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Effect of the processing on the properties of biopolymer based composites filled with wood flour

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ABSTRACT: Wood-polymer composites (WPCs) are well known today in the field of industrial applications, because of several advantages they can grant if compared with mineral filler-polymer composites. These advantages regard the low cost of wood based fillers, the reduced specific weight, the lower hazards for production workers in case of inhalation, the special aesthetic features, environmental issues. The scientific literature reports studies regarding polymer matrices like, for instance, polyethylene and polypropylene, in combination with several natural-organic fillers. However, a limit of these composites is represented by the fact that there is not a full biodegradability: this, in fact, regards only the filler, therefore the environmental performance is lower than expected. To overcome this limit, it is necessary to replace the traditional, non-biodegradable polymer matrices (typically, polyolefins) with biodegradable ones. An available solution consists in the use of biodegradable polymers like, for instance, those belonging to the Mater-Bi® family.

In this work, a biodegradable polymer matrix, coming from the Mater-Bi® class, was mixed with a natural-organic filler, wood flour. The investigation was focussed on the comparison between different processing ways (single-screw extruder, twin-screw extruder; compression moulding, injection moulding), in particular the relationships between the mechanical properties and the processing method, the filler type, the humidity content of the components were analyzed. Morphological characterization was also carried out by SEM analysis, in order to get further indications regarding some different behaviours shown by the processing methods and the fillers.

Key words: Wood, Biodegradable Polymers, Mechanical Properties

1 INTRODUCTION

The advantages granted by the use of wood flour-polymer composites in several applications (automotive industry, indoor furnishing, packaging, etc.) [1,2] have widened both the academic and industrial interest in this class of materials. They regard basically the low cost of wood based fillers, the reduced specific weight (in comparison to mineral fillers) the environmental issues (reduction in the use of non-renewable sources), the aesthetic features (the composites can look quite similar to some kinds of wood), the reduced hazard for production workers in case of inhalation [3,4].
Literature reports a number of combinations of natural fibres [5] (especially wood) and polymers (polyolefins in prevalence). However, the main shortcoming of these composites is due to the fact that there is not a full biodegradability (which concerns only the filler). A straightforward solution is then to replace the classic polymers used with biodegradable ones. These can be synthetic (polyesters, polyester amides, polyvinyl alcohol) or come from renewable sources (like vegetable ones, or even wastes). The latter class includes, for instance, starch and starch derivatives, polylactic acid (PLA), polyhydroxyalcanoates (PHA), cellulose, etc [6]. Starch is a polysaccharide extracted from a number of natural sources (corn, in prevalence); it is a thermoplastic polymer which can be processed using the typical polymer processing techniques, and reacts with water degrading through hydrolysis. Novamont Mater-Bi® comes from corn starch which is subjected to chemical modifications to improve and tailor its characteristics.

There are not many studies on biocomposites based on Mater-Bi. The "Z" grade was investigated in conjunction with flax-derived cellulose pulp and sisal, while the "Y" grade was studied in combination with sisal, flax and myscanthus [6].
In the present work, therefore, a biodegradable polymer matrix, coming from the Mater-Bi® class, was mixed with the most widespread natural-organic filler, wood flour. A comparison was carried out between different processing ways (single-screw extruder, twin-screw extruder; compression moulding, injection moulding) and the relationships between the mechanical properties and the processing method, the filler type, the humidity content of the components were investigated. Morphological characterization was also carried out by SEM analysis, in order to get further indications on the properties of the composites.

