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Additional Information

**“Development and characterization of a new natural fiber reinforced thermoplastic  
(NFRP) with Cortaderia Selloana (Pampa grass) short fibers”**

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A handwritten signature in blue ink, appearing to be 'A. Jorda-Vilaplana', is located in the lower-left quadrant of the page.

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## **Abstract**

In this work, fully bio-based thermoplastic composites are manufactured with bio-based polyethylene (from sugarcane) and short fibers coming from Cortaderia Selloana (CS) wastes. These wastes are characterized by high cellulose content, which can provide high stiffness to the polymeric matrix. The effect of Cortaderia Selloana short fibers on thermal properties has been evaluated by differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA). The effect of the filler load on mechanical properties has also been evaluated by tensile and impact tests as well as the effects of different coupling agents. Fiber-matrix interactions have been studied by scanning electron microscopy (SEM). The addition of 15-30 wt. % Cortaderia Selloana short fiber leads to high elastic and flexural modulus without remarkable changes in thermal degradation of the polymer composite.

**Keywords:** Cortaderia Selloana; natural fiber reinforced plastics (NFRP); Bio-based polyethylene; mechanical properties.

## **1.- Introduction.**

In recent decades, society has raised awareness regarding the need to preserve the environment. The way of life of modern societies based on consumption is responsible for the high production of wastes from both the manufacturer and the final consumer. This generates significant environmental and pollution problems. Governments in developed countries have the need to implement laws to protect the environment against the generation of waste and allow their controlled management [1, 2]. This is the main aim in the research on biodegradable and environmentally friendly materials.

It is evident that within the industrial sector the largest volume of waste generated will correspond to polymeric materials, mainly for their wide range of applications and their short life cycle, in areas of high consumption of materials like packaging, household goods, toys, leisure, construction, automotive, medical and catering, etc. Taking into account the petrochemical origin of most polymeric materials and their non-biodegradable nature, these become an interesting material to replace. Petroleum depletion is another factor that positive influence on the interest in developing new polymeric materials from renewable sources. Currently polymeric materials from

renewable sources (bio-based polymers) are marketed both thermoset and thermoplastic nature. Their properties are virtually the same as petroleum-based polymers, but derived from natural oils, sugars, proteins, starches, polysaccharides, etc. These biopolymers are perfect substitutes for traditional polymers, in any industry and application [3-6].

One of the technological applications with high demand for traditional polymers is as composites matrices. Most industrial composites are constituted by two components: the matrix and the reinforcing fiber. This structure is responsible for its virtually impossible recyclability and therefore it has become an environmental problem.

Thus, the possibility of obtaining materials that do not damage the environment promotes the use of reinforcement fibers from natural origin [7]. Several studies have analyzed the so-called "green composites" made from lignocellulosic fibers such as jute, sisal, flax, sunhemp, pineapple, flax, kenaf, wood, rice or seaweed and different polymer matrices [8-10]. In addition, optimizing the composition of these composites together with its low cost, has led to a new family of composite materials called Natural Fiber Reinforced Plastics (NFRP) and Wood Plastic Composites (WPC, being wood and wood wastes the lignocellulosic component of the composite) with highest growth in the plastics industry in recent years [11, 12].

Just in the USA, WPC have generated 5.3 billion dollars in revenue and more than 1 billion tons of production. The largest consumer of WPC is USA with 73% of the total, followed by China with 15%. The main applications are in the building industry, which represents almost 70% of total consumption on items such as handrails, railings, deckings, outdoor benches, fences, wall coverings or window frames [12-14]. The European Union has a demand on WPC of about 5% of the global production, mainly for applications in the automotive sector such as side interior panels, dashes, roofs, seats, acoustic panels or glove boxes [15-18].

The most widely used polymer matrix in these commercial WPC is polyethylene (PE), both low density (LDPE) and high density (HDPE), followed by the PP and PVC. In USA the PE use represents 83% of the total, while the PP and PVC represents 9% and 7% respectively.

These thermoplastic matrices give the composite versatility and easy processing. Furthermore, compared with natural wood, WPC have greater dimensional stability and longer service life without maintenance [16, 19].

