20 and 100 µm.

The final conclusion is that the combination of compounding and injection moulding affects the morphological properties of the fibres and the variability.

Mechanical properties

Fibre bundle tensile testing

The mechanical properties of the compound depend on which matrix is used and on the fibre properties like the aspect ratio of the fibres, fibre strength, fibre stiffness and the bonding between fibre and matrix. In this context the mechanical properties are determined for the flax fibre bundles (technical fibres). In literature fibre stiffness for flax fibre bundles is found to be in a range which is as wide as 10 to 100 GPa with 70 GPa most frequently published[9,28]. For the strength in literature the value is in the range between 350 and 1500 MPa with 700 MPa most frequently published[9]. The results for flax used in this study are given in Table 4.

Table 4. Measured strength and stiffness properties of the flax fibre bundles.

Gauge length	Strength		E-modulus*	
[mm]	[MPa]	COV [%]	[GPa]	COV [%]
75	769	27.8	43.5	9.9

* Between 0.15% and 0.35% strain

Tensile testing of composites

The mechanical properties in tension of the flax composite depend on the initial properties of both the matrix material and the flax fibres, and the fibre matrix interaction.

For the flax fibres not only the mechanical properties but also the variation in morphological properties[29] play an important role.

Figure 7 shows the tensile stress-strain curve for three of 13 samples of the 20 wt.-% flax fibre reinforced composite. The mechanical properties are presented in Table 5. The secant modulus at 0.2% strain is significantly higher for the flax composite, over 50%, compared to the virgin PP matrix material. The tensile strength also increases over 20%. This can be attributed to the presence of the fibres with an aspect ratio beyond 15. Since both the elastic modulus and the tensile strength of the fibres are higher than the matrix and the aspect ratio exceeds 15, the fibres have a reinforcing effect in the composite. The scatter plots in Figure 6(e) and (f) show that there are also a high number and volume of fibres with an aspect ratio beyond 30. The higher the aspect ratio of the fibres the more they contribute to the reinforcement of the composite[30]. Because the fraction of fibres having an aspect ratio higher than 15 is only 4.9% in number or 8.9% in volume of all fibres in the compound the expected reinforcement is lower than could be expected knowing that the measured properties for the technical fibres are respectively 769 MPa for strength and 43 MPa for stiffness for a 20wt.-% fibre filling. Despite the fibre shortening due to fibre breakage, the disentangling of the fibre bundles has an important effect on the stiffness of the composite. As explained previously in Table 2 and Figure 6, processing causes not only fibre breakage but also disentangling of the fibre bundles into technical and elementary fibres which result in more fibres (see Figure 6 and Table 2) with a high aspect ratio.

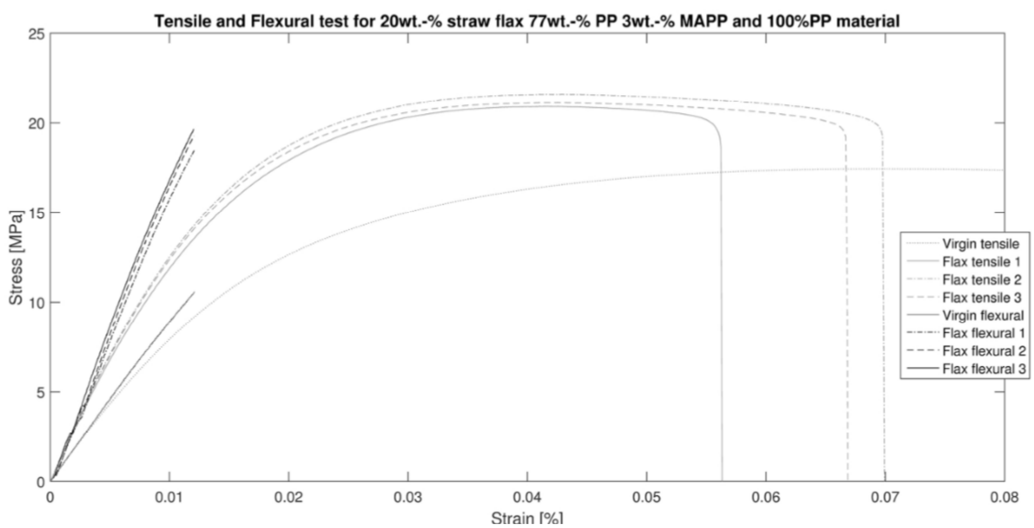

Figure 7. Tensile and flexural stress-strain curves for different samples of the 20wt.- % flax - 3wt.-% MAPP 77 wt.-% PP compound and one sample of virgin PP.

