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Mechanical properties of polypropylene composites based on natural fibres subjected to multiple extrusion cycles.

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Abstract

The influence of multiple extrusion cycles on the behaviour of natural fibres reinforced polypropylene was studied. Composites were fabricated with 20 wt.% of flax fibres. Final fibres dimensions (length and diameter) were measured by means of optical microscopy. Mechanical properties of matrix and composites were measured after each extrusion cycle. It was observed that the elastic modulus increased by fibres incorporation. The elastic modulus of the matrix was higher after the first process cycle than that of the virgin material, mainly due to chain scission. In the next cycles, the modulus kept constant. On the other hand the elastic modulus of the composite after a single extrusion step was lower than that predicted by the Halpin – Tsai model probably due to a poor mixing and to low adhesion at the fibre/matrix interphase. In the following two steps, modulus increased because the better fibre dispersion was observed. For the final two extrusion cycles, the slow decrease in this property was correlated with the darkening and poor organoleptic properties observed as a result of thermal degradation.

Keywords: re-use, natural fibres, polypropylene, mechanical models, extrusion cycles.

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Introduction

Environmental policies of developed countries are increasing the pressure on manufacturers to consider the environmental impact of their products. Therefore, the interest in using natural fibres as a reinforcement of polymeric based composites is growing, mainly due to its renewable origin. This is particularly true in the case of the automotive industry, where many parts can be produced massively by using natural fibres as reinforcement. This kind of industries is trying to replace progressively the traditional materials for others with lower environmental impact. [1-2].

Natural fibres show many advantages (low cost, low density and high specific properties, biodegradabily and non-abrasion) compared to the traditional ones [3]. However, there are many drawbacks that reduce its potential use as reinforcement in polymers. Those disadvantages are: a) incompatibility with hydrophobic polymeric matrices (especially thermoplastic ones), b) tendency to form aggregates during processing, c) low resistance to processing temperatures and d) poor resistance to moisture.

There are many publications [4-11] dedicated exclusively to short fibre/thermoplastic polymer composites, however there are not many publications related to the effect of extrusion cycles on the properties of this kind of composites [12]. The aim of this work was to study the thermo – mechanical degradation suffered by the matrix and the fibres during extrusion cycles and the effect of that on the final properties of obtained composites. The feasibility of re-use of composite materials was also determined.

Experimental

1.   Materials

A commercially isotactic polypropylene (Basell C30G - Italy) was used as a matrix. Its main properties, extracted from its technical datasheet, are shown on **Table 1**.

Natural flax fibres, obtained by a retting process of flax plants, from Finflax (Finland) were used as reinforcement. Some of its properties are also summarized on **Table 1** [13].

2. *Composites Preparation*

Flax fibres were cut to an approximate length of 10 mm in order to make its incorporation in the extruder easier (length (*L*): 10.44 ± 4.58 mm; diameter (*d*): 0.47 ± 0.16 mm and *L/d*: 23.91 ± 11.57), and vacuum dried in an oven at 70 ºC for 24 h. Drying of fibres is a fundamental step since the presence of water within their structure reduces the adhesion between them and the matrix.

Blending of materials was carried out in a twin-screw extruder (Bausano MD30 – Terni, Italy). All processing parameters were fixed: temperature profile (160, 180, 190, 185, 190, 190 ºC); screw speed (25 rpm); screw configuration; and residence time. Materials were extruded and pelletized five times. In order to differentiate the effects on the fibres to that on the neat matrix unfilled polypropylene was also extruded and pelletized five times. The name of the samples were indicated by PP for polypropylene and COM for composites and a number which indicates the extrusion cycles step (for example., PP0 means pure polypropylene matrix before processing whereas COM1 is referred to the composite after the first cycle of extrusion)

Materials extracted after each extrusion step were compression moulded. The temperature – pressure – time cycle applied in all cases was similar and it is shown in **Figure 1**. In order to reduce the residual thermal stresses within the plates, a thermal treatment (100 kg/cm^2 at 100 ºC for 10 minutes) was applied.