The main drawback of WPC is the lack of interaction between the lignocellulosic fiber and the polymer matrix. Due to the different hydrophilic behavior of the fiber and the polymer matrix, there is not a good adhesion/interaction between them. The mechanical behavior of the composites depends directly on the matrix-fiber interface [20, 21]. A solution to this problem is the surface pretreatment of the fibers to improve adhesion to the matrix through the use of so-called coupling agents. Recent studies demonstrate the effectiveness of alkaline treatments coupled with isocyanates, acrylates, silanes and peroxides, which enhance matrix-fiber interaction [17, 19, 22, 23].

In the present study we used natural fiber from the plant *Cortaderia selloana* usually called Pampa Grass. Pampas grass is a hemicryptophyte that develops in clumps up to 3 meters high. It presents inflorescence in white-gray panicles of 60x15 cm high. This kind of natural fiber has been selected to manufacture the composite because it allows revalue the large volume of waste generated by pruning this plant. In addition Pampa grass is an allochtoon gramineae, which is considered "invasive species" with strong dominance that can become invasive on important native ecosystems. In addition, it drastically reduces biodiversity, changes the landscape and competes with native species. The most effective method for control is cutting or pruning, which generates large amounts of waste, with no use or value.

This work aims to characterize a new NFRP for uses in the WPC industry based on the use of *Cortaderia selloana* as reinforcing lignocellulosic material into a bio-based high-density polyethylene (HDPE) thermoplastic matrix. The first objective is to analyze the effects of compatibilizers to improve fiber-matrix interface adhesion. Secondly, the influence of the relative amounts of fiber added to the polymeric matrix has been studied.

## **2.- Experimental.**

### **2.1.- Materials.**

The HDPE used in this work as composite matrix was bioPE SHA7260 supplied by Braskem (Braskem Ideasa, Sao Pablo, Brasil). This HDPE is characterized by a minimum of 94 wt% bio-based content measured following the ASTM D6866 standard as indicated by the manufacturer. The melt flow index is the 20 g/10 min at 190 °C and the density is 0.955 g cm<sup>-3</sup>, making it a suitable polymer for injection molding.

The lignocellulosic reinforcing component was *Cortaderia Selloana* collected from locally pruning and harvesting on Valencia, Spain. Fig. 1 shows the typical *Cortaderia Selloana* plant. Different coupling systems were used to evaluate their influence on fiber-matrix interactions: a maleated polyethylene copolymer, polyethylene-graft-maleic anhydride (PE-g-MA) CAS 9006-26-2, and previous silanization with a hydrophobic silane, trimethoxy-propyl silane CAS 1067-25-0. Both coupling agents were supplied by Sigma-Aldrich (SIGMA-ALDRICH Chemie GmbH, Steinheim, Germany). Sodium hydroxide, 99% purity, was provided by Scharlab (Shacharlab, SL, Barcelona, Spain) and was used for alkali treatment.

## **2.2.- Composites processing.**

Firstly *Cortaderia selloana* fibers were cut in an industrial mill to obtain short fibers. The final length of the fibers was in the 1-4 mm range. After this they were washed with distilled water and then dried in oven at 60 °C for 24 hours. Different formulations were prepared for manufacturing as described below in Table 1 together with the corresponding designations. The four different formulations were extruded in a twin-screw extruder with the temperature in each of the four heating zones of 155, 160, 165 and 170 °C (from hopper to die) respectively and a screw speed of 30 rpm; finally the extruded material was pelletized. Rectangular samples (80x10x4 mm<sup>3</sup>) and standard tensile test samples were obtained by injection molding using an injection machine Meteor (Meteor 270/75, Mateu and Sole, Barcelona, Spain) at an injection temperature of 190 °C.

**Table 1.-** Composition and designation of the four different formulations of Bio-PE/*Cortaderia selloana* composites.

## **2.3.- Thermal characterization.**

Thermal transitions were studied by differential scanning calorimetry (DSC) using a Mettler-Toledo 821 (Mettler-Toledo Inc., Schwerzenbach, Switzerland). Samples ranging 5-10 mg in weight were used. A heating program from 30 to 300 °C at a heating rate of 10 °C min<sup>-1</sup> in an air atmosphere was used for all composites. Thermogravimetric analysis (TGA) was carried out in a TGA/SDTA 851 (Mettler-Toledo Inc., Schwerzenbach, Switzerland) and the temperature

ranged from 30 °C to 900 °C with a heating rate of 20 °C min<sup>-1</sup>. TGA tests were performed in nitrogen environment at a flow rate 60 mL min<sup>-1</sup>.

