DEVELOPMENT OF NATURAL FIBRE REINFORCED POLY(LACTIC ACID) BIOCOMPOSITES

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Abstract

In our work we have produced natural resource based biodegradable polymer composites from Poly(Lactic Acid) (PLA) and plant fibres by using film stacking technology. We have investigated the effect of the compression molding parameters on the fibre-matrix adhesion and on the mechanical properties. It was found that significant improvements were reached due to the increased pressure during compression molding.

1. Introduction

Due to the strict environmental regulations interest has been shown for development of natural resource based biodegradable polymers and composites in the plastic industry [1-5]. Most of the research is related to the Poly(Lactic Acid) (PLA) due to its good mechanical properties. PLA is a thermoplastic biopolymer based on the lactic acid which can be obtained from different carbohydrate sources for example maize, sugarbeet, rice, etc. It can be processed with the conventional thermoplastic technologies. Based on the mechanical properties PLA can be compared to polystyrene (PS) or polyethylene-terephthalate (PET). Although PLA has excellent mechanical properties, it also has some drawbacks. It behaves rigidly and it is sensitive to moisture content [6-8]. Despite its mechanical properties PLA is mainly used as a raw material of packaging, but the development of medical [9] and automotive [10] applications have already begun. Making composites is the best way to improve the mechanical properties of PLA, in order to be able to use it as renewable resource-based engineering materials in the future. The biodegradability and the environment friendly features of the PLA can be preserved by the application of natural fibres. In the recent years many types of natural fibres were used as reinforcement for PLA [11-16].

Granuper et al. [13] compared the reinforcing effect of different types of natural fibres. They prepared 40 wt% cotton, hemp, kenaf and lyocell composites with compression molding. They achieved significant improvements on the mechanical properties, namely the tensile strength increased by 20-50 MPa and the modulus by 3-4 GPa respectively. Bledzki et al. [14] made a comparative study on natural fibre reinforced, injection molded PP, PLA, PHBV composites with 30 wt% fibre content. Significant improvements were reached, the maximum tensile strength was 92 MPa (PLA/Cellulose) and the maximum modulus was 9.6 GPa (PLA/Jute). Also impact (Charpy) tests were made and PHBV/Cellulose composite was found to have the highest impact strength with 33 kJ/m^2 at 23°C and 24 kJ/m^2 at -30°C. Baghaei et al. [15] made PLA/hemp biocomposites with compression molding from a special co-wrapped hybrid yarn prepregs. The effect of fibre content (10-20-35-45 wt%) and wrapping density on the mechanical properties were analyzed. The mechanical test showed that the tensile and flexural properties increased with the fibre content and the wrapping density. The composites with 45 wt% fibre content and 250 turns/m wrapping density had the best mechanical properties. Tensile and flexural strength increased up to 59 MPa and 124 MPa respectively. The SEM images shown that the adhesion between the fibre and the matrix was not fully appropriate, therefore with additional fibre treatment the mechanical properties can be further increased.

In bio-based composites basalt fibre can also be used as a bioinert reinforcing material. Tábi et al. [16] used untreated and silane treated chopped basalt fibers. Significant improvements were reached during the research. It was found that the silane treated basalt fibers have better adhesion with PLA, and the mechanical properties increased by the increasing fibre content. The optimum fibre content was determined, namely the PLA/40 wt% basalt was found to have the best properties with tensile and flexural strength of 124 MPa and 185 MPa, and with tensile and flexural modulus of 8 and 12 GPa respectively. Also thermal tests were carried out, and due to the fibers the storage modulus and the HDT temperature were increased.

2. Materials and methods

Film extrusion grade PLA 4032D was purchased from NatureWorks. The natural fibre based, plain weaved jute, cotton and flax fabrics were purchased from Dél-Alföldi Műszaki Konfekció Ltd. (Hungary). In our previous work the drying characteristic of PLA and the fabrics were analyzed, and the optimum drying conditions were determined. Accordingly PLA was dried at 100°C for 6 hours before the film extrusion, and the fabrics were dried at 120°C for 24 hours. The properties of the raw materials can be seen in Table 1.

