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Manufacture of compression moulded PLA based biocomposites: a parametric study

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Abstract.

In this study, an analysis of the fabrication parameters that influence on the strength of biodegradable composites is presented. The effect of heating temperature, pressure, number of plies, fibre, and matrix on the tensile strength was studied. Biodegradable composites were manufactured using film stacking and compression moulding process. The strengths of biocomposite based on two different polylactic acid (PLA) matrix, and reinforced with four different woven fibres of jute, flax and cotton were compared. The aim of this work is the development of an optimized manufacture process to reduce costs and production time of high strength biocomposites. As a result, a tensile strength higher than 100 MPa was obtained with a preheating time of 2 minutes and 3 minutes of heating under pressure. This result means a significant reduction of production time that can lead to reduce the manufacture cost of these materials.

Keywords: Biocomposites; Compression moulding; Fabrics/textiles; Mechanical properties

1. INTRODUCTION

Composite materials combine lightweight and high specific stiffness and strength, these properties make composites suitable for a wide range of applications in industry [1-3].

During the last years, several researchers have introduced natural fibres as reinforcements to develop biodegradable composites [4,5]. The use of biocomposites as an alternative to traditional composites materials can lead to a reduction of the environment impact. Thus, numerous researchers are trying to improve manufacture processes reducing production costs and increasing their mechanical properties.

Some of the conventional composites fabrication techniques can be applied to manufacture biocomposites. Thermoplastic injection moulding uses a screw-type plunger to force the plastic material into a mould [6]. All Cellulose-Composites (ACC) are obtained by the solution of the superficial cellulose of the fibres, and this cellulose acts as matrix of the fibres when dried. ACC are characterised by a total cohesion between matrix and reinforcement [7,8]. Other fabrication procedures are heating under vacuum [9], RTM (resin transfer moulding) [6,10,11], pultrusion [6] or electrospinning for bio-nanocomposites [12]. However, compression moulding after a film stacking of fibre and matrix plies is one of the most popular processes to manufacture biocomposites [13-17].

Satyanarayana et al. [18] found compression moulding as the better option to obtain a composite based on natural fibres in terms of mechanical properties. This method also uses small amounts of energy, so the green concept is reinforced. The main parameters that influence on the mechanical properties of biocomposites manufactured using compression moulding are fibre material, matrix type, fibre quantity, heating temperature, pressure and time.

Therefore, numerous parameters influence on the compression moulding method, and many researchers have manufactured biocomposites using different values of these parameters. For example, Ochi [14] manufactured a PLA composite reinforced with kenaf fibres by compression moulding with a previous prepreg of the fibres.

Temperature was 160°C with a 5 minutes preheating and a hot pressing during 10 minutes with 10 MPa pressure, obtaining 223 MPa tensile strength. Jang et al. [13] manufactured PLA based biocomposites reinforced with coconut fibres at 200°C, obtaining a tensile strength of 65 MPa. Song et al. [15] kept at 170°C during 5 min and applied 2.45 MPa pressure to obtain a 65 MPa tensile strength biocomposite made of hemp fibres and PLA matrix. Porras and Maranon [16] manufactured a biocomposite reinforced with bamboo fibres by compression moulding after an extrusion of the PLA matrix with a temperature of 160°C and a pressure of 556.7 kPa resulting in 77.55 MPa tensile strength. Graupner et al. [17] tested mixtures of different fibres and PLA, obtaining a 81.8 MPa tensile strength with a 40% lyocell and 60% PLA biocomposite, applying 4.2 MPa of pressure during 20 min at 180°C. Bodros et al. [19] followed a compression moulding after film stacking, using flax, glass fibre, hemp and sisal, applying different conditions depending on the fibre and the matrix.

However, very few studies have analysed the influence of the main manufacture parameters on biocomposites strength. Ochi [14] studied the influence of kenaf reinforcement quantity, determining optimum strength of biocomposites with around a 70% volume fraction of kenaf fibres. Jang et al. [13] studied the reinforcement quantity parameter applying different treatments to the fibres, but with a maximum reinforcement volume fraction of 10%. Song et al. [15] tested different woven architectures of hemp, founding that twill woven provided higher strength than plain woven. Bodros et al. [19] compared biocomposites reinforced with flax fibres using

different matrices: PHB (Polyhydroxybutyrate), PBS (Polybutylene succinate), PLA, PLLA (poly-L-lactide), and PP. PLA and PLLA provided the better mechanical properties in terms of stiffness and strength. In addition, they showed that flax/PLLA biocomposites can be competitive with the glass-fibre/polyester composite.

