LEMONGRASS PLANT LEAF AND CULM AS POTENTIAL SOURCES OF REINFORCEMENT FOR BIO-COMPOSITES

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Abstract: *A possible source of natural reinforcement for bio-composites can be represented by lemongrass plant (Cymbopogon flexuosus), a clumped and perennial grass which belongs to the Poaceae family. This plant is extensively used for several applications such as pharmacology, food preservation and cosmetics but, to the best of our knowledge, few papers were published on its use as source for reinforcement of composites and no one article was focused on the comparison between lemongrass leaves and culms as potential source of natural reinforcement. To this aim, a preliminary investigation on leaf and culm fibers was carried out to compare their physical and chemical features as well as their tensile properties. Furthermore, bio-composites based on a biodegradable starch-derived matrix (MaterBi®) and lemongrass leaf and culm particles were manufactured via extrusion and compression molding. For both fillers, two compositions (i.e., 10% and 20 wt.%) were investigated in terms of morphological and mechanical properties.*

Keywords: natural fibers; biodegradable polymers; lemongrass; bio-composites

1. Introduction

The use of natural fibers as reinforcement for bio-composites has received a great interest both from the academic world and several industrial fields since the 1970s. This attention is motivated by serious environmental issues correlated both to the unsustainable manufacturing processes of synthetic fibers and plastics, and to the limited recyclability of "traditional" composites and their end of life disposal options.

In addition to the most widely investigated and used natural fibers such as flax, jute, sisal and hemp, several researchers focused their attention in the last decade on the use of less common fibers, extracted from local plants, due to several beneficial aspects such as low cost, wide availability and quite good properties [1].

Indeed, the high demand for natural fibers requires discovering new lignocellulosic reinforcements with adequate properties for composite reinforcement.

Driven by this impulse, more than 40 papers were published on this topic just in the past two years. For instance, new natural fibers extracted from Chrysanthemum morifolium stem [2], Calotropis gigantea fruit bunch [3], Aristida adscensionis [4], Eleusine indica grass [5], Stipa obtusa and Jarava ichu leaves [6] and Strelitzia reginae plant [7] were recently investigated.

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In such a context, we focused our attention on lemongrass plant (Cymbopogon genus) with the aim of comparing the leaf and the culm (i.e., stem) of this perennial grass as possible source of reinforcement for bio-composites.

Cymbopogon genus grows worldwide comprising more than 55 species. Among them, Cymbopogon flexuosus and Cymbopogon citratus are the most important ones with the latter widely cultivated to extract their essential oils used in perfumery as well as in food industry [8-9]. Other important applications of lemongrass plant are due to its antiseptic, antibacterial, antimicrobial, antifungal, and anti-inflammatory properties in pharmacology [10-11].

To the best of our knowledge, just a limited research was focused on the use of lemongrass as reinforcement of composites [12-13]. Furthermore, no investigation was addressed to compare lemongrass leaf and culm as potential source of natural reinforcement to date.

To fill this gap, a preliminary investigation was performed out allowing to evaluate which part of this plant (i.e., leaf or culm) is the best one to obtain natural reinforcement having promising features. In particular, the chemical composition of leaf and the culm lemongrass fibers was investigated through standard methods. The main properties of these fibers were assessed by means of thermogravimetric analysis (TGA), scanning electron microscope (SEM), Fourier transform-infrared spectroscopy (FT-IR) and helium pycnometer analysis. Fifty tensile tests were performed on both fibers and the two-parameter Weibull statistical model was applied to interpret statistically the experimental data.

Furthermore, a biodegradable starch-derived matrix (MaterBi®) was reinforced with lemongrass leaf and culm particles (i.e. < 500 μm). For both fillers, two bio-composites were manufactured at varying the particles weight content (i.e., 10% and 20 wt.%) through extrusion and compression molding and their rheological, morphological and mechanical properties were investigated.