Properties of PLA 4032 D	
Density	1.24 cm^3
Tensile Strength	103 MPa
Tensile Modulus	3.45 GPa
Melting Point	155-170 °C
Area weigth of the fabrics	
Jute	400 g/m^2
Flax126	126 g/m^2
Flax200	200 g/m^2
Cotton P695	120 g/m^2
Cotton 9/10	375 g/m^2
Cotton 350h/P	480 g/m^2

Table 1. Properties of the raw materials

In the first step 100 μm thick PLA film was extruded by a sheet extrusion machine (Labtech Scientific LCR 300) (Figure 1.). The extrusion parameters can be seen in Table 2. After the film extrusion a layered structure were made from PLA films and natural fibre based fabrics with 30 wt% fibre content. The composites (Figure 2.) were made by compression molding with two various processes. In the first pressing method the layered structure was pressed at 190°C and 150 bar in one step with Collin P200E press for 5 minutes. In the second processing technology a Schwabentan Polystat 300S press was used where the applied pressure was twice as high as in the first way. The final pressure was built up in 4 stages (0-100-150-300 bar). Each pressure stage was held for 2 minutes, and the pressing temperature was the same as in the first method. Finally, the effect of compression molding process was analyzed on the mechanical properties of the biocomposites by using tensile, flexural test and scanning electron microscopy. Tensile and flexural tests were performed by using a Zwick Z020 universal testing machine. The tests were performed at room temperature and at a relative humidity of 40% by using a cross-head speed of 5 mm/min, and 5 specimens were tested for each composite. The Scanning Electron Microscopy (SEM) was made on the natural fibre reinforced PLA composites by using a JEOL JSM 6380LA type microscope.

Figure 1. PLA film extrusion

Film extrusion parameters	
Extruder zone 1.	220°C
Extruder zone 2.	225°C
Extruder zone 3.	230°C
Extruder zone 4.	235°C
Extruder zone 5.	240°C
Die zone 1.	245°C
Die zone 2.	240°C
Die zone 3.	245°C
Srew rotational speed	17 rpm
Pull roller 1. speed	2.5 rpm
Pull roller 2. speed	3 rpm
Reeling speed	8 rpm

Table 2. PLA film extrusion parameters

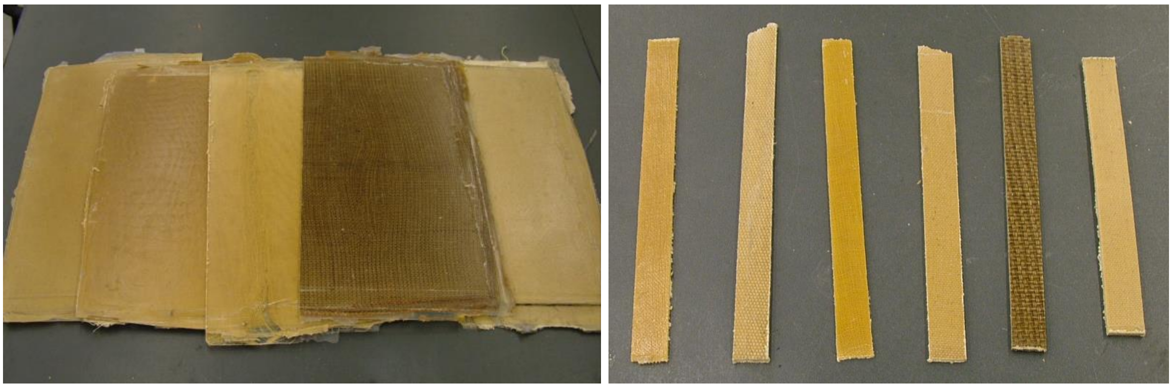

Figure 2. PLA/natural fibre composites made with compression moulding (left) and the test specimens (right)

3. Results and discussion

Figure 3. shows the mechanical properties of the different fibre reinforced PLA composites. According to these results, significant improvements were also reached with the simple, one step compression molding technology. It was found, that the PLA/Flax126 composites had the best tensile properties, and the PLA/Cotton 350h/P composites had the best flexural properties.