2 EXPERIMENTAL

2.1 Materials, Processing and Sample Preparation

The Mater-Bi used was kindly supplied by Novamont and the composition is proprietary. However, according to previous studied concerning the Mater-Bi family [6,7], it is known that this class of materials is usually composed by a starch fraction and a syntethic, biodegradable polymer fraction, plus additives.
The wood flour, kindly supplied by La.So.Le. (Udine, Italy) was utilized in two different size classes: "150/200" (average particle diameter 150-200 μm, L/D ≈ 2.8) and "35" (average particle diameter 350-500 μm, L/D ≈ 3.9), respectively labelled as "SDF" and "SDC".
The materials underwent some pre-treatments before processing: the wood flour was always dried in an oven at 70°C overnight; some samples were prepared with the "as-it-is" polymer (and were labelled as "humid") while other were prepared with the polymer having underwent a thermal treatment (in a vacuum oven at 70°C overnight, plus one hour at 90°C) before compounding, in order to assess the effect of water molecules on the polymer during processing.
The materials were then mixed and processed according to several different techniques, as follows:
- Mixing in a Brabender PLE330 batch mixer, at a temperature of 140°C and a speed of 30 rpm, and a residence time which varied according to the time needed to achieve a constant torque.
- Extrusion through a twin-screw, counter-rotating intermeshing compounder (Brabender, D = 45mm, L/D = 7) at a temperature of 140°C and a speed of 30 rpm, residence time 1.5-1.8 minutes.
- Extrusion in a single-screw extruder (Brabender, D=19mm, L/D= 25), followed by a slot die, in order to obtain cast films with the aid of a Collin Univex calendering system. The speed was set at 30 rpm and the temperature profile from 70°C to 115°C. The estimated residence time was 3.7-4 min.
- Extrusion through an industrial twin-screw, corotating extruder (OMC, D = 19mm, L/D = 35) with a speed of 100 rpm and a temperature profile ranging from 90°C to 125°C. The estimated residence time was 2.25-2.75 min.
All the obtained materials were then pelletized and compression-molded using a Carver laboratory press set at 140°C for 4 minutes, in order obtaining the specimens for tensile tests, except for the materials coming from the single screw extruder, where the samples were directly cut out of the films.
In addition, the materials obtained from the corotating extruder were also molded through injection molding, by means of a Sandretto μ30 press, operating at 140°C, with a pressure of 900 bar and an estimated residence time of 2'-4'. The obtained injection-molded specimens were subjected to tensile tests as well.
Tensile tests according to ASTM D882 were performed using an Instron 3365 universal testing machine. At least seven replicates were tested.
Finally, in order to understand and explain some phenomena emerged from the characterization, SEM analysis was carried out by means of a Philips ESEM XL30 apparatus.

3 RESULTS AND DISCUSSION

The outcomes of the mechanical tests for most of the samples and the processing methods are reported in Tab. 1 and 2, showing respectively the data for the neat polymer (humid) and the SDF composites (humid), where E=elastic modulus, TS=tensile strength, EB=elongation at break.

Table1. Mechanical properties of the neat polymer

Virgin, humid			
Processing	E, MPa	TS, MPa	EB, %
Mixer	421	16.7	165
Single screw-TD	378	15.3	366
Single screw-MD	333	26.5	443
Twin screw corot.	373	27.4	1081
Injection mold.	200	15	421
Compounder	312	17.8	450

First, let us the consider mixing, which is the preliminary study processing technique. The addition of the filler leads, as predictable [5] to a stiffening effect, as revealed by the increase of the elastic modulus, but also a decrease of tensile strength and elongation at break.

The analysis of the results of the different processing techniques shows that the corotating twin screw extruder gives the best compromise of mechanical properties, since it grants higher values of the elongation at break, keeping also relatively high tensile strength and elastic modulus. However, very interesting properties were also shown by cast film extrusion (single screw, machine direction) and injection molding. The relatively high tensile strength is probably due to the flow-induced orientation of the particles that these processing techniques impart to the material. On the other hand, injection molded samples show a reduced elastic modulus. This can be explained considering the degradation the polymer can undergo in the process, because of the high residence times; this effect overcomes the opposite one due to fiber orientation, and therefore the ductility is greater.

Table2. Mechanical properties of composite

15%SDF, humid			
Processing	E, MPa	TS, MPa	EB, %
Mixer	543	12.8	5.8
Single screw-TD	530	13.6	6.9
Single screw-MD	491	17.4	13.4
Twin screw corot.	569	12.9	5.8
Injection mold.	306	17.7	12.4
Compounder	542	14.3	7.1

Furthermore, it was observed that the drying pre-treatment (not reported here), on average, causes an increase of the elastic modulus of the materials, and a decrease of the elongation at break. This is probably due to the loss of the plasticizing effect of water. However, these effects are small.