#### **2.4.- Mechanical characterization.**

Tensile and flexural properties of the different samples of bio-HDPE/CS were measured with a universal test machine Ibertest Elib 30 (Ibertest S.A.E., Madrid, Spain) with a load cell of 5 kN and crosshead speed of 10 mm min<sup>-1</sup> (ISO 527) for tensile test, and 5 mm min<sup>-1</sup> crosshead speed for flexural test (ISO 178). Hardness of bio-HDPE/CS composites was measured with a Shore D hardness durometer model 676-D (J. Bot Instruments, Barcelona, Spain) according to impac

#### **2.5.- Fractography by scanning electron microscopy (SEM).**

Fractured surfaces from impact tests of bio-HDPE/CS composites were observed with a scanning electron microscope (SEM) model Phenom (FEI Company, Eindhoven, Netherlands). Before observation, samples, mounted on a conductive adhesive tape, were covered with a thin gold-palladium layer. The coating process lasted 120 seconds using a sputter-coater Emitech mod. SC 7620 (Quorum Technologies Ltd, East Sussex, UK).

#### **2.6.- Water uptake characterization of bio-HDPE/CS composites.**

Water uptake of bio-HDPE/CS composites was determined with samples sizing 80 x 10 x 4 mm<sup>3</sup>. Three different samples were tested to obtain average values. Prior to the water uptake test, HDPE/CS composite samples were dried at 80 °C for 4 h. After this, samples were immersed in distilled water at room temperature for a period of 60 days. The percentage water uptake was calculated by using the following expression:

$$\text{Water Uptake (\%)} = \frac{(M_f - M_i)}{M_i} \times 100 \quad (1)$$

Where  $M_f$  is the final weight after a certain immersion period and  $M_i$  is the initial weight of the sample before immersion.

### 3.- Results and discussion

#### 3.1.- Effect of compatibilizing system on the bio-HDPE/CS composite properties.

Figure 1 shows the DSC calorimetric curves of the bio-HDPE/CS composites using different compatibilizing system. As it can be observed, the compatibilizing system does not significantly modify the melting temperature ( $T_m$ ) and the degradation onset. bio-HDPE from natural resources shows a melt peak at 135 °C, and this is decreased to 132.8 °C with the addition of 15 wt% of CS fiber without treatment, probably due to the nucleating effect of the filler. Compatibilizing system maintain the melt peak between 132 °C and 133 °C, so that they have no effect on  $T_m$ . The same applies to the degradation temperature, as no significant changes are observed. Furthermore, the addition of CS fiber without treatment and with PE-g-MA, has no influence on the thermal behavior of bio-HDPE/CS composite [24].

**Figure 1.-** Comparative plot of the differential scanning calorimetry thermograms (DSC) of unfilled bio-HDPE and bio-HDPE/CS composites with different compatibilizing system.

Regarding to results obtained by thermogravimetric analysis of bio-HDPE/CS composites, Figure 2 shows similar behavior for the different compatibilizing system used. Degradation of unfilled bio-HDPE occurs in the 350-520 °C range and above 520 °C the amount of residual char is very small, since the degradation of polyethylene generates large amount of gaseous products by its organic nature. With regard to *Cortaderia selloana*, thermogravimetric curve is the typical of a lignocellulosic material: at a relative low temperatures (50-150 °C), residual moisture evaporation takes place. At intermediate temperatures ranging from 220 °C to 350 °C decomposition of low molecular weight species (hemicelluloses, glycosidic bonds typical of cellulose and other volatile compounds) are removed and a remarkable weight loss takes place (about 40-50 wt.%). Finally, thermal degradation of cellulose occurs and lignin degradation proceeds slowly with final residual char of about 20 wt.%.

