



Research paper

The suitability of banana leaf residue as raw material for the production of high lignin content micro/nano fibers: From residue to value-added products



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ABSTRACT

Each year the agri-food industry produces millions of tonnes of lignocellulosic residue, which could be valorized for its composition. Taking into account the economic cycle, this valorization is increasingly necessary and important. The Canary Islands' banana (*Musa acuminata* var. *Dwarf Cavendish*) is an example of this. Its annual production is around 400,000 tons, which translates into 320,000–400,000 tons of banana leaf waste or residue. This residue can represent an important source of raw material for the production of value-added products, such as lignocellulosic micro/nanofibers (LCMNF) for different applications. However, when using a residue as raw material for value-added products, the maximum exploitation rate has to be achieved in terms of producing the minimum amount of sub-residue. In this sense, the present work aims to produce LCMNF from pulp made of banana leaf residue and to assess their reinforcement potential and production costs for the papermaking sector. For the first time, high-lignin content LCMNF will be produced with the same reinforcing capacity than CMNF prepared from woody and bleached pulp, using methodologies under the umbrella of bioeconomy, waste valorization and sustainable growth. The obtained results showed that the presence of high contents of lignin and hemicelluloses promotes the fibrillation, leading to LCMNF with the same reinforcing potential than those obtained from wood and oxidative methods. In addition, the obtained LCMNF had lower production costs than the aforementioned and presented higher yield in terms of raw material utilization. However, further research must be still developed on decreasing production costs thereof.

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1. Introduction

The bioeconomy encompasses the production of renewable biological resources and their conversion into food, feed, bio-based products and bioenergy. Its ultimate aim is to help keep society competitive, innovative and prosperous by providing sustainable, smart and inclusive economic growth and jobs. This means, in general terms, an economy based on biomass derived fuel, chemicals and materials sustainably sourced and produced (EFIB, 2016). However, the use of any type of biomass can rebound on J-curve effects difficult to address. For instance, if the use of natural resources from wood is increased, deforestation could be promoted, leading to a non-sustainable development where the consumption of wood

would be higher than natural afforestation. In this sense, even wood being a renewable resource that could be converted into value-added products, many efforts would have to be performed in order for it to be sustainable.

Annual crops as well as agricultural residues are receiving increasing attention in recent years as alternative raw materials for the production of cellulose fibers (Villar et al., 2009; Marrakchi et al., 2011). Generally, these raw materials have lower lignin content than wood raw materials (Marques et al., 2010). They have shorter growth cycles with a moderate risk level and they also are annually renewed. These characteristics give these raw materials a great annual potential in cellulose.

To not complete the exploitation of the available natural resources, such as biomass, is something that today's society cannot afford, neither from the economic nor the environmental point of view. Each year the agri-food industry produces millions of tonnes of lignocellulosic residues, which could be valued for their

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composition. Taking into account the economic cycle, this valuation is increasingly necessary and important. The Canary Islands' banana (*Musa acuminata* var. *Dwarf Cavenish*) is an example of this. Its annual production is around 400,000 tons. Considering that the ratio of produced fruit vs. quantity of residue generated (pruning) can be placed around 0.8–1 (Rodríguez et al., 2010; Jiménez and Rodríguez, 2010), in-between 320,000–400,000 tons of lignocellulosic waste from banana exploitation is generated each year.

These residues should be treated for their total or at least partial elimination. If not, they can produce serious environmental issues to be addressed such as soil contamination, pests generation, interference with the harvest, etc. (Rodríguez et al., 2010). The burning of this waste in the field, or in the best of cases, in power plants, means a loss of money and resources. Meanwhile, it also produces CO₂ emissions into the atmosphere, at a faster rate than the plant assimilation speed.

Papermaking industry has been historically aware of environmental issues and increasing the efficiency of their processes such as wasting less water and energy and introducing paper recycling in their production processes. Moreover, papermakers and paper technology research institutions are constantly searching innovative ways to enhance mechanical properties of paper without causing much structural damages to fibers, increasing thus their lifespan. In this sense, cellulose nanofibers (CNF) deserve a special mention.

The use of CNF as an additive for papermaking slurries has been already assessed by several authors (Ahola et al., 2008; Brodin et al., 2014; Delgado-Aguilar et al., 2015a; Delgado-Aguilar et al., 2015b; Eriksen et al., 2008; González et al., 2012). CNF suspensions are formed by the smallest structural unit of cellulose fibers, the microfibril, also called nanofibril or nanofiber in some literature, depending if length or diameter is referred (Klemm et al., 2011). CNF can have diameters between 3 and 5 nm, though CNF suspensions usually contain aggregates thereof diameters can reach 100 nm. The addition of CNF into papermaking slurries improves the mechanical strength of papers, reduces porosity and increases paper's density; besides, CNF addition reduces or even eliminates the need of mechanical refining of paper slurries, which reduces considerably the lifespan of fibers (Delgado-Aguilar et al., 2015d).