All specimens were machined from these plates. The finishing of each coupon was carefully examined in order to check for existing flaws or imperfections.

3. *Testing Methods*

Melt Flow Index (*MFI*) tests were carried out in accordance to ASTM D1238 standards. It was measured with a plastometer VP-05 at the following conditions: temperature 230°C (for the matrix) and 190ºC (for the composites due to the thermal sensitivity of the fibres), time of preheating without loading 4 min, time of preheating with loading 10 min, load 2.16 kg (for matrix and composites with 1, 2 and 3 extrusion cycles) and 5 kg (for composites with 4 and 5 extrusion cycles), nozzle diameter 2 mm.

Optical microscopy and size distribution analysis of fibres. Fibres were extracted from the composites by means of polypropylene dissolution using xilene as diluent (140 ºC) in a soxhlet equipment. Extracted fibres were dried and dispersed into vaseline. Fibre length and

diameter were measured using an optical microscope Olympus SZH 10 and digital image processing software (Image Pro v 4.5, 1993-2001 Media Cybernectic, Inc).

Tensile tests of bone-shaped composite specimens were carried out in Universal Testing Machine, an INSTRON 4467 at a crosshead speed of 5 mm/min (in accordance to ASTM D638 recommendations). Three specimens of each kind of material were tested.

Three point bending tests were performed in a Universal Testing Machine INSTRON 4467. The crosshead speed was chosen according to ASTM D790 standard. Three specimens for each material were tested.

Izod Impact tests were carried out by following ASTM D256 recommendations in an instrumented FRACTOVIS of Ceast impact test machine. Notched specimens were used. The test speed was set at 1 m/s was Due to high data dispersion usually observed in this kind of tests, five samples of each kind of material were tested.

Falling weight impact tests were carried out in an instrumented FRACTOVIS of Ceast impact test machine in order to study the in–plane response of materials. Compression moulded plates were directly used. Load was set in order to ensure the complete breakage of plates. At least two samples of each kind of material were tested.

Isothermal thermogravimetric measurements were performed on the fibres and the polypropylene separately in a Shimadzu TGA-DTGA instrument. Processing temperature conditions were replicated. For the extrusion step, an approximate temperature of 230 ºC was selected in order to take into account the shear forces effect within the material (according to previous experience). For the case of compression molding, the temperature was well known (190 ºC). Therefore, temperature programs were run at 190ºC and 230ºC for 4 hours. Tests were carried out under nitrogen atmosphere (20 ml/min) in order to prevent any thermoxidative degradation

X-Ray Diffraction (XRD) and **Differential Scanning Calorimetry (DSC)** were performed on unfilled materials in order to evaluate the changes on the crystallinity. From **XRD** curves, the relation between crystalline peaks and amorphous phase was calculated. On the other hand, **DSC** tests were carried out from 25 to 250 ºC at a heating rate of 10 ºC/min under nitrogen atmosphere. The heat of fusion was estimated from these curves.

Fracture surfaces of tensile test specimens were observed by using a Philips **Scanning Electron Microscope (SEM).** Surfaces were gold coated in order to make them conductive. Different zoom levels were applied in order to the different deformation/fracture mechanisms exhibited by materials under study.

All tests were carried out at room temperature.

Mixing Index (MI)**.** In order to estimate the mixing quality of the blends obtained after extrusion, mixing indexes were calculated. This index was calculated according to equation 1 [14]:

$$MI = \sqrt{\frac{\sum_{i=1}^{N}(x_i - \overline{x})^2}{(N-1)\cdot\mu\cdot(1-\mu)}} \qquad (1)$$