Therefore, there is a need of parametric studies to optimize the results of compression moulding manufacturing process. In this work, two different PLA matrices were reinforced with jute, flax, and cotton fibres to analyse the influence of temperature, number of plies, and pressure. The optimal manufacture process has led to a fully biodegradable biocomposite with a tensile strength higher than 100 MPa. The main contribution of this paper is the significant reduction of production time, 2 minutes of preheating and 3 minutes of heating under pressure, that can lead to reduce the manufacture cost of these materials.

2. FABRICATION PROCESS

Biocomposites were manufactured using jute, cotton and flax fibres as reinforcement of PLA matrix applying compression moulding method.

2.1. Materials

Four woven fibres were used as reinforcement: plain weave jute (J); basket weave cotton (C), basket weave flax (BF); and plain weave flax (PF). No chemical pre-treatment was applied to woven fibres. The four woven materials were subjected to uniaxial tensile tests to obtain their Young modulus and tensile strength, Table 1. Tensile strength ranges from 116.5 MPa of cotton woven fibres to 271.62 MPa of basket weave flax. Tensile tests were conducted as described below in section 2.3.

PLA is a thermoplastic resin that was acquired in pellets form. Two kinds of PLA polymer were used: 3260HP and 10361D, both provided by Natureworks LLC.

3260HP PLA is aimed to an extrusion process; its typical application is to obtain final products as cups, plates or cutlery. 10361D PLA is specifically aimed as a binder of natural fibres. PLA properties were obtained from [18]. The PLA density is 1.24 g/cm^3 and the melt temperature is $145\text{-}170^\circ\text{C}$. Mechanical properties of PLA are shown in Table 1.

2.2. Compression moulding method

Compression moulding method is schematically shown in Figure 1. First, the PLA pellets are placed between two thermoheated plates at a temperature of 185°C . A universal test machine Servosis ME-404/100 + PCD-1065 with a limit load of 900 kN was used to apply 2 MPa pressure. A 0.5 mm thickness uniform film was obtained.

Then, the matrix films are stacked alternatively with woven plies. The stacked plies are placed between the thermoheated plates. After a pre-heating time, pressure was applied using the same universal test machine.

The values of temperature, pressure, and number of plies were modified to analyse their influence on the biocomposite tensile strength. Temperature of compression ranged between 175°C and 200°C , pressure from 0.8 MPa and 40 MPa, and number of plies from 2 to 4.

The quantity of reinforcement was stated in a weight ratio of 65%, this parameter was not analysed in this work because Ochi [14] studied its influence on biocomposite strength founding that this is the optimum value for compression moulding procedures. Ochi [14] also found that natural fibres can be degraded at temperatures higher than 180°C , but if this temperature is applied during a short time period, fibres do not present damage. In this work, preheating time is set to 2 minutes, heating under pressure time in 3 minutes. These are the minimum times to assure PLA melting, its homogenous

distribution in the laminate, and a slowly application of pressure to avoid fibre misalignment.

2.3. Mechanical testing

Tensile tests were performed in a universal test machine Instron 8516. The cross-head speed was set to 0.5 mm/min. Samples were cut in rectangular specimens of 120 x 30 mm. The thicknesses of the samples depend on the number of plies and the type of woven fibre. Tensile strength was determined with the peak force of the force-displacement curve.

3. RESULTS AND DISCUSSION

The results of the tensile tests are shown in Table 2. The modification of the different parameters led to 21 different configurations, four specimens were tested for each configuration. The influence of the main parameters is analysed in the next sections.

3.1. Type of fibre and number of plies

The comparison of the tests no. 1-8 can illustrate the influence of fibre and number of plies on tensile strength. These specimens were manufactured at the same heating temperature (185°C) and pressure (32 MPa), and PLA matrix was 10361D.

Figure 2 shows a graphical representation of the results. The best results were obtained with the basket weave flax (BF), following by plain weave flax (PF), jute (J), and cotton (C). These results are in agreement with the fibre strengths shown in Table 1. Thus, the biocomposite tensile strength is directly related to the fibre strength.

The analysis of the influence of number of plies shows that there is more cohesion in bilayer biocomposites than the four layers ones, with the exception of cotton. This is consistent with the cohesion problems in composite materials. However, the difference between two and four layers is smaller than standard variation for flax woven

biocomposites. Consequently, basket weave flax seemed to be the best option to reinforce biocomposites independently of the number of layers.

