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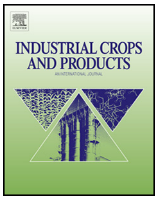

A comprehensive approach for obtaining cellulose nanocrystal from coconut fiber. Part II: Environmental assessment of technological pathways

Diego Magalhães do Nascimento[a], Amanda Ferreira Dias[b], Celso Pires de Araújo Junior[a], Morsyleide de Freitas Rosa[c], João Paulo Saraiva Morais[d], Maria Cléa Brito de Figueirêdo[c,*]

[a] *Federal University of Ceará (UFC), Department of Chemical, Bloco 940, CEP 60455-760 Fortaleza, Ceará, Brazil*
[b] *State University of Ceará (UECE), Campus do Itapery, CEP 60455-900 Fortaleza, Ceará, Brazil*
[c] *Embrapa Tropical Agroindustry, Rua Dra Sara Mesquita 2270, Planalto do Pici, CEP 60511-110 Fortaleza, Ceará, Brazil*
[d] *Embrapa Cotton, Rua Oswaldo Cruz 1143, Centenário, CEP 58428-095 Campina Grande, Paraíba, Brazil*

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ABSTRACT

Recently, development of methods for nanomaterial production from natural fibers, especially those from the crop residues in agroindustry, has rapidly increased because of the high availability of residues and the possibility to add value to them. The environmental assessment of such methods is important to continually improve the performance of new nanomaterials throughout the innovation process, especially when it is easier and cheaper to implement modifications in the product design. Unripe coconut fibers are a byproduct of the coconut water industry. These fibers can be extracted from unripe coconut husks, a renewable and abundant source of lignocellulose in the tropical regions. However, recent studies have reported high environmental impacts associated with the method of cellulose nanocrystal extraction from this biomass, mostly related to increased water and energy consumption, use of chlorine-based chemical reagents, and low yield. The aim of the present study was to investigate the environmental impacts of the methods for cellulose nanocrystal extraction from coconut fibers, and to determine the most environmentally sustainable method using Life Cycle Assessment (LCA). Detailed descriptions of these extraction methods have been included in Part I of this study. As cellulose nanocrystal extraction methods allow lignin recovery, this material was analyzed considering two aspects: as a byproduct from the cellulose nanocrystal extraction process, and as a power source for the system. Results indicate that cellulose nanocrystals obtained using the high power ultrasound method cause lower environmental impacts amongst all the assessed categories. The use of lignin as a power source for the biorefinery system does not demonstrate significant differences in relation to its use as a byproduct chemical in other technological applications. The present study reinforced the feasibility of applying LCA to continuously improve the environmental performance of nanomaterials throughout the innovation process, since LCA studies guide the choice of raw materials and technological pathways that result in reduced environmental impacts.

1. Introduction

Cellulose nanocrystals are considered as renewable nanomaterials with several high value-added applications, e.g., enzymatic immobilization (Edwards et al., 2013), controlled release of quimioterapics (Dong et al., 2014), and reinforcement agent for films and nanocomposites (Arrieta et al., 2014; Medeiros et al., 2014; Saralegi et al., 2013).

Cellulose nanocrystals are an alternative to carbon nanotubes as reinforcement agents in nanocomposites, due to their wide availability, low cost, biocompatibility, and biodegradability (Moon et al., 2011). Cellulose nanocrystals confer different properties to conventional cellulose fibers, such as a high surface area, Youngís modulus around 150 GPa, a high aspect ratio (Length/Width), surface rich in hydroxyl groups (-OH), and a high viscosity (Charreau et al., 2013).

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* Corresponding author.
E-mail address: mariacleabrito@gmail.com (M.C.B.d. Figueirêdo).

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Nanocellulose can be extracted from industrial plant fibers obtained mostly from the agroindustrial residues or byproducts (Rosa et al., 2010), and such residues are globally available at low cost. In Brazil and other tropical regions, many fibers from fruit husks and peels can be used as raw material for the extraction of cellulose nanocrystals, such as coir fibers extracted from unripe coconut husks.

The Brazilian production of unripe coconuts in 2015 was estimated as 1,575,094 t (IBGE, 2015). This production resulted in the generation of huge amounts of husks that, if not used, could reduce the lifetime of landfills, threaten community health, and be harmful for the environment.

Rosa et al. (2010) described the first cellulose nanocrystal extraction method using coconut fibers as the cellulose source, while Figueirêdo et al. (2012) analyzed the environmental impact of these cellulose nanocrystals in comparison to that of the cellulose nanocrystals obtained from cotton fibers. This environmental study showed coconut fiber processing for cellulose nanocrystals resulted in higher environmental impacts, mainly because of higher water and energy consumption, chlorine-based chemical reagents, and low yields. Since coconut fiber is lignin-rich, it naturally yields a lower value than lignin-poor fibers. To increase the overall environmental performance, Figueirêdo et al. (2012) indicated the need to increase the yield, to reduce the use of water and energy, and to recover as much of other components, such as lignin and hemicellulose, as possible.

