High-Performance-Tensile-Strength Alpha-Grass Reinforced Starch-Based Fully Biodegradable Composites

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Though there has been a great deal of work concerning the development of natural fibers in reinforced starch-based composites, there is still more to be done. In general, cellulose fibers have lower strength than glass fibers; however, their specific strength is not far from that of fiberglass. In this work, alpha-fibers were obtained from alpha-grass through a mild cooking process. The fibers were used to reinforce a starch-based biopolymer. Composites including 5 to 35% (w/w) alpha-grass fibers in their formulation were prepared, tested, and subsequently compared with those of wood- and fiberglass-reinforced polypropylene (PP). The term "high-performance" refers to the tensile strength of the studied composites and is mainly due to a good interphase, a good dispersion of the fibers inside the matrix, and a good aspect ratio. The tensile strength of the composites showed a linear evolution for fiber contents up to 35% (w/w). The strain at break of the composites decreased with the fiber content and showed the stiffening effects of the reinforcement. The prepared composites showed high mechanical properties, even approaching those of glass fiber reinforced composites.

Keywords: Fiber-reinforced composites; Tensile strength; Fully biodegradable composites

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INTRODUCTION

It is commonly accepted that biopolymers and bio-fibers have the potential to replace or reduce the consumption of fossil fuel-derived plastics. In addition, to facilitate the recycling of these materials, such replacement should lead to reductions in energy consumption, pollution, and CO_2 emissions. Hence, fiber-reinforced composites are often described as eco-friendly materials. However, the mechanical properties and/or the price of such composites have rendered them inadequate for many applications to date. In general, cellulosic fibers have inferior strength compared to their synthetic counterparts, such as aramid, carbon, and glass fibers.

Due to their lower density, some natural fibers have a specific strength not far from those of fiberglass. On the other hand, bio-based matrices (such as polylactic acid or polyhydroxybutyrate) remain more expensive than petroleum-derived polymers. Nevertheless, thanks to their price/performance balance, ongoing innovations are offering new commercial opportunities for bio-based polymers and their composites in several applications.

Compared to lactones, butyrates, valerates, and other monomers used for the synthesis of biodegradable polymers, starch is an inexpensive product whose transformation into a thermoplastic polymer can be accomplished in a rather straight-forward manner (Coombs and Hall 1998; Teixeira *et al.* 2007; Averous and Halley 2009; Dogossy and Czigany 2011; Gandini 2011). As a result, thermoplastic starch (TPS) is considered one of the most promising biopolymers for large-scale applications. During the last few decades, TPS has received considerable attention among the scientific world (John and Thomas 2008; Belhassen *et al.* 2009; Cañigueral *et al.* 2009; Chaleat *et al.* 2012). As a result, numerous works have been published evaluating their mechanical and thermal properties, water uptake behavior, and/or degradation kinetics in an attempt to adapt these parameters to the technical demands of potential applications (Angles and Dufresne 2001; Carvalho *et al.* 2005; Ayadi *et al.* 2011). In recent years, some companies have started the mass production and commercialization of several starch-based biodegradable polymers. Nevertheless, research is still needed to properly adapt the mechanical properties, water uptake, and/or biodegradation kinetics of TPS to any particular application (Fowler *et al.* 2006). In the particular case of mechanical properties, the most common way to increase them is through the utilization of reinforcing elements. Given the eco-friendly character of the TPS matrix, the use of biodegradable fibers such as those obtained from natural plants seems the most adequate option (Faruk *et al.* 2012).

Esparto (*Stipa tenacissima*), also known as alpha-grass, is a highly fibrous herbaceous plant native to Spain and North Africa. In these regions, alpha-fibers have been used since ancient times to prepare ropes, basketry, and espadrilles due to their toughness and resistance. Nevertheless, at present, the most common utilization of esparto-grass comes from the production of fibers for high-quality lightweight paper (Nada *et al.* 2002; Belkhir *et al.* 2013). Despite the large number of works published regarding the use of fibers from plants as reinforcing agents, the research devoted to alpha-fibers remains very limited (Abdelmouleh *et al.* 2007; Paiva *et al.* 2007; Girones *et al.* 2012; Valente Nabais *et al.* 2013). In this work, we attempted to take advantage of the high toughness of esparto fibers to use them as the reinforcing element of a thermoplastic matrix. Esparto-grasses were submitted to mechanical and chemical processes to optimize the preparation and isolation of alpha-fibers. After their characterization, alpha-fibers were used to prepare fully biodegradable fiber-reinforced thermo-plastic composites, and their tensile strength was evaluated. The obtained results were compared with that of glass fiber / polypropylene (PP-FG) and stone groundwood / polypropylene (PP-SGW) composites. The Kelly-Tyson model was used to model the micromechanic properties of the composites, specifically the interfacial shear strength, the orientation factor, and the intrinsic tensile strength of the fibers.