**Figure 2.-** Comparative plot of the thermogravymetric curves (TGA) of *Cortaderia selloana* (CS) fiber, unfilled bio-HDPE and bio-HDPE/CS composites with different compatibilizing system.



Bio-HDPE/CS composites show the typical degradation steps of both components, but in the thermograms we can observe two main different steps: the first one corresponds to a temperature range 250-350 °C and it is attributable to degradation of the reinforcement fiber. The second main step is located in the 350-500 °C temperature range, which corresponds to the degradation of polyethylene. In general terms, the compatibilizing system does not affect the thermal stability at high temperatures although some delay is achieved by silane and PE-g-MA compatibilizing system. As expected, the residual char in bio-HDPE/CS composites increases from 2.05 wt.% (raw unfilled bio-HDPE) up to values in the 5-6 wt.% range due to char formation from lignocellulosic component [19, 25].

Figure 3 and Figure 4 show the effect of the treatment on tensile and flexural properties respectively. In general terms, very small differences are detected. With regard to tensile modulus, the maximum values are reached for untreated bio-HDPE/CS (559 MPa). This value is higher to unfilled bio-HDPE with typical values of 373 MPa. It is important to remark that all composites (with same *Cortaderia selloana* content and different compatibilizing system) are stiffer than unfilled bio-HDPE. By analyzing tensile strength results, we observe similar values in the 17.4 – 20.1 MPa for untreated fiber (bio-HDPE/CS-UT) and fiber subjected to NaOH treatment followed by silanization respectively (bio-HDPE/CS-Silane). Unfilled polyethylene shows a tensile strength of about 19.6 MPa and, as we can see, composites show similar tensile strength values independently of the compatibilizing system. Nevertheless, elongation at break is highly affected by presence of the reinforcing filler. Unfilled bio-HDPE is characterized by high plastic deformation with elongation at break values of about 520%. Nevertheless all composites are characterized by a high embrittlement process with very low elongation at break values, even with different compatibilizing system, with values around 1.5%. This is due to the high stiffness of the lignocellulosic particles if compared to polyethylene and, mainly, due to lack of interactions between the particles and surrounding polymer matrix.

**Figure 3.-** Tensile properties of bio-HDPE/CS with different compatibilizing system: a) Bio-HDPE/CS-UT; b) Bio-HDPE/CS-NaOH; c) Bio-HDPE/CS-MA; d) Bio-HDPE/CS-Silane.

This fact can be easily observed in Figure 4, which shows detailed images corresponding to fractured surfaces. We can clearly observe absence of matrix-filler continuity even with the use of compatibilizing system. We observe typical gaps around the particle perimeter. These gaps are representative for low particle-matrix interactions. Lead to stress concentration phenomena thus leading to microcracks formation and subsequent crack growth. And promote early fracture of the composite. All composites break in a typical fragile way, almost without plastic deformation. The use of the different compatibilizing system does not show remarkable differences if compared to composite with untreated fibers [9, 18, 26].

**Figure 4.-** SEM images (2000x) of fractured surfaces from tensile tests of bio-HDPE/CS composites with different compatibilizing system: a) Bio-HDPE/CS-UT untreated fiber, b) Bio-HDPE/CS-NaOH; c) Bio-HDPE/CS-MA; d) Bio-HDPE/CS-Silane.

With regard to flexural properties, similar behavior to tensile properties can be observed. The flexural modulus of unfilled bio-HDPE is increased by 60% up to values of 1290 MPa for bio-HDPE/CS-UT composite without previous treatment. With respect to flexural strength an average percentage increase of 13% is detected for all composites with values in the 46-47 MPa range. Once again, in the case of *Cortaderia selloana* fibers, the compatibilizing system do not lead to remarkable differences in composites' response.

**Figure 5.-** Flexural properties of Bio-HDPE/CS with different compatibilizing system: a) Bio-HDPE/CS-UT; b) Bio-HDPE/CS-NaOH; c) Bio-HDPE/CS-MA; d) Bio-HDPE/CS-Silane.