2. Materials and Methods

2.1 Fibers

Lemongrass plants were collected in the area of Bangkok, Thailand. After collecting the raw plant, the culm was separated from the leaves. Both parts were first washed several times with tap water to remove dirt and other impurities and then dried at room temperature for 24 hours. Afterward, fibers were extracted from culms and leaves by mechanical separation.

Fifty tensile tests were carried out on both fibers by using a Universal Testing Machine (U.T.M.) model Z005 by Zwick-Roell, equipped with a load cell of 200 N, bonding each single fibers onto a paper frame before clamping to the screw grips of the U.T.M. According to ASTM D3822 standard, the strain rate and gauge length were set equal to 2.5 mm/min and 30 mm, respectively. Before tensile test, the diameter of each fiber was measured at three different random locations along its length thorough a Leica optical microscope model MS5 and the apparent cross-section area was measured by considering it as perfectly circular [14]. Furthermore, due to the large variability in the mechanical properties of natural fibers, a statistical approach (i.e., two-parameter Weibull distribution) was used to interpret the experimental data.

The cellulose and the hemicellulose contents of lemongrass fibers were evaluated through Kushner and Hoffer method [15] and NFT 12-008 standard, respectively. The lignin amount was measured using APPITA P11s-78. Ash content was determined according to ASTM E 1755-61.

The fibers chemical structure was also analyzed via Fourier-transform infrared spectroscopy (FTIR) using an infrared spectrometer model Spectrum II by PerkinElmer. The analysis was performed in transmission mode in a wavenumber range from 400 cm^{-1} to 4000 cm^{-1} with a scan rate equal to 4 cm^{-1}.

The thermogravimetric analysis (TGA) was performed by using a thermobalance TG/DTA model SDT Q600 by TA instruments. Fiber samples were placed in an alumina crucible heated from 30 °C to 1000 °C at a heating rate of 10 °C/min in an inert atmosphere.

The experimental density was evaluated by using a helium pycnometer by Thermo Electron Corporation model Pycnomatic ATC and an analytical balance model AX 224 by Sartorius. The morphological analysis of leaf and culm lemongrass fibers was performed through Scanning Electron Microscopy (SEM) investigation by using a FEI microscope model Quanta 200, operating at 10 kV. Before the observations, all samples were coated with a thin layer of gold and rubbed upon a 25 mm diameter aluminum disc.

2.2 Composites

The biodegradable polymer used as matrix for bio-composites is a sample of Mater-Bi® (Mater-Bi HF51L2 by Novamont SpA) with melt flow index (190 °C/2.16 kg), melting temperature and density equal to 4.5 g/10 min, 150°C and 1.21 g/cm^3, respectively. After drying at room temperature for 24 hours the fresh culms and leaves, they were ground by means of a grinding machine. After that, lemongrass particles were sieved to obtain a fraction with maximum size equal to 500 µm.

Before the composites preparation, both the Mater-Bi and lemongrass particles were dried under vacuum at 60 °C for 12 hours with the aim of protecting the polymeric matrix by hydrolytic scission phenomena during processing. For both lemongrass particles, two bio-composites (i.e., containing 10 and 20 wt.% of lemongrass) were manufactured via extrusion and compression molding. In particular, bio-composites particles were first prepared by using a modular co-rotating twin screw extruder (OMC Italy, D = 19, L/D = 35). For the sake of comparison, neat Mater-Bi matrix was processed under the same conditions. The thermal profile adopted was 120–130–140–150–160–170–170 °C while the rotational speed was set to 180 rpm. The molten material obtained from the extruder die was cooled on line in a water bath, pelletized and used for the further manufacturing step.

Afterwards, the so obtained pellets were dried under vacuum at 60°C for 12 hours before the preparation of the samples for the mechanical, rheological and morphological characterizations were prepared via compression moulding by using a Carver Laboratory press (T = 160 °C, P = 27 bar, time = 3 min).

The bio-composites will be identified in the next with the codes L10, L20, C10 and C20, were the letter (i.e., L or C) indicates the lemongrass part (i.e., leaves or culms) and the number (i.e., 10 or 20) the particle weight content.