EXPERIMENTAL

Materials

Esparto or alpha-grass (*Stipa tenacissima*), with an initial length of about 30 cm, was kindly provided by Mas Clarà de Domeny, S. L. (Catalonia, Spain). A starch-based polymer with a density of 1.3 g/cm^3 (Mater-bi® YI014U/C, TPS from here on), supplied by Novamont (Novara, Italy), was used as a biodegradable thermoplastic matrix.

For comparison purposes, polypropylene composites reinforced with softwood fibers were also prepared. The materials used were as follows: polypropylene Isplen

PP090 G2M (Repsol, Madrid, Spain); stone-ground wood (SGW) pulp derived from *Pinus radiata* (Zubialde S.A, Aizarnazabal, Spain); and maleic anhydride-grafted polypropylene (MAH-PP) Epolene G3015 (Eastman Chemical Company, Tennessee, USA). E-glass fiber (FG) was produced by Vetrotex (Chamberly Cedex, France) and provided by Maben, S.L. (Banyoles, Spain).

Sodium hydroxide (Merck KGaA, Darmstadt. Germany) and anthraquinone (BASF AG, Ludwigshafen, Germany) were used for the treatment ("cooking") of the esparto fibers. Decahydronaphthalene (Decalin) (Fisher Scientific, Hampton, USA) was used to dissolve the polymer matrix and recover the reinforcing fibers from the composites. Chloroform was procured from Fisher Scientific. Acetone (reagent grade; 95% purity) from Sigma Aldrich was used as received.

Methods

Characterization of esparto-grass

The determination of the basic chemical composition of esparto-grass was conducted following TAPPI standard protocols. The amounts of extractives, ash, lignin, hemicellulose, and alpha-cellulose were measured according to TAPPI methods T204, T211, T222, and T203, respectively.

Esparto-grasses were submitted to chemical treatments to eliminate the extractives and a small portion of lignin. Plants were soaked in a thermostatic bath set at 98 ± 1 ºC. Then, 5% sodium hydroxide and 0.1% anthraquinone (based on the dry fiber content) were added to the reactor and the mixture was stirred for different periods of time, ranging from 150 to 270 min. Although the elimination of extractives is quickly accomplished, due to the relatively mild reactive conditions applied, the elimination of soluble lignin and hemicellulose is expected to be time-dependent.

Defibration of alpha-fibers

Prior to use as a reinforcing element, the fiber bundles need to be separated to enhance the interface with the polymer matrix. This was accomplished with the help of a Sprout-Waldron hammer mill. Fiber aliquots were collected and analyzed through an automatic fiber morphology analyzer (MorFi Compact Analyser - Techpap SAS, Grenoble). A diluted aqueous suspension (1 wt% consistency) of fibers was analyzed during 2 to 5 min, and the length of the fibers was evaluated, considering an amount of individual fibers in the range of 2500 to 3000 units. A minimum of two samples were analyzed. The analyzer offers complete fiber, fines, and shive morphology characterization, but only the fiber length and fiber diameter distribution were used for the present work.

Composite Preparation

A 20% (w/w) alpha-fibers composite blend was produced by means of a Brabender internal mixer for 10 min at 80 rpm rotor speed and at 175 ºC. Additional composite blends comprising 5 to 35% (w/w) alpha-fibers were produced by means of a Gelimat intensive kinetic mixer for 2 min at 2500 rpm, until a discharge temperature of 175 ºC was reached. Composites were granulated in a blade mill provided with a 10-mm mesh and kept in an oven at 80 ºC until required to prevent moisture absorption.

Composite materials comprising 30 to 50 wt% PP-SGW and 20 to 40 wt% PP-FG with coupling agent were obtained. The materials were prepared in a Brabender internal mixing machine. The working parameters were 80 rpm for SGW and 20 rpm for FGsized, during 10 min at a temperature of 180 ºC. The MAH-PP was added to the plastograph with the PP pellets.