A decrease of 98% in strain at break (see Table 5) and the higher coefficient of variation for this property can be attributed to the presence of larger particles in the compound (D > 100 µm) with low aspect ratio (L/D < 15). These particles behave as impurities in the sample and induce micro cracks in the tensile bar causing fracture which results in a low strain at break.

Flexural testing

The results for the flexural modulus, measured at a deflection of 1 mm using a three point bending flexural test with a span width of 64 mm, are presented in Table 5. In Figure 7 the flexural stress-strain curves are shown for three of 13 samples of the 20 wt.-% flax fibre reinforced composite. The flexural stiffness for the flax composite is even more than 70% higher than for the virgin compound. In literature similar increases for scutched and hackled fibres are reported[4,9,14]. As shown in the fibre morphology measurements, the flax fibre material used consists of several fractions of fibres (aspect ratio > 15, dust particles with aspect ratio < 15…) and different fibre materials (shives, fibre bundles, technical fibres, elementary fibres, dust particles), leading to high variability in morphological properties of the flax inside the compound. Nevertheless, comparing with the results from literature, good results for stiffness can be achieved with this straw flax material.

When looking at the coefficient of variation for the flexural modulus a small decrease can be observed compared to the coefficient of variation for the tensile modulus. An explanation for this can be found in the integration over the cross-section of the beam when applying a tensile load. This is not present in flexural loading since in this study for a limited deflection of 1mm, only the outer layers are subjected to the forces. This indicates that the influence of the high variability in the fibre morphology is lower for the flexural properties compared to the tensile properties due to the different loading of the sample. When comparing the modulus for tensile and flexural test, an increase of 12% can be found. In the outer layers, the orientation of the fibres is higher compared to the rest of the test specimen. Figure 8 shows a micro computed tomography image for the cross section of the tensile bar indicating this high degree of alignment of the fibres in the outer layers. In the core of the sample, the fibres are more randomly oriented. This preferred orientation near the surface of the beam is caused by the high shear forces that occur during the injection moulding process[11,20]. The higher the fibre alignment in the loading direction, the better the mechanical properties are. So since in flexural testing only the outer layers are loaded compared to the complete cross section which is subjected to a load in tensile testing, with highly oriented zone (outer layers) and zones (middle of the test specimen) with a lower fibre orientation in loading direction, the modulus is higher for the flexural test.

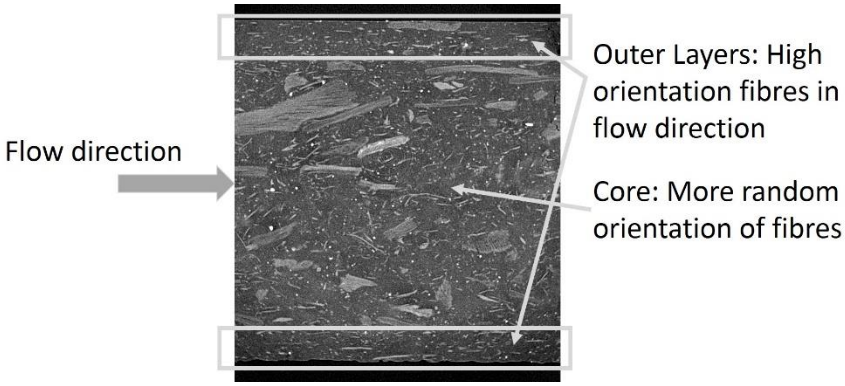

Figure 8. Cross section slice of tensile bar from micro computed thomography.

Table 5. Mechanical properties for the 20wt.-% retted straw flax 77wt.-% PP Daplen EE050AE and 3wt.-% MAPP compound.