CNF can be produced from many raw materials (Nechyporchuk et al., 2016), such as wood (Brodin et al., 2014; González et al., 2012), waste paper (Wang et al., 2013), fruit plants, palms and trees (Zhao et al., 2015; Rambabu et al., 2016) and straw from annual plants (Alemdar and Sain, 2008; Montañó-Leyva et al. (2011); Espinosa et al., 2016). In addition, many efforts have been performed to find a low-energy consumption method for the production of cellulose micro/nanofibers. In this sense, several methodologies have been developed with the purpose of treating fibers before fibrillating them and, thus, reducing the energy required in homogenizers, microfluidizers and other fibrillation equipment such as grinders. However, many of the methodologies require the use of chemicals that raise the production costs until unaffordable levels for many industrial sectors, including papermaking. In addition, sometimes the chemicals used for fiber pretreatment are not recoverable and their environmental impact is high. This places these methodologies out of the bioeconomy framework described at the beginning of the present introduction. Nonetheless, when using homogenizers or microfluidizers, if the fibers have not been previously treated, pressure chamber clogging is a big issue. In order to overcome this situation, some researchers have started to assess the role of lignin during fibrillation processes. Osong et al. (2013) mentioned that high temperature homogenization could improve the fibrillation by affecting the softening of lignin and, in addition, affecting thus the swelling behavior of the fibers. This effect was also found by Ferrer et al. (2012), where the effect of residual lignin in fibers

was assessed for the production of lignocellulosic micro/nanofibers (LCMNF).

The aim of the present work is to provide added value to the banana leaf residue through its utilization as raw material for the production of LCMNF and to assess its potential application in papermaking, reaching the same properties as when fresh wood is used. Moreover, the present work also intends to produce these LCMNF only by mechanical methods, taking profit of the high lignin content of this waste during fibrillation process.

2. Materials and methods

2.1. Raw material characterization

The Agricultural Cooperative of Tenerife FAST, Spain, provided the banana leaves (*Musa acuminata* var. *Dwarf Cavenish*). This raw material is a high herbaceous plant (2–16 m high), composed of long fibers strongly overlapped forming a pseudo-stem. Banana-trees generally produce large leaves (almost 2 m long and 30–60 cm wide) that were used in this work.

The content of α -cellulose, holocellulose, lignin, ashes, ethanol extractable, hot water solubles, 1% NaOH solubles, Kappa number and viscosity were determined according to TAPPI standards T-9m54, T-222, T-203os61, T-211, T-204, T-257, T-212, T-236 cm and T230-om-94, respectively.

2.2. Pulping

The raw material was pulped in a 15 L batch reactor under Specel[®] process conditions, (100 °C \pm 1 °C, 150 min, 7% (o.d.m) NaOH as reagent and a liquid/solid ratio 10:1). Pulping yield was calculated according to the following equation:

$$\text{Yield (\%)} = \frac{W_i}{W_f} \cdot 100$$

Where W_i is the initial dry weight (raw material) and W_f is the dry weight of the pulp after treatment.

After the pulping process, cooked material was washed in order to remove residual cooking liquor and then the cooked material was dispersed in a pulp desintegrator at 1200 rpm during 30 min. After that, the pulp was passed through Sprout-Bauer refiner, and using a Sommerville equipment with a netting of 0.14 mm mesh size, the uncooked material was separated.

2.3. Banana leaf pulp characterization

The resulting pulp was chemically characterized in the same way as the raw material. In addition, the morphology of fibers was determined using a MorFi Compact equipment (TechPap, Grenoble).

2.4. LCMNF production

In the mechanical treatment, the banana leaves residue soda-Specel[®] pulp was refined in a PFI mill (NPFI 02 Metrotec SA) according to ISO 5264-2 until achieving a drainage rate of 90° SR. Then, 1% aqueous suspension was prepared and passed through a high pressure homogenizer (Panda Plus 2000) following the next sequence: 4 times at 300 bar, 3 times at 600 bar and 3 times at 900 bar (Espinosa et al., 2016). The energy consumption of the homogenizer during the production of LCMNF was carried out with an energy measuring equipment (Circutor CVM-C10), which gives values of the energy consumption of the equipment or, what is the same, the energy required from the electrical grid.

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