# Sustainable production of nanofiber cellulose from sugarcane trash:

# Quality and life cycle assessment

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## Abstract

One of the most important challenges for sustainable production is the development of green and ecofriendly processes in chemistry and engineering domains. ~~sugarcane~~ Sugarcane trash (ST) generated in the sugar obtaining process ~~is an excellent biomass in sugar producing that~~ is a valuable feedstock for the production of cellulose nanofiber (CNF). Four different routes, namely ATH, ALH, OTH, and OLH were designed to optimize CNF production from ST. The performed characterization of the CNF samples showed good results in terms of purity and crystallinity. Fourier transform infrared spectroscopy (FTIR) illustrated a good correlation with CNF for characteristic functional groups. X-ray diffractograms (XRD) showed 64.28, 69.52, 60.54, and 68.81% crystallinity for OTH, OLH, ATH, and ALH, respectively. The ~~higher~~ highest yield of cellulose was observed with the OTH method (0.98 g/g ST) whereas OTH and OLH one~~method~~s provided high CNF yields as well (0.34 g/g ST)~~, and a higher yield of CNF with OTH (0.98 g/g ST)~~. ~~Fourier transform infrared spectroscopy (FTIR) illustrated a good correlation with CNF for characteristic functional groups. X-ray diffractograms (XRD) showed 64.28, 69.52, 60.54, and 68.81% crystallinity for OTH, OLH, ATH, and ALH, respectively.~~ Environmental parameters associated to the CNF obtaining processes were evaluated as well to target eco-friendly production of CNF from ST and to evaluate the environmental impact, including EcoScale (ES) and Life Cycle Analyses (LCA). EcoScale analysis awarded scores 84 for the OTH method, and 74, 67, and 74 for OLH, ALH, and ATH~~, respectively. However, eco-efficient production of CNF should be developed, within this context, life cycle assessment (LCA) was applied to target eco-friendly production of CNF from ST, and the~~LCA results showed that ATH method offered relatively ~~less~~ lower environmental impact in terms of all impact categories. ~~The sustainability analysis was employed to assess the environmental impact of the process of CNF production, in all methods.~~ Finally, the comparison between ~~The results of comparing~~ CNF ~~to~~ and other carbon nanomaterials~~, it was~~ shown that, in particular, CNF produced via ~~the~~ CNF method had lower environmental impacts.

**Keywords:**

Ecoscale, global warming potential, human toxicity, resources, ecosystem

## 1. Introduction

Lignocellulosic biomass wastes, from agricultural or forestry activities, has been already positioned as a competitive resource for the obtaining of different goods, materials, commodity chemicals and energy. The focus nowadays is on the development of ~~Recently,~~ green and ~~the~~ sustainable processes for the conversion of ~~waste~~ the lignocellulosic biomass residues though different routes~~o commodity chemicals , and bio-based biomass is widely recognized~~ (Sheldon, 2014). A lot of lignocellulosic biomass wastes are generated in agricultural activities ~~through agriculture~~ in Iran; for example, sugarcane waste, which includes two parts, sugarcane bagasse (SB) and sugarcane trash (ST~~), ~~). SB is produced in large quantities every year in Iran, and mainly used as a fuel, to produce ~~Medium~~medium-density fiberboard (MDF)~~,~~ and for animal feeding.~~, and~~ ST is produced ~~due~~ during the~~to~~ green harvesting of sugarcane, and represents about the 25-30% of the ~~it is a good source of renewable biomaterials. Approximately 25-30 % of the~~ processed sugarcane ~~yields~~ weight, that is more than 7,600 t / year in Iran, ~~tops and leaves, i.e., 250 - 300 kg ST per ton of sugarcane,~~ based on FAO reports ~~7561703 kg sugarcane produce per year in Iran~~ (FAO, 2020). That means an amount about 1,900 – 2,300 t /year of ST are generated and partially used to generate mulch for soil protection (Yadav, Prasad, Singh, & Srivastava, 1994) and to improve the crops yield by producing compost to increase the soil organic matter content and, thus, the soil fertility (Goyal, Dhull, & Kapoor, 2005)

~~Additionally, the sugarcane sector has experienced several changes over the years in Iran, especially the phase-out of leaves and tops burning before harvesting due to a variety of environmental issues. Furthermore, the amount of 189425.75- 2268510.9 kg ST produce yearly, and the rest remains as waste. ST can be used in the form of mulching on soil surface that it may have benefit on the soil quality , and crop yield (Yadav, Prasad, Singh, & Srivastava, 1994), another application of ST is producing compost to improve soil organic matter level , and long term soil fertility (Goyal, Dhull, & Kapoor, 2005).~~ However, ST can be potentially converted into ~~the~~ products with high added-value~~value-added~~.

Cellulosic materials, with at least one dimension in the nanometer range, are referred to as nano-~~-~~celluloses, ~~that are fascinating biopolymers and sustainable raw materials. Nano~~ Nano-cellulosic materials are the most important natural polymers and source of renewable materials (Klemm et al., 2018)~~.~~ ~~Nano-cellulose~~ that can be used in a wide range of applications, such as packaging (Azeredo, Rosa, & Mattoso, 2017; Khan, Huq, Khan, Riedl, & Lacroix, 2014), paper production (González et al., 2012), or medical applications (Duan et al., 2018; Yan, Hu, Yang, Zhang, & Zhao, 2018).

Various treatments, whether chemical or mechanical, can be used to remove the hemicelluloses, lignin, fat, wax and pectins that are surrounding the cellulose structure. Several methods have been reported in the literature to address the mechanical treatment for manufacturing CNFs from biomass, such as the use of high-pressure homogenizers (Muhamad et al., 2019), ~~ or~~ micro-fluidizer (Borrega & Orelma, 2019), ball milling (Nagarajan, Balaji, & Ramanujam, 2019), steam explosion (Cherian et al., 2010), ultra-soni~~fi~~cation~~,~~ or high-speed blenders (Sofla et al., 2019; Uetani & Yano, 2010). These cited methods have proved a ~~the advantage of~~ high~~er~~ consistency processing~~,~~ but they ~~seem to be~~are ~~more~~ energy demanding, and this is a significant concern regarding the production of CNF. The impediments of the high energy consumption in high-pressure homogenizers/micro-fluidizers have been alleviated by using various pretreatment methods, which have been necessary for commercial exploitation of CNF production (Klemm et al., 2018).

~~Various treatments can be used to remove hemicellulose, lignin, fat, wax, and pectin surrounding the cellulose structure, whether chemical or mechanical~~. Alkali or acid treatments, ionic liquid~~,~~ and enzymatic hydrolysis~~, are~~ classified as chemical processes, are the most used ones at the moment for~~. Generally,~~ CNFs producing~~are produced by chemical treatments~~ (Abraham et al., 2011; Mishra, Ha, Verma, & Tiwari, 2018; Song et al., 2018). Nevertheless, ~~T~~these methods involve the use of considerable quantities of ~~energy and~~reactants and solvents, such as acids, and large amounts of water for the several washing steps to remove the chemicals and solubilized compounds and neutralize the acids (Nascimento et al., 2018). ~~While showing the effective results, chemical pretreatment demands several washing steps to remove the chemicals and solubilized compounds.~~ The e~~E~~nergy demand for the pretreatment steps on an industrial scale is a bottleneck as well in the producing of ~~nano-cellulose~~ nano-cellulose under a ~~more~~ cost-effective perspective. ~~, energy efficient, and environmentally friendly.~~

Recently, there has been an intense research effort to produce ~~nanocellulose~~nano-cellulose under more sustainable conditions (Chaker, Mutjé, Vilar, & Boufi, 2014; Pinto, Bernardes, & Rezende, 2019; Zhai, Kim, Kim, Choi, & Kim, 2018). ~~For example, r~~Recent research presents the results of combining ~~suggests that the combined~~ mechanical and chemical methods with the aim of ~~reduce~~ reducing the ~~amount of~~process energy consumption (Klemm et al., 2018). ~~As~~ Junior et al.~~,~~ reported an integrated process focused on reducing steps in the CNF obtaining~~, it is possible to obtain nanocellulose by steam explosion method and by other methods that did not undergo the alkaline treatment, suggesting a probable decrease of steps in nanocellulose obtaining~~ (Júnior, Borsoi, Hansen, & Catto, 2019). Pinto et al., reported that it is possible to produce ~~nanofiber cellulose~~CNF using TEMPO-mediated oxidation without a mechanical defibrillation step (Pinto et al., 2019). Furthermore, CNF produced using TEMPO -mediated oxidation reduced the process environmental impact. ~~Additionally, the use of acids needs a large amount of water for neutralization, this problem limits industrialization of CNF production. Scientists are developing a green~~

~~strategy for the production of CNF. For example, a~~An interesting strategy for a more eco-friendly ~~,~~ and sustainable production of CNF is the use of natural organic acids, like citric acid, which is used for the extraction of ~~a~~ the non-cellulosic part of the lignocellulosic biomass (Minjares-Fuentes et al., 2014; Oliveira et al., 2016; Vriesmann, Teófilo, & de Oliveira Petkowicz, 2012). Lime juice, rich in citric acid, is ecofriendly and ~~economical acid since it~~its use reduces the environment impacts t~~hat~~ caused by the processing of more corrosive acids. It also reduces the amount of water needed for neutralization. Ravindran et al.~~, for the first time~~, reported for the first time that it is possible to produce NFC via a green-cost effective route using lime juice followed by ball-milling of the hydrolyzed pulp for ~~obtaining the~~ NFC production (Ravindran, Sreekala, & Thomas, 2019).

In this study, four ~~different routs~~routes ~~designed~~ with different reaction conditions for the production of NFC have been designed~~production and~~ and~~,~~ the related productivity and sustainability ~~were~~ have been compared. ~~Nanofiber cellulose obtained by subjecting cellulose pulp fibers to two different pre-treatments, and two different way of nanofibrilation differ in using TEMPO oxidation method, and using natural acid like lime juice. Furthermore, apart from the LCA analysis , and energy consumption of the scenarios of nanofiber producing from ST; S~~scenario 1, named OTH, ~~define~~ included~~as~~ an Organosolv pre-treatment, TEMPO oxidation ~~,~~ and High-pressure homogenization~~er; (OTH), scenario~~ Scenario 2, named OLH, included ~~define as~~an Organosolv pre-treatment~~,~~ followed by Lime juice hydrolysis (OLH~~),~~); S~~scenario 3, named ATH, consisted of Alkaline pre-treatment, TEMPO oxidation~~,~~ and High-pressure ~~homogenizer~~ homogenization (ATH). Finally, S~~, and s~~cenario 4, named ALH, included~~ define as~~an Alkaline pre-treatment, Lime juice hydrolysis ~~,~~ and High-pressure homogenization~~er~~ (ALH) Figure. 1 ~~show~~ summarizes ~~of~~ the ~~4~~ four used S~~s~~cenarios for ~~nanofiber cellulose~~NFC production.

Regarding the environmental impact assessment, f~~F~~or the best of our knowledge, this is the first study that presents a complete ~~focuses on~~ LCA of the CNF production process from ST. LCA is considered as a useful tool to quantify and understand the definite impact of one method or process on nature, society and economy, which are considered concrete pillars for sustainability. Eco scale was employed for the comparison of the Scenarios based on safety, economic and ecological features. The used software for the analysis was Simapro (8.8.1 version). The main objective of this study was to gain a better understanding of the initial production process by identifying the weakest points of the process in terms of environmental impact.~~which scenarios in the process had the largest contribution to environmental burden~~. For this purpose, ~~production of NFC from ST,~~ the evaluation of energy consumptions and environmental impact ~~in~~ s of the different methods for NFC extraction ~~have been done~~was performed ~~by~~ for the four defined scenarios.

~~LCA considered as a tool to quantify and understand the definite impact of the method or process on nature, society, and economy, which are considered concrete pillars for sustainability. Eco-scale employed for comparison of the processes based on safety, economics, and ecological features.~~

## ~~2.~~ Material and Method

### 2.1. Chemicals and raw material~~:~~

Green sugarcane trash (ST) was collected from, ~~cleaned from~~ Debal Khazai Agro-Industry of Ahvaz, Iran and cleaned~~.~~ ST was oven-dried (70° C, 24 h), milled and powdered; the fraction passing 60 mesh size was selected as raw material for the process. Sodium hydroxide, 2,6,6-tetramethylpiperidine-1-oxyl (TEMPO), ~~Ethanol~~ethanol, $NaClO_2$, NaClO, ~~Sodium~~ sodium ~~Bromide~~bromide, ~~Lime~~ lime juice, ~~Acetic~~ acetic acid and ~~, Glacial~~ glacial acetic acid were used in the different Scenarios. ~~Environmental sustainability analysis performed with EcoScale, LCA tools, and Simapro (8.8.1 version).~~

### 2.2. Experimental part~~:~~

CNF obtaining implied the extraction of the cellulosic fraction from ST followed by the transformation of cellulose into CNF by different routes. ~~was achieved by extracting of cellulose from ST followed by conversion of cellulose to CNF. The pre-treatments executed with different methods; chemical pre-treatment, and organosolv methods have been employed for removing lining and hemicellulose from ST.~~ CNF production was optimized with four different methods to find optimal strategies for comparison between them in terms of energy and environmental impacts. ~~Different~~ The different routes for CNF production are explained in detail in the following sections and ~~they~~ are depicted in Figure 1.

#### 2.2.1 Scenario 1 (OTH)~~:~~

A three-step ~~ST~~ delignification process was implemented for the obtaining ~~extraction~~ of CNF from ST, with organosolv, TEMPO oxidation~~,~~ and mechanical treatment. In OTH (Organosolv-TEMPO oxidation-~~Homogenizer~~Homogenization) method, 50 g of ST at a 1:10 solid to liquid ratio ~~was~~ were treated with a 1:1 (v/v) ethanol/water solution in a PARR reactor at 160°C for two hours, as previously described (de Oliveira et al., 2017). The resulting pulp was mixed with a 1% (w/w) NaOH solution and rinsed with distilled water until ~~achieving~~ neutral pH. The ~~resulting~~ generated cellulose was bleached with 500 mL of an aqueous solution comprising acetic acid (25 % wt.) and~~,~~ $NaClO_2$ (2 ~~wt%~~% wt.) for 180 min at 70~~.~~ °C under vigorous stirring as previously described (Pinto et al., 2019). 5g of ~~Bleached~~ bleached cellulose hydrated in ultrapure water (500 mL) for 24 h, were mixed with ~~followed by the addition of~~ TEMPO (0.08 g, 0.5 mmol), and sodium bromide (0.5 g mmol). Then, the oxidation started by the addition of specific volumes (15.6 or 78.0 or 156.0 mL per gram of cellulose) of a 12% (w/v) NaClO solution. The bleached fibers were stirred at room temperature. The TEMPO-oxidized cellulose was abundantly rinsed with

ultrapure water by centrifugation until constant conductivity was reached in water (Pinto et al., 2019). Finally, the pulp was ~~submitted to a~~treated in a homogenizer to reach better quality of CNF. After mechanical treatment, the CNF was dried using a freeze drier.

**2.2.2 Scenario 2 (OLH)~~:~~**

In this method (Organosolv-Lime juice hydrolysis-Homogenizer), the raw material (ST) wa~~i~~s mixed with the solvent (ethanol-water, 50:50 w/w) in a pressurized reactor (~~T: 1~~60~~°~~°C, ~~time: 1~~20 min, liquid/solid ratio10:1 w/w). Once reaction time finished, the pressure reached during the reaction was reduced down to atmospheric value in a flash operation that ~~allows~~ allowed recovering a stream composed by ethanol and water that ~~is~~ was condensed~~,~~ and recycled. The remaining cellulose was washed with distilled water and bleached. The bleached cellulose solution was diluted with distilled water, filtered~~,~~ and dried. The extracted cellulose ~~is~~ was subjected to depolymerization in 100 ml of lime juice (the pH of lime juice ~~is~~ was 2.5), and stirred at 1000 rpm for two h at 70~~°~~°C. Then the pulp was washed several times with distilled water to remove the excess acid.

In order to improve the dispersion of nanofibers, the samples were submitted to mechanical treatment in a high-pressure homogenizer. The pressure in the first and second stages was 500 and 50 bar, respectively. CNF suspensions were passed through the homogenizer five to seven times.

**2.2.3 Scenario 3 (ATH)~~:~~**

In ATH (Alkaline-TEMPO oxidation-Homogenizer) method, 20 g~~rams~~ of dried ST were treated with 10% sodium hydroxide solution and stirred for two h at 75 °C. Then, the obtained pulp was~~ and~~ washed with distilled water several times until rich neutral pH~~,~~ and dried. For bleaching, 20 g~~rams~~ of cellulose ~~will~~ were treated with 500 mL of an aqueous solution comprising acetic acid (25 ~~wt%~~% wt.) and~~,~~ $NaClO_2$ (2 ~~wt%~~% wt.)~~, and~~ for 180 min at 70~~°~~ °C under vigorous stirring as previously described. A ~~a~~fter that, cellulose fibers were dried at 50~~°~~ °C for 24 hours~~,~~ and milled (Pinto et al., 2019). For NFC production, 5g of ST were hydrated in ultrapure water (500 mL) for 24 h, followed by the addition of TEMPO (0.08 g, 0.5 mmol), and sodium bromide (0.5 g mmol). Then, oxidation started by the addition of a 12% (w/v) NaClO solution. The bleached fibers were stirred at room temperature. Finally, TEMPO-oxidized cellulose was rinsed with ultrapure water by centrifugation until constant conductivity reached in water (Pinto et al., 2019). In order to improve the dispersion of nanofibers, the samples were submitted to mechanical treatment in a two-stage high-pressure homogenizer. The pressure in the first and second stage was 500 and 50 bar, respectively. CNF suspensions were passed through the homogenizer five to seven times. CNF not submitted to homogenization used for comparison purposes.

**2.2.4 Scenario 4 (ALH)~~:~~**

In this method (Alkaline-Lime juice hydrolysis-Homogenizer), for reducing the chemical load in the extraction of CNF from ST, natural organic acids were used for hydrolysis. Firstly, ST was treated with 10% NaOH at 75 ~~°~~ºC for an hour ~~with~~ under stirring. After the given time, the remaining pulp was collected and the remaining sodium hydroxide (NaOH) was washed out by ~~resuspending~~suspending the pulp in 5 L of water~~,~~ and collecting it on the sieve again; this procedure was repeated until pH 6.5 was measured in the pulp~~,~~ and then it was filtered. Then the delignified residue was dried~~,~~ and bleached with 500 mL of an aqueous solution comprising acetic acid (25 ~~wt%~~% wt~~),~~ .) and $NaClO_2$ (2 ~~wt%~~% wt.) ~~, and~~ for 180 min at 70~~°~~ ºC under vigorous stirring as previously described (Pinto et al., 2019). The bleached pulp was diluted with distilled water, filtered~~,~~ and dried. The extracted cellulose ~~is~~ was subjected to depolymerization in 100 ml of lime juice and stirred ~~with~~ at a speed of 1000 rpm for 2 h at 70~~°~~ ºC. Then, the suspension was washed several times with distilled water to remove the excess acid. In order to improve the dispersion of nanofibers, the samples were submitted to mechanical treatment in a two-stage high-pressure homogenizer.

**2.3 Characterization of ST**

The chemical c~~C~~haracterization of ST was performed ~~with~~ following the NREL protocol for the determination of structural carbohydrates~~,~~ and lignin (Sluiter et al., 2008). Solvent extractives ~~performed~~ were measured with the T 204 cm-97 protocol (Tappi Standard, 2004)~~,~~ and the inorganic matter content ~~ash content~~ determination was performed ~~with~~ following the T 211 om-93 protocol (Tappi Standard, 2007).

**2.4. Functional groups, relative crystallinity of the fibers, and SEM images**

FTIR spectra were obtained with an FTIR spectrometer in the wavenumber range from 4000 $cm^{-1}$ to 400 $cm^{-1}$ using the KBr disk method. The relative crystallinity of the fibers of CNF samples ~~were~~ was investigated by X-ray diffraction. The analysis was performed in an X-ray diffractometer with a scanning range between ~~°~~5º and ~~°~~40º (~~°~~2º Th). The calculation of the relative crystallinity of the fibers was performed according to the method described by Segal, Creely, Martin and Conrad (1959). The crystallinity index (Icr) percentage was measured as indicated in Equation 1:

$$Icr = \left(\frac{I_{200} - I_{am}}{I_{200}}\right) 100 \quad (1)$$

where $I_{200}$ (22.5°) is the intensity of the peak corresponding to cellulose and $I_{am}$ (16°) is the intensity of the peak of the amorphous fraction.

SEM images of the samples recorded on a field emission scanning electron microscopy (FE-SEM) with an accelerating voltage of 5 kV.

**2.5 Calculation of energy demand**

The energy demand integrated for lime juice hydrolysis, TEMPO oxidation, and mechanical treatment were calculated ~~from the following~~as indicated in E~~-e~~quation 2:

$$\text{Energy} = \text{Power (w)} \times \text{Time(h)} \quad (2)$$

The energy consumption for the process was expressed as kilowatt-hours per kilogram of the dry weight of CNF.

### 2.6 Eco-Scale and Life Cycle Assessment

#### 2.6.1 Eco-Scale analysis

An organic product involves not only a relatively efficient reaction but also environmental issues. Therefore, to evaluate the quality of the preparation process, it is essential to examine the safety and ecology. EcoScale is a post-synthesis tool that measures the environmental responsibility of the a synthesized product by considering six critical parameters, such as the yield of the product, the price, the safety to in the use of chemicals (reactants, and products), the technical set up, processing conditions (temperature, /time), and the mode of product purification steps of the product (Van Aken et al., 2006). These six parameters that affect the quality of the product of a reaction are taken into consideration and the tool represents measurementa scale covers from 0 to 100; the ideal reaction has would have an EcoScale value of 100 it meanings that the reaction efficiency is 100%. If the EcoScale score isbe more than 75, the process is categorized as excellent process, ; the values in between 50 and 75 is are considered as acceptable process, and thewhile processes with EcoScale score less lower than 50 are cathegorized asis an inefficient process ones (Katakojwala et al, 2019). Ecoscale EcoScale evaluateions are based ond by penalty points (Table 4), and the EcoScale score is calculated as shown in Equation 3by:

$$\text{EcoScale} = 100 - \text{sum of individual penalties} \quad (3)$$

#### 2.6.2 LCA

The applied methodology for the performedis LCA is based on the guidelines of ISO series 14044 (ISO 2006, ISO 2012) ,), and 14045, which provide a framework for carrying out the study that consists of four phases, namely (1) the goal and, and scope, (2) the inventory analysis, (3) the impact assessment phase, , and (4) the interpretation phase.

##### 2.6.2.1 Objectives and scope

LCA main aim has been thes are comparison of the full range of environmental impacts associated todue to CNF production. This study has been designed as a “cradle-to-grave” concept, and the boundaries of the system have been well defined for the analysis, including ST transferring ST to the factory, production of nanofiber cellulose, and waste management. The first phase of the LCA study is the definition of the goal and scope, the goal and scopewhich in this case, as it has been previously mentioned, in this case is the evaluation of the environmental impacts for of the CNF production processes through the used of chemicals, organosolveorganosolv pretreatmentss, and mechanical treatment means using ST as the raw material.

The LCA performed for optimizing the environmental performance of CNF production through the identification of hotspots, and to perform a comparative LCA on the different processing methods and environmental performance useful for industrial uses.

The functional unit for producing ~~nanofiber cellulose~~NFC from ST was considered as 1 kg of output product. Weighting in LCA was done to express~~make~~ the indicators dimensionless, in order to~~and provide~~ offer an easy comparison of the different Scenarios~~the possibility of comparing them, and a better understanding of the results~~. The normalized indicators or results ~~are~~ were made dimensionless by the selected weighting components. Normalization helps a better understanding of the results of the manufacturing system in the study.

One of the critical issues in LCA is the multi-product system allocation, which allows the partitioning of the environmental~~,~~ and energy burdens associated with a multi-output process to its products and co-products. During the LCA modeling of CNF from ST, the "cut-off" approach was applied as a default~~,~~. In~~in~~ this approach, outputs (in this case output is ST) subjected to recycling were considered inputs to the ~~next~~ following life cycle~~,~~ and no environmental burdens or environmental impacts derived from the recycling process were allocated to the waste. The ST is then free of any load of environmental impact.

**2.6.2.2 Inventory analysis**

In this stage, systems modeling has been done by the compilation of all foreground data, that is all input and output data for the ~~nanofiber cellulose~~NFC ~~production~~ obtaining process from ST including the production sectors and the modelling of the environmental processes. The weighing of the effect groups was examined as well. ~~The data related to the modelling of the production system called foreground data (all inputs , and outputs in a manufacturing process), whereas~~Furthermore, the background data was determined as well from databases, including ~~includes~~ energy~~,~~ and materials added to the foreground system as aggregated data sets ~~that found in databases and resource~~.

**2.6.2.3 Life cycle impact assessment**

Life cycle impact assessment (LCIA) quantifies the environmental impact of inputs~~,~~ and the emissions at different stages of a product life cycle. The impact categories considered for this study have been the following: global warming potential, ozone depletion, human toxicity, photochemical oxidant formation, particulate matter formation, ionizing radiation, climate change ecosystems, terrestrial acidification, freshwater eutrophication, freshwater ~~ecotoxicity,~~and ~~marin~~marinee ecotoxicity, transformation, metal depletion, and fossil depletion. Those categories allow the determination and identification of the potential environmental impacts.

**2.6.3 Sensibility analysis**

Sensibility is an essential phase in bioenergy LCA studies that shall be thoroughly investigated before drawing conclusions and recommendations. In many studies, most of the inventory data used are secondary data, obtained from estimation models or literature~~;~~. T~~t~~herefore, it is recommended to perform a sensitivity analysis of the most important parameters, as indicated by ISO14044 standards (ISO 2006b). Sensitivity

analysis investigates the influence of input changes on the results of a model by verifying the extent to which the final result of the study depends on a given choice or assumption. A sensibility analysis is carried out with the purpose of identifying the most promising systems improvements (Pianosi et al., 2016).

## 3. Results and discussion

### 3.1 Proximate analysis of ST

The ST ~~containd~~contained 34.58% of cellulose, 21.63% of hemicelluloses, and 21.16% of lignin ~~(by~~ weight percentages) measured according to NREL protocols (Sluiter et al. 2008, 2012). Table 2 shows the detailed ~~of~~ carbohydrate composition~~ ,~~ and lignin percentages in terms of soluble and insoluble lignin~~,~~. ~~These~~ The obtained results ~~agree~~ are in concordance with those reported by Jutakanoke et al. (2012) and Pathak et al., (2020)~~, they reported that ST contains 37% cellulose, 23% hemicellulose , and 20% lignin (w/w) on a dry weight basis (Jutakanoke et al., 2012).~~. ~~Pathak et al., 2020 also reported that the cellulose, hemicellulose, and lignin contents of sugarcane trash were 35.0 ± 0.4%, 30.3 ± 0.5%, and 17.0 ± 0.7%, respectively; their results also are similar to this study.~~ The sligh differences in the published compositions may be due to the different varieties of sugarcane~~ and different procedures applied for analysis~~. The total extractives content in ST was 1.26 %, out of which were 0.34% ethanol-soluble, and rest 0.92% were water-soluble; extractives include aromatic phenolic compounds, terpenes, saturated, and unsaturated higher fatty acids, flavonoids, and proteins (Franco et al, 2013).

### 3.2 Isolation of nanofiber cellulose

#### 3.2.1 Cellulose extraction

Cellulose was extracted from ST by an organosolv treatment with ethanol (in Scenarios 1 and 2) or alkaline process with NaOH (In Scenarios 3 and 4) as the first step of the ~~followed by the production of~~ NFC production process~~ from extracted cellulose~~. The obtained ~~products~~ yields ~~by the alkaline and organosolv processes~~for each Scenario are presented in Table ~~2~~3. As it can be seen, the cellulose-rich solid fraction yield was slightly lower for the alkaline treatment (0.30 ± 0.02 g/g ST) compared to the organosolv one (0.34 ± 0.06 g/g ST )indicating that, by~~ alkaline treatment reagent~~this process, ~~produced~~ a stronger delignification was achieved. ~~In method 1, and 2 (alkaline treatment), the obtained cellulose yield was 0.30 ± 0.02 g/g ST. Scenario 2 and 3 (organosolv) yielded relatively higher cellulose 0.34 ± 0.06 g/g ST.~~ Removing lignin is a key factor in effective cellulose extraction, and it can be considered that an effective delignification was achieved by both treatments even if the ~~strong~~ alkaline ~~treatment~~ one~~can~~ was stronger. ~~remove lignin content from ST~~. ~~The alkali pretreatment was carried with NaOH in methods 1 and 2 for removing the lignin content from ST, in method 3 and 4, the organosolv pretreatment had significant influence on the performance of cellulose production.~~

### 3.2.1 ~~Convert to~~ NFC obtaining

The highest yield of NFC from cellulose was observed in the OTH ~~method~~Scenario~~, CNF produced was~~ (0.96 ± 0.01 g/g cellulose) followed by the ~~. In~~ OLH ~~method production of CNF was~~one (0.93 ± 0.01 g/g cellulose), a~~nd in~~ ATH (0.90 ± 0.01 g/g cellulose) and ALH ~~the amount of CNF was 0.9 ± 0.01, and~~ (0.89 g/g cellulose~~), respectively~~. The quantitative yields are listed in Table 3. CNF yield was calculated ~~with concerning~~considering the total amount of ST used~~,~~ and the productivity obtained in the four Scenarios ~~of~~ (OTH, OLH, ATH, and ALH) w~~ere~~as ~~0.32 ± 0.02~~, 0.32± 0.02, 0.31 ± 0.02, 0.27 ± 0.02~~,~~ and 0.26± 0.02 g/g ST, respectively. ~~The highest yield of cellulose and CNF was observed in OTH method, and the quantitative yields are listed in table 3~~. From the obtained results it could be concluded, on the one hand, that the differences in the yields of the four Scenarios is moderate, with a maximum difference of 0.07 g of CNF per g of cellulose between the best and the worse Scenarios in terms of yield. This quantity is not negligible but not determinant by itself as well. On the other hand, the organosolv treatment offered the highest yields in OTH and OLH processes even if the alkaline treatment use allowed the obtaining of higher quantities of cellulose to be used for NFC obtaining. This fact could be associated to the performance of the subsequent applied treatments, such as bleaching. The use of the alkaline treatment, with a stronger delignification power, combined with the subsequent bleaching could have resulted in a lower final yield of NFC. Furthermore, the comparison between the yields obtained by TEMPO or lime juice treatments indicated slightly higher results in terms of yield for the TEMPO process.

### 3.3 Characterization of CNF

#### 3.3.1 FTIR

Figure~~.~~ ~~1~~ 2 illustrates the FTIR ~~spectra~~ spectrum of CNF for all ~~methods~~Scenarios; FTIR can characterize the functional groups of natural fiber. The main absorbance b~~,~~ands emerged between 800 and 1800 $cm^{-1}$ region, and resulted from the typical vibrations of protein~~,~~ and polysaccharide structures present in these samples, such as cellulose~~,~~ ~~and~~ as well as the remaining hemicelluloses or lignin (Edi Syafri et al., 2018). The adsorption peak near 3340–3367 $cm^{-1}$, observed in all spectra, represents the stretching vibration of OH (Sain & Panthapulakkal, 2006). A broad absorption b~~,~~and between 3600 $cm^{-1}$~~,~~ and 3000 $cm^{-1}$ ~~is~~ is associated to~~due to~~ the vibration of hydrogen-bonded hydroxyl groups in the structure of CNF in all method~~s~~, and it indicates hydrophilic nature in all samples (Li, Wang, Li, Cheng, & Adhikari, 2014). The peak at 2924 $cm^{-1}$ ~~is~~ was due to the stretching vibration of saturated CH in cellulose, hemicellulose~~s,~~ and lignin (Alemdar & Sain, 2008). The pe~~a~~ek at 1035 $cm^{-1}$ indicates the frequency of C=H and C-O groups of cellulose (Sun, 2004). Previews studies showed that the increase in the intensity of this peak was ~~attributes~~ attributed to~~for~~ the ~~removing~~ removal of non-cellulosic parts ~~bound~~ around the cellulose. The

adsorption peak at 1630 $cm^{-1}$ is principally associated with the bending vibration of absorbed water molecules. The blending vibration of CH gets reflected in two peaks, at 1371 $cm^{-1}$, due to asymmetric bending vibrations (Sain& Panthapulakkal, 2006; Xiao, Sun, & Sun, 2001). No ~~peek~~ peak was observed at 1245 $cm^{-1}$ (lignin's aromatic ring vibration), and 1640 $cm^{-1}$ (OH group water absorption) for all method, ~~it indicates~~ indicating that all methods were successful in the ~~removing~~ lignin removal (Mahardika et al., 2018). No significant differences were found between the spectra of CNF in all scenarios.

### 3.3.2 XRD

Fig~~.~~ure 3 shows the X-ray diffraction pattern of fibers for all ~~methods~~Scenarios. Higher crystallinity is the key factor that determines better reinforcement with the polymer matrix. The two major peaks that appeared in the diffractogram ~~are~~ were 2θ=16º, and 22.5°. ~~Higher crystallinity is the key factor that determines better reinforcement with the polymer matrix.~~ There was an increase in the height of the intensity peak of 2θ=22.5°. The h~~H~~igh~~est~~ of these peaks is a measure of the crystallinity index (Icr) of the fiber (Abral, Putra, et al., 2018; W. Chen et al., 2011). Values of Icr for each sample were 69.52, 68.81, 64.28, and 60.54 for OLH, ALH, OTH, and ATH, respectively.

### 3.3.3 SEM

Scanning (SEM) of the ST is used for the investigation of the structure of the fibers from all methods. The morphology of four CNF samples is portrayed in ~~figure~~Figure~~.~~ 4. The FE-SEM images at a similar resolution indicates that the particles' size in the OLH is larger than the other particles. In ALH, and OLH methods acid hydrolysis using lime juice followed by high pressure homogenizer shows CNF with a nanoscale size~~,~~ . ~~this~~ This result ~~agrees~~are in good concordance with those reported by Ravindran et al (2019). This observation showed that the TEMPO oxidation and high-pressure homogenizer facilitated the effective hydrolysis of glycosidic linkages in the long cellulosic chains in ATH and OTH. Due to the aggregation of individual cellulose fibers, similar FE-SEM images were obtained for other authors (Chen, Lee, & Abd Hamid, 2017; Qua, Hornsby, Sharma, & Lyons, 2011). From the FESEM result, it ~~is clear~~could be concluded that ~~although~~ the size of CNF from ALH and OLH is larger than the one ~~CNF~~ from OTH and ATH ~~but~~ and that the use of ~~using~~ lime juice for the hydrolysis followed by high-presser ~~homogenizer~~ homogenization resulted in a ~~have been a~~ successful route for the CNF production.

### 3.2.1 EcoScale

The EcoScale score results for the four ~~CNF methods~~Scenarios were 82 for OTH, ~~-~~74 for OLH and ATH and ~~,~~67~~, 82, and 74~~ for ~~ATH,~~ ALH~~,~~ . ~~OTH~~, ~~and OLH respectively~~ (Table 4). The scores for ~~ATH,~~ OTH, OLH and ~~OLH~~ ATH methods are ~~falling~~ classified as~~under the~~ excellent category, ~~and~~ whereas ALH

method is ~~falling under~~considered as ~~the~~ acceptable category. The ~~different~~ differences in the obtained score ~~was observed due~~can be attributed to the yield of the products, cost~~s,~~ and operational conditions (temperature/time) ~~from the individual~~of each method~~s~~. The yield from ~~ATH~~ OTH and ~~OTH~~ ATH methods was 84%, and ~~for ALH~~ 69~~%, and~~ for OLH ~~69%~~and ALH. No penalty points were ~~observed in~~associated to the cost for all routes because of the inexpensive used raw material~~s~~ (agricultural wastes). No plenty point was observed in safety parameters for OTH~~,~~ and OLH methods because the chemicals used in these processes are non-toxic in nature but~~,~~ in ATH~~,~~ and ALH Scenarios, the plenty point was 10 due to the use of~~using~~ sodium hydroxide, as the ~~because~~ dissolution of sodium hydroxide is highly exothermic. The penalty points for temperature and reaction condition category were considered based on the used operating conditions in terms of temperatures, time~~,~~ and purifications. Plenty point of these parameters scored the highest ~~amount~~ value for OTH and OLH methods ~~in compared to other methods~~. ALH got the least EcoScale in ~~compare~~ comparasion with~~to~~ other routes due to lower yield and the use~~using~~of toxic material~~s~~, such as sodium hydroxide.

### 3.2.2 Life cycle impact assessment (LCIA)

In this study, life cycle impact assessment (LCIA) approach was applied for the~~to~~ evaluation of the environmental effects of the obtained products and processes using the inventory results. The inventory data for all the CNF production routes are provided in Table 1. ~~In LCA~~The methodology was established by considering mass~~,~~ and energy balances, the data of ~~a~~ chemical compound~~s~~, and the processes in the system ~~only incorporated~~ excluding the previous impacts of its formation~~, and this would become a feasible approach in future of CNF production~~. LCA approach involved inventory data with the environmental impact categories. For the environmental impact assessment, the methods were selected based on the ILCD handbook (EC, 2011).

The quality assessment of CNF was executed followed by LCIA assessment using contribution analysis to identify the environmental hotspots. Meanwhile, sensitivity analysis and a comparison with other studies were included to consider the limitation and recommendations to use in further process development.

#### 3.2.2.1 Midpoint method, Endpoint method and IPCC 2013 GWP 100a/V1.03 method

The results of quantification of impact categories are provided in ~~table~~ Table 5 for all routes for the production of 1 kg of CNF from ST~~,~~. It is worth to mention ~~that~~that the impact categories quantified are based on the ILCD 2011 Midpoint + method (EC, 2011). Climate change, Terrestrial acidification, Human toxicity, Photochemical oxidant formation, Freshwater ecotoxicity, ~~Marin~~Marine~~e~~ ecotoxicity, ~~Ionising~~Ionizing radiation, Water depletion, Metal depletion~~,~~ and Fossil depletion were considered for the relative analysis. In comparison, OLH ~~has appeared~~presented the highest impact on all the impact

categories~~,~~ and OTH showed a higher impact in comparison to ATH~~,~~ and ALH~~;~~. In the case of~~in~~ OLH a higher impact is ~~due to using~~associated to the use of more electricity for the ~~organosolve~~organosolv pretreatment, ~~using ethanol,~~ and higher energy consumption ~~for~~ in the lime juice hydrolysis. The e~~E~~ffect of the CNF production methods on human health, ecosystem~~,~~ and resources was quantified by the Midpoint method, and the results are shown in Table 6. In the group of all inputs, ST presented a lower effect on all the impact categories due to the cut-off approach, because ST is a waste of sugar~~,~~ and bioethanol production from sugarcane. A major impact on human toxicity, ~~marin~~marinemarine ecotoxicity~~,~~ and climate change was observed in all methods~~;~~. T~~t~~hese impacts occurred due to the electricity consumption needed in different unit operations~~,~~ as well as the use of~~and~~ acetic acid for the bleaching. As depicted in Table 6, the chemicals, such as ethanol used on OTH~~,~~ and OLH for the ST pre-treatment ~~of ST~~, showed ~~the~~ a potential impact on the ecosystem and resources. Regarding these observations, the electricity was the most effective input on environmental sustainability~~,~~ and ATH method ~~illustrated~~ presented the lowest~~lower~~ impact on all the impact categories ~~than ALH, OTH, and OLH,~~ associated to electricity consumption~~in electricity area~~. However, the influence of OTH ~~,~~and OLH methods on human health, ecosystem ~~,~~and resources ~~can~~ could be potentially reduced by the recovery of ethanol in organosolv pre-treatment ~~to achieve minimum ethanol consumption for the processing of ST.~~(Teramoto, Lee, & Endo, 2008).

In the IPCC GWP 100a method, Sankey~~snaky~~ diagrams are presented ~~is supplied~~ for ~~in~~ the input materials (~~figure~~ Figure 4 and ~~table~~ Table 7). ~~Sankey diagrams is a special kind of diagrams that the arrows show the quantity of the flow and provide an appropriate understanding the contribution of each input on the environmental impacts.~~ Sankey diagrams emphasize the major transfers or flow within a system. They help locate the most important contributions to a flow. They often show conserved quantities within defined system boundaries. Sankey diagrams of OLH and OTH represented 69%~~,~~ and 70 % of the GWP ($CO_2$ kg eq.) contributed by electricity and ethanol, respectively, which were main energy inputs (~~figure~~ Figure 4). Electricity utilized in various unit operations was the major contributor towards environmental impact in all methods. Acetic acid, which used in the bleaching, ~~indicated~~ supposed the highest contribution in ATH and ALH. The chemical inputs, such as sodium hydroxide and inorganic chemicals showed insignificant contribution to GWP.

The performance of CNF production methods was evaluated using the IPCC GWP 100a method. Table 7 showed the share of each input in terms of $CO_2$ equivalents. The results showed that the ALH method~~,~~ had ~~a~~ the ~~lower~~ lowest impact ~~than other methods~~ in terms of GWP. Although acid hydrolysis of ST with lime juice in the ALH method implied longer operation times ~~used with more time~~ than the other routes, ~~but~~ the use of chemicals, ~~ethanol~~solvents~~,~~ and ~~high amount of~~ energy ~~consumption~~ was reduced in comparison with OTH and OLH Scenarios.~~(regarding organosolv in OTH, and OLH) reduced, this result illustrated that~~

~~electricity had a potential impact on the environment~~. OLH method presented in general the better results, as a ~~is~~a mineral acid-free scheme ~~and uses~~with the lowest ~~less~~ energy use for CNF production. ~~The~~ This green method ~~has lower impacts~~presented the lowest ~~on~~ environmental impacts~~,~~ as well as~~and more~~a more cost-effective rote. ~~;t~~These results agree with the ones published by Ravindran et al., 2019.

#### 3.2.2.2 Water Consumption

Water consumption for all methods is illustrated in T~~t~~able 2. The results show that OLH~~,~~ and OTH methods presented the lowest~~have lower~~ water consumption ~~than other methods~~. Two different pre-treatments were employed to ST to remove the non-cellulosic polysaccharides ~~CNF~~. ATH method had the highest water consumption due to the applied alkaline treatment~~,~~ and TEMPO oxidation. ~~,~~ T~~t~~he amount of water required in the standard chemical pre-treatment was at least 60 L for neutralizing the pH after the alkaline treatment with 10% sodium hydroxide ~~, , and~~ . Furthermore, 15 L were required to wash out ~~an~~ the oxidizing agent with acetic acid ~~,~~ and sodium. A n~~N~~eutralization of the pH is necessary to enable buffering of the oxidant solution to pH 5, at which chlorite works effectively for oxidizing non-cellulosic polymers~~,~~ and preserving cellulose (Kantouch, Hebeish, & El-Rafie, 1970). ~~—~~By replacing 10% sodium hydroxide with ~~organosolve~~organosolv pre-treatment, the first washing step ~~can~~ could be ~~removed~~ completely removed, ~~thus~~ saving 30 L of water ~~or~~ and avoiding the use of strong acid for neutralization. On the other hand, t~~T~~he using of natural acids, such as lime juice, is more ecofriendly and economic~~al that chemical materials since it can protect environment from the impacts caused by chemicals~~. It also reduces the amount of water needed for neutralization and it is~~,~~ safe for the operators ~~and~~ as it presents no health issues. ~~-~~By replacing TEMPO oxidation ~~lime juice hydrolysis with~~ by lime juice hydrolysis ~~TEMPO oxidation~~, the washing step before homogenizing the pulp ~~can~~ could be potentially reduced, saving 30 L of water or avoiding the use of expensive material, such as TEMPO. These results agree with the ones published by Perzon et al, 2019).

### 3.3 Sensitivity analysis

The following aspects have a strong influence on environment: electricity, ethanol~~,~~ and acetic acid cosnumptions. A sensitivity analysis ~~is~~ based on what-if scenarios (Börjeson, Höjer, Dreborg, Ekvall, & Finnveden, 2006) was conducted to assess the~~ment~~ actual influence of each of the mentioned aspects. Results are ~~showed~~ shown in Figure 5.

Regarding the use of acetic acid in the bleaching step, there is a possibility to eliminate ~~using acetic acid~~it, as Trilokesh et al., 2019 indicated in their research, where they applied $NaClO_2$ for the bleaching and ~~they~~ reported that this method resulted in a decrease of the ~~ease to decrease~~ costs~~,~~ and environmental impacts. Furthermore, with the purpose of reducing the environmental impacts, four scenarios were defined without acetic acid (ALH.WAC, ATH.WAC, OTH.WAC, OLH.WAC), and the results ~~illustrate~~ are included in

~~figure~~ Figure 6-a. ~~this~~ This methods have potentially reduced the environmental impacts in four crucial categories including, GWP, climate change, human toxicity, and ~~marin~~marine ecotoxcity.

The influence of the electricity ~~aspect~~ consumption was tested and compared in another four scenarios defined as ALH.E, ATH.E, OTH.E and OLH.E. They were compared with the baseline scenarios and the obtained results are presented in fFigure 6-b ~~another four scenarios were defined (ALH.E, ATH.E, OTH.E OLH.E); the~~ . It could be concluded that~~results showed that~~ the use of electricity from biomass instead of electricity from mazut~~,~~ and natural gas could significantly reduce the environmental impacts. The mentioned change would mean~~It means~~to use renewable energy instead of fossil-origin one ~~converting a fossil-based energy to biomass energy~~ for heat production. This fact had an effect on ~~the~~ environmental impacts, especially on the GWP, climate change, human toxicity, and ~~marin~~marine ecotoxity categories.

~~Recovery~~ The recovery of solvent, in particular the ethanol used in the organosolv process by distillation, is another possibility to optimize the process of CNF production. ~~through distillation of ethanol and water solvent the separation of water and ethanol is possible (Kleinert, 1971).~~ The ethanol recovery by distillation has a minimum amount of environmental impact and energy. Figure 6-c showes~~d~~ the results of two scenarios with the recovery of ethanol (OTH.ER, OLH.ER) and its comparison with the ~~, comparison defined scenarios, and~~ baseline ~~scenarios~~ ones. It could be concluded ~~indicate~~ that the environmental impacts in OTH.ER and OLH.ER are much ~~less~~ lower than the ones of OLH and OTH.

### 3.4 ~~Compare~~ Comparison with other studies

In this part, the LCA results of CNF production are compared to previous studies for CNF and CNC. Many studies ~~are also compared to~~present LCA results of ~~celluloseic~~cellulosic materials with carbon nanotubes, carbon nanofiber~~,~~ and graphene, which ~~they can utilize~~are typically used as reinforcement in composite materials. Alkaline treatment and direct bleaching followed by mechanical treatment, ~~have~~ has previously been used by Berglond et al. The highest environmental impacts were ~~due~~ associated to the energy demand for the alkaline treatment, which caused a higher electricity consumption for the pre-treatment in ~~compare~~ comparison with the~~to~~ direct bleaching. This result indicates that alkaline pre-treatment caused high energy consumption. In another study by Arvidson et al three methods for CNF production ~~have been used~~are presented; enzymatic pretreatment, carboxymethylation~~,~~ and no pre-treatment. The results indicate that CNF production by the carboxymethylation method had the ~~higher~~ highest environmental impacts caused due to the use of solvents. ~~they also showed that the enzymatic and no pre-treatment methods have lower environmental impacts~~. In general, the ~~Overall~~ pre-treatment step has been found as a high energy consumption unit in the CNF production process. In a study by Li et al., the TEMPO oxidation and the homogenization resulted in ~~a~~ lower impacts than the carboxymethylation. In the other study, Turk et al.,

showed that the TEMPO oxidation caused considerable energy consumptions and environmental impact. ~~However, there is some opportunities to employ strategies for reduction of energy, for example, solvents recovery, can potentially reduce the environmental impact~~. ~~Recovery~~ The solvents recovery ~~of solvent can~~ could decrease the energy consumption ~~for~~ of the reaction ~~indirectly~~ (de Figueirêdo et al., 2012). ~~using~~ Furthermore, using better insulation~~,~~ and heat recovery during the production, ~~and~~ as well as obtaining bioenergy from biomass~~burning~~ ~~biomass for generated~~to produce electricity are other opportunities to optimize the reaction process (do Nascimento et al., 2016).

~~Carbon nanofiber is a nanomaterial that can be used as reinforcement in nanocomposites; however, as Berglond et al., showed the GWP was higher than natural nanomaterials (Khanna et al.,2008).~~

Gavankar et al. studied the energy consumption for carbon nanotubes~~,~~. ~~t~~Their results showed that the energy use for carbon nanotubes obtaining was~~is~~ higher than the one ~~energy use~~ of nanomaterialsproducing from biomass. For graphene, Arvidsson et al., and Pizza et al. calculated the energy consumption of graphene~~,~~ and their results indicates that graphene ~~has~~ had high energy demand in ~~compare~~ comparison to bio-based materials. Berglond et al., showed, the GWP was higher than natural nanomaterials (Khanna et al.,2008).

### 3.4 Future studies~~:~~

The results of this study indicated that CNF production has high environmental impacts. The most important recommendation to CNF producers and researchers is to scale up the process. Although there have been many promising achievements at laboratory or pilot scale, there are several challenges to solve in order to be able to produce cellulose-based nanocomposites at the industrial scale. ~~Process of production at lab scale can look different than production at an industrial scale plant, because of the fact that equipment, heat, and energy recovery of an industrial plant are not comparable to lab scale~~. Therefore, for evaluating this kind of assessment at the industrial scale, a theoretical scale-up of the production process seems to be the strategy to head for. The simulation of a process before establishing of an industrial scale unit has a lot of advantages. Identifying energy saving opportunities is a complex problem for the industry because of the existing interconnections between processes units, which is typical at the industrial scale (Wang et al., 2019) (Peduzzi, Boissonnet, Haarlemmer, & Maréchal, 2018). A scale-up framework for nano cellulosic material has been done by Piccinno et al~~.,~~ .. T~~t~~hey reported higher efficiencies of large-scale production in comparison to~~than~~ lab scale (Piccinno et al., 2018b). In the scale up of the process, it ~~can~~ could be possible to apply some strategy to reduce of environmental impacts and energy consumption. For example, using by-products as inputs for other processes~~,~~ in large-scale processes (Katakojwala & Mohan, 2020).

## 4. Conclusion

This present study depicted a sustainable approach ~~with the lowest environmental impacts~~ for a cleaner production of CNF from ST. The optimizing methods performed by solvent recovery, electricity generation from~~based on~~ biomass~~,~~ and ~~omitting~~ avoiding the use of acetic acid ~~to minimize the energy consumption and chemical use that~~ were identified as the main contributors to the environmental impact. During the application of optimizing scenarios, the environmental impact decreased in all impact categories, such as GWP, climate change, ~~marin~~marine ecotoxity, and human health, and the energy consumption, chemical, and water use significantly decreased ~~simultaneously~~. Altogether, the use of ST for CNF production offers a promising way for having eco-efficient process, the main recommendation from this study is using of bio-based electricity for heating, bioethanol instead of ethanol, which have a reduced environmental impact and total energy demand. ~~For future development, it would be an interesting issue to conduct LCA and simulation for identifying energy saving and reduction of environmental impacts opportunities~~.

## 5 References


1. Abraham, E., Deepa, B., Pothan, L., Jacob, M., Thomas, S., Cvelbar, U., & An, ~~andjiwala~~Andjiwala, R. 2011. Extraction of ~~nanocellulose~~nano-cellulose fibrils from lignocellulosic fibres: A novel approach. Carbohydrate Polymers, 86(4): 1468-1475.
2. Abral, H., Soni Satria, R., Mahardika, M., Hafizulhaq, F., Affi, J., Asrofi, M., H, andayani, D., Sapuan, S. M., Stephane, I., & Sugiarti, E. 2019. Comparative study of the physical, and tensile properties of jicama (Pachyrhizus erosus) starch film prepared using three different methods. ***Starch-Stärke***, 71(5-6): 1800224.
3. Alemdar, A., & Sain, M. 2008. Isolation, and characterization of nanofibers from agricultural residues–Wheat straw~~-~~, and soy hulls. Bioresource technology, 99(6): 1664-1671.
4. Arvidsson, R., Kushnir, D., S, andén, B. A., & Mol, ander, S. 2013. How to make policy-relevant life cycle assessments of future products? Lessons learned from nanomaterials. Paper presented at the 6th International Conference on Life Cycle Management, Gothenburg, 25-28 August.
5. Azeredo, H. M., Rosa, M. F., & Mattoso, L. H. C. 2017. ~~Nanocellulose~~Nano-cellulose in bio-based food packaging applications. Industrial Crops, and Products, 97: 664-671.
6. Berglund, L., Noël, M., Aitomäki, Y., Öman, T., & Oksman, K. 2016. Production potential of cellulose nanofibers from industrial residues: Efficiency, and nanofiber characteristics. Industrial Crops, and Products, 92: 84-92.

7. Börjeson, L., Höjer, M., Dreborg, K.-H., Ekvall, T., & Finnveden, G. 2006. Scenario types, and techniques: towards a user's guide. Futures, 38(7): 723-739.

8. Borrega, M., & Orelma, H. 2019. Cellulose Nanofibril (CNF) Films, and Xylan from Hot Water Extracted Birch Kraft Pulps. Applied Sciences, 9(16): 3436.

9. Chaker, A., Mutjé, P., Vilar, M. R., & Boufi, S. 2014. Agriculture crop residues as a source for the production of nanofibrillated cellulose with low energy dem, and. Cellulose, 21(6): 4247-4259.

10. Chen, W., Yu, H., Liu, Y., Chen, P., Zhang, M., & Hai, Y. 2011. Individualization of cellulose nanofibers from wood using high-intensity ultrasonication combined with chemical pretreatments. ***Carbohydrate Polymers***, 83(4): 1804-1811.

11. Chen, Y. W., Lee, H. V., & Abd Hamid, S. B. 2017. Facile production of nanostructured cellulose from Elaeis guineensis empty fruit bunch via one pot oxidative-hydrolysis isolation approach. Carbohydrate polymers, 157: 1511-1524.

12. Clavreul, J., Guyonnet, D., & Christensen, T. H. 2012. Quantifying uncertainty in LCA-modelling of waste management systems. Waste Management, 32(12): 2482-2495.

13. de Figueirêdo, M. C. B., de Freitas Rosa, M., Ugaya, C. M. L., de Souza, M. d. S. M., da Silva Braid, A. C. C., & de Melo, L. F. L. 2012. Life cycle assessment of cellulose nanowhiskers. Journal of Cleaner Production, 35: 130-139.

14. de Oliveira, J. P., Bruni, G. P., Lima, K. O., El Halal, S. L. M., da Rosa, G. S., Dias, A. R. G., & da Rosa Zavareze, E. 2017. Cellulose fibers extracted from rice, and oat husks, and their application in hydrogel. Food chemistry, 221: 153-160.

15. do Nascimento, D. M., Almeida, J. S., Vale, M. d. S., Leitão, R. C., Muniz, C. R., de Figueirêdo, M. C. B., Morais, J. P. S., & Rosa, M. d. F. 2016. A comprehensive approach for obtaining cellulose nanocrystal from coconut fiber. Part I: Proposition of technological pathways. Industrial Crops-, and Products, 93: 66-75.

16. Duan, B., Lu, A., Deng, H., Du, Y., Shi, X., & Zhang, L. 2018. Fabrication of cellulose nanofibers from waste brown algae, and their potential application as milk thickeners. Food Hydrocolloids.

17. EC, 2011. European Commission: International Reference Life Cycle Data system (ILCD) h, andbook- recommendations for life cycle impact assessment in the European context. https://eplca.jrc.ec.europa.eu/uploads/JRC-Reference- Report-ILCD-H, andbook-Towards-more-sustainable-production-, andconsumption-for-a-resource-efficient-Europe.pdf.

18. FAO,2020. Food-, and Agriculture Organization. www.FAO.org

19. Fazeli, M., & Simão, R. A. 2019. Preparation, and characterization of starch composites with cellulose nanofibers obtained by plasma treatment–, and ultrasonication. Plasma Processes, and Polymers: e1800167.

20. Franco, H. C. J., Pimenta, M. T. B., Carvalho, J. L. N., Magalhães, P. S. G., Rossell, C. E. V., Braunbeck, O. A., Vitti, A. C., Kölln, O. T., & Rossi Neto, J. 2013. Assessment of sugarcane trash for agronomic, and energy purposes in Brazil. Scientia Agricola, 70(5): 305-312.

21. Gavankar, S., Suh, S., & Keller, A. A. 2015. The role of scale, and technology maturity in life cycle assessment of emerging technologies: a case study on carbon nanotubes. Journal of Industrial Ecology, 19(1): 51-60.

22. González, I., Boufi, S., Pèlach, M. A., Alcalà, M., Vilaseca, F., & Mutjé, P. 2012. Nanofibrillated cellulose as paper additive in eucalyptus pulps. BioResources, 7(4): 5167-5180.

23. Goyal, S., Dhull, S., & Kapoor, K. 2005. Chemical, and biological changes during composting of different organic wastes, and assessment of compost maturity. Bioresource technology, 96(14): 1584-1591.

24. ISO 14044, 2006. Environmental Management: Life Cycle Assessment: Requirements, and Guidelines.

25. ISO 14045, 2012. Environmental Management, Eco-Efficiency Assessment of Product Systems. Principles, Requirements, and Guidelines.

26. Júnior, M. A. D., Borsoi, C., Hansen, B., & Catto, A. L. 2019. Evaluation of different methods for extraction of ~~nanocellulose~~nano-cellulose from yerba mate residues. Carbohydrate Polymers, 218: 78-86.

27. Jutakanoke, R., Leepipatpiboon, N., Tolieng, V., Kitpreechavanich, V., Srinorakutara, T., & Akaracharanya, A. 2012. Sugarcane leaves: Pretreatment, and ethanol fermentation by Saccharomyces cerevisiae. Biomass, and Bioenergy, 39: 283-289.

28. Kantouch, A., Hebeish, A., & El-Rafie, M. 1970. Action of sodium chlorite on cellulose, and cellulose derivatives. Textile research journal, 40(2): 178-184.

29. Katakojwala, R., & Mohan, S. V. 2020. Microcrystalline cellulose production from sugarcane bagasse: Sustainable process development, and life cycle assessment. Journal of Cleaner Production, 249: 119342.

30. Khan, A., Huq, T., Khan, R. A., Riedl, B., & Lacroix, M. 2014. ~~Nanocellulose~~Nano-cellulose -based composites, and bioactive agents for food packaging. Critical reviews in food science, and nutrition, 54(2): 163-174.

31. Khanna, V., Bakshi, B. R., & Lee, L. J. 2008. Carbon nanofiber production: Life cycle energy consumption, and environmental impact. Journal of Industrial Ecology, 12(3): 394-410.

~~32. Kleinert, T. N. 1971. Organosolv pulping, and recovery process: Google Patents.~~

~~33.~~32. Klemm, D., Cranston, E. D., Fischer, D., Gama, M., Kedzior, S. A., Kralisch, D., Kramer, F., Kondo, T., Lindström, T., & Nietzsche, S. 2018. ~~Nanocellulose~~Nano-cellulose as a natural source for groundbreaking applications in materials science: Today's state. Materials Today, 21(7): 720-748.

~~34.~~33. Li, M., Wang, L.-j., Li, D., Cheng, Y.-L., & Adhikari, B. 2014. Preparation, and characterization of cellulose nanofibers from de-pectinated sugar beet pulp. Carbohydrate Polymers, 102: 136-143.

~~35.~~34. Mahardika, M., Abral, H., Kasim, A., Arief, S., & Asrofi, M. 2018. Production of ~~nanocellulose~~nano-cellulose from pineapple leaf fibers via high-shear homogenization, and ultrasonication. Fibers, 6(2): 28.

~~36.~~35. Minjares-Fuentes, R., Femenia, A., Garau, M., Meza-Velázquez, J., Simal, S., & Rosselló, C. 2014. Ultrasound-assisted extraction of pectins from grape pomace using citric acid: a response surface methodology approach. Carbohydrate polymers, 106: 179-189.

~~37.~~36. Muhamad, M., Hornsby, P., Carmichael, E., Zakaria, M., Seok, Y. B., Mohamed, S., & Sharma, S. 2019. Characterisation of Cellulose Nanofibres Derived from Chemical, and Mechanical Treatments. Paper presented at the MATEC Web of Conferences.

~~38.~~37. Nagarajan, K., Balaji, A., & Ramanujam, N. 2019. Extraction of cellulose nanofibers from cocos nucifera var aurantiaca peduncle by ball milling combined with chemical treatment. Carbohydrate polymers, 212: 312-322.

~~39.~~38. Oliveira, T. Í. S., Rosa, M. F., Cavalcante, F. L., Pereira, P. H. F., Moates, G. K., Wellner, N., Mazzetto, S. E., Waldron, K. W., & Azeredo, H. M. 2016. Optimization of pectin extraction

from banana peels with citric acid by using response surface methodology. Food Chemistry, 198: 113-118.

~~40.~~39. Parsaee, M., Kiani, M. K. D., & Karimi, K. 2019. A review of biogas production from sugarcane vinasse. Biomass, and bioenergy, 122: 117-125.

~~41.~~40. Pathak, P., Gupta, A., Bhardwaj, N. K., Goyal, A., & Moholkar, V. S. 2020. Impact of mild , and harsh conditions of formic acid-based organosolv pretreatment on biomass fractionation of sugarcane tops. Biomass Conversion, and Biorefinery: 1-14.

~~42.~~41. Pianosi, F., Beven, K., Freer, J., Hall, J. W., Rougier, J., Stephenson, D. B., & Wagener, T. 2016. Sensitivity analysis of environmental models: A systematic review with practical workflow. Environmental Modelling & Software, 79: 214-232.

~~43.~~42. Peduzzi, E., Boissonnet, G., Haarlemmer, G., & Maréchal, F. 2018. Thermo-economic analysis, and multi-objective optimisation of lignocellulosic biomass conversion to Fischer–Tropsch fuels. Sustainable Energy & Fuels, 2(5): 1069-1084.

~~44.~~43. Pianosi, F., Beven, K., Freer, J., Hall, J. W., Rougier, J., Stephenson, D. B., & Wagener, T. 2016. Sensitivity analysis of environmental models: A systematic review with practical workflow. Environmental Modelling & Software, 79: 214-232.

~~45.~~44. Piccinno, F., Hischier, R., Seeger, S., & Som, C. 2015. Life cycle assessment of a new technology to extract, functionalize, and orient cellulose nanofibers from food waste. ACS Sustainable Chemistry & Engineering, 3(6): 1047-1055.

~~46.~~45. Piccinno, F., Hischier, R., Seeger, S., & Som, C. 2016. From laboratory to industrial scale: a scale-up framework for chemical processes in life cycle assessment studies. Journal of Cleaner Production, 135: 1085-1097.

~~47.~~46. Pinto, L. O., Bernardes, J. S., & Rezende, C. A. 2019. Low-energy preparation of cellulose nanofibers from sugarcane bagasse by modulating the surface charge density. Carbohydrate Polymers, 218: 145-153.

~~48.~~47. Pizza, A., Metz, R., Hassanzadeh, M., & Bantignies, J.-L. 2014. Life cycle assessment of nanocomposites made of thermally conductive graphite nanoplatelets. The International Journal of Life Cycle Assessment, 19(6): 1226-1237.

~~49.~~48. Qua, E., Hornsby, P., Sharma, H., & Lyons, G. 2011. Preparation, and characterisation of cellulose nanofibres. Journal of Materials Science, 46(18): 6029-6045.

~~50.~~49. Ravindran, L., Sreekala, M., & Thomas, S. 2019. Novel processing parameters for the extraction of cellulose nanofibers (CNF) from environmentally benign pineapple leaf fibers (PALF): Structure-property relationships. International journal of biological macromolecules, 131: 858-870.

~~51.~~50. Sain, M., & Panthapulakkal, S. 2006. Bioprocess preparation of wheat straw fibers, and their characterization. Industrial Crops, and Products, 23(1): 1-8.

~~52.~~51. Segal, L., Creely, J. J., Martin Jr, A., & Conrad, C. 1959. An empirical method for estimating the degree of crystallinity of native cellulose using the X-ray diffractometer. Textile research journal, 29(10): 786-794.

~~53.~~52. Sheldon, R. A. 2014. Green, and sustainable manufacture of chemicals from biomass: state of the art. Green Chemistry, 16(3): 950-963.

~~54.~~53. Sluiter, A., Hames, B., Ruiz, R., Scarlata, C., Sluiter, J., Templeton, D., & Crocker, D. 2008. Determination of structural carbohydrates, and lignin in biomass. Laboratory analytical procedure, 1617: 1-16.

~~55.~~54. Sofla, M. R. K., Batchelor, W., Kosinkova, J., Pepper, R., Brown, R., & Rainey, T. 2019. Cellulose nanofibers from bagasse using a high-speed blender, and acetylation as a pretreatment. Cellulose, 26(8): 4799-4814.

~~56.~~55. Song, Y., Chen, W., Niu, X., Fang, G., Min, H., & Pan, H. 2018. An Energy-Efficient One-Pot Swelling/Esterification Method to Prepare Cellulose Nanofibers with Uniform Diameter. ChemSusChem, 11(21): 3714-3718.

~~57.~~56. St, andard, T. 2007. T 211 om-07. Ash in wood, pulp, paper, and paperboard: combustion at 525 C. Tappi St, andard.

~~58.~~57. St, andard, T. 2007. T 211 om-07. Ash in wood, pulp, paper, and paperboard: combustion at 525 C. Tappi St, andard.

~~59.~~58. Sun, X.-F., Sun, R.-C., Su, Y., & Sun, J.-X. 2004. Comparative study of crude, and purified cellulose from wheat straw. Journal of agricultural, and food chemistry, 52(4): 839-847.

~~60.~~59. Tappi, T. 2004. 204 cm-97, Solvent extractives of wood, and pulp. TAPPI test methods, 2005.

~~61.~~60. Teramoto, Y., Lee, S.-H., & Endo, T. 2008. Pretreatment of woody, and herbaceous biomass for enzymatic saccharification using sulfuric acid-free ethanol cooking. Bioresource Technology, 99(18): 8856-8863.

~~62.~~61. Trilokesh, C., & Uppuluri, K. B. 2019. Isolation, and characterization of cellulose nanocrystals from jackfruit peel. Scientific reports, 9(1): 1-8.

~~63.~~62. Uetani, K., & Yano, H. 2010. Nanofibrillation of wood pulp using a high-speed blender. Biomacromolecules, 12(2): 348-353.

~~64.~~63. Uetani, K., & Yano, H. 2010. Nanofibrillation of wood pulp using a high-speed blender. Biomacromolecules, 12(2): 348-353.

~~65.~~64. Van Aken, K., Strekowski, L., & Patiny, L. 2006. EcoScale, a semi-quantitative tool to select an organic preparation based on economical, and ecological parameters. Beilstein Journal of Organic Chemistry, 2(1): 3.

~~66.~~65. Vriesmann, L. C., Teófilo, R. F., & de Oliveira Petkowicz, C. L. 2012. Extraction, and characterization of pectin from cacao pod husks (Theobroma cacao L.) with citric acid. LWT-Food Science, and Technology, 49(1): 108-116.

~~67.~~66. Yadav, R., Prasad, S., Singh, R., & Srivastava, V. 1994. Recycling sugarcane trash to conserve soil organic carbon for sustaining yields of successive ratoon crops in sugarcane. Bioresource technology, 49(3): 231-235.

~~68.~~67. Yan, X. 2012. Bioethanol, and biodiesel as alternative transportation fuels in China: Current status, future potentials, and life cycle analysis. Energy Sources, Part A: Recovery, Utilization, and Environmental Effects, 34(12): 1067-1075.

~~69.~~68. Zhai, L., Kim, H. C., Kim, J. W., Choi, E. S., & Kim, J. 2018. Cellulose nanofibers isolated by TEMPO-oxidation, and aqueous counter collision methods. Carbohydrate polymers, 191: 65-70.

**Table 1**. Input in terms of materials, chemicals, water ~~consumption, ,~~ and energy consumptions for four designed methods.

| | Units | ATH | ALH | OTH | OLH |
|---|---|---|---|---|---|
| ST residue | g | 20.0 | 20.0 | 20.0 | 20.0 |
| Yield | % | 0.30 | 0.30 | 0.34 | 0.34 |
| Sodium hydroxide | g | 10.0 | 10.0 | - | - |
| Sodium bromide | g | 0.16 | - | 0.17 | - |
| Sodium hypochlorite | g | 3.90 | - | 4.08 | - |
| Sodium chlorite | g | 2.00 | 2.00 | 2.00 | 2.00 |
| TEMPO | g | 0.02 | - | 0.02 | - |
| Acetic acid | L | 25.0 | 25.0 | 25.0 | 25.0 |
| Lime juice | L | - | 0.08 | - | 0.08 |
| Ethanol | L | - | - | 0.20 | 0.20 |
| Water | L | 60.0 | 60.0 | 30.0 | 30.0 |
| Energy | MJ | 0.06 | 0.07 | 0.15 | 0.10 |

**Table 2.** Analysis of moisture, ash, lignin ~~,~~ and carbohydrates

| | This study % |
|---|---|
| Moisture | 7.53 |
| Ash | 9.01 |
| Extractive | |
| Extractive in water | 0.92 |
| Extractive in ethanol | 0.34 |
| Lignin | |
| Soluble lignin | 0.04 |
| Insoluble lignin | 21.2~~16~~ |
| Carbohydrates | |
| Glucose | 34.6~~58~~ |
| Acetate | 1.03 |
| Arabinose | 1.72 |
| Xylan | 19.3 |

**Table 3.** Q~~The q~~uantitative yields for all scenarios

| ~~Method~~Scenario | Cellulose production | NFC production (g/g cellulose ) | NFC production (g/g ST) |
|---|---|---|---|
| OTH~~AHT~~ | 0.34 ± 0.06 ~~0.30 ± 0.02~~ | 0.93 ± 0.01~~0.90 ± 0.01~~ | 0.31± 0.02~~0.27± 0.02~~ |
| OLH~~ALH~~ | 0.34 ± 0.06 ~~0.30 ± 0.02~~ | 0.96 ± 0.01~~0.89 ± 0.01~~ | 0.32± 0.02~~0.26± 0.02~~ |
| ATH~~OHT~~ | 0.30 ± 0.02 ~~0.34 ± 0.06~~ | 0.90 ± 0.01~~0.93 ± 0.01~~ | 0.27± 0.02~~0.31± 0.02~~ |
| ALH~~OLH~~ | 0.30 ± 0.02 ~~0.34 ± 0.06~~ | 0.89 ± 0.01~~0.96 ± 0.01~~ | 0.26± 0.02~~0.32± 0.02~~ |

**Table 4.** EcoScale score for four CNF production methods.

| Parameters | Penalty points | | | |
|---|---|---|---|---|
| | OTH~~ATH~~ | OLