Life Cycle Assessment of Cellulose

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The huge plastic production and plastic pollution are considered important global issues due to environmental aspects. One practical and efficient way to address them is to replace fossil-based plastics with natural-based materials, such as cellulose. The applications of different cellulose products have recently received increasing attention because of their desirable properties, such as biodegradability and sustainability.

nanocellulose life cycle assessment cradle-to-grave low carbon materials cradle-to-gate

1. Introduction

Due to environmental issues caused by the consumption of fossil resources and their depletion, the concept of sustainable development using environmentally innocuous materials is being adopted [1][2][3]. Cellulose is the most abundant biodegradable polymer available, having a total production capacity of 10^{11}–10^{12} tons each year [4]. The gross domestic product induced by the commercialization of nanocellulose, as a form of cellulose materials, is estimated to be around $600 billion worldwide by 2020 [5]. The U.S Department of Energy predicts that renewable sources will supply 50% of necessary chemicals by 2050 [6]. Therefore, research on different aspects of cellulose products, their application in various industries, and their production techniques have increased. Due to the increasing consumption of cellulose products in recent years, one of the required fields of this research, as well as the purpose of this study, is the evaluation of environmental effects arising from the extraction of cellulose, fabrication processes of cellulose products, cellulose products use, and their end-of-life disposal. In this regard, life cycle analysis (LCA) is a powerful tool for assessing the cumulative environmental impact attributed to all the steps from extraction-manufacture-use-dispose of cellulose, in other words, cradle to grave. Based on these analyses, measures can be taken to minimize the environmental impact and develop low-carbon cellulosic materials [7][8].

Payen first discovered cellulose in 1838, and its molecular formula was determined to be $C_6H_{10}O_5$ by elemental analysis. The structure of this polymer is a linear syndiotactic homopolymer formed by D-anhydroglucopyranose units (AGUs), which are joined by glycosidic bonds (as shown in Figure 1) [9]. The primary source of cellulose is plants such as wood, hemp, cotton, and linen, and it has been used as an energy source and construction materials for thousands of years [10][11][12]. Moreover, cellulose is produced from many microorganisms such as fungi and bacteria having the same chemical makeup as was studied by Brown in 1886 [13][14]. Bacterial cellulose differs from a plant in a degree of polymerization, purity, and characteristics [15][16][17][18]. It is clear that due to having three hydroxyl groups in the monomer structure, the formation of hydrogen bonds has a noticeable influence on directing the crystalline regions and, as a result, controlling the physical properties [19][20]. The presence of these hydroxyl groups also eases cellulose's chemical modification processes to obtain the cellulose derivatives that are useful for manufacturing new biopolymers with various applications [21][22][23].

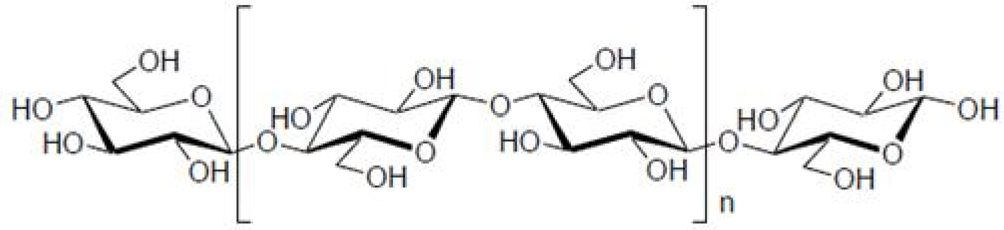

Figure 1. Structure of cellulose.