Test specimens were molded in a Meteor-40 injection-molding machine (Mateu&Soler, Barcelona, Spain) using a steel mold complying with ASTM: D3641 specifications.

Mechanical characterization

Composite specimens were placed in a conditioning chamber (Dycometal) at 23 ºC and 50% relative humidity during 48 h, according to ISO D618 protocol standards. Tensile tests were carried out with an Instron 1122 universal testing machine following ASTM D638 regulations.

Fiber extraction from composite and morphological analysis

Injection-molded composites were successively refluxed in chloroform, acetone, and decahydronaphthalene (decalin) to eliminate all the components of the polymeric matrix. After each treatment, fibers were filtered and washed several times in the hot solvent. Fiber aliquots were collected and analyzed using an automatic fiber morphology analyzer (MorFi Compact Analyser - Techpap SAS, Grenoble).

RESULTS AND DISCUSSION

Chemical Characterization of the Esparto-Stalks

Knowing the chemical composition of the fibers allows the design of chemical treatments to modify them, with the objective of increasing the performance of the fibers as reinforcing elements. In the present case, the objective was to obtain an excellent interphase between fiber and matrix by creating fibers capable of good interaction with the starch-based polymer. Table 1 shows the basic composition of the esparto-stalks before treatment. The TAPPI T211 protocol could identify the salt forms of some extractives as ash content, and thus such components would be accounted for twice. Hence, the values presented account for more than 100%. It seems that analyzed esparto-grass contained significant amounts of extractives and lignin. Furthermore, the contents of gamma- and alpha-cellulose were high and in line with other reported analyses (Marrakchi *et al.* 2011).

Table 1. Basic Constituents of Esparto-Grass

	Ash	Extractives	Alpha-cellulose	Gamma-cellulose	Lignin
Content (%)	3.2	7.6	45.3	24.1	23.4

Effect of Esparto-Grass Cooking Time on the Tensile Strength of Composites

Prior to the final evaluation of the tensile strength of the composite materials, esparto-grass was submitted to different cooking times, as the natural stalks had no ability to interact chemically. TPS reinforced with 20% alpha-fiber composite material was prepared by means of a Brabender internal mixer. The main objective behind the cooking of esparto-grass is the elimination of the extractives and ash, thus obtaining neat fibers with high cellulose content and a surface capable of chemical interaction. Because both cellulose and starch are based on glucose, it was hypothesized that higher cellulose content might enhance the interaction with the starch-based polymeric matrix.

Esparto-grasses were submitted to sodium hydroxide treatment for 150, 210, and 270 min to reduce their extractives, ash, and lignin/hemicellulose contents to various degrees. After defibration of the bundles obtained during this process, fibers were used to prepare composites based on TPS reinforced with 20% alpha-fibers (in %w/w).

Table 2 shows the main mechanical properties of TPS reinforced with 20% alpha-fibers obtained before and after submitting esparto-grass to an alkali solution cooking for different periods of time. For comparison purposes, the mechanical properties of the neat TPS matrix have also been included.

Table 2. Tensile Properties of TPS and Alpha-Fiber-Reinforced TPS Composites

Reaction time (min)	Tensile strength (MPa)	Young's Modulus (GPa)	Tensile strain at break (%)
0	26.9 (2.4)	4.8 (0.2)	0.9 (0.2)
150	54.7 (2.7)	5.1 (0.2)	1.8 (0.1)
210	54.1 (2.2)	4.5 (0.2)	2.0 (0.1)
270	52.0 (2.5)	4.9 (0.2)	1.8 (0.2)
Neat TPS	25.6 (1.8)	2.5 (0.1)	3.4 (0.1)

The use of untreated fibers as reinforcement slightly changed the tensile strength of the neat matrix (Table 2). On the other hand, the use of treated fibers noticeably increased the tensile strength of composites. The tensile strength showed a slight tendency to decrease in the course of continued treatment, particularly after 4 h. Nevertheless, in general, the mechanical parameters evaluated remained fairly constant, and no major statistical differences were detected.

It must be noted that the incorporation of 20% alpha-fibers in the TPS matrix more than doubled the tensile strength of the polymer, which went from about 26 MPa for the neat polymer to 52 to 55 MPa for the composites. The influence of fiber addition on the Young's modulus was equally important, with increases of about 50%. As expected, the increased rigidity caused by the reinforcing fibers also caused a considerable diminishment in the capacity of composites to sustain plastic deformation (Lopez *et al.* 2012b).