Tensile tests were performed on composites according to ASTM D638 standard, with the same U.T.M. used for fiber characterization, equipped with a load cell of 5 kN. Five Dumbbell samples for each composite were tested by setting the crosshead speed equal to 5 mm/min for elongation percentage values up to 8% and then equal to 50 mm/min.

The morphological observation of the fractured surfaces of Dumbbell samples was carried out by using the same scanning electron microscope used for the fiber characterization.

3. Experimental results

3.1 Fibers

The typical stress-strain tensile curves of leaf and culm fibers, shown in Figure 1, evidence the brittle behavior of both fibers. By comparing these curves, it can be noticed that the mechanical properties of lemongrass fibers are function of the part of the plant from which they have been extracted. In particular, leaf fibers show higher strength and stiffness than culm fibers, even though the latter reach break at greater elongation percentages.

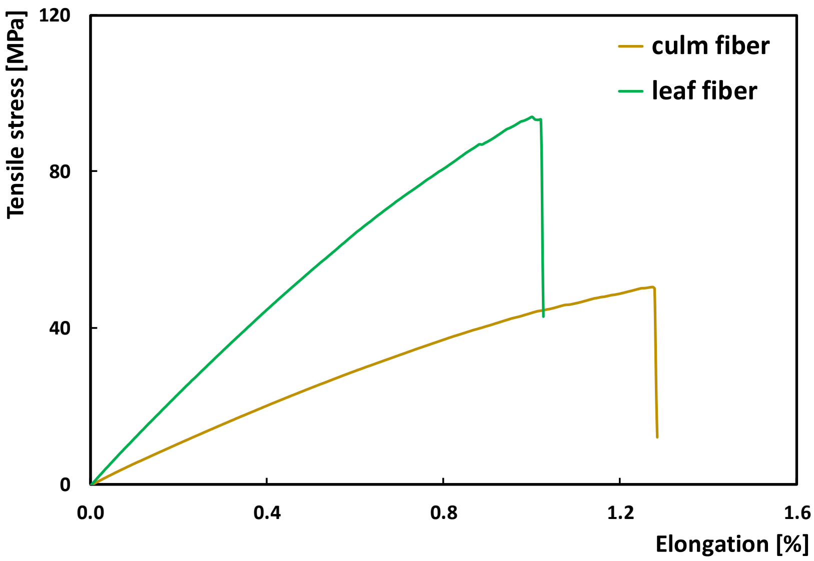

Figure 1. Tensile stress-strain curves of culm and leaf fibers

The statistical analysis of the mechanical results through Weibull model provided a reasonable approximation of the experimental data, furnishing the Weibull shape and scale parameters (i.e., the characteristic values of the distribution) for both properties (i.e., tensile strength and modulus). In particular, it was shown that the tensile strength and modulus of leaf fibers are about 55% and 76% higher in comparison to culm fibers, respectively. Vice versa, the elongation at break of culm fibers is just slightly higher than that of leaf fibers.

The chemical analysis of leaf and culm fibers showed that both fibers contain similar amounts of α-cellulose (i.e., 45.5% and 44.2% for leaf and culm fibers, respectively), hemicellulose (i.e., 29.1% and 28.1%) and lignin (i.e., 17.0% and 17.3%, respectively). These compositions cannot justify the noticeable mismatch between the mechanical properties of leaf and culm fibers. The only remarkable difference is the ash content, about 2% lower in leaf fibers, which explains at least partially the better mechanical properties of leaf fibers than culm ones.

The FTIR results shown in Figure 2 confirm that the compared fibers can be considered similar in terms of chemical composition.

