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Study of the fibre morphology stability in polypropylene-flax composites

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ABSTRACT

The objective of this paper is to understand the impact of the fibre morphology and process parameters on the final stability and microstructure of polypropylene-flax fibres composites. In a first part, we showed that after various extrusion or injection cycles, fibre length and aspect ratio are quite similar, no matter the initial length. This point is crucial for the choice of initial fibre length and fibre incorporation easiness. Moreover, the values of elastic modulus and stress at break have revealed the importance of the fibre dispersion to obtain performing mechanical properties. In the second part, we studied the influence of the vegetal fibre volume loading on the fibre morphology and on the composite mechanical properties. The aspect ratio of the plant fibre after extrusion and injection was measured. After a good stabilization for low fibre loadings, it decreases, from a critical threshold around 30-vol %, as fibre content increases due to the shear rate increase. In the same time, we showed that the mechanical properties of the injected composites are optimal around this threshold. Finally, the study of the injected composites morphology evidenced differences in orientation between flax and glass fibres, which could explain the shrinkage properties of the different composites.

1. Introduction

Glass fibres make up more than 95% of the reinforcements used in the composite industry [1]. The use of agricultural resources for the manufacturing of structural materials has been practiced for centuries (cellulose, mud, concrete …) and has incited a renewed interest in recent years, especially in the automotive industry [2]. This use is justified by environmental and especially economic considerations. Anything that can be done to limit human impact on the environment and to liberate it from its dependence on oil is currently strongly encouraged. In this context, composites made with natural fibres are developed to replace some conventional composites based on synthetic fibres. Indeed vegetal fibres are renewable resource materials requiring low non renewable energy to be produced and can store carbon dioxide due to the photosynthesis phenomenon [3,4]. Moreover, they have a low density allowing for the reduction of environmental impact of vehicles [5]. At end of life, they are biodegradable which can contribute to a healthy ecosystem [6,7] and, unlike to glass fibres, can provide energy once incinerated (19.47 MJ/kg during their combustion against −1.7 MJ/kg for glass fibres) [8]. These environmental advantages are combined with high mechanical properties [9,10] showing that vegetal fibres are good candidates for the substitution of glass fibres in thermoplastic reinforcement. CO_2 emissions over a lifecycle are thus reduced [11]. Natural fibres, unlike glass fibres have less impact on the health of composite manufacturers (irritation of the skin, lung cancer) [12]. However, some manufacturing processes producing natural fibres are known to produce large quantities of dust which can be harmful in the long term for the production staff [13]. Moreover, the use of vegetal fibres for composite reinforcement could have some limits, due to their poor thermal resistance or their water sensibility.

Our study focuses on flax fibres. Flax is grown primarily for its fibre content. This spring culture is harvested about one hundred days after being sown. The processing steps are highly dependent on climate and consist in sequence of uprooting the whole plant, laying on the ground for retting, turning it for the homogenization of retting, drying of the stems and then scutching. The flax fibres obtained in this way are then cut to the desired length.

Thermoplastic composites reinforced with plant fibres have good mechanical properties and good recycling properties [14–17]. However, the mechanical performances of such composites are not yet equal to thermoplastic composites reinforced with glass fibres. Plant fibres have a great interest to be coupled with biodegradable matrixes like PLLA or PHA, due to the composite biodegradability; nevertheless, for intensive automotive applications, these matrixes

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exhibit some limits due to their poor thermal stability. For these reasons, this work focus on polypropylene flax fibres reinforced composites.

Different fibre parameters can influence the mechanical properties of polypropylene composites reinforced with vegetal fibres. Their differences of structure, microfibrillar angle, aspect ratio and chemical composition may lead to differences in mechanical properties [18]. Absence or presence of fibre bundles is also a major factor to understand to behaviour of the composite. Similar conclusions have been highlighted by Klason et al. [19] on various thermoplastics reinforced by wood or cellulose flour; they showed the importance of the reinforcement homogeneity, to obtain good mechanical properties. In the case of glass fibres, short and long fibres give composites with different behaviour: at molten state, long fibres are able to structure as a network inducing yield stress [20]. At solid state, bundles can be observed for long fibres with mechanical properties strongly different compared to composites made with short fibres [21]. Thus, bundles can modify the damage mechanisms [22]. For composites made with short glass fibres, they are not organized in bundles or networks and they do not exhibit yield stress at molten state. The fibre content is also a parameter which influences mechanical properties [23–26]. It is important to determine the optimum fibre volume fraction to obtain materials with good properties [27]. The mechanical properties increase linearly with fibre content. However, some authors have reported a stabilization of properties from a certain amount of fibre (40%).

The aim of this paper is to understand the importance of the initial fibre length or volume fraction and the process parameters on the composite final mechanical and dimensional properties. In the first part of this work, the consequences of the thermo mechanical degradation induced by different extrusion or injection cycles on the fibre morphologies and composites mechanical properties were highlighted. The second part of the study was dedicated to the study of influence of the fibre loading on the fibre morphological parameters and on the plant fibre composite mechanical properties. To conclude, these results have been then compared to glass fibres composites morphology and the impact of the fibre loading and orientation on shrinkage has been measured.

2. Experimental

2.1. Materials

The polypropylene (PP) used in this study is PPC 10642 supplied by Total Petrochemicals with an MFI of 44 g/10 min (at 230 °C and 2.16 kg). A polypropylene modified with maleic anhydride (PPgMA) was also used as a coupling agent to improve the fibre/matrix adhesion. Maleic anhydride grafted polypropylene is widely used as a coupling agent in polypropylene composites reinforced with natural fibres [28]. This agent supplied by Arkema (Orevac CA 100) with an MFI of 10 g/10 min (at 190 °C and 0.325 kg) was added to the matrix at a 4%-wt loading. In order to compare flax fibre filled PP and regular glass fibre reinforced PP, a 13.2-vol% chemical coupled glass fibre reinforced PP-homopolymer named POLYFORT® FPP 30 GFC and supplied by A. Schulman has been characterized and studied.