2. Life Cycle Assessment (LCA) of Cellulose

LCA is a useful tool for assessing environmental impacts related to the extraction of raw material, manufacturing, use of final products, and disposal. In other words, all the stages from the fabrication of products by using raw materials (cradle) to end-of-life (EOL) disposal methods of final products (grave) constitute LCA. Since cellulose-derived monomers and their derivatives, the first and second groups, are converted to chemicals and other polymers, their LCA is not performed. Therefore, the LCA of nanocelluloses and their products will be elaborated on in the subsequent sections. Due to some challenges such as data unavailability, inconsideration towards end-of-life treatments, and limitations related to the lab-scale processes, there are only a few LCA studies related to cellulose products, i.e., cradle-to-grave. Therefore, we characterized our LCA studies into two categories of cradle-to-gate and cradle-to-grave; their detailed information is shown in Table 1. Figure 2 shows a schematic of all the stages of the life cycle of nanocellulose products. The LCA for each form of cellulose can be investigated in terms of cumulative energy demand (CED), ozone depletion (kg CFC-11 equivalents), terrestrial acidification (TA, kg SO_2 equivalents), eutrophication (kg of phosphorus equivalent for freshwater, kg nitrogen equivalent for marine), water depletion (WD, cubic meters), human toxicity (HT, kg of 1,4-dichlorobenzene equivalent) and fossil fuel depletion (kg Oil equivalents), and climate change (CC) in the following sections. The evaluation of climate change in the life cycle, which is related to GHG emissions, resource depletion, global warming potential (GWP, kg CO_2 equivalents), and waste generation, is done to develop low-carbon materials. The main tools that are used for assessing the environmental impacts include CED (SimaPro v1.08 software, PRé Sustainability, Amersfoort, The Netherlands), International Panel on Climate Change (IPCC) (World Meteorological Organization, Geneva, Switzerland) with a time frame of 100 years (IPCC7 GWP 100a v1.02), Eco-Indicator-99 (PRé Sustainability, Amersfoort, The Netherlands) (EI99, SimaPro v2.08 for human health, ecosystem quality and resources), CML2001 (the Institute of Environmental Sciences, Leiden, The Netherlands) and ReCiPe (endpoint or midpoint for different parameters) (RIVM, Utrecht, The Netherlands). In the ReCiPe method, the life cycle inventory results transform to a limited number of indicator scores, where each indicator score illustrates the relative severity of an environmental impact category. Eco-indicator 99 identifies 11 environmental impact categories into three environmental damages endpoints, including human health, ecosystem quality, and resources.

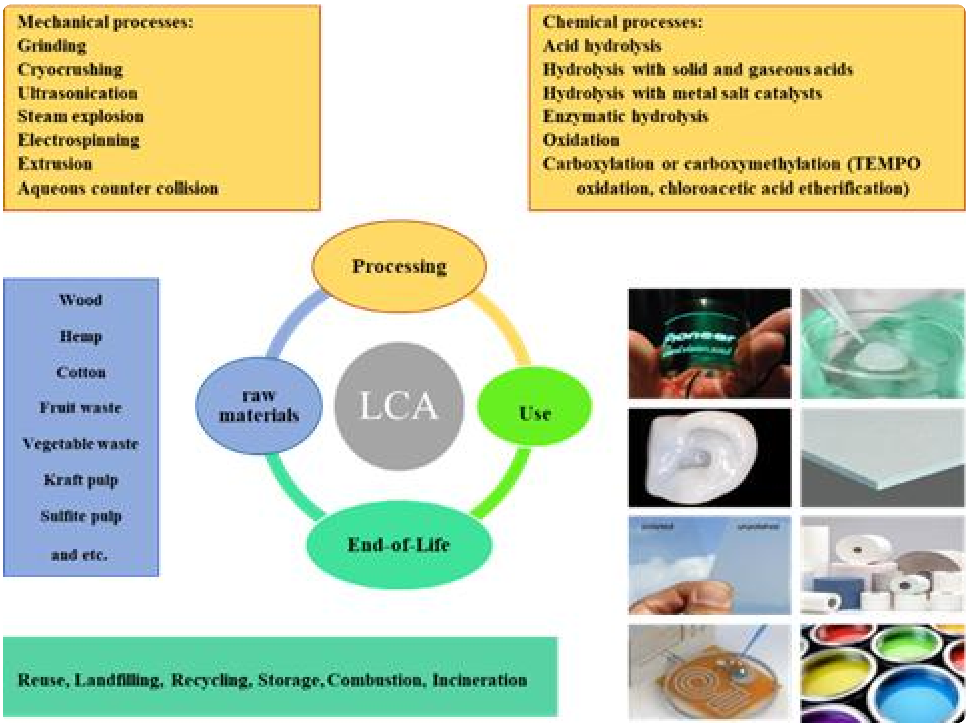

Figure 2. The life cycle of nanocellulose products, including raw materials, processing, use, and end-of-life stages.

Table 1. A summary of LCA findings of nanocellulose products, including cumulative energy demand and climate change from cradle-to-gate.