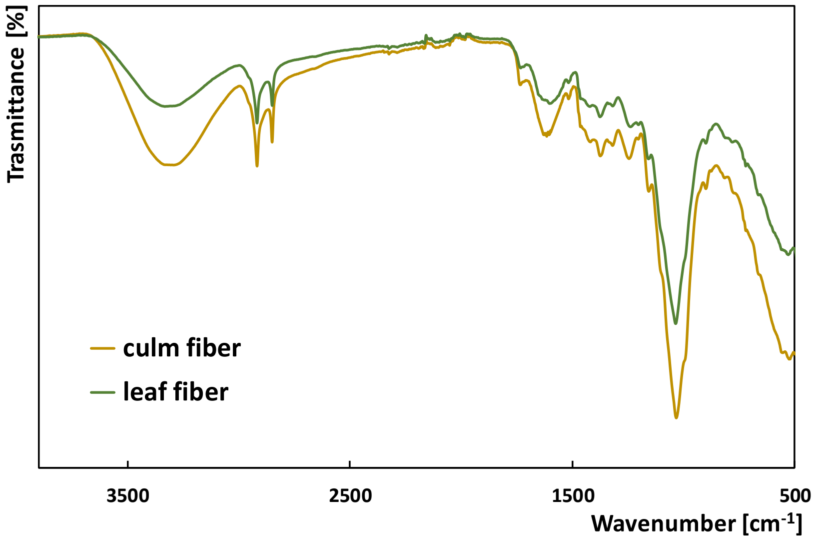

Figure 2. FTIR spectra of culm and leaf fibers

In particular, by comparing these spectra, it is possible to notice that only two peaks are noticeably different. The first one, at about 3325 cm^{-1}, is due to the O-H stretching vibration and hydrogen bond of the hydroxyl groups [16]. The second one, located at about 1600 cm^{-1}, is strictly correlated with the presence of water in the fibers [17]. Both peaks are greater for culm fibers, thus indicating that the latter contain a larger amount of water in comparison to leaf fibers.

Basically, the thermogravimetric analysis confirm this finding. Overall, both leaf and culm fibers experienced three different decomposition stages at around 100, 250 and 310°C, related to the water vaporization, hemicellulose degradation and α-cellulose decomposition, respectively. However, and more interestingly, it was shown that the weight loss found for culm and leaf fibers at 150 °C are respectively equal to about 12% and 8%, thus indicating that a greater amount of water is absorbed in culm fibers in comparison to leaf ones.

The experimental densities measured though helium pycnometer are equal to 1.02 g/cm^3 and 1.14 g/cm^3 for culm and leaf fibers, respectively. The higher density evidenced by leaf fibers meaning that the latter are characterized by a more compact structure in comparison to culm fibers, as clearly shown by observing the SEM micrographs in Figure 3.