where N is the number of samples evaluated, x_i is the measured reinforcement content in sample "i" and μ is the original reinforcement content (in our case, 13% v/v; calculated from the weight % content and the densities of each component). In order to estimate the reinforcement content in a particular sample, an image analysis was performed. Backlighting images of different plates were taken, and the total area was then divided into small sub-areas ("i" samples). For each sub-area, the area of dark (reinforcement agglomerates) and translucent (polypropylene matrix) zones were selected and measured by means of an image analysis software. Finally, the relation between agglomerates and matrix was estimated. A good mixing quality is reached when good dispersion and good distribution of both components are obtained. Due to the highly hydrophilic characteristics of natural fibres and the counter hydrophobic behaviour of polypropylene matrix, there is a natural tendency of agglomerate formation when no compatibilizer is added. Therefore, only bad distribution effects were analyzed (how well agglomerates were dispersed within the matrix). It was found that well dispersed fibres did not influence the overall mixing index analysis since they increase both factors in the equation (x_i and $\overline{x}$), being their difference constant.

Results

Melt flow index studies showed that an important degradation took place in the matrix during sequent extrusion cycles.

Viscosity was calculated as follow [15].:

$$\eta_{ap} = \frac{4.86\cdot\rho\cdot P}{MFI} \qquad (2)$$

where P is the applied charge, ρ is the material's density and *MFI* is the measured Melt Flow Index.

Figure 2 show the decrease on its apparent viscosity. Viscosity is related to the mean molecular weight of the polymer. Therefore, a decrease in the viscosity is related to a decrease in the average length of polypropylene chains. This occurs, mainly due to chain scission, as stated by Canevarolo [16].

As it was mentioned before, melt flow index analysis could not be done on composites corresponding to 1, 2 and 3 extrusion cycles. Results for those subjected to 4 and 5 extrusion cycles are presented in **Table 2**. These results should not be directly compared to those obtained for polypropylene, since test conditions were considerably different.

An improvement in all studied mechanical properties (uniaxial tensile, three point bending and Izod impact) with increasing extrusion cycles was observed. This is due to the better mixing of components (distribution and dispersion).

The elasticity modulus (tensile tests) of the composites was at all stages higher than that of the neat polypropylene (**Figure 3.a**.). However, the strength of the composites was lower to that of the matrix for the corresponding number of extrusion cycles (**Figure 3.b.**), but it also showed an improvement with increasing number of cycles.

The same tendency was observed for flexural tests (**Figure 4.a**.). However, materials corresponding to 2 and 3 extrusion cycles showed a better performance, reaching similar values than those subjected to 4 and 5 cycles. The flexural modulus of the neat polypropylene remained almost constant. Bending strength, considered as the outermost fibre's tension at break (as shown on **Figure 4.b**.), was at all stages lower for the composite than for the unfilled polypropylene.

The energy (per unit area) absorbed on impact tests is shown on **Figure 5**. It was higher for composites than for matrix due to the energy dissipation mechanism related to the fibres such as: axial splitting, transverse microcracking and multiple elementary fibres fracture [17-18]. Values corresponding to materials extruded 1 and 2 times showed a high dispersion. This is mainly due to deficient mixing (in terms of distribution and dispersion of reinforcement within the matrix). So, high energy values are related with the impact on a reinforcement rich zone, and low ones indicate impact on a matrix rich (low fibre) content zone. The dispersion on the energy values decreased with increasing extrusion cycles showing an improvement in mixing quality. For both the composite and the unfilled polypropylene, impact energy remains approximately constant (excluding initial cycles).

Energy absorbed on falling weight impact tests was higher for the composite than that of neat polypropylene (**Figure 6**). This behaviour was related to the pull-out and longitudinal

splitting mechanisms involved in the fracture of composites with poor fibre/matrix and fibre/fibre interphases. However, the tendency was not as clear as in the case of Izod impact tests. This is mainly due to the fact that breakage is reached by means of two different mechanisms: circumferential and radial ones. Also, the amount of material subjected to deformation is different in both cases. On Izod impact tests, crack runs through the specimen's depth, but along a previously established zone (due to pre-cracking) whereas on falling weight tests (**Figures 7.a and 7.b**.), cracks run along radial paths starting at the impact point. Low propagation energy is needed for radial cracks to run need (fragile). At the tip of each radial crack, several perpendicular cracks can be generated: circumferential, which consumed higher amount of energy to propagate. [17].