The flax fibre used in this study is from the Marilyn variety and comes from a 2003 harvest cultivated in Normandy (France). Flax fibres selected are representative and have known and reproducible properties. Each step in the development of the fibre is mastered and controlled. To get the fibres, the different steps are: pulling the plants and dew retting on the ground, scutching to extract the fibres and then cutting at three different lengths: 0.5, 1 and 2 mm. The scutching and cutting steps initiate the plant fibre bundle division; this individualization will continue during the processing phase and could be optimized according to the chosen processing way or the possible end of life recycling [14].

2.2. Composite processing

To study the influence of fibre content on fibre morphology and after molding shrinkage, we have made reinforced composites at different volume fractions (3, 6.1, 13.2, 20, 32.4, 36.9 and 46.7%). Flax fibres were dried under vacuum at 60 °C during 12 h and PP was then extruded with PPgMA. Composites were extruded using a single screw extruder at a temperature of 190 °C and an extrusion speed of 20 rpm. The single screw extrusion machine is a FAIREX extruder. The length and the diameter of the screw are 600 and 20 mm, respectively inducing an L/D around 30. The approximate residence has been measured by using colour polymer; for an extrusion speed of 20 rpm, it is 1′15″. After a granulation step, normalized samples were injected on an injection molding press Battenfeld 80 tons with a mold temperature of 30 °C and a barrel temperature of 190 °C; the nozzle die diameter of the injection molding press is 2 mm. The injection pressure was 1100 bars and the cooling time was 20 s. Thus, we obtained randomly dispersed short fibres composites. Mold shrinkage was determined in flow direction, on tensile specimens using ASTM D955-96, 48 h after the injection molding. The shrinkage results are averages from ten measurements on tensile specimen 48 h after injection.

2.3. Morphological analysis

To measure fibre aspect ratio (ratio of length to diameter) we used an optical microscope Leica (Microsystems, Wetzlar, Germany) and the image analysis software LEICA QWIN, we measured the length and diameter of around 2 00 fibres and the aspect ratio before processing, after extrusion and injection. The values were obtained after an extraction of the PP matrix in boiling xylene during 24 h. To determine the orientation and distribution of fibres in the composites, observations of polished surfaces of samples injected were performed in the flow direction and in the transverse direction as shown on Fig. 1. These photographs were taken using a scanning electron microscope Jeol JSM 6460LV. The tensile fracture surfaces were analysed by scanning electron microscopy (SEM). The samples were sputter-coated with a thin layer of gold in an Edwards Sputter Coater and analysed with a Jeol JSM 6460LV electron scanning microscope.

2.4. Tensile characterizations

Tensile tests on composites were performed on a tensile machine type MTS Synergie RT/1000 at room temperature with a sensor strength of 10 kN. The speed of the ram was 1 mm/mn. An

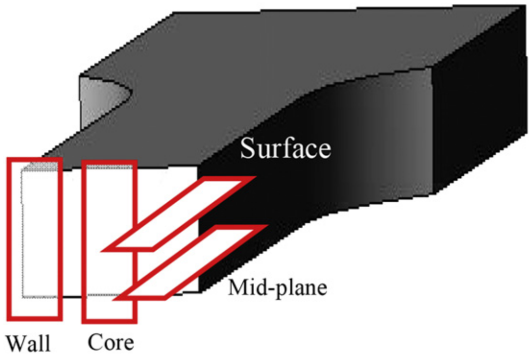

Fig. 1. Schema of cutting planes.

extensometer with a nominal length of 10 mm was used. For the virgin PP-g-MA, the strain of the specimens is above the limit of the extensometer. The extensometer was used only in the elastic area of specimens to determine the Young's modulus. The values presented in this report are averages from five reproducible tests.

2.5. Longitudinal shrinkage measurement

Longitudinal shrinkage measurements were performed on injected specimens. According to the EN ISO 294-4 European norm, the samples length was measured 48 h after injection molding and compared to the initial mold length.

The longitudinal shrinkage $S\%$ was obtained by using the following formula:

$$S\% = \frac{(L_0 - L)}{L_0} \times 100$$

where L_0 is the initial mold length and L the sample length 48 h after injection molding.

3. Results and discussion

The key point of this paper is the optimization of the process and fibre parameters, in order to obtain PP plant fibre composites with a high level of mechanical performances. We used a PP matrix due to its low-cost, good mechanical strength and Young's modulus, excellent fatigue properties or for its good temperature resistance and easiness of processing. For all of these reasons, this material is commonly used in automotive industry. In this way, we search to define the better fibre length, process cycle parameters and volume fraction, and so as to improve the composites properties. The first experimental section is dedicated to the impact of the stages of extrusion and injection on the fibres dimensions and the composites mechanical properties.

3.1. Effect of initial fibre length and extrusion process

It is now obvious to notice that processing has a direct impact on fibres dispersion and morphology; in some cases, bundles of fibres can be observed which means that the dispersion of fibres is not complete [18]. The question is to know if two passages in the extruder make more single fibres and better reinforcement. Composites with shorter initial fibre length have been prepared by using single screw extrusion. In the literature, polymer and vegetal fibres are mixed with a twin-screw extruder. Nevertheless, some authors [29] highlight single screw extrusion interest in order to reduce the shear rate and preserve the fibre integrity; in this study, single screw was preferred to twin-screw extruder for this reason. Most of the composites had large content of fibres and we had difficulty to add large amounts of fibre into the twin-screw extruder. Of more our objective is to mix the matrix and the fibres by respecting at best the integrity of fibres. Bigg [30] clearly showed that single screw does less damage to glass or carbon fibres than a twin-screw.