Researchers/Type of Cellulose	Production Method	CED Value	GWP (kg CO$_2$ eq)	ME/FE (kg N eq/kg p eq)	TA (kg SO$_2$ eq)	Fossil Fuel Depletion (kg Oil eq)	Human Toxicity (kg 1,4-DB eq)	WD (kg or m^3 H$_2$O)	Key Assumption Made
Hohenthal et al./CNF [24]	Enzymatic + HPH	—	1.2–3.1	0.015–0.016	0.008–0.045	0.3–0.75	—	50	Enzymatic pretreatment has more yield and lower wastewater. Energy consumption of the TEMPO oxidation reaction is more in that process.
	TEMPO oxidation+ HPH	—	1.0–1.8	0.018–0.024	0.005–0.0065	0.25–0.5	—	158	
	TEMPO oxidation+ mechanical refinement	—	0.75–1.0	0.014–0.015	0.0045–0.005	0.20–0.25	—	120	

Li et al./CMF [5]	TEMPO oxidation + Sonication + Centrifuge purifying (TOSO)	145.9 MJ	980 (per kg NC)	—	—	—	—	—	Weight loss does not have a significant influence on LCA results. Both chemical modification processes (TO, CE) create similar anionic surfaces. The products of two mechanical disintegration processes (SO, HO) are the same. The batch processing capacity ratio of the HO process to the CE process is assumed three. Washing does not influence four different fabrication routes. Energy recovery of the incineration/combustion process was not considered because of complexity. Solvent evaporation was considered negligible.
	TEMPO oxidation + Homogenization (TOHO)	34.7 MJ	190 (per kg NC)	—	—	—	—	—	
	Chloroacetic acid etherification + Sonication + Centrifuge purifying (CESO)	176.1 MJ	1160 (per kg NC)	—	—	—	—	—	
	Chloroacetic acid etherification + Homogenization (CEHO)	64.9 MJ	360 (per kg NC)	—	—	—	—	—	
Piccinno et al./CNF [25]	MFC liberated (Enzymatic + homogenization) +	32.2 MJ for production	1.5–1.6 (10 g of MFC)	—	—	—	—	(0.201 for MFC	All processes are performed in one place because of the lack of

	Coating MFC with GripX + Wet spinning by adding Sodium Alginate (route 1a).	of 10 gr MFC						liberation) 0.253 l/gr	transport between the various partners.
	MFC liberated (Enzymatic + homogenization) + Wet spinning by adding Sodium Alginate (without coating) (route 1b)			—	—	—	—	(0.201 for MFC liberation) 0.255 l/g	
	MFC liberated (Enzymatic + homogenization) + electrospinning by adding PEO as a carrier polymer (route 2)			—	—	—	—	(0.201 for MFC liberation) 0.205 l/g	
Piccinno et al./CNF [26]		—	—	—	—	—	—	—	——
Arvidsson et al./CNF [27]	Enzymatic pretreatment+ microfluidization	87 MJ/kg	0.79	—	0.0078	—	—	240	Neglecting the microbicide input due to low mass input toward produced CNF.
	Carboxymethylation pretreatment + microfluidization	1800 MJ/kg	99	—	0.18	—	—	1000	The contribution of heat losses to the overall CED is neglected.
	Without pretreatment +	240 MJ/kg	1.2	—	0.0069	—	—	130	

	homogenization treatment								
Figueiredo et al./CNC [28]	EUC system	15.943 MJ for the extraction of raw materials	1.086412	0.000320/0.000134	—	—	0.291122	131 L/g	Transportation of coconut husks was not considered due to the installation of these units in the vicinity of companies extracting coconut water. The transportation of fibers and chemicals are neglected because of lab-scale processes.
	EC system	1.8 MJ for the extraction of raw materials	0.122171	0.000065/0.000024	—	—	0.034797	138 L/g	
Nascimento et al./CNC [29][30]	Extraction of CNC with dilute sulfuric acid (CNH1)	—	—	—	—	—	—	—	Lignin was burned and used as a power source for cellulose nanocrystal extraction.
	Extraction of CNC with concentrated sulfuric acid (CNH2)	—	—	—	—	—	—	—	
	Extraction of CNC with ammonium persulfate (CNO)	—	—	—	—	—	—	—	
	Extraction of CNC with high powered ultrasound (CNU)	—	0.207	5.68×10^{-5}/3.03×10^{-5}	0.00045	—	0.0477	0.0023	
Hervy et al./BC/epoxy	BC/EP	—	~13.8	—	—	~270 MJ	—	—	BC is produced by A.xylinum under certain
	CNF/EP	—	~8.50	—	—	~145 MJ	—	—	

(BC/EP) and CNF/epoxy (CNF/EP) composites [88]	conditions, which are specified in the reference [88]. Purification of BC also was performed in a specific state. The influence of additional epoxy resin during the process on the LCA results was not significant. Materials and energy losses during the processing of the epoxy/BC and CNF composites were assumed to be negligible. The environmental impacts related to transportation were disregarded. The energy requirement for fibrillating kraft pulp to CNF was determined according to the work of Josset et al. [89]. GaBi software was used for the production model of BC and CNF nano papers.