Scanning Electron Microscopy images show that the main deformation among polypropylene specimens (**Figure 8.a**) took place by means of shear effects being predominant the shear tearing mechanism. For composites (**Figure 8.b**) low interfacial adhesion (void formation close to the fibres and completely clean fibres) and fibre extraction (cavities originated by completely extracted fibres) can be observed.

The mechanical degradation of flax fibres was studied by means of optical microscopy. **Figure 9** shows the reduction in the dimensions of the fibres. It was observed that mechanical degradation took place in both dimensions: length and diameter, and the major changes were observed in the first extrusion step. Additional extrusion cycles did not cause any significant further degradation. Final length is close to 2 % of the original value, while the final diameter is approximately 12 % of its original value.

In all cases, the aspect ratio (length to diameter relation) was lower than the critical value needed for full load transfer (from the matrix to the fibres) to take place. This has a very important incidence on mechanical properties of the composites and in the resulting energy dissipation mechanisms as it was described on impact properties section. When aspect ratio (*L/d*) is lower than the critical value, pull out is the preferred mechanism. On the other hand, when the aspect ratio exceeds the critical *L/d* value, fibre breakdown become the predominant mechanism. For the studied system (flax-polypropylene), it has been found that the critical *L/d* ratio is close to 33 [13].

As it is well known [19], each natural fibre is a composite material itself, in which soft lignin and hemicellulose act as a matrix and stiff cellulose microfibrils acts as reinforcement. Microfibrills form hollow cells and they are helically wound along the fibre axis.

Natural fibres are a complex mix of organic materials and, as a result, thermal treatments produce a variety of chemical and physical changes that can be studied by means of TGA

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(thermogravimetric) measurements. The thermal decomposition of natural fibres at 200 ºC or higher produces volatiles, and the obtained composites exhibit low density and poor mechanical properties [3, 13]

TGA measurements (**Figure 10**) showed that the major mass loss (related to thermal decomposition) took place during the former instances of high temperature exposition (i.e. extrusion step). It can be seen, as well, that polypropylene suffers no (or at least minimum) thermal decomposition at selected processing temperatures.

Another important factor is the crystallinity of the matrix: an increase on the matrix crystallinity leads to higher mechanical properties. Therefore, in order to analyse possible changes on polypropylene crystallinity and the effect of that on the mechanical properties, the content of crystalline phase was calculated: X–Ray Diffraction technique (XRD) shows high data dispersion between different samples, possibly due to the methods itself, which only analyses a small superficial area of the sample. Differential Scanning Calorimetry (DSC) was used for measuring and comparing the heats of fusion of the samples. It was found that it remained almost constant for all number of extrusion cycles. The percent crystalline phase was calculated by considering the heat of fusion of a 100 % crystalline polypropylene [20] and it was around 47%.

Results for mixing indexes are shown on **Figure 11**. An important improvement on mixing quality with increasing number of extrusion cycles was observed. This result can be attributed to the shear stress and elongation during the twin-extrusion process that conduct to a better dispersion and distribution of the fibres into the PP matrix.

Discussion

The mechanical properties of composites depend on several factors, which act simultaneously. Thus, according to obtained results, it would be appropriate to analyse the effect of several factors that could affect the final performance of the studied materials:

1. Mechanical and thermal degradation of the matrix: Polypropylene degradation

During the extrusion process, molecular chain scission takes place due to shear forces. Higher molecular weigth chains are more likely to be affected by this effect, as a result of a greater probability of finding chain parts at both sides of shear stress [16]. This mechanical degradation has an important effect on the viscosity of the neat matrix, as it was determined

on melt flow index tests, especially when shear forces are higher. Therefore, it was expected that fibres incorporation lead to an increase on the matrix mechanical degradation. When fibres are incorporated into the matrix, viscosity increases and shear stresses developed in the extruder become higher and this can affect the mechanical response of the matrix. Chain scission could be more important in this case conducting to lower mechanical properties.

Concerning thermal decomposition, accounted as mass loss, it was found that the polypropylene matrix suffered low or no effects. It was also determined that the matrix crystallinity remained almost unchanged.