First, fibres were cut at 2 mm, 1 mm, and 0.5 mm. The average lengths for a sample of 100 fibres were measured at 1.98 ± 0.18 mm, 1.07 ± 0.14 mm, and 0.42 ± 0.07 mm. These fibres have been blended with PP and extruded once or twice before injection. Fig. 2 shows the effects of initial fibre length and number of extrusion or injection cycles on average fibre length (a) or aspect ratio (b).

We observe that the first extrusion drastically reduces the fibre length, particularly for the longer ones (−77%). The decrease in fibre length after compounding is always important with 1 mm fibres (−67%) but highly reduced with 0.5 mm fibres (−4%). After this first extrusion, to a lesser extent, the following steps will also reduce the fibre length. There is a tendency towards a plateau, which in this study is estimated around 185 μm. Whatsoever the initial length, the final one is around this plateau showing that the shear rate has no effect on the fibre length under this critical length. Due to the difficulty to introduce long fibres, this phenomenon is interesting information for the compounding step. The study of aspect ratios of the fibres provides further information. The first extrusion step separates the individual fibres inducing aspect ratios greater for shorter fibres (Fig. 2b). Then, the second extrusion and the injection step reduce the aspect ratio again. Due to the conjugated reducing in length and diameter, the aspect ratio decrease is more moderate than those of length. In the case of short fibres, the first process step has a positive effect by increasing the aspect ratio thanks to the bundles division associated to a poor length decrease. After the injection, fibre length and aspect ratio are quite similar no matter the initial length.

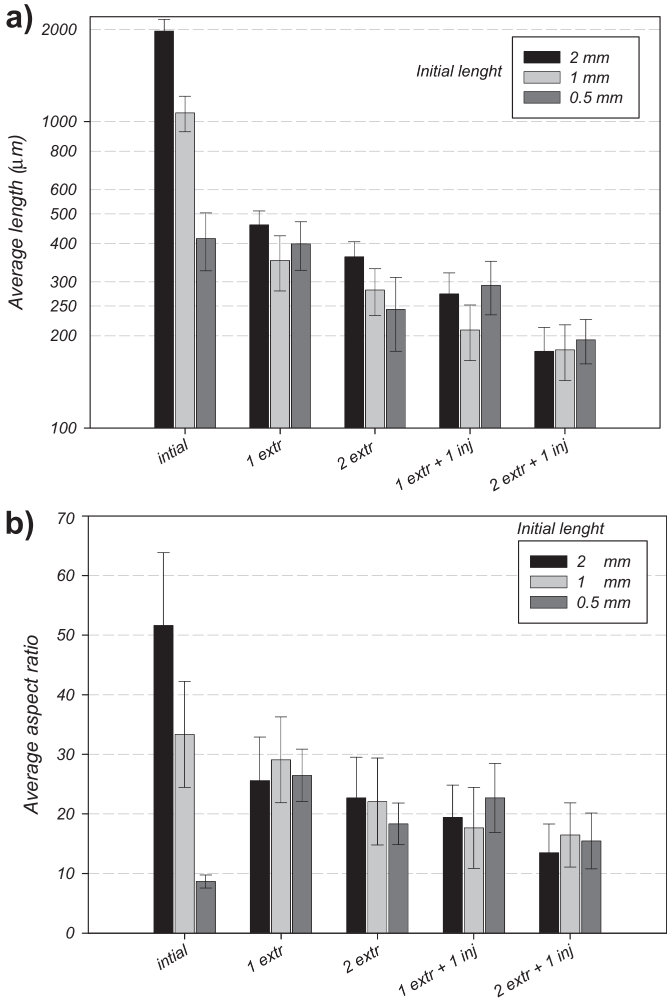

Fig. 2. Effect of initial fibre length and number of extrusion or injection cycles on average fibre length (a) and aspect ratio (b) (fibre morphologies obtained on PP/13.2-vol% flax).

Nevertheless, we find that mechanical properties are quite different. Fig. 3 shows the effect of initial fibre length and number of extrusion passage on Young's modulus (Fig. 3a) and stress at break (Fig. 3b). For 1 or 2 extrusions, the longer fibres (2 mm) give the lower Young's modulus. We can suppose than, due to their high initial length, these fibres are subjected to more important shear rate than shorter ones, inducing a structure alteration of the cell walls. A previous work of our team evidenced an important