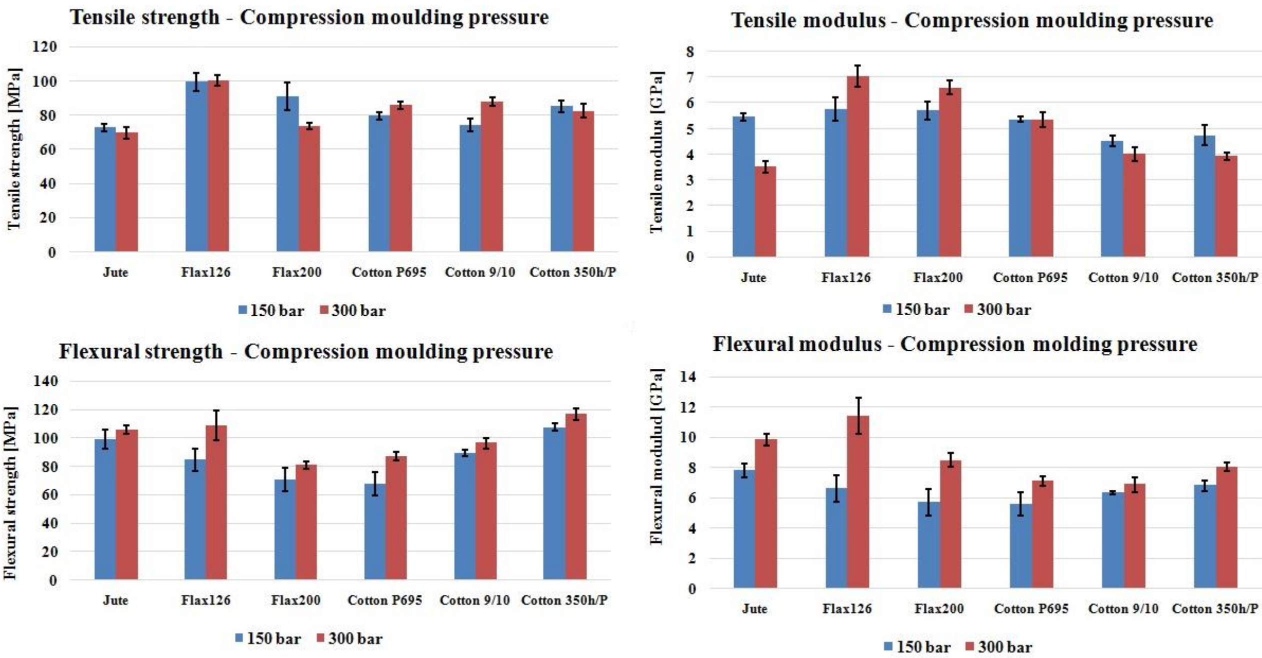

Figure 3. Mechanical properties of the composites

SEM test were performed, and as it can be seen on Figure 4. the PLA is not correctly wetted the fibers. On this basis, we conclude that the 150 bar and the 5 minutes are not enough for preparing composites with good adhesion. Based on the SEM results, the one step compression molding process has been changed to a modified 4 step method. As it can be seen, the mechanical properties increased by the 4 step process. The PLA/Flax126 composites were found to have the best combination of tensile and flexural properties (Figure 3), namely the tensile and flexural strength were 100.1 MPa and 108.9 MPa, the tensile and flexural modulus were 7 GPa and 11.4 GPa respectively. The second SEM tests (Figure 5) show, that due to the higher pressure, and longer pressing time, the adhesion between the PLA and the fibers is improved which caused significant increase on the mechanical properties.

During the tests, it was found that the application of finer fabrics with lower area weight resulted in better mechanical properties, and higher increase due to increased pressure. As it can be seen in Figure 4. and 5., the adhesion between fibers and PLA were better in the case of finer fabrics (PLA/Flax126, PLA/Flax200).