	Virgin unfilled PP			20%Fl-77%PP-3%MAPP		
	Mean	sd.	CV %	Mean	sd.	CV %
Maximum Tensile stress [MPa]	17.6	0.4	2.3	21.4	0.4	1.9
Secant 0.2% Modulus [MPa]	939	18	1.9	1436	96	6.7
Strain at break [%]	359	12	3.3	6.3	0.7	11.1
Flexural Modulus [MPa]	926	16	1.7	1607	49	3.0

Summary and conclusions

This research work has developed a procedure to quantify the morphological characteristics of short flax fibre reinforced compounds in successive processing steps. This study shows that the variability of the initial straw flax fibres is very large and that it changes strongly in each processing step. Due to its composition of different reinforcement constituents like shives, fibre bundles, technical and elementary fibres, fibre morphology of straw flax exhibits a high degree of variability already prior to processing in the composite material. Both the compounding and the injection moulding steps strongly affect the morphological properties of the fibres (length and width). They also affect the variability of the fibres because of the shortening and disentanglement of the fibres into elementary and technical fibres. During the compounding step a first disentangling of the fibres takes place which results in a decrease of the fibre width of 19% and 59% respectively in number and volume mean. In the injection moulding step fibres disentangle even further but the effect is less pronounced when compared to compounding. The injection moulding step mainly affects the fibre length because of fibre breakage. A decrease of almost 40% of the fibre length, both for number and volume mean, is found compared to the fibre length after compounding. Even after processing a high degree of variability is still present in the fibre morphology. Nevertheless, it is observed that the coefficient of variation decreases somewhat and shifts towards smaller particles both in width and length caused by fibre breakage and disentanglement. This also explains the increase of particles with low aspect ratio.

Due to the presence of the fibres and the presence of larger parts (aspect ratio < 15 and width > 100 µm), the strain at break decreases significantly with 98% compared to the virgin material. The strength increases with over 20% and the stiffness with 50% for the secant 0.2% modulus and 70% for the flexural modulus.

Straw flax can successfully be used as fibre filling in natural fibre reinforced PP compounds to improve mechanical properties in spite of the high scatter in morphological properties. Not only the mechanical properties are similar to scutched or hackled flax fibres but also the compounding step and the cutting of the flax fibres is less difficult compared to the further processed flax fibres like scutched flax.

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Declaration of Conflicting Interests

The author(s) declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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References

1. Joshi S V., Drzal LT, Mohanty AK, et al. Are natural fiber composites environmentally superior to glass fiber reinforced composites? Compos Part A Appl Sci Manuf 2004; 35: 371–376.
2. Eberle R, Franze H. Modeling the use phase of passenger cars in LCI. SAE technical paper 982179, 1998
3. Steuernagel L, Meiners G, Ziegmann D. Recycling of fiber reinforced thermoplastics - Natural fibers vs. Glass fibers. In: TexComp11. Leuven, 2013.