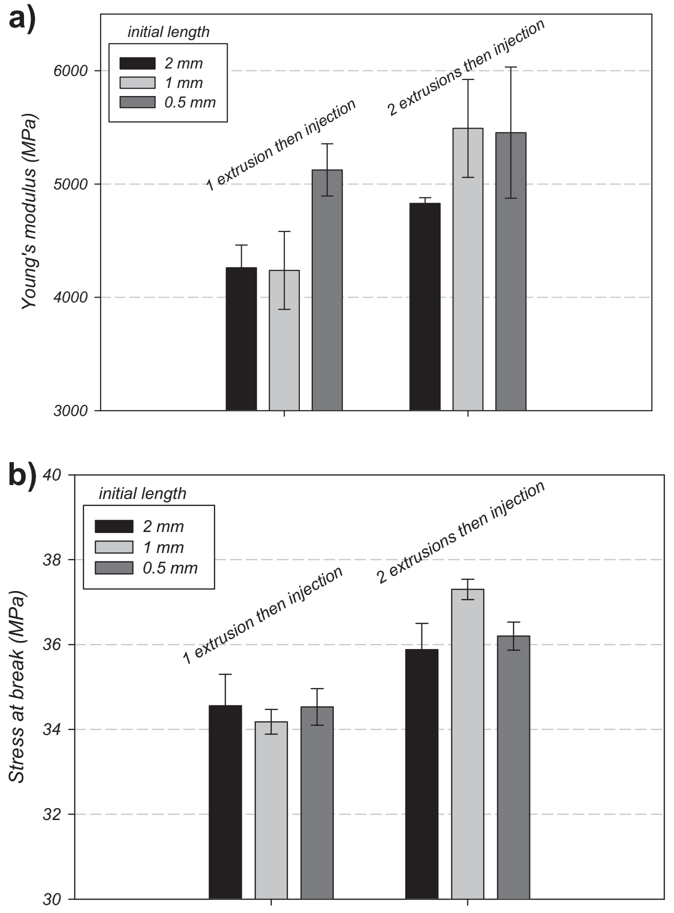

Fig. 3. Effect of initial fibre length and number of extrusion passage on Young's modulus (a) and stress at break (b) (fibre morphologies obtained on PP/13.2-vol% flax).

decrease of cell wall mechanical properties of 4 mm fibres, after a compounding and injection cycle [31].

The effect of the second extrusion stage is also well shown. The second extrusion slightly reduces the fibre aspect ratio but induces a higher Young's modulus and stress at break. Despite a smaller average aspect ratio, the Young's modulus after 2 extrusions is higher than with only one extrusion. This second extrusion improves fibre dispersion in the matrix. At the wall, several photos taken with SEM have been put together and analysed to get a black and white picture through the thickness of the sample. Pictures obtained for fibre initial length of 0.415 mm after one and two extrusions are shown on Fig. 4. Numbers of particules have been automatically counted. The number of particles is 1612 for one extrusion and 2038 for two extrusions. Even if some bundles are always remaining, it shows that the second extrusion has a strong effect on fibre dispersion. Similar results have been observed for fibre initial length of 1.068 mm and 1.976 mm. However, after two extrusions, some bundles remain and the dispersion of single fibres is not fully achieved. Fibre dispersion is an important factor to get good mechanical properties and it will be done in the processes of mixing and shaping. The degradation of bonds and pectic cements between the individual fibres is carried out mainly during the retting and extraction of the fibres; so, the ability of fibres to disperse depends on the level of retting which must be well understood and controlled. The process of dispersion and degradation of the fibres in a melted thermoplastic under flow must also be better understood.

These different results highlight the interest of a reprocessing for the fibres division and the optimization of the plant fibre composites mechanical properties; nevertheless, the use of two successive extrusion steps is not sui to an industrial productivity goal. In the next part, we will study the impact of the volume fraction fibre on the aspect ratio and on the mechanical properties to define an optimal volume fraction.

3.2. Impact of the fibre volume fraction on the fibre aspect ratio and composites mechanical properties

To better understand the process of reinforcement induced by the flax fibre volume fraction in the polymer, we studied the fibres morphology. We focus our attention on the fibre lengths, aspect ratio, distribution in the cross section and their orientation. All of these parameters participate to the reinforcement. Specimens with various flax fibre volume fractions have been analysed to determine these parameters and a comparison with glass fibres composites has been done. The end of this section is dedicated to the shrinkage measurement which highlights the impact of the fibre distribution and orientation.

Fig. 5 shows the evolution of the length, diameter and aspect ratio (length/diameter) of flax fibres after extrusion and after injection for different volume fractions. The initial length of the fibre was 1.98 ± 0.18 mm with an average aspect ratio of 51.8 ± 12.2. We can notice an important decrease of the flax fibres length (Fig. 5a) with the fibre content after extrusion as well as injection molding. The addition of fibres in the matrix increases the viscosity of the material and then increases shear stress during processing due to the large amount of fibre which involves more fibre ruptures. As evidenced in previous work [31], the main fibre length degradation occurs after the compounding step, due to the important initial length. The extrusion process causes preferentially damage to the longer fibres. This length decrease depends on the initial fibre length and the viscosity of the polymer matrix [32].

In the same time (Fig. 5b), the extrusion process induces a slight decrease of the aspect ratio. The aspect ratio plays a role in the properties of load transfer between the fibre and the matrix [33]. This decrease after the extrusion process is more significant than after the injection cycle due to the presence of many big bundles at the beginning of the processing. These bundles are due to the flax stems morphology. In the stems, the elementary fibres are located at the periphery, and assembled by bundles of several dozens. These elementary fibres are linked by pectic cements [34] which are damaged during the retting phase in order to separate the fibres, this separation process could be carried on during the extrusion or injection steps thanks to the important shear rate suffered by fibres. These results have also been observed by Arbelaiz et al. [25] or Bourmaud et al. [14]. After one extrusion and injection cycle, the fibre diameters are between 19 and 24 μm (Fig. 5a) meaning that fibres are still made with several single fibres. A complete individualization could probably be obtained by using more aggressive process parameters, but in this case an important cell wall damage would happen [31].

We can notice that, after a slight decrease of the aspect ratio for low fibre content, a threshold could be highlighted around a volume fibre fraction of 30%. We can consider this threshold as the critical aspect ratio; this aspect ratio depends on fibre intrinsic characteristics, matrix properties and quality of fibre/matrix adhesion. Above this critical ratio, the best reinforcement and mechanical properties, such as strength and toughness, are obtained. In literature, some papers determine a vegetal fibre critical aspect ratio for PP-flax [35] or PP-hemp [36]. In the case of PP-flax [35], Le Duc et al. found a critical aspect ratio of 27 while our value is only around 18 due to the lower length of our fibres (2 mm against 10 for Le Duc et al. fibres). From this threshold, the decrease of the aspect ratio is much more important after extrusion as well