On the other hand, the impact resistance is not remarkably affected by the use of different compatibilizing system as it can be observed in Table 2. It is important to remark that unnotched HDPE does not break due to the high plastic deformation that can support. All results showed in Table 2 are obtained, thus, using notched sample. Noteworthy that sample Bio-HDPE/CS-UT, carried out using a CS filler without compatibilizing treatment, obtained the lowest values of impact resistance. This due to the poor compatibility between polymeric matrix and the used cellulosic filler. On the other hand, when different compatibilizing systems are used this impact resistance are increased, even above to Bio-HDPE. Nevertheless this increment is

reduced (around 9%). As to the different compatibilizing systems used, negligible differences are obtained, thus the compatibilizing system chosen does not play a key role on impact resistances as observed previously with flexural and tensile properties.

**Table 2.-** Impact resistance values of Bio-PE/CS composites with different compatibilizing system.

These results show that the different compatibilizing system do not play a key role in improving the overall performance of bio-HDPE/CS composites. This fact could be directly related to the physical morphology of the fibers themselves. Figure 6a shows the typical appearance of untreated *Cortaderia selloana* fibers with a feather like morphology, with very thin outstretched fibers characterized by very high surface area. Figure 6b corresponds to the same fibers subjected to alkali treatment with NaOH. We can clearly see the difference in morphology between the untreated and NaOH-treated fibers. In the case of the NaOH-treated fibers we can see that the linear fiber shapes have disappeared and new aggregates can be detected with more compact fibers and, subsequently, with lower surface area due to the aggregation state. Although some fiber-matrix interactions can be achieved by the different compatibilizing system, the treatment leads to breakages of the feather-like structure and low surface area is obtained thus leading to lower fiber-matrix interactions. For this reason, we evaluate the influence of the *Cortaderia selloana* content in composites without previous treatment and/or compatibilizer as we have corroborated that the different compatibilizing system do not improve mechanical and thermal behaviour of bio-HDPE/CS composites.

**Figure 6.-** Physical morphology of *Cortaderia selloana* fibers (10x): a) without treatment; b) alkali treatment with NaOH.

### **3.1.- Effect of *Cortaderia selloana* content on bio-HDPE/CS composite properties.**

Differential scanning calorimetry (DSC) results are summarized in Table 3. The melting peak is slightly reduced as the fiber content increases due to the nucleating effect provided by the lignocellulose component. With regard to the onset degradation temperature it is important to

note the remarkable increase in thermal stability as the *Cortaderia selloana* content increases. The onset degradation temperature of the raw unfilled bio-HDPE is located at about 232.5 °C and this is remarkably increased up to values of 265 °C for composites containing 30 wt.% CS. DSC study reveals a clear stabilizing effect against thermo-oxidative processes due to presence of intrinsic antioxidant components in *Cortaderia selloana*.

**Table 3.-** Effect of *Cortaderia selloana* content on thermal properties of Bio-HDPE/CS composites obtained by differential scanning calorimetry (DSC).

Although addition of *Cortaderia selloana* leads to a clear thermal stabilization effect at moderate temperatures, thermal degradation of bio-HDPE/CS composites is slightly reduced at moderate-high temperatures due to lignocellulosic components degradation [24]. Thermogravymetric analysis (TGA) of bio-HDPE/CS composites is shown in Figure 7. *Cortaderia selloana* degradation mainly occurs in the 250-350 °C range with a slight weight loss around 100 °C due to removal of residual water and quite residual ash at high temperatures (close to 20 wt.%). With regard to unfilled bio-HDPE, degradation occurs in the 350-500 °C range in a single step process with very low char formation. With regard to bio-HDPE/CS composites, degradation occurs in two clearly detectable stages. The first stage is located in the 250 °C-350 °C and corresponds to degradation of lignocellulosic components and other organic constituents [27, 28]. The second stage is directly related to polyethylene degradation and we can clearly see the stabilizing effect provided by *Cortaderia selloana* fibers as the onset degradation temperature of the second stage is higher than unfilled polyethylene. In addition, as expected, the weight loss of the first degradation step is directly related to the total amount of *Cortaderia selloana* fibers. Moreover, the residual char increases with the total amount of *Cortaderia selloana* changing from 2.05 wt.% (composites with 7.5 wt.% CS) up to 11.4 wt.% for composites containing 30 wt.% CS.

**Figure 7.-** Thermogravymetric (TGA) curves of raw *Cortaderia Selloana* fiber and bio-HDPE/CS composites with different *Cortaderia Selloana* content.

The effect of the fiber content on mechanical response can be observed in Figure 8. As the fiber content increases, both the tensile and flexural modulus increase. In the case of tensile modulus, it increases by 86% for composites containing 30 wt.% CS with regard to unfilled HDPE. The flexural modulus increases up to twice the initial value of unfilled HDPE for composites with 30 wt.% CS. Addition of short dispersed fiber leads to a dramatic decrease in plastic deformation. The modulus can be defined as a ratio between stress and deformation (in tensile and flexural conditions). As we can see, the strength values are not highly reduced but the elongation at break is reduced to minimum values. For this reason, the stress/elongation ratio increases thus indicating more stiffness; so that, addition of *Cortaderia selloana* fibers leads to a clear reinforcing effect [19, 24]. With regard to tensile and flexural strength it is important to remark that the fiber content does not produce important changes in their values.

**Figure 8.-** Mechanical properties of bio-HDPE/CS composites in terms of the wt.% of *Cortaderia Selloana* content.

The evolution of the elongation at break is similar to that observed in the previous section with a clear embrittlement effect due to a dramatic decrease in elongation at break values. The total content on reinforcement fiber does not have significant effects on elongation at break and very low values, near 1.5%, are obtained for all composites. Figure 9 shows the fractured surfaces from tensile tests for different fiber content. As indicated previously, morphologies are characterized by lack of interaction among the fiber-matrix interface. Obviously, as the fiber content increases, the total amount of fibers in the fractured surfaces is more evident.

**Figure 9.-** SEM images (500x) of fractured surfaces from tensile tests of bio-HDPE/CS composites with different *Cortaderia Selloana* content: a) 7.5 wt.%; b) 15 wt.%; c) 30 wt.%.

### **3.2 Water uptake of bio-HDPE/*Cortaderia* fibre composites**

Figure 10 shows the time dependences of water uptake for the different bio-HDPE/CS composites using various coupling agents. bio-HDPE absorbed only 0.021 wt% of water after 60 days, due to its hydrophobic character. It was found that the composite of unfilled bio-HDPE with untreated fiber exhibits very low water uptake even after 60 days. Comparatively, the fibers with coupling

agents show a higher water absorption levels. The composite unfilled bio-HDPE with fibers pre-treated with NaOH and PE-g-MA submits values of water uptake of 6.2 wt%, while in composite of unfilled bio-HDPE with untreated fiber is only 0.844 wt%, after 60 days. For the other fiber treatment studied, also increases the amount of water absorbed. These results show that in the bio-HDPE/CS system studied, coupling agents worsen the water absorption behavior of the material. This phenomenon is due to the certain natural hydrophobic character of the Cortaderia selloana fibers. While NaOH bath favors a more hydrophilic behavior of the composite material as shown in Figure 10. These considerations show that the composite unfilled bio-HDPE with untreated Cortaderia fiber unfilled bio-HDPE has the best behavior in water uptake phenomenon.

**Figure 10.-** Variation of the water uptake as a function of the treatment time for bio-HDPE/CS composites with different compatibilizing system

The water uptake of the bio-HDPE/CS composites is a function of amount of fiber as shown in Figure 11. Observed graphically as to lower fiber content, the amount of water uptake is low after 60 days of immersion. During this test time, the composite with 7.5 wt% and 15 wt% of fiber presents 0.384 wt% and 0.844 wt% of water, respectively. What is the same, with twice the amount of fiber, water uptake is almost twice. While high amounts of fiber (30 wt%) the composite behavior is different, showing high water uptake (5.35 wt%) to the same time test. This involves six times the sample of fiber 15 wt%, and 14 times for 7.5 wt% respectively. These results may be due to the low interfacial adhesion between the fiber and the matrix, which increase the rate of diffusion of water. So that the greater quantity of fiber in the composite, more water absorbency occurs because the voids formation on the fiber-matrix interface.

**Figure 11.-** Variation of the water uptake as a function of the treatment time for bio-HDPE/CS composites with different Cortaderia Selloana content.

#### **4.- Conclusions.**

Addition of Cortaderia selloana short fiber to bio-based high density polyethylene leads to a remarkable increase in stiffness while strength (tensile and flexural) is not highly affected

even with relatively high fiber load (30 wt.%). Elastic and flexural moduli for composites with 15-20 wt.% *Cortaderia selloana* are almost twice the values of unfilled HDPE, thus leading to stiff materials with potential use as wood substitute in applications in which structural properties are not required. The main advantage of bio-HDPE/CS composites is a high bio-based content provided by polyethylene from ethanol together with an agroforestral waste from *Cortaderia selloana*, natural intrinsic stabilization properties against thermo-oxidation due to *Cortaderia selloana* natural antioxidants, low maintenance and a water uptake typical of other Wood Plastic Composites (WPC). The use of compatibilizing system based on alkali treatment coupled to maleated copolymer or hydrophobic silane treatment do not provide better properties. It is important to remark that untreated *Cortaderia selloana* fibers have a feather-like structure which gives high surface area and this morphology is lost after a wet- treatment with silanes. Moreover *Cortaderia Selloana* it is an invasive species in Mediterranean Area. It forms dense populations which may exclude the presence of other plants. Diminishes the quality of pastures and increases the risk of fire. It has been suggested starting the plant as a preferred method of disposal and the use of herbicides (glyphosate) although it should evaluate potential impact on amphibian populations. importantly can influence the dynamics of river systems and control is essential to keep the channels in good condition. Authors consider that using *Cortaderia Selloana* as filler a revaluation of cellulosic waste may be carried out, giving an economical and environmental solutions respect the elimination of this species.

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### Figure legends

**Figure 1.-** Comparative plot of the differential scanning calorimetry thermograms (DSC) of bio-HDPE and bio-HDPE/CS composites with different compatibilizing system.

**Figure 2.-** Comparative plot of the thermogravymetric curves (TGA) of Cortaderia selloana (CS) fiber, bio-HDPE and bio-HDPE/CS composites with different compatibilizing system.

**Figure 3.-** Tensile properties of bio-HDPE/CS with different compatibilizing system: a) Bio-HDPE/CS-UT; b) Bio-HDPE/CS-NaOH; c) Bio-HDPE/CS-MA; d) Bio-HDPE/CS-Silane.

**Figure 4.-** SEM images (2000x) of fractured surfaces from tensile tests of bio-HDPE/CS composites with different compatibilizing system: a) Bio-HDPE/CS-UT untreated fiber, b) Bio-HDPE/CS-NaOH; c) Bio-HDPE/CS-MA; d) Bio-HDPE/CS-Silane.

**Figure 5.-** Flexural properties of Bio-HDPE/CS with different compatibilizing system: a) Bio-HDPE/CS-UT; b) Bio-HDPE/CS-NaOH; c) Bio-HDPE/CS-MA; d) Bio-HDPE/CS-Silane.

**Figure 6.-** Physical morphology of Cortaderia selloana fibers (10x): a) without treatment; b) alkali treatment with NaOH.

**Figure 7.-** Thermogravymetric (TGA) curves of raw Cortaderia Selloana fiber and bio-HDPE/CS composites with different Cortaderia Selloana content.

**Figure 8.-** Mechanical properties of bio-HDPE/CS composites in terms of the wt.% of Cortaderia Selloana content.

**Figure 9.-** SEM images (500x) of fractured surfaces from tensile tests of bio-HDPE/CS composites with different Cortaderia Selloana content: a) 7.5 wt.%; b) 15 wt.%; c) 30 wt.%.

**Figure 10.-** Variation of the water uptake as a function of the treatment time for bio-HDPE/CS composites with different compatibilizing system

**Figure 11.-** Variation of the water uptake as a function of the treatment time for bio-HDPE/CS composites with different Cortaderia Selloana content.

### **Table legends**

**Table 1.-** Composition and designation of the four different formulations of Bio-PE/*Cortaderia selloana* composites.

**Table 2.-** Impact resistance values of Bio-PE/CS composites with different compatibilizing system.

**Table 3.-** Effect of *Cortaderia selloana* content on thermal properties of Bio-HDPE/CS composites obtained by differential scanning calorimetry (DSC).