Figs. 1 and 2 show the SEM micrographs of the neat polymer, respectively humid and dry, coming from the corotating extruder. It can be observed that the humid shows smaller and more evenly distributed clusters, and an overall improved smoothness.

With regard to the different size classes, it can be stated that, once more, the differences are relatively small. However, on average, SDF imparts slightly higher resistance if compared to SDC. This can be explained assuming that SDF has a better dispersion within the matrix, in comparison to SDC. The hypothesis was proved by calculating the "mixing index" of some samples, according to the methods described by Kalyon et al [8]. For instance, in the case of twin screw extruded samples (humid), it is 0.97 for SDF and 0.91 for SDC; in the case of injection molded samples, on the other hand, it was 0.95 for both SDF and SDC. The dry samples showed lower values of the mixing index, indicating a worse dispersion of the filler. All these outcomes confirm and explain the results of mechanical tests.

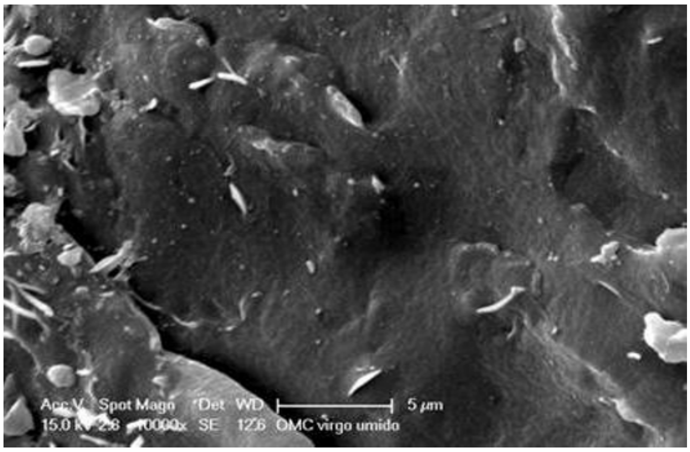

Fig. 1. SEM micrograph of a humid virgin sample

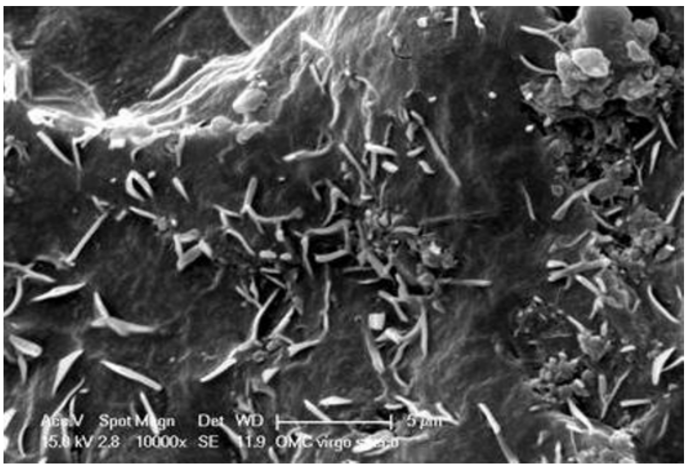

Fig. 2. SEM micrograph of a dry virgin sample

Finally, figs. 3-6 show some SEM micrographs of SDC and SDF filled samples, coming from mixing and injection molding, respectively, with clear differences in the morphology.