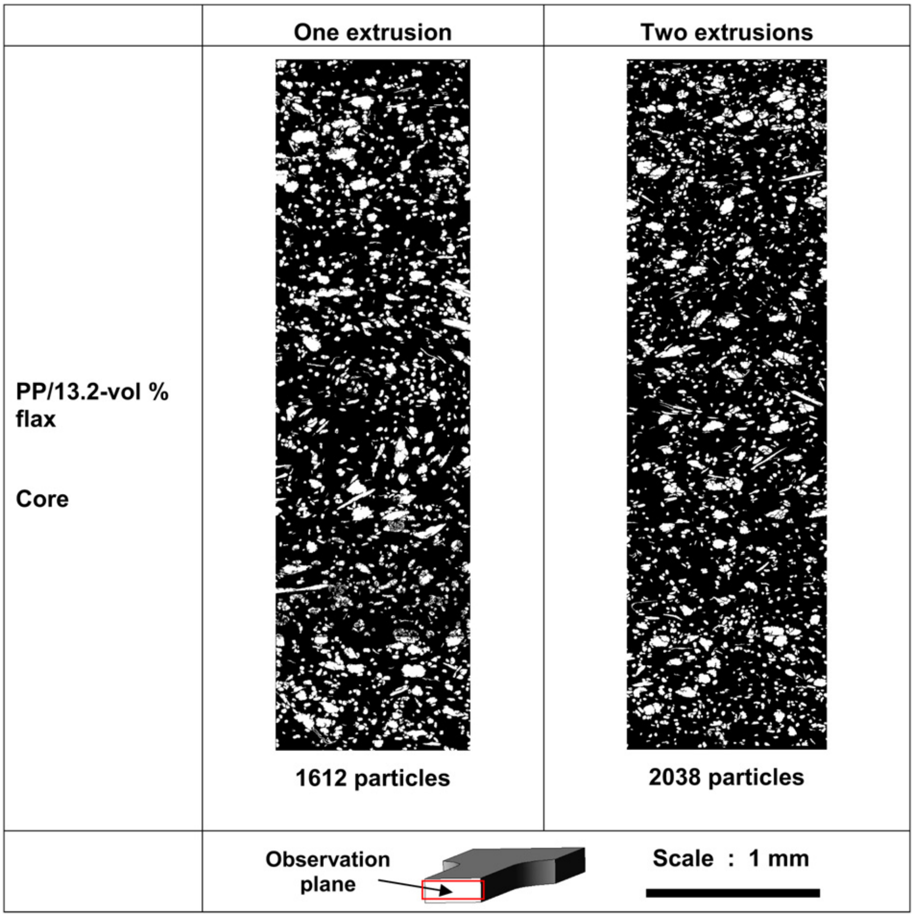

Fig. 4. Particles at the wall through the thickness for one and two extrusions (on PP/13.2-vol% 2 mm flax fibre).

as injection. We can explain this phenomenon by the important fibre length decrease due to the important shear rate induced by the high fibre volume fraction.

In complement, and for the various composites made with flax fibre, the effect of fibre content on the mechanical properties was investigated. The Young's modulus and the tensile strength of polypropylene composites reinforced by flax fibres are higher than the values obtained for the polypropylene matrix, highlighting the strengthening mechanisms induced by plant fibres. The Young's modulus, the tensile strength and elongation at break of polypropylene composites reinforced with different weight fractions of flax fibres are presented in Fig. 6. The values of elastic moduli increase with fibre concentration. This trend has also been confirmed by John et al. [37], Baiardo et al. [38] and Joseph et al. [39]. In contrast, the tensile strength increases with the fibre until it reaches a maximum value at a mass fraction of about 32% (threshold on Fig. 6a). From this value, the tensile strength stabilizes, due to the concentration of strain within the matrix between fibres. Arbelaiz et al. [40] also show that resistance increases with the fibre up to 30–40% by mass. With high fibre content, the value of resistance decreases linearly (Fig. 6b), suggesting a drop in load transfer between matrix and fibres. This behaviour has also been showed by other authors for polypropylene composites reinforced with lignocellulosic fibres [25,26,33]. This trend can also be explained by a strong decrease of fibre aspect ratio for volume fraction above the threshold (Fig. 5b). Moreover, the elongation at break of PP-flax fibre composite decreases significantly compared to the value recorded for the pure matrix and stabilized from a mass rate of 30% (Fig. 6b). This drop is generally explained by the concentration of deformation between fibres.

The composite reinforced polypropylene with 13.4% by volume of fibre glass has a modulus of elasticity of 6200 MPa. The value of the polypropylene composite/flax being equivalent volume of 4260 MPa, the PP/glass is twice as efficient as the PP/flax. Properties of flax fibre reinforced PP have been compared to a composite made with glass fibre for the same fibre weight fraction. Young's modulus of composite with glass fibre is more than twice that obtained with flax fibre. However, there is still the issue of fibre matrix bound and fibre orientation and dispersion, which will be discussed in the next section.

Bledzki et al. [41] shows that the load transfer between fibres and matrix does not depend only on the intrinsic properties of fibre and matrix but also on parameters of geometry (aspect ratio) and arrangement of fibres in the matrix as the fibre distribution. In an injection process, the viscosity of the fluid is strongly linked to the microstructure induced by the flow conditions, thus affecting the physical properties of the final product [42]. Thus the mechanical properties can be explained by the dispersion and orientation of fibres in the polypropylene matrix.

These results show clearly the impact of the fibre volume fraction on the fibre morphology and on the composites mechanical properties. A limit threshold could be defined around 32%-vol; from these values the stress at break and the aspect ratio begin to decrease inducing a lower reinforcement. This fibre volume fraction could be assimilated to an optimal value for plant fibre composites.