2. Fibres degradation

It is worth known that reinforcement effects depend on the aspect ratio (*L/d*). Fibres were subjected to high shear forces during extrusion (intensive mixing) and their dimensions, length an diameter, were significantly reduced.

Obviously, a reduction in fibres length is detrimental for attaining high load transfers. Although it is an undesirable effect, it takes place during extrusion. There was also an important reduction on fibres diameter (defibrillation) that helped keeping the aspect ratio high. [21]. However, length reduction was much higher than the reduction in the diameter, leading to an overall reduction on the aspect ratio (1/6 of its original value).

The mechanical degradation of flax fibres took place mainly during the first extrusion step. Additional extrusion cycles did not cause significant further degradation. This degradation was important and was in part responsible for low strength found on obtained composites. Short fibres are not capable of bridging propagating cracks because each of its ends is closer to the edge of the crack than the critical value. Thus, short fibres are more likely to be pulled out from the matrix. This is consistent with the results found on the impact tests, where composites showed higher values of absorbed energy than that of the unfilled matrix. SEM micrographs (**Figure 8.b.**) also confirmed this result important (fibres pulled out).

Regarding thermal degradation, an important mass loss (12 %) was observed for flax fibres. This degradation could be macroscopically detected through poor organoleptic properties (such as colour, smell, etc.). Decomposition owed mainly to lignin and hemi cellulose degradation. As natural fibres consist mainly in cellulose fibrils embedded on a lignin – hemicellulose matrix, its degradation leads to defibrillation, which accounts for the important fibre diameter reduction.

3. Agglomerate formation – Matrix-fibre interface

Bad fibre dispersion and distribution owes mainly to a great difference between the surface polarity of the polypropylene and the natural fibres. Natural fibres are hydrophilic in nature and present polar groups on its surface (hydroxyl) which are repelled by the non polar hydrophobic polypropylene surface [3]. Polar groups on natural fibres are attracted to each other, leading to agglomerate formation.

This tendency to form agglomerates could cause an undesirable effect on the mechanical properties. Fibre agglomerates act more like a discontinuity within the matrix structure than as a reinforcing material. When extrusion cycles increased, composites became more homogeneous. However, small agglomerates were still present but they were not visible to the nude eye.

The type and characteristics of fibre-matrix bonding also affect the mechanical behaviour of a composite. In this work, all composites showed an important amount of fibres pull out. This mechanism was promoted mainly by two factors. Firstly, after extrusion, fibres aspect ratio was strongly reduced. Therefore, most of the fibres located on the way of the propagating crack had at least one of its ends at a smaller distance than the critical length required for pull out mechanism to show up ($L_c/4$) [21]. Secondly, a moderate adhesion between fibres and matrix was observed (extracted fibres showed completely clean – resin free surface). Therefore, there is a strong interdependency of these two factors in order to attain high absorbed energy values due to pull out effects. If the interfacial bonding is too high, not many fibres will be extracted, but they will consume an important amount of energy during the process. But if the interfacial bonding is poor, most of the fibres will be pulled out from the matrix, consuming small amounts of energy individually but an important one collectively.

Theoretical models

There are a great number of models for predicting the mechanical properties of composites [21]. The simplest one assumes an iso-strain condition. This means that both, the fibres and the matrix are subjected to the same strain ($\varepsilon_F = \varepsilon_M = \varepsilon_C$, where *F*, *M* and C stand for fibre, matrix and composite respectively). Consequently, mechanical properties are determined by the simple rule of mixtures:

$$\sigma_C = \sigma_F \cdot V_F + \sigma_M \cdot (1 - V_F) \tag{3.a}$$

$$E_C = E_F \cdot V_F + E_M \cdot (1 - V_F) \tag{3.b}$$

where σ is the tensile strength; V_F is the reinforcement's volume fraction and E is the elastic modulus.