The results suggest that a treatment of 150 min in alkali solution assured the elimination of enough ash and extractives to generate a good-to-excellent interface between the esparto-grass fibers and the thermoplastic matrix; further reaction times had a slightly negative impact, if any, on the capacity of fibers to reinforce TPS. The reason behind this observation remains unclear, although the increasing difficulties encountered in dispersing the alpha-fibers with the Brabender internal mixer might play an important role. The higher hydroxyl group content of fibers as the treatment progressed may lead to the establishment of stronger fiber-fiber interactions through hydrogen bonding.

In light of these results, a set of experiments was developed. A cooking time of 150 min was established to prepare the esparto-grass pulps, and a new set of composite materials was prepared by means of a Gelimat multikinetic mixer.

Effect of Alpha-Fiber Content on the Tensile Properties of Composites

Table 3 shows the tensile properties of TPS reinforced with 5, 15, 25, and 35% alpha-fibers treated in alkali solution for 150 min. The mechanical properties of the neat matrix are also included for comparison.

Table 3. Main Tensile Properties of Starch-Based Composites Reinforced with 5 to 35% w/w Alpha-Fibers

Fiber weight percentage (% w/w)	Fiber volume fraction (% v/v)	Tensile strength (MPa)	Strain at break (%)	Matrix tensile strength at composite failure (MPa)
0	0	25.6 (1.8)	3.4 (0.1)	25.6 (1.8)
5	0.047	32.0 (2.3)	2.8 (0.2)	25.3 (1.5)
15	0.142	46.8 (2.5)	2.1(0.1)	23.9 (2.1)
25	0.238	58.5 (2.2)	1.6 (0.2)	21.3 (2.3)
35	0.335	70.1 (2.3)	1.4 (0.2)	19.6 (2.6)

As can be seen in Table 3, the incorporation of the alpha-fibers caused a large and progressive increase in the tensile strength of composites. This continuous trend suggests that, even at 35% fiber content, a good dispersion of the reinforcing fibers was achieved during compounding.

The tendency of cellulosic fibers to self-aggregate, particularly delignified fibers, usually leads to the formation of fiber bundles within the composite that causes a decrease in their reinforcing efficiency. This effect is particularly problematic for composites based on conventional hydrophobic petroleum-derived polymers such as polypropylene (PP), polyethylene (PE), or polystyrene (PS) (Vilaseca *et al.* 2004). Although the treatment applied to alpha-grass was rather mild, it was expected that their inherently high alpha-cellulose content might cause some dispersion problems. However, such problems did not appear, and the efficiency index of alpha-fibers remained rather constant throughout the range of weight percentages evaluated.

The reason behind this behavior might be found in the high chemical resemblance between the alpha-fibers (with alpha-cellulose as the main component) and the thermoplastic matrix used (starch-based). This resemblance could allow the establishment of strong interactions between the two components of the composite, generating a good fiber-matrix adhesion. As a result, fibers not only can effectively act as reinforcing elements (as evidenced by the evolution of tensile strength), but the limitation of inter-fiber interactions also favors the dispersion of the reinforcement within the composite.

As expected for any fiber-reinforced composite, the presence of reinforcing elements led to a reduction in the capacity of the material to sustain plastic deformation. Thus, the tensile strain at break was limited to about 1.5% when the weight percentage of alpha-fibers was 25 to 35%.

It was also observed that the contribution of the matrix to the tensile strength of the composite material decreased with increasing fiber content. That behavior is in line with other studies (Vallejos *et al.* 2012).

Comparison with "Traditional" Fiber-Reinforced Composites

In previous works, the mechanical properties of polypropylene and its fiber-reinforced composites were analyzed (Girones *et al.* 2007; Mendez *et al.* 2007; Lopez *et al.* 2012a). Table 4 displays the tensile properties of composites prepared and tested following the same conditions used in this work. The only noteworthy difference with the materials evaluated here comes from the utilization of maleic anhydride-grafted polypropylene, a coupling agent that has been extensively used in fiber-reinforced polypropylene composites (Franco-Marquès *et al.* 2011).

Table 4. Tensile Properties of Polypropylene Composites Reinforced with SGW and Fiberglass (Lopez *et al.* 2012a)

Fiber	Fiber weight percentage (% w/w)	Tensile strength (MPa)	Strain at break (%)
None	0	27.6 (0.5)	9.3 (0.2)
SGW	30	47.8 (0.4)	5.0 (0.1)
	40	56.7 (0.7)	4.5 (0.2)
	50	60.4 (0.5)	4.4 (0.2)
Fiberglass	20	67.9 (0.9)	4.7 (0.2)
	30	79.8 (1.2)	4.4 (0.2)
	40	101.2 (0.7)	3.7 (0.2)

The tensile strength of the TPS matrix used in this work was about 7% lower than that of PP. However, even without a coupling agent, alpha-fiber-reinforced TPS composites exhibited higher tensile strength than softwood-reinforced PP. The intrinsic mechanical properties of the two reinforcing fibers may have played a role, although this has not been determined (this is thought to be difficult for such small fibers). Nevertheless, this enhancement was attributed to the better fiber-matrix stress transfer caused by the higher affinity of fibers for the starch-based polymer rather than polypropylene. After all, both starch and cellulose are composed of glucose units. Hence, a good fiber matrix interaction can be expected between these two components. In clear contrast, the highly hydrophobic structure of PP has been considered to be responsible for the lack of performance of wood plastic composites, a problem that has required designing coupling agents to enhance the fiber-matrix interphase. It seems that the hydrogen bonds created between the high quantity of OH groups present on the alpha-fiber surface and in the TPS created a similar or better interface than that of PP-SGW, where the proposed coupling mechanism is hydrogen bonding and also covalent ester linkage generated by the chemical reaction of the anhydride groups of the MAPP and the hydroxyl groups of the fiber surface (Mendez *et al.* 2007)

If weight fraction is considered, the results obtained suggest that composites based on TPS reinforced with alpha fibers can have considerably higher strength than PP-SGW composites. In fact, even with the incorporation of an efficient coupling agent, SGW- reinforced polypropylene materials require higher fiber weight content to attain the same strength as alpha-TPS composites. In addition, TPS composites reinforced with alpha fibers had tensile strengths not too far from those of fiberglass. This result might suggest that they could effectively replace such materials in some applications. However, if the same graph is reproduced, considering the volume fraction and not the fiber weight content, results are not as straightforward (Fig. 1).

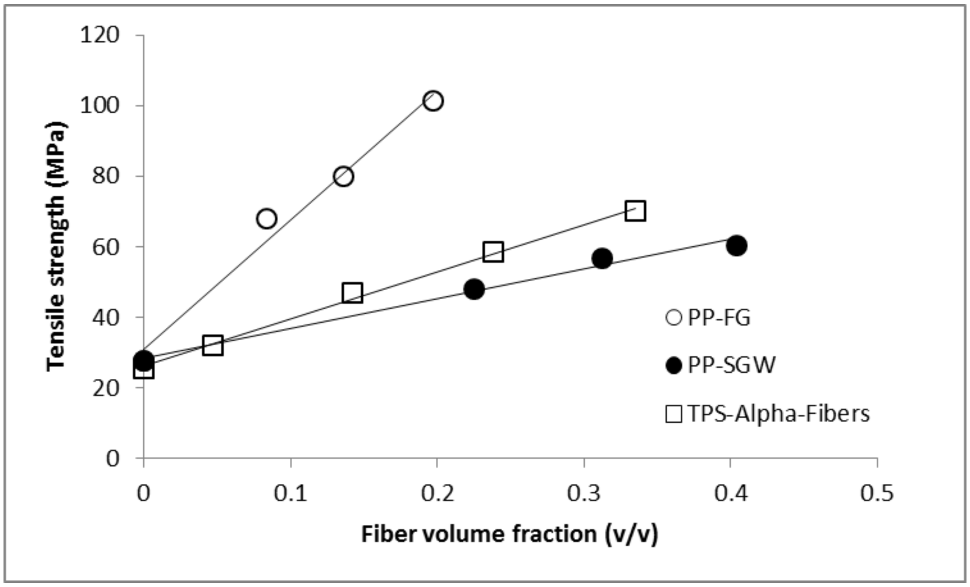

Fig. 1. Comparison of the tensile strength of alpha-fiber-reinforced TPS composites with softwood- and fiberglass-reinforced PP as a function of fiber volume

The mechanical properties of TPS-alpha composites did not differ from those of SGW-PP (the densities of SGW and PP are 1.33 and 0.905 g/cm^3, respectively). In addition, the mechanical performances of fiberglass composites were still far from being reached. Nevertheless, the materials prepared seem suitable for less mechanically demanding and/or temporary applications, thus taking full advantage of their biodegradable character. In that sense, if a tensile strength of 60 MPa is required, the equivalent fiber contents would be as follows: 17% wt FG, 26.5% wt alpha-fibers, and 47.5% wt SGW.

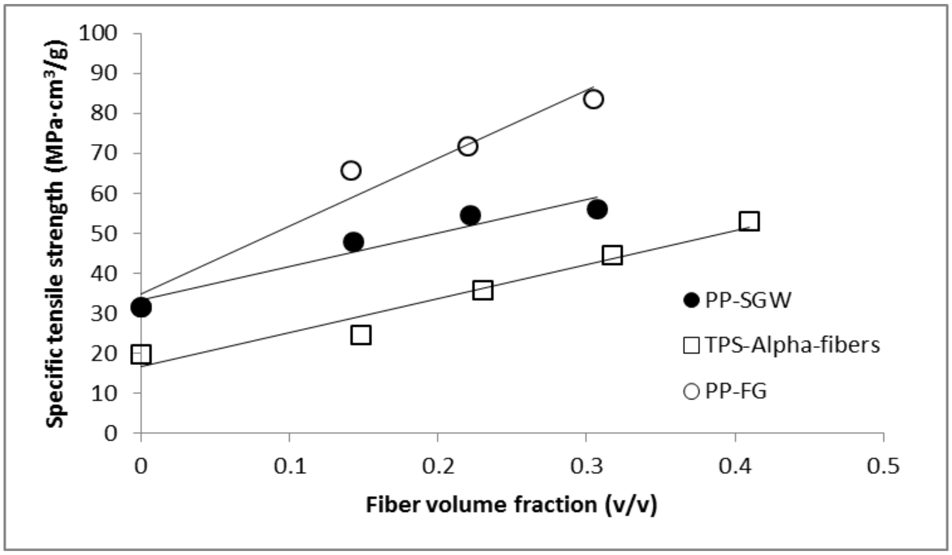

Fig. 2. Comparison of the specific tensile strength of alpha-fiber-reinforced TPS composites with softwood- and fiberglass-reinforced PP as a function of fiber volume

The advantages of the specific properties of natural fiber-based composites over FG-reinforced composites have been discussed (Baroulaki *et al.* 2006; Nedjma *et al.* 2013). In that sense, the densities of alpha-fibers, SGW, and FG are 1.375, 1.33, and 2.45 g/cm^3, respectively. On the other hand, TPS-based composites are penalized by its density: 1.3 g/cm^3. Figure 2 shows the evolution of the specific tensile strength of the composite materials against the volume fraction of the fibers.

While the regression line of the PP-SGW composites remained virtually unchanged (primarily due to a mean density close to 1), the slope of the regression line of the PP-FG composites decreased (the density evolved from 0.905 to 1.2) and the regression line of the TPS-alpha-fiber composites moved downwards with slight changes in its slope (the density changed slightly, to around 1.3).

Micro-Mechanics

The mechanical behavior of composite materials reinforced with semi-aligned fibers can be predicted by the modified rule of mixtures (Thomason 2002; Vilaseca *et al.* 2010) (Eq.1). The parameters σ_t^c, σ_t^f, and σ_t^{m*}represent the composite, fiber, and matrix tensile strengths at failure, respectively; V^f is the volume fraction of the reinforcement, and f_c is a compatibility factor. The compatibility factor can be expressed as the product of an orientation factor (χ_1) and an interface factor (χ_2) (Fu and Lauke 1996).

$$\sigma_t^C = f_c \cdot \sigma_t^f \cdot V^f + \sigma_t^{m*} \cdot (1 - V^f) \tag{1}$$

According to the literature, for well-bonded composites, the compatibility factor (f_c) is commonly around 0.2 (Sanadi *et al.* 1994). On the other hand, the orientation factor depends primarily on the characteristics of the machinery and the melt characteristics. Using the same equipment as in this study, it was determined in previous works that, using specific injection-molding equipment, the orientation factor was in the vicinity of 0.3 for well-bonded composites (Rodriguez *et al.* 2010; Lopez *et al.* 2011; Vallejos *et al.* 2012).

Assuming these values as correct, the length and interface factor has been hypothesized to be $\chi_2 = f_c / \chi_1 = 0.66$. In addition, by applying the modified rule of mixtures in combination with the data in Table 3, the intrinsic tensile strength of alpha fibers (σ_t^f) was determined to be 874.95 MPa.

The interfacial shear strength (τ) is an important property of fiber-reinforced composites, as it evaluates the capacity of the material to effectively transfer stresses from the matrix to the reinforcement and consequently determines the fiber failure mechanism during fracture (breakage or pull-out) (Hassan *et al.* 2011). Its measurement can be accomplished either experimentally by pull-out test or through mathematical models (Lopez *et al.* 2012c):

$$\tau = \frac{d^f \cdot \sigma_t^f}{4 \cdot l^f \cdot (1 - \chi_2)} \tag{2}$$

Thus, the mathematical characterization of τ requires the mean length of the fibers within the composite (Li *et al.* 2009; Vallejos *et al.* 2012).

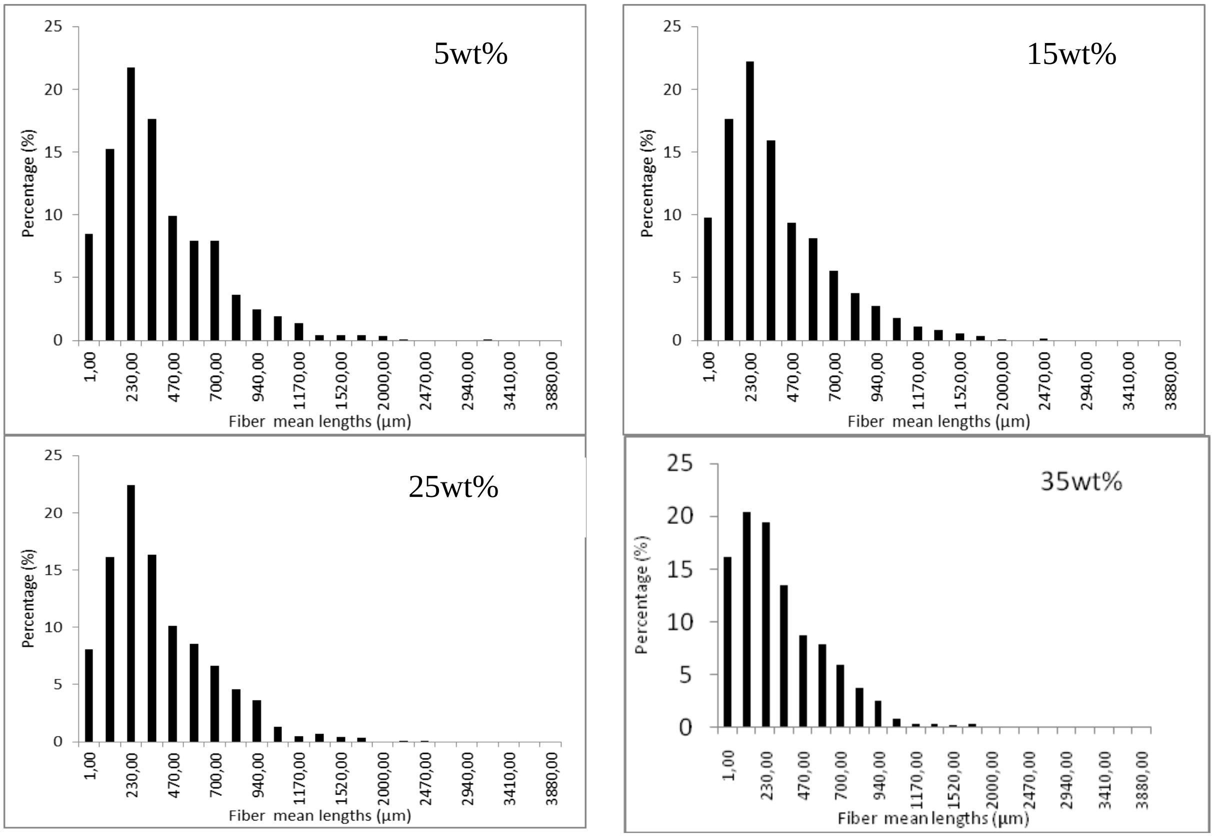

Fig. 3. Evolution of the length distribution of the fibers with reinforcement content

For this purpose, a diluted aqueous suspension of fibers was eluted through a capillary, and the mean fiber length was determined from the analysis of more than 3000 fibers. Figure 3 shows the length distribution of the reinforcing alpha fibers inside the composites. The mean weighted length of fibers was found to be 955, 898, 844, and 768 µm for materials comprising, respectively, 5 to 35 wt% reinforcement.

The weighted fiber length (l^f) experienced a noticeable reduction when the weight fraction of reinforcement increased from 5 to 35%. The diameter of the fibers (d^f) was found to be almost independent of fiber content, with a mean value of 17.3 µm. Consequently, the aspect ratio (l^f/d^f) progressively diminished as the fiber content increased. Nevertheless, the average aspect ratio remained above 40 for all the materials tested. This result is in agreement with the high reinforcing capacity of the fibers throughout the range of fiber content evaluated.

With these experimental values, it was possible to compute the interfacial shear strength of the composites, which turned out to be 11.9, 12.6, 13.5, and 14.8 MPa, respectively. The values obtained are near 14.8 MPa, the value obtained by applying the Von Misses criteria (Pegoretti *et al.* 1996).

Kelly and Tyson (1965) proposed a model to characterize the tensile strength of composites reinforced with semi-aligned fibers:

$$\sigma_t^C = \chi_1\left(\sum_i\left[\frac{\tau\cdot l_i^f\cdot V_i^f}{d^f}\right]+\sum_j\left[\sigma_t^f\cdot V_j^f\left(1-\frac{\sigma_t^f\cdot d^f}{4\cdot\tau\cdot l_j^f}\right)\right]\right)+(1-V^f)\cdot\sigma_t^{m*} \qquad (3)$$

This large equation can be simplified as $\sigma_t^C = \chi_1(X+Y)+Z$, where X, Y, and Z represent respectively the contributions of subcritical fibers, supercritical fibers, and the matrix to the composite's strength (Vallejos *et al.* 2012; Serrano *et al.* 2013). From this model, the orientation factor can be calculated as:

$$\chi_1 = \frac{\sigma_t^C - Z}{X+Y} \tag{4}$$

Table 5 shows the orientation factors obtained using the interfacial shear strength (τ) values computed through the Kelly-Tyson model.

Table 5. Experimental Values for the Tensile Properties of Reinforced Composites and Polymeric Matrix

Fiber %	σ_t^c *(MPa)*	σ_t^{m*} *(MPa)*	τ *(MPa)*	*X+Y* *(MPa)*	χ_1
5	31.9	25.3	11.9	29.3	0.267
15	46.8	23.9	12.6	86.9	0.302
25	58.5	21.3	13.5	148.3	0.285
35	70.1	19.6	14.8	215.8	0.264

The values of the orientation angle were quite close to the assumed 0.3 value. The mean value for the orientation factor was 0.28. The same calculations were performed using Von Misses τ, and the obtained value for the orientation angle was 0.27. Taking into account that $\chi_1 = \cos^4\theta$, the obtained orientation factors imply mean orientation angles of 38.7º and 38.39 º, respectively.

CONCLUSIONS

1. Alpha grass (esparto) was found to be a suitable source of fibers for reinforcement of starch-based composites. The fibers were submitted to a mild cooking process to eliminate part of the extractives and obtain a good fiber-matrix interface. It was found that cooking processes that lasted more than 150 min had little effect on the reinforcing ability of the fibers. The obtained interface was rated as excellent.
2. The fibers were used to reinforce a starch-based biopolymer. Composites including 5 to 35% (w/w) alpha-grass fibers in their formulation were prepared and tested. The tensile strength of the composites showed a linear evolution against 5 to 35% fiber contents. The strain at break of the composites decreased with increasing fiber content and showed the stiffening effects of the reinforcement. The fibers extracted from the matrix showed a mean fiber length and diameter of 866 µm and 17.3 µm, respectively, thus having an aspect ratio of around 50, which ensured a good stress transfer with the polymeric matrix.
3. Alpha-fiber-reinforced TPS composites showed comparable performance with fiberglass-reinforced polypropylene composites, adjusting the fiber content and

considering the tensile strength the main concern. This shows the potential of fully biodegradable composites to be used for technical and engineering purposes.

4. The micromechanics study showed that the interface between alpha fibers and the TPS was excellent, with a coupling factor of 0.2 and orientation factor of around 0.3.

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