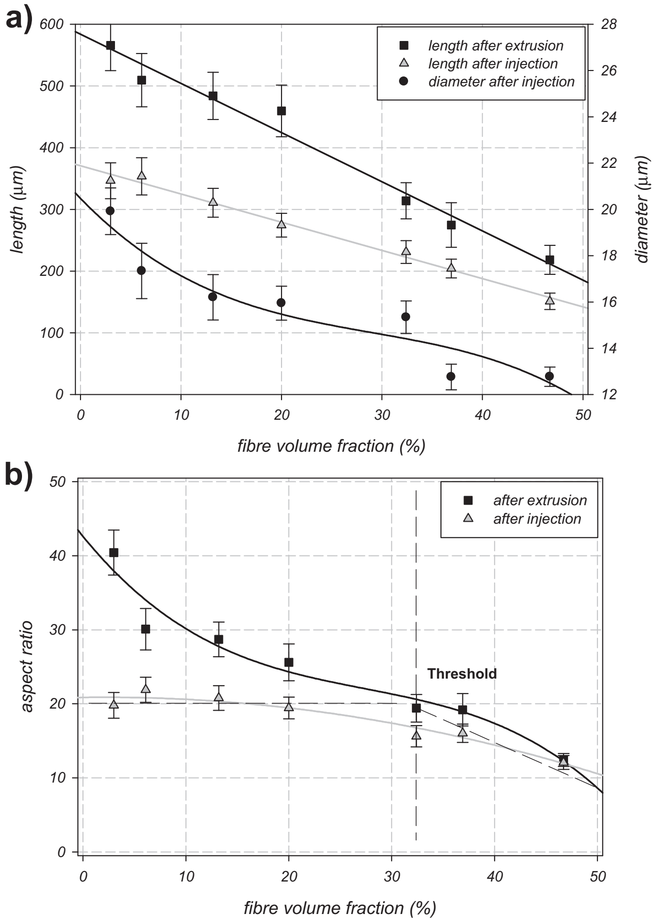

Fig. 5. Evolution of the length, diameter a) and aspect ratio b) of fibres with 2 mm fibre content.

In the last part of this work, we will study the flax fibre composite microstructure. In order to obtain significant length and aspect ratio, we focused on 13.2-vol% flax fibre composites.

3.3. Study of the composites morphology and mold shrinkage

A large number of pictures have been taken to observe fibre distribution and orientation. Samples have been polished according to the schema shows on Fig. 1, then pictures have been taken with SEM. Figs. 7 and 8 present microscopic observations in the plane perpendicular to the flow direction of tensile specimens reinforced with different volume fractions of flax fibre. These observations were made at two different levels through the thickness of the specimen, at 1 mm depth from the skin and at the centre.

The orientation of synthetic fibres and the existence of a skin-core effect on injected parts have been demonstrated in numerous publications [43–46]. However, as shown on Fig. 7, and due to stiffness and morphology differences, the transition between each area with high degrees of orientation is less clear for flax fibres than for glass fibres. As evidenced in a previous work [18], near to the skin, the glass fibres are highly orientated in the flow direction (Fig. 7), highlighting the formation of a frozen layer, or fountain effect, of the polymer during the mold filling. In the centre of the core layer, the glass fibres exhibit an orientation which is perpendicular to the flow, particularly at the centre of the part; this phenomenon is due to a divergent flow. In the case of PP/flax fibres, the skin-core effect is always present, and a relative good alignment of the flax fibre with the flow direction can be noticed in the skin layer (Fig. 8). Nethertheless, this orientation is less obvious than for the PP/glass fibres. The difference in longitudinal stiffness of flax fibres (54 GPa) and glass (72 GPa) for a same fibre volume fraction may partly explain this observation; another explanation could be fibre morphology and the presence of bundles, since the