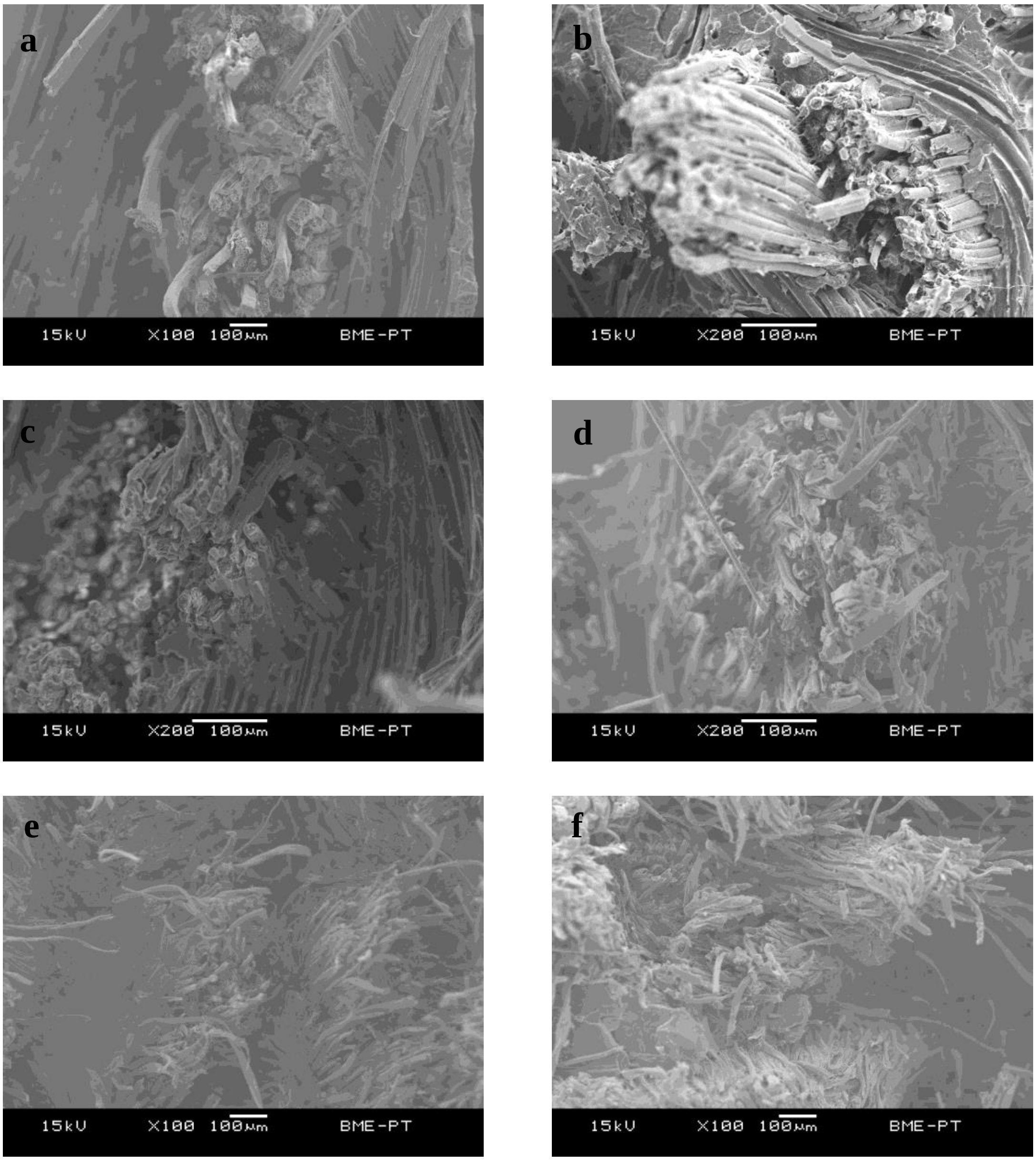

Figure 4. SEM images of the composite made with 150 bar a) PLA/Jute, b) PLA/Flax126, c) PLA/Flax200, d) PLA/Cotton P695, e) PLA/Cotton 9/10, f) PLA/Cotton 350h/P