The efficiency of all electrical appliances was assumed to be 100%.

The durability of epoxy composites containing BC and CNF, PLA, and GF/PP composites were considered to be the same [88].

2.1. LCA from Cradle-to-Gate

Hohenthal et al. evaluated the environmental impact of CNF for the first time. Their study was based on the cradle-to-gate LCA for the production of one-ton CNF using sulfite pulp as raw material and three different processing routes shown in Table 1 and two laboratories and one pilot-scale study. LCA was performed using the ReCiPe method and included GWP, eutrophication, TA, water depletion, and fossil fuel depletion (Table 1) [25][26][27][28][29][30]. Besides, a difference in electricity consumption between chemical processes was investigated. Table 1 shows that there is a considerable difference in wastewater between three different processing routes. Moreover, enzymatic pre-treatment has more yield and consumes more energy amongst chemical pre-treatment processes. In contrast, the TEMPO oxidation reaction’s energy consumption is less than other strategies and has a lower yield.

Li et al. studied the Cradle-to-gate LCA for 10 g of MFC from kraft pulp as raw materials on a laboratory scale. The fabrication processes included chemical treatment followed by mechanical techniques [90,91]. In this regard, chemical pre-treatments included 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO) oxidation and chloroacetic acid etherification [92–95]. Additionally, homogenization and sonication processes were selected as mechanical disintegration processes [90,91,96]. For samples treated by the sonication process, a centrifuge purifying process was required. Therefore, there were four possible routes of MFC production. CED (with SimaPro v1.08 software) and GWP (IPCC 7 GWP 100a v1.02) were assessed as two important environmental impacts. Furthermore, three main categories of environmental impacts, including human health, ecosystem quality, and resources, were assessed by Eco-Indicator 99 (EI 99, SimaPro v2.08) method and different egalitarian (E), hierarchist (H), and individualist (I) perspectives, which are long, medium, and short time horizon, respectively. The obtained results for

CED and GWP are summarized in Tables 1 [5]. Generally, they observed that the chloroacetic acid etherification and sonication processes require more energy for chemical and mechanical processes, respectively. Therefore, TOHO and CESO processes require a minimum and maximum energy, respectively. Besides, the GWP trend is similar to CED because CO_2 is emitted by using fossil fuels. The results of EI 99 show that human health has more importance in short- term perspective, while resources become more important for hierarchist and egalitarian perspectives. It is worth mentioning that ecosystem quality stays the same factor for all three perspectives. Moreover, among different nanocellulose production methods, the TOHO and CESO routes have the lowest and highest environmental impacts in each perspective.

In another study performed by Piccinno et al., the environmental impact for cradle-to-gate LCA of 1 g CNF was evaluated. The authors extracted CNF from waste carrot (carrot or carrot pomace) and considered three routes for fabricating CNF in the laboratory (as explained in Table 1). In this study, the impact assessment’s different scenarios included GWP, CED, ecosystem quality, human health, and resources by ReCiPe midpoint and endpoint indicators with the hierarchist perspective [82]. The results of ReCiPe endpoint indicators of three routes show that electrospinning has a higher impact on the environment than wet spinning due to the smaller scale, lower yield (60%), and mainly the high-energy consumption during this process. In wet spinning (route 1a and 1b), the liberation of MFC was considered as the most energy-consuming stage. A closer look at the MFC liberation stage shows that the enzymatic treatment is the main contributor to environmental impact because a lot of energy is needed for heating and stirring the mixture at 40 °C for 24 h. Besides, this stage has the highest share of wastewater. On the other hand, according to the results of ReCiPe midpoint indicators for route 1a, the liberation of the MFC has the highest environmental impact. Besides, acetone usage in the solvent exchange and GripX production has a high potential for photochemical oxidant formation and terrestrial ecotoxicity, respectively.