4. Arbelaiz A, Fernández B, Ramos JA, et al. Mechanical properties of short flax fibre bundle/polypropylene composites: Influence of matrix/fibre modification, fibre content, water uptake and recycling. Compos Sci Technol 2005; 65: 1582–1592.
5. Le Duigou A, Pillin I, Bourmaud A, et al. Effect of recycling on mechanical behaviour of biocompostable flax/poly(l-lactide) composites. Compos Part A Appl Sci Manuf 2008; 39: 1471–1478.
6. Bourmaud A, Baley C. Rigidity analysis of polypropylene/vegetal fibre composites after recycling. Polym Degrad Stab 2009; 94: 297–305.
7. Arbelaiz A, Cantero G, Fernández B, et al. Flax fiber surface modifications: Effects on fiber physico mechanical and flax/polypropylene interface properties. Polym Compos 2005; 26: 324–332.
8. Duc A Le, Vergnes B, Budtova T. Polypropylene/natural fibres composites: Analysis of fibre dimensions after compounding and observations of fibre rupture by rheo-optics. Compos Part A Appl Sci Manuf 2011; 42: 1727–1737.
9. Bos HL, Müssig J, van den Oever MJA. Mechanical properties of short-flax-fibre reinforced compounds. Compos Part A Appl Sci Manuf 2006; 37: 1591–1604.
10. Le Moigne N , Van Den Oever M, Budtova T. A statistical analysis of fibre size and shape distribution after compounding in composites reinforced by natural fibres. Compos Part A Appl Sci Manuf 2011; 42: 1542–1550.
11. Barkoula NM, Garkhail SK, Peijs T. Effect of Compounding and Injection Molding on the Mechanical Properties of Flax Fiber Polypropylene Composites. J Reinf Plast Compos 2008; 29: 1366–1385.
12. Van sumere C. The biology and processing of flax fibres. Belfast: M Publications, 1992.
13. Bos HL. The potential of flax fibres as reinforcement for composite materials. 2004.
14. Puch F, Hopmann C. Experimental investigation of the influence of the compounding process and the composite composition on the mechanical properties of a short flax fiber–reinforced polypropylene composite. Polym Compos 2015; 36: 2282–2290.
15. Quijano-Solis C, Yan N, Zhang SY. Effect of mixing conditions and initial fiber morphology on fiber dimensions after processing. Compos Part A Appl Sci Manuf 2009; 40: 351–358.
16. Kelly A, Tyson W.R. Tensile properties of fibre-reinforced metals: Copper/tungsten and copper/ molybdenum. J. Mech. Phys. Solids 1965; 13; 329-350.
17. Aslan M, Chinga-Carrasco G, Sørensen BF, et al. Strength variability of single flax fibres. J Mater Sci 2011; 46: 6344–6354.
18. Andersons J, Poriķe E, Spãrninš E. Modeling strength scatter of elementary flax fibers: The effect of mechanical damage and geometrical characteristics. Compos Part A Appl Sci Manuf 2011; 42: 543–549.
19. Andersons J. Spârniņš. Stiffness and strength of flax fiber/polymer matrix composites.Polym Compos 2006; 27: 221-229.
20. Joseph P.V, Joseph K, Thomas S. Effect of processing variables on the mechanical properties of sisal-fiber-reinforced polypropylene composites. Comos Sci Technol 1999; 59(11):1625-1640
21. Vanaerschot A, Cox BN, Lomov S V., et al. Stochastic framework for quantifying the geometrical variability of laminated textile composites using micro-computed tomography. Compos Part A Appl Sci Manuf 2013; 44: 122–131.

22. Vanaerschot A, Cox BN, Lomov S V., et al. Stochastic multi-scale modelling of textile composites based on internal geometry variability. Comput Struct 2014; 122: 55–64.
23. Olave M, Vanaerschot A, Lomov S V., et al. Internal geometry variability of two woven composites and related variability of the stiffness. Polym Compos 2012; 33: 1335–1350.
24. A.B.V. Algemeen Belgisch Vlasverbond http://www.vlasverbond.be/. (2015).
25. Graupner N, Rößler J, Ziegmann G, et al. Fibre/matrix adhesion of cellulose fibres in PLA, PP and MAPP: A critical review of pull-out test, microbond test and single fibre fragmentation test results. Compos Part A Appl Sci Manuf 2014; 63: 133–148.
26. CELABOR. CELABOR http://www.celabor.be/ (2015).
27. Charlet K, Jernot JP, Eve S, et al. Multi-scale morphological characterisation of flax: From the stem to the fibrils. Carbohydr Polym 2010; 82: 54–61.
28. Baley C. Influence of kink bands on the tensile strength of flax fibers. J Mater Sci 2004; 39: 331–334.
29. Cocca M, Avolio R, Gentile G, Di Pace E, Errico M.E, Avella M. Amorphized cellulose as filler in biocomposites based onpoly(ε-caprolactone). Carbohydr Polym 2015; 118: 170-182.
30. Avolio R, Graziano V, Pereira Y.D.F, Cocca M, Gentile G, Errico M.E, Ambrogi V, Avella M. Effect of cellulose structure and morphology on the properties of poly(butylene succinate-co-butylene adipate) biocomposites, Carbohydr Polym 2015; 133: 408-420.