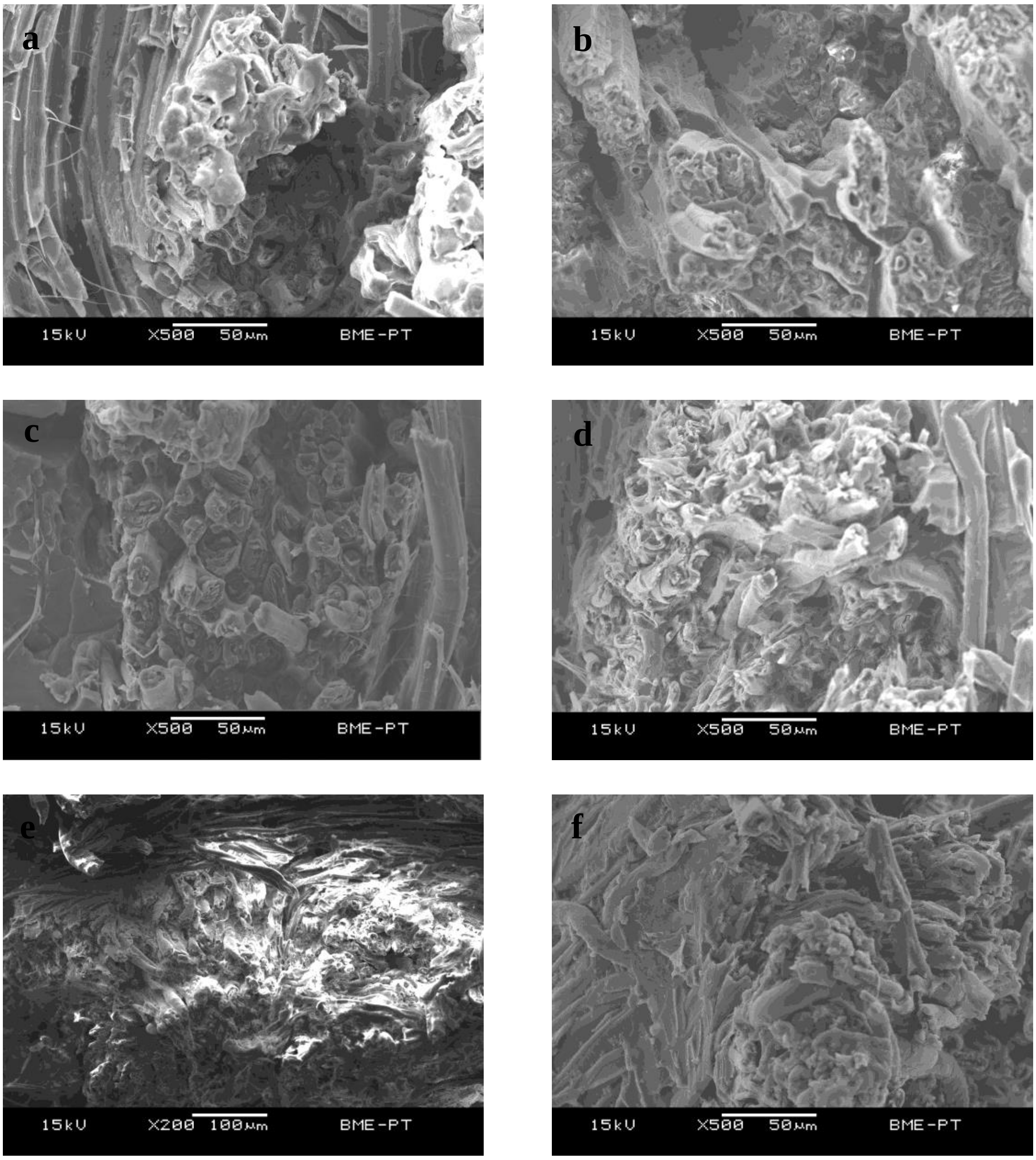

Figure 5. SEM images of the composite made with 300 bar a) PLA/Jute, b) PLA/Flax126, c) PLA/Flax200, d) PLA/Cotton P695, e) PLA/Cotton 9/10, f) PLA/Cotton 350h/P

4. Summary

In our work, different natural fiber reinforced PLA biocomposites were prepared by film-stacking. The effect of the fiber reinforcement and the compression molding pressure on the mechanical properties were analyzed and it was found that, the tensile and flexural strength and modulus of neat Poly(Lactic Acid) (PLA) increased by using 30wt% natural fibre reinforcement up to 80-90 MPa and 7 GPa respectively. The investigation of the mechanical properties of the biocomposites revealed that the pressing parameters have significant effect, due to the increased pressure and the 4 step method further improvements were reached with a flexural strength of 110 MPa and a flexural modulus of 11,4 GPa. The best combination of the tensile and flexural properties was reached with PLA/Flax126 composites.

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