In comparison with the production of 10 g of MFC, which was studied by Li et al., the total energy consumption for the enzymatic treatment was lower than HO and SO processes in Li et al. study because, in the wood pulp production process, chemicals were responsible for a significant portion of CED, while in this process, electricity contributes to about 95% of the CED. About GWP, the impact of the TOHO route (1.9 kg CO_2 eq) was close to the process of this study.

In another study by Piccinno et al. on the impact assessment of CNF, the authors applied the scale-up framework to address the limitations of the lab-scale processes. In this regard, the authors only evaluated the Cradle-to-gate LCA for 1 kg of spun yarn process with the GripX coating (route 1: MFC liberated (enzymatic + homogenization) + coating MFC with GripX + wet spinning by adding sodium alginate). They examined different systems resulting in several scenarios, including different types of starting materials (carrot, carrot pomace), enzyme deactivation procedures (with heat or with a bleaching agent, ClO2), with or without heat, and solvent recovery, solvent exchange, or drying of the acetone. The authors evaluated the LCA of these routes based on depletion of resources, damage to human health, and ecosystem quality by applying the ReCiPe method (endpoint and midpoint) with the hierarchist perspective. The findings related to these scenarios show that: 1) using carrot pomace as a starting material reduces every step, such as transport, 2) using bleaching agent for deactivating the enzymes is preferable than heat deactivation, 3) solvent recovery in producing GripX has a considerable advantage [83].

In another study by Arvidsson et al., Cradle-to-gate LCA for 1 kg of CNF manufactured by wood pulp was studied by three different production methods (as shown in Table 1). For the manufacture of CNF, four different types of pulp were used, which contain elementary chlorine-free sulfate (ECF), totally chlorine-free sulfate (TCF), unbleached sulfate, and chlorine-bleached sulfite pulp. The environmental impact was studied in CED, GWP, TA, and WD, using the CED (SimaPro v1.08 software) ReCiPe method. The results of these categories are summarized in Table 1 [84]. According to Table 1, the environmental impact of the carboxymethylation route is significantly higher than other routes. To be more specific, for the carboxymethylation route, CED is higher compared to other routes because of the use of chemicals such as ethanol, isopropanol, and methanol. Besides, the pretreatment stage was the main contributor in GWP, TA, and WD. For the enzymatic route, pulp production has the main share in CED and GWP; while water usage in the washing stage and phosphate production, enzymatic treatment has more impact in WD and TA. The treatment process contributes more CED, WD, and GWP than pulp production for the no pretreatment route. However, the share of pulp production in TA is more considerable than in the treatment process.

In comparison with the study by Li et al., generally, the environmental impacts from the enzymatic and no pretreatment methods are lower than that of the TOHO process, which has the lowest environmental impact in that study.

Figueiredo et al. studied the Cradle-to-gate LCA of 1 g CNCs for the first time. The authors produced CNCs through acid hydrolysis from two different raw materials, namely, unripe coconut fibers (EUC process) and white cotton fibers (EC process) on a laboratory scale. Figure 3a,b presents the system boundary for EUC and EC processes, respectively. The authors evaluated the environmental impact, including CC, WD, HT, and eutrophication, using the ReCiPe method. In the ReCiPe method, climate change expresses the results according to the IPCC. The obtained values for each parameter are summarized in Table 4. The EUC system considers extra environmental impact than EC system except for WD. To elucidate, water consumption in turbines at hydropower plants to produce energy in the systems is the main contributor to WD. Furthermore, due to copper’s use in cables that distribute electricity, the EUC system generates more toxic substances and nutrients, leading to human toxicity and freshwater and marine eutrophication. In the EC system, the production of cotton on farms has a significant share in eutrophication.