In a similar way, the iso-stress model assumes that the fibres and the matrix are subjected to the same stress level, with perfect bonding between them, resulting in different strains for each component. Mechanical properties are determined by means of a reciprocal rule of mixture:

$$\frac{1}{\sigma_C} = \frac{V_F}{\sigma_F} + \frac{(1 - V_F)}{\sigma_M} \tag{4.a}$$

$$\frac{1}{E_C} = \frac{V_F}{E_F} + \frac{(1 - V_F)}{E_M} \tag{4.b}$$

The two models described above represent the upper and lower bounds for the real composites behaviour. However, many models have been suggested that lie among these boundaries (Hill, Chamis, Halpin – Tsai, Nielsen, etc.) [22].

Within these models, a powerful one yet simple to apply, is the Halpin – Tsai model. Mechanical properties estimated by using this model agree quite well with experimental results for a great variety of reinforcement geometries, such as fibres, ribs, plates, etc. The mechanical properties of the composites are determined by using eq 5.a and 5.b, where p is the desired property, and η is a parameter representing the reinforcement geometry.

$$\frac{p}{p_m} = \frac{1 + \xi \cdot \eta \cdot V_F}{1 - \eta \cdot V_F} \tag{5.a}$$

$$\eta = \frac{(p_F / p_M) - 1}{(p_F / p_M) + \zeta} \tag{5.b}$$

All models are appropriate for unidirectional composites. When applying them to randomly oriented discontinuous fibres composites, some approximate solutions may be useful. These solutions take into account the longitudinal and transverse properties of the components.

Tsai and Pagano [22] related the elastic modulus value for randomly oriented discontinuous fibres composite materials to the elastic modulus of oriented fibres (eq 6)

$$E_{com} = \frac{3}{8}E_{11} + \frac{5}{8}E_{22} \tag{6}$$

where E_{11} is the elastic modulus in the longitudinal direction and E_{22} in the transverse direction. Therefore, values for E_{11} and E_{11} were calculated by means of the Halpin Tsai model (applying two correction factors, η_L (7.c) and η_T (7.d)) as shown in eq 7.a and 7.b.

$$E_{11} = \left(\frac{1 + 2 \cdot (L_f / d_f) \cdot \eta_L \cdot \alpha \cdot V_f}{1 - \eta_L \cdot \alpha \cdot V_f} \right) \cdot E_m \tag{7.a}$$

$$E_{22} = \left(\frac{1 + 2 \cdot \eta_T \cdot \alpha \cdot V_f}{1 - \eta_T \cdot \alpha \cdot V_f} \right) \cdot E_m \tag{7.b}$$

where

$$\eta_L = \frac{(\beta \cdot E_f(d_f)/E_m) - 1}{\beta \cdot E_f(d_f)/E_m + 2 \cdot (L_f / d_f)} \tag{7.c}$$

$$\eta_T = \frac{(\beta \cdot E_f(d_f)/E_m) - 1}{\beta \cdot E_f(d_f)/E_m} \tag{7.d}$$

Two correction factors were used. The first one, α, was applied to the reinforcement volume fraction and could be related to mixing quality. It was estimated by calculating the mixing index as described ($\alpha = 1 - MI$).

In order to take into account the effect of the mechanical degradation of the fibres on their properties, their elastic modulus was estimated by using the Grifith's model [23]

$$E_f(d_f) = A + \frac{B}{d_f} \tag{8}$$

Where A and B are constants and d_f is the diameter of the fibre.

By using the calculated values for α and the average value of each dimension of the fibre (L, d, and L/d), and taking β value as a fitting parameter, the model was applied.

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The experimental values of the Young's modulus of the polypropylene matrix were used for model prediction. The modulus of composites was estimated by applying the model in four different cases (always by using equation 6):

a)   Considering a constant value for the modulus of the fibres;

b)   Calculating the modulus of the fibre from the Grifith's model (equation 8)

c)   Similar to b) and with the mixing factor: α

d)   Similar to c) and with the fitting parameter β

Results are shown in **Figure 12.** For the a) case important disagreements were observed. It can be seen that a horizontal line was found for Halpin-Tsai model. By computing the change of the fibre modulus with the diameter (Grifith's model) (case b) the model prediction did not fit properly the experimental values. Taking into account the mixing factor (case c) the line becomes a curve. However, all experimental values were within 85 – 90 % of predicted ones. The fibres modulus parameter values (case d) which fit the experimental ones, is close to 0.12. The thermal degradation of the natural fibres leads to a reduction in its elastic modulus due to the damage suffered by some of its component (morphological changes). However, the modulus of the fibres after degradation should be close to 6 GPa. We considered that it is not possible to obtain such value and that another effect which could be taken into account is the fibre/matrix interphase, which is not perfect in our case.

Conclusions

The feasibility of re-use of polypropylene – flax composite was analysed. This study was useful in order to analyse the influence of mixing on thermo-mechanical degradation suffered by the matrix and the fibres and the effect of these factors on the mechanical properties of final composites.

Polypropylene suffered a significant mechanical degradation due to intense chain scission (melt flow index – apparent viscosity). However, this degradation had a low influence on its mechanical properties, and so that, on the mechanical properties of the composites. On the other hand, the crystallynity remained almost unchanged during extrusion.

Obtained composites showed an acceptable mechanical response. Their elastic modulus was at all stages higher than that of the neat polypropylene. Flax fibres acted as preferred sites for crack formation and propagation due to their small size (aspect ratio lower than the critical

value) after the significant mechanical degradation suffered. On the other hand, these materials exhibited a good impact performance

It could be pointed out that three extrusion cycles are enough for attaining a good dispersion and distribution of the fibres. However, the use of compatibilizers could contribute to obtain a better fibre dispersion (reducing the tendency to form aggregates).

Finally, it is important to note that the application of mathematical models for predicting the mechanical properties of natural fibre composites is particularly difficult for this type of reinforcement due to the poor fibre/matrix interphase (which is not considered on the model). Nevertheless, the application of a micromechanical model allow to say that the mixing quality and the fibre modulus at each stage have a great influence on the modulus of the composite, higher than that of the aspect ratio and diameter of the fibre.

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Table captions

Table 1. Matrix and fibers properties

Table 2. Melt Flow Index results for composite materials

Figure Captions

Figure 1. Compression moulding cycle

Figure 2. Polypropylene apparent viscosity as a function of extrusion cycle.

Figure 3. a) Tensile elastic modulus and b) Tensile strength of matrix and composites as a function of extrusion cycle.

Figure 4. a) Flexural modulus and b) Flexural strength of matrix and composites as a function of extrusion cycle.

Figure 5. Energy per unit area absorbed during Izod impact test as a function of extrusion cycle.

Figure 6. Energy per unit thickness absorbed during falling weight tests as a function of extrusion cycle.

Figure 7. Radial and circumferential cracks under falling weight tests on a) polypropylene plates and b) composite plates.

Figure 8. SEM micrographies of fracture surface for: a) Polypropylene and b) Composites.

Figure 9. Natural fibres size distributions as a function of extrusion cycle.

Figure 10. Thermogravimetric analysis results.

Figure 11. Mixing Index results as a function of extrusion cycle.

Figure 12. Elastic modulus results: experimental and predicted values.

Matrix properties (PP)		Fibres properties (flax)	
Tm (ºC)	168 – 172	Single fibre tensile strength (MPa)	600
Tg (ºC)	-15 – -18	Single fibre Young´s Modulus (GPa)	48.8
Density (g/cm^3)	0,905	Density (g/cm^3)	1.44
Yield stress (MPa)	31	Surface energy polar component (mJ/m^2)	6.25
Elongation at yield (%)	10 – 12	Total surface energy (mJ/m^2)	20.57
Young Modulus (GPa)	1.00 – 1.15	Single fibre tensile strength (MPa)	600

T = 190 ºC / 5.0 kg.	**Melt Index (gr/10 min)**	$\eta_{apparent}$ **(Pa.s)**
COM 4	1.79 ± 0.21	13200 ± 1750
COM 5	5.37 ± 0.16	4400 ± 128