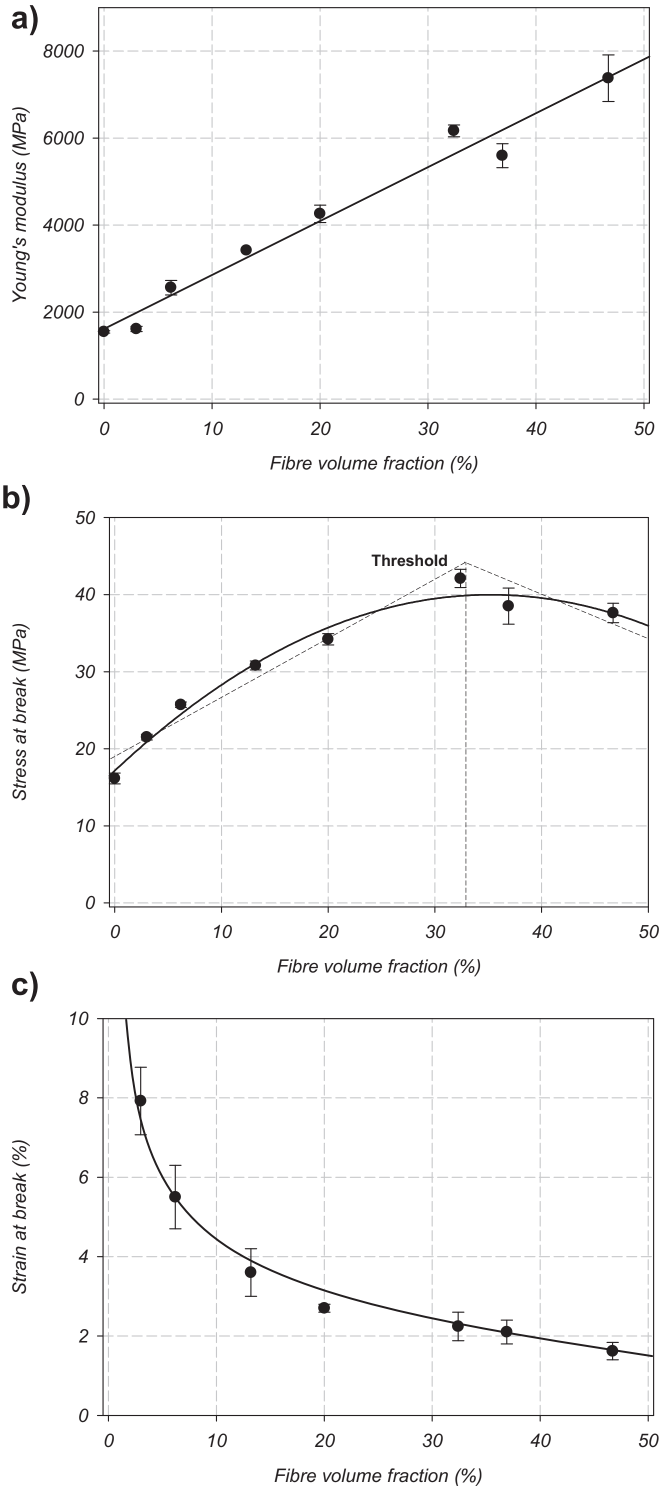

Fig. 6. Evolution of the Young's modulus (a), stress (b) and strain (c) at break of composites flax fibre with 2 mm fibre content.

fibres with high aspect ratio are prone to bending. In the skin layer, the fibres seem to be less aligned in the centre of the specimen. As already noticed in PP/glass fibres, this phenomenon is less obvious on the edge probably because of the decrease in speed in these areas.

This fibre orientation could have an influence on the parts dimensional properties. Fig. 9 shows the evolution of the longitudinal shrinkage of PP composites according to the nature and the fibre loading rate. We can notice the fibre loading interest, compared with virgin PP. In the case of flax fibre, the longitudinal shrinkage decrease with the loading rate, from 0.7 to 0.36%, corresponding to 3 to 36.9%-vol, then it stabilizes around 0.35%, whatever the flax content. This value could be compared with the results of Santos et al. [47]. They compared the shrinkage of virgin and curaua fibre loaded PA; they found an important decrease of the shrinkage with the vegetal fibre incorporation. The shrinkage value for the PP-glass composite is lower than with flax fibre (0.22% against 0.35%). We can explain this difference by the fibre orientation. In the case of glass, as explained previously, fibres close to the skin are well orientated with the flow direction (Fig. 8) but the phenomenon is less clear for flax fibres, inducing a higher shrinkage value. The lower wall orientation of the flax fibres induces a limit into the shrinkage value; in order to increase the flax fibres potential as dimensional stabilizer, an optimization of the molding parameters and of the mold geometry have to be developed. In a forthcoming work, we will study the morphology and mechanical properties of injected unidirectional flax fibre composites.