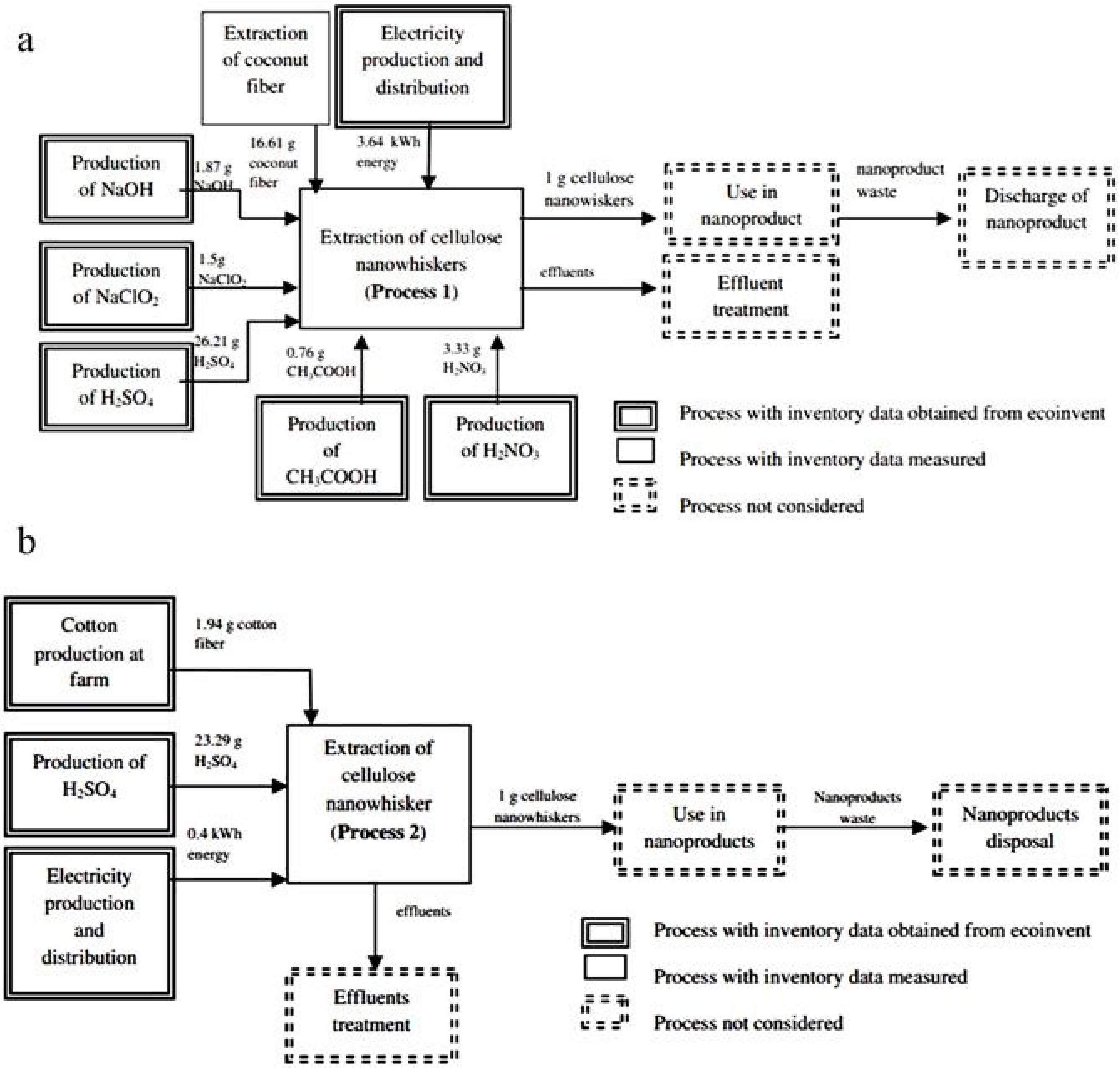

Figure 3. (**a**) EUC system (raw material: unripe coconut fiber), (**b**) EC system (raw material: cotton fiber) [28].

Additionally, the energy demanded by two process routes were compared with each other and other nanomaterials such as CNTs and carbon nanofibers. It is observed that the EUC system also demands more energy than the EC system. The extraction process in both systems is the main contributor to energy consumption. Compared to the production of 1 g carbon nanofibers, the production of 1 g CNC in the EC system has a lower impact on CC and HT, whereas the EUC system is affected at the same level [28]. The energy demand to make 1 g nanowhiskers in the EC system is lower than 1 g carbon nanotube and carbon nanofiber.