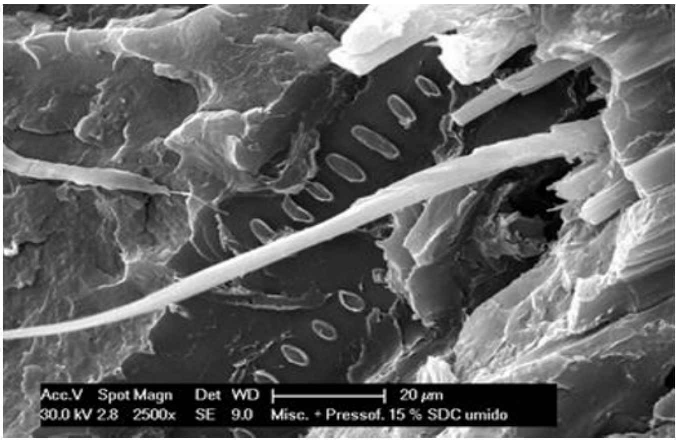

Fig. 3. SEM micrograph of a mixed SDC sample

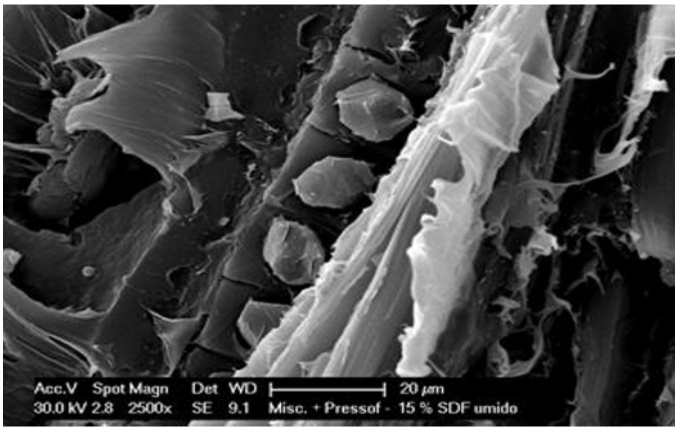

Fig. 4. SEM micrograph of a mixed SDF sample

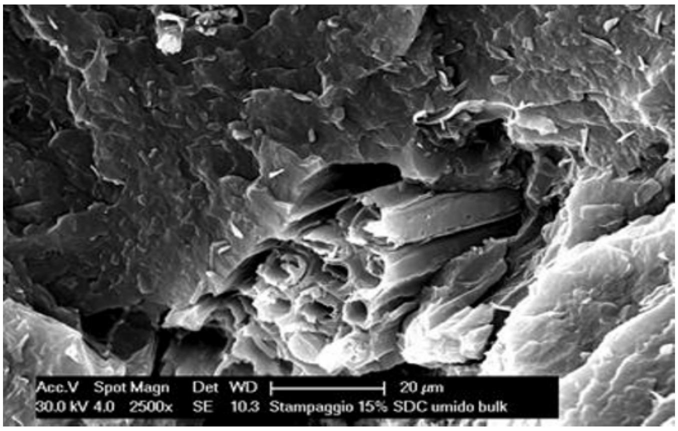

Fig. 5. SEM micrograph of an injection molded SDC sample

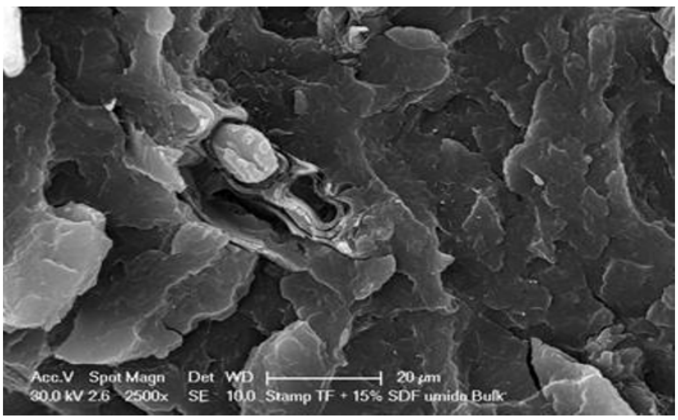

Fig. 6. SEM micrograph of an injection molded SDF sample

In particular, it is easy to observe the improvements achieved with the injection molding, in agreement with the results of mechanical tests.

4 CONCLUSIONS

A biodegradable, starch-based biopolymer was added with two different size classes of wood flour.

The addition of the natural filler caused an increase of rigidity, in particular a significant increase of the elastic modulus and a sharp decrease of the elongation at break. This effect was slightly more marked in the case of the dry samples. With regard to the different size classes, SDF imparted, on average, a slightly higher resistance.

Single screw extrusion followed by cast film production gave the most interesting results in the case of the composite materials (while, for the neat polymer, corotating twin screw extrusion showed to be the preferable compromise). Also injection molding allowed to obtain relatively high tensile resistance, but lower rigidity.

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