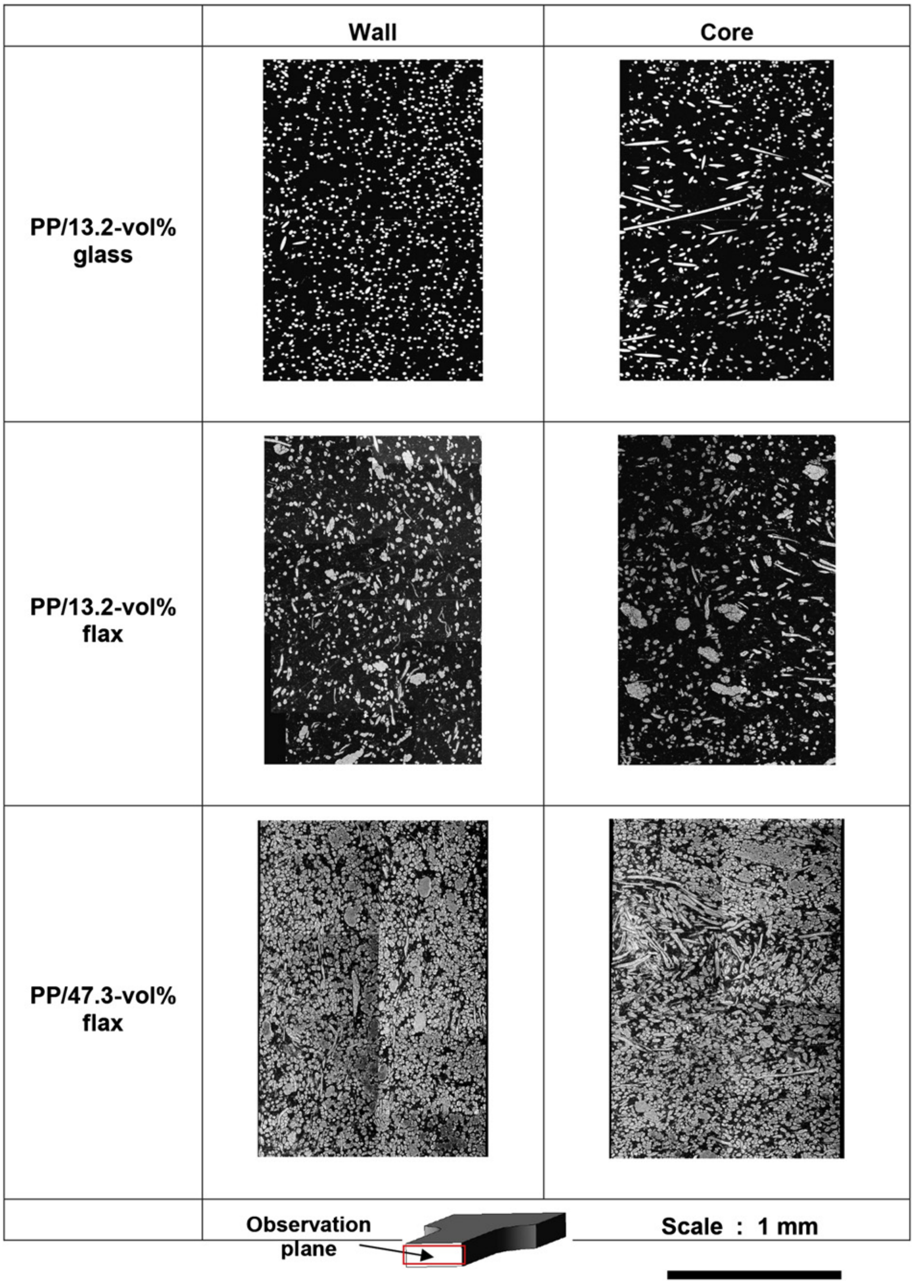

Fig. 7. Observation of the skin-core structure viewed in the thickness of a section of PP/2 mm flax fibre reinforced at 13.2 and 47.3%-wt and PP/glass 13.2%-wt.

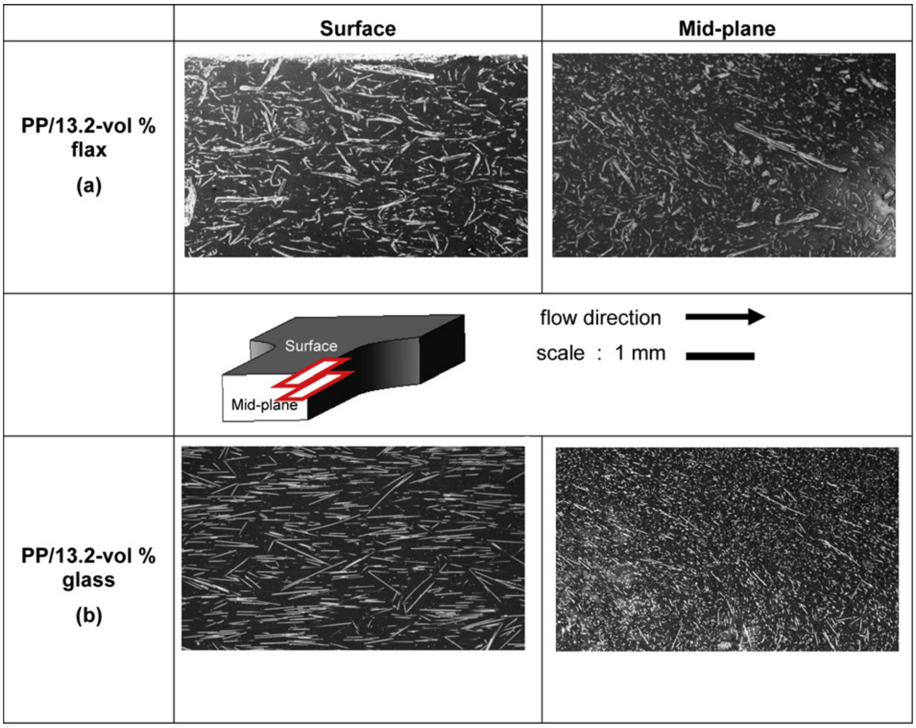

Fig. 8. Microscopic observation of microstructures of composites PP/2 mm flax fibre reinforced to 13.2-vol% (a) and PP/glass (b) 13.2-vol% in the direction of flow.

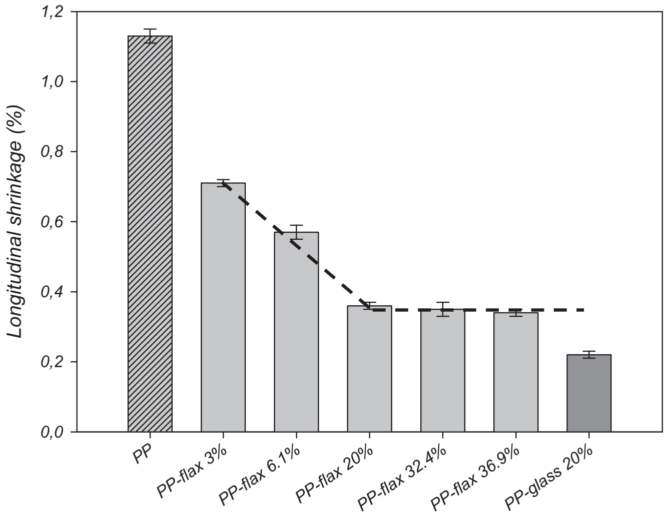

Fig. 9. Evolution of the longitudinal shrinkage of PP composites according to the nature and the fibre volume fraction.

4. Conclusion

We investigated the effects of pertinent parameters like reinforcement content, initial fibre length or number of process cycles on the final composites morphological or dimensional properties. We evidenced an aspect ratio decreases for high volume contents of flax fibres. The improvement of fibre content increases the collisions between fibres and the compound viscosity inducing higher shear stress during processing.

Compared with glass fibres composites, plant fibres ones exhibit clear difficulty to be aligned in the flow direction. Microscopic observations in the flow direction and transverse to the flow showed differences in fibre orientation between the flax and glass. The flax fibres and glass do not exhibit the same behaviour in the polypropylene matrix. These fibres orientation differences have an impact on shrinkage evolution; the incorporation of flax fibres highly improves the shrinkage values compare to virgin PP, but, even for high loading rates, the shrinkage doesn't reach glass composites performances.

Each step of the process decreases fibre length, particularly the first step of extrusion. In some cases, this first step also increases the aspect ratio showing that extrusion also participates in breaking up bundles. A second step of extrusion improves a final property, showing the importance of the process used to make pellets before injection molding. The effect of retting and the behaviour of fibres in processing operations need to be studied to refine our control of the development of natural fibre reinforced composites.

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