In this context, the aim of the present study was to comparatively evaluate the environmental performance of four cellulose nanocrystal extraction methods, as proposed in Part I of this work (Nascimento et al., 2016), using the extraction process described by Rosa et al. (2010) as a reference, and the evaluation performed by Figueirêdo et al. (2012). The four studied methods recovered lignin from the coconut fibers and used the following alternatives to hydrolyze cellulose: i) diluted sulfuric acid (CNH1); ii) concentrated sulfuric acid (CNH2); iii) ammonium persulfate (CNO); and iv) high powered ultrasound (CNO).

2. Methodology

Description of the cellulose nanocrystal extraction methods as well as the methods used to determine the crystallinity, thermal stability, yield, and aspect ratio have been included in Part I of this study. The procedures to assess the environmental impacts of cellulose nanocrystals obtained from these methods have been further detailed.

2.1. Life cycle assessment (LCA) of cellulose nanocrystals produced by different methods

The environmental assessment performed in this study used the LCA approach described in the ISO standard methods 14040:2006 and 14044:2006.

2.1.1. Functional unit and scope of the study

The product system is *cradle to gate*, considering processes from the extraction of coconut fiber to the extraction of cellulose nanocrystals, as well as the production of electricity and chemical inputs for the methods (Fig. 1). Since unripe coconut husks are still considered residues, the coconut crop production was not considered in this study. The adopted functional unit is the production of 1 g of cellulose nanocrystal.

2.1.2. Allocation criterion

The evaluated cellulose nanocrystal extraction methods generate lignin as a byproduct. To evaluate the environmental impact of the cellulose nanocrystals, mass and economical allocation were performed for the unit processes that are common to lignin and cellulose nanocrystal production: gridding and pulping (Fig. 2).

Table 1
Data used for mass and economic allocation.

		Cellulose nanocrystal extraction methods[a]			
Outputs	Unit	CNH1	CNH2	CNO	CNU
Cellulose nanocrystal	g	1.0	1.0	1.0	1.0
Lignin	g	1.3	2.4	2.2	0.9
Mass allocation					
Cellulose nanocrystal	%	44	29	31	53
Lignin	%	56	71	69	47
Economic allocation					
Cellulose nanocrystal(11 $/g)[b]	%	86	77	78	90
Lignin (1.4 $/g)[c]	%	14	23	22	10

[a] CNH1: extraction with diluted acid; CNH2: extraction with concentrated acid; CNO: extraction with ammonium persulfate; CNU: extraction with high power ultrasound.

[b] Values based to Cowie et al., 2015.

[c] Values in accordance to Technology, Pure Lignin Environmental (2009).

Economic allocation considered the price projection for both cellulose nanocrystals and lignin, since the commercialization of such products began three years ago and the market is still growing. Allocated values for each product, using mass and economic criteria, are presented in Table 1.

2.1.3. Data collection

The unit processes related to the extraction of cellulose nanocrystals are presented in Fig. 2. Lignin production covers the processes of gridding, pulping, and recovering. For cellulose nanocrystals, milling and pulping are also needed, as well as other processes, depending on the method applied to hydrolyze cellulose (CNH1: extraction with diluted acid; CNH2: extraction with concentrated acid; CNO: extraction with ammonium persulfate; and CNU: extraction with high power ultrasound). The detailed descriptions of these processes are in Part I of the present study.

Data regarding the consumption of water, electricity, and reagents, as well as the concentration of pollutants in liquid effluents from cellulose nanocrystal extraction were measured in the laboratory, at Embrapa Tropical Agroindustry, in 2014.

In Part I of this study, biodegradability and potential production of methane in effluents from the pulping processes were evaluated. The generated methane was considered as a power source, after burning, with the generated energy used in the pulping process. As the effluent from bleaching presented low methane production potential, the polluting load of this effluent was assessed. The parameters from the bleaching effluents, analyzed based on the methods of Eaton et al. (1998), Silva and Oliveira (2001), and Gouveia et al. (2009), were: chemical oxygen demand (COD), biochemical oxygen demand (BOD), Kjeldahl total nitrogen (KTN), total phosphorus, furfural, and hydroxymethylfurfural (HMF).

Inventory data related to the production of chemical reagents and energy were obtained from ecoinvent 3.1 (Goedkoop et al., 2009). Data related to the fractionation of unripe coconut husks was obtained from Rosa et al. (2010). Data related to the production of ammonium persulfate [$(NH_4)_2S_2O_8$] (APS) were not found in the database used, and was replaced by the ammonium sulfate (NH_4SO_4) inventory from ecoinvent, which was considered a similar chemical. Since the inventory data from Figueirêdo et al. (2012) was based on evaluation using a previous version of ecoinvent (v.2) and Simapro (*7.2*), a new evaluation was performed using the version ecoinvent 3.0 and Simapro 8.0.3.

2.1.4. Impact assessment

By applying a hierarchical version of the ReCiPe method, at the midpoint level, the following environmental impact categories