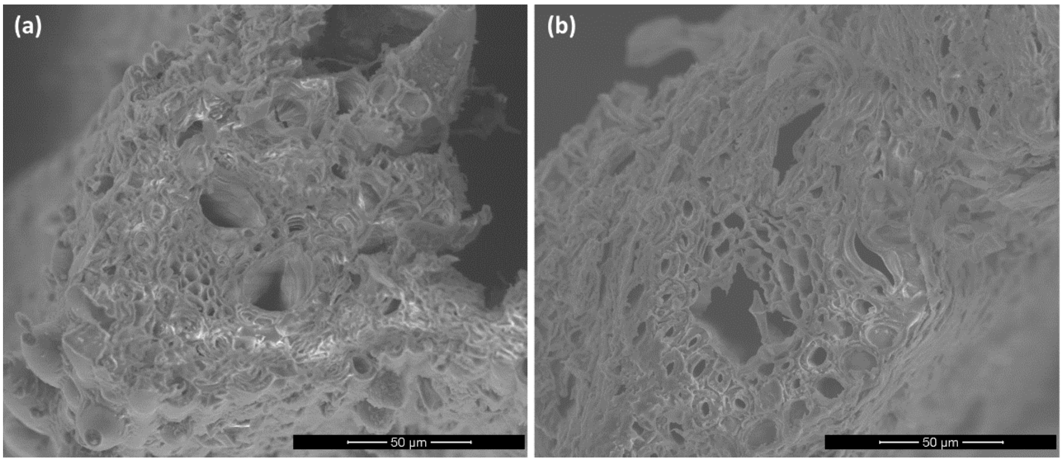

Figure 3. Cross section morphology of (a) leaf and (b) culm fibers

3.2 Composites

The tensile properties of neat matrix and bio-composites are reported in Table 1. All the composites show higher tensile moduli than Mater-Bi. In particular, the tensile modulus strongly increases on increasing the filler content. Furthermore, for both the compositions, the composites incorporating leaf particles exhibit higher stiffness than those containing culm particles. In particular, the L10 and L20 show tensile moduli about 81% and 186% higher than neat matrix, whereas the C10 and C20 show tensile moduli about 53% and 124% higher than Mater-Bi. It is worth noting that, the addition of relatively low content of lignocellulosic particles used in this study, led to remarkable increments of the tensile stiffness of the composites. Despite the positive effect on tensile modulus, the addiction of lemongrass particles led to a reduction in the tensile strength and elongation at break of all the composites compared to neat matrix. However, the reduction is less pronounced for composites incorporating the leaf particles. The decrease in the tensile strength of the bio-composites can be ascribed to the premature failure of the samples, which is expected when rigid particles are loaded in polymer matrices.

Table 1: Tensile properties of bio-composites.

	Strength [MPa]	Modulus [MPa]	Elongation at break [%]
Neat matrix	13.4 ± 0.5	62.5 ± 3.8	665.8 ± 79.8
L10	8.3 ± 0.4	113.3 ± 3.2	196.9 ± 45.4
L20	7.2 ± 0.1	178.9 ± 5.9	51.4 ± 14.9
C10	6.9 ± 0.3	95.7 ± 4.7	146.3 ± 44.8
C20	5.9 ± 0.1	140.0 ± 5.3	51.0 ± 15.5

The higher mechanical performances exhibited by bio-composites incorporating leaf particles can be probably attributed to the better morphology shown by these materials in comparison with the bio-composites incorporating the culm particles, as suggested by analysis of SEM micrographs. In particular, the latter exhibited a worse adhesion between the matrix and the filler, as clearly visible in the micrographs reported in Figure 4.

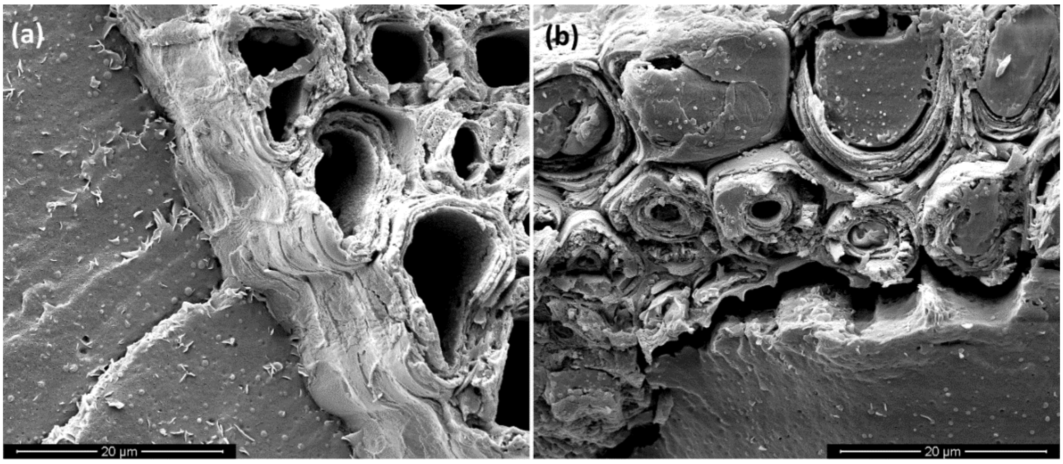

Figure 4. SEM micrographs of nitrogen-fractured surfaces of biocomposites (a) L10 and (b) C10.

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