In another study of CNC production, Nascimento et al. evaluated the Cradle-to-gate LCA to produce 1 g of CNC from four different CNC extraction methods after extraction of coconut fiber on a lab-scale (as shown in Table 1). These methods were applied for recovering lignin by four other chemicals used to hydrolyze cellulose. Categories consisted of CC, TA, eutrophication (FE and ME), HT and WD were evaluated as environmental impact criteria via the ReCiPe method (at the midpoint level) with a hierarchical version. The results related to these assessments are summarized in Table 1 [29][30]. These results show that the CNU route has the lowest resource consumption and emission loads for producing 1 g CNC. Besides, this route has the highest yield among all the other routes. The usage of a high concentration of H_2SO_4 to speed up the hydrolysis of the amorphous domains leads to a decrease in the yield in the CNH2 method. Moreover, high reaction time, high demand for equipment use, and high selectivity of ammonium persulphate are the main reasons for the lower yield of the CNO method. However, the fabrication of CNC from the CNU route has more environmental impacts than the fabrication of CNF studied by Arvidsson et al. and Piccinno et al.

2.2. LCA from Cradle-to-Grave

To complete the investigation of environmental impact, it is essential to evaluate all stages from the extraction of raw materials (cradle) to EOL of cellulose products (grave). In this regard, Hervy et al. studied cradle-to-gate and cradle-to-grave LCA of the epoxy/BC and CNF composites for the first time. The authors used PLA and reinforced polypropylene/30 wt% glass fiber (GF/PP) as benchmark materials for comparison. Figure 4 shows the system boundaries for the epoxy/BC and CNF composites’ life cycle, neat PLA, and GF/PP composites. Final products were considered to be used as automobile parts. Depending on the waste density, landfills, incineration to recover energy, and recycling were selected as the end-of-life treatments for the plastic wastes. GWP and abiotic depletion of fossil fuels (ADF) were used to assess the environmental impact via the CML2001 (April 2013 version) method developed by the Centre for Environmental Science in Leiden University.

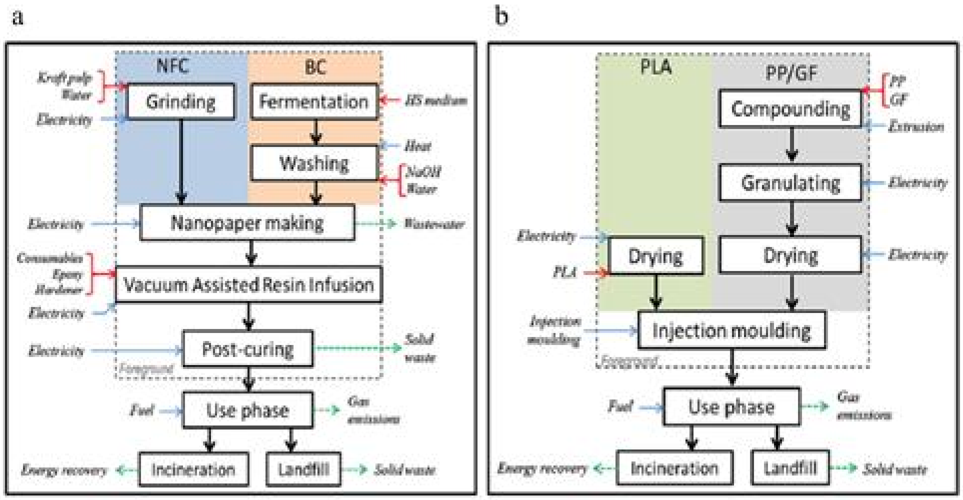

Figure 4. System boundaries for the life cycle of the (**a**) epoxy composites containing BC and CNF, (**b**) pure PLA and GF/PP composites. Red arrow: raw material required, blue: energy consumption, green arrow: material and energy wastes [31].

Considering the results in Table 1, for BC/EP composites, ADF was more than pure PLA and GF/PP composites. However, their study showed that the amount of BC/EP composites required were less due to the higher tensile modulus. Besides, BC/EP composites have the highest GWP compared with other materials. On the other hand, CNF/EP composites showed the same results compared to pure PLA and GF/PP composites, except that their values were less than BC/EP composites. The reason should be the higher amount of CNF needed to reinforce the epoxy.

Generally, although the evaluation of cradle-to-gate LCA of epoxy composites containing BC and CNF shows higher environmental impacts than pure PLA and GF/PP, the cradle-to-grave LCA of the composite containing 60 vol.% nanocellulose was lower than that of the pure PLA and GF/PP composites. To be more specific, in comparison with neat PLA and GF/PP, neat PLA has a higher GWP among all composites. In contrast, GF/PP composites have the lowest values. The cause attributed to these results is that the mass required to achieve performance is less for GF/PP composites.

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