3.2. Type of PLA matrix

The two PLA matrix (3260HP and 10361D) were compared in tests no. 9 and 10. These specimens were manufactured with basket weave flax plies at the same conditions of pressure (32 MPa) and temperature (185°C).

Results show that 3260HP PLA matrix lead to a higher tensile strength, 116.33 MPa, than PLA 10361D matrix, 104.70 MPa. These results are in contradiction with the supplier recommendations because 10361D is specifically designed for natural fibres binder, while 3260HP is make for extrusion processes. However, the higher properties obtained with 3260HP matrix are only valid for two layers laminates, it was not able to spread into the inner layers when it was applied to a four layer biocomposite because temperature distribution was not uniform. Thus, 10361D PLA was selected because it was able to provide high tensile strength independently on laminate thickness.

3.3. Heating temperature

The influence of heating temperature, from 175°C to 200°C, was analysed through comparison of tests no. 10-15. These results are represented in Figure 3. These specimens were manufactured from 10361D PLA matrix reinforced with basket weave flax woven plies, and the applied pressure was 32 MPa.

Heating temperatures over 200°C produced fibre damage due to overheating. On the other hand, the matrix was not completely melted for heating temperatures below 175°C for short processing times. Thus, the temperature must be high enough for matrix melting, but not so high to produce fibres damage. As Figure 3 shows, specimens manufactured with heating temperatures between 180°C and 185°C presented the

maximum tensile strength. However, some of the specimens produced at 175°C and 180°C were discarded because of the presence of delaminations. The tensile strength corresponding to 180°C heating temperature shown in Figure 3, 110.92 MPa, was obtained as the average of intact specimens, however many other specimens were manufactured but not tested due to delaminations. Consequently, 185°C was chosen as the optimum heating temperature.

3.4. Pressure

The effect of pressure was studied comparing the tests no. 10, 16-21. The heating temperature was stated at 185°C to manufacture 10361D PLA based biocomposite reinforced with basket weave flax fibres. Results are graphically represented in Figure 4. Tensile strength increases with manufacture pressure until 8 MPa, then there is plateau until a manufacture pressure of 32 MPa, finally tensile strength decreases with pressure. Pressure over 32 MPa produced fibre breakage, while pressure under 8 MPa mean lack of cohesion in the biocomposite. Thus, there is a wide range of pressure, between 8 MPa and 32 MPa, that can be used to produce biocomposites with similar mechanical properties.

4. OPTIMIZED MANUFACTURING METHOD

A review of the results obtained by other authors manufacturing biocomposites by compression moulding with natural fibres and biodegradable matrices has been included in table 3 to analyse the significance of the present results. The results are also graphically represented in Figure 5. This comparison must be carefully analysed because even when the same manufacturing method is applied to the same type of matrix and fibres there are many uncertainties as fibre content or fibre properties. For example, flax fibres raised in different countries can lead to different mechanical properties.

In the case of biocomposites reinforced with flax fibres, the present method showed the higher tensile strength and the minimum production time. It should be noticed that these results were obtained with the maximum value of reinforcement ratio, but the possibility to manufacture biocomposites with high fibre weight ratios can also be considered an advantage of the present method.

Considering biocomposites based on jute fibres, several authors [23-25] have achieved better results in term of tensile strength. However, the manufacturing time was much higher than the limit of 5 minutes applied in the present work. The biocomposites manufactured with cotton fibres showed higher strength than those reported by Graupner [28] and the present production time is lower.

Table 3 also includes the biocomposites reinforced with kenaf fibres because these fibres can lead to higher values of strength. However, the production time required was higher than the time used in this work.

CONCLUSIONS

The influence of heating temperature, pressure, type of fibre and matrix, and number of plies is studied in this work. The results of 21 tests configurations are analysed, leading to the next conclusions:

- Biocomposite strength is directly related to fibre strength. This conclusion is not obvious because implies similar adhesion between the matrix and the different fibres. Thus, biocomposites manufactured with basket weave flax fibres presented the higher tensile strength.
- The influence of matrix is related to the number of plies. For two-ply biocomposites the higher strength was obtained with 3260HP PLA matrix. However, this matrix presented spreading problems for higher number of plies

due to the non-uniform temperature distribution. Thus, 10361D PLA matrix was selected as the best option.

- A heating temperature of 185°C was found as the optimum value. Higher heating temperatures produce less-strength biocomposites because fibres are damaged due to overheating. While, matrix melting is not complete for lower heating temperatures and short processing times.
- A wide range of pressure during compression moulding, from 8 MPa to 32 MPa, led to similar values of tensile strength. Lower pressure implies lack of cohesion in the biocomposite, and higher pressure produce fibre breakage.

The present method can produce a biocomposite with a tensile strength higher than 100 MPa in a production time of 5 minutes, 2 minutes of preheating and 3 minutes of heating under pressure. This result implies a significant reduction of production time leading to a reduction of the manufacture cost of biocomposites.

However, more studies must be carried out to improve this method. The conclusions are stated only from the tensile strength point of view. There are other mechanical and physical properties, as biodegradability, that should be studied to verify the quality of biocomposite manufactured with the present method.

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Figure captions

Figure 1. Scheme of the manufacturing process to obtain the biocomposite from natural fibres woven plies and PLA in pellets.

Figure 2. Tensile strength obtained for different number of plies and fibres: basket weave flax (BF), plain weave flax, plain weave jute (J), and basket weave cotton (C).

Figure 3. Tensile strength as a function of heating temperature.

Figure 4. Tensile strength as a function of the pressure applied in compression moulding method.

Figure 5. Tensile strength as a function of the processing time. Comparison of present model and literature review for different fibres and matrices.

Table captions

Table 1. Mechanical properties for natural fibres woven plies and PLA matrix.

Table 2. Tensile strength of biocomposite manufactured with different fibres, matrix, temperature, pressure, and number of plies.

Table 3. Comparison of the present results and the literature review in term of processing time and tensile strength.

<i>Material</i>	<i>Tensile strength (MPa)</i>	<i>Young modulus (GPa)</i>
BF fibres	271.62	7.84
PF fibres	225.39	8.5
J fibres	136.18	7.385
C fibres	116.5	12.34
PLA [17]	54.27	3.18

<i>Test no.</i>	<i>Temperature (°C)</i>	<i>Pressure (MPa)</i>	<i>PLA</i>	<i>Fiber</i>	<i>Plies</i>	<i>Tensile strength (MPa)</i>	<i>Desv. (MPa)</i>
1	185	16	10361D	BF	2	105.95	4.54
2	185	16	10361D	BF	4	102.00	4.55
3	185	16	10361D	PF	2	98.93	1.58
4	185	16	10361D	PF	4	94.83	4.48
5	185	16	10361D	J	2	73.30	3.86
6	185	16	10361D	J	4	64.43	2.44
7	185	16	10361D	C	2	58.28	0.29
8	185	16	10361D	C	4	59.93	4.84
9	185	32	3260HP	BF	2	116.33	2.42
10	185	32	10361D	BF	2	104.70	4.44
11	175	32	10361D	BF	2	102.76	0.88
12	180	32	10361D	BF	2	110.92	6.32
13	190	32	10361D	BF	2	95.32	4.69
14	195	32	10361D	BF	2	96.80	4.52
15	200	32	10361D	BF	2	99.13	7.32
16	185	0.8	10361D	BF	2	83.75	1.91
17	185	4	10361D	BF	2	95.73	5.31
18	185	8	10361D	BF	2	106.94	4.47
19	185	16	10361D	BF	2	105.95	4.54
20	185	24	10361D	BF	2	103.55	4.51
21	185	40	10361D	BF	2	90.42	3.76

<i>Author</i>	<i>Material</i>	<i>Tensile Strength (MPa)</i>	<i>Production time (min)</i>	<i>Reinforcement weight ratio (%)</i>
present	Flax/PLA	106.94	5	65
Bodros et al. 2007 [19]	Flax/PLA	100	8	30
Nassiopoulos and Njuguna, 2015 [20]	Flax/PLA	72	5	40
Barkoulaa et al. 2010 [21]	Flax/PHB	41	17.5	40
Yun et al. 2010 [22]	Flax/PLA	24	35	35
Plackett et al. 2003 [23]	Jute/PLA	100.5	12	40
Khondker et al. 2006 [24]	Jute/PLA	84	10	38
Arifuzzaman et al. 2013 [25]	Jute/PLLA	81	10	65
present	Jute/PLA	73.3	5	65
Tao et al. 2009 [26]	Jute/PLA	50	9	30
present	Cotton/PLA	59.93	5	65
Graupner, 2008 [27]	Cotton/PLA	41.2	20	40
Ochi, 2008 [14]	Kenaf/PLA	223.3	10	70
Graupner and Müssig, 2011 [28]	Kenaf/PLA	82	20	40
Graupner and Müssig, 2011 [28]	Kenaf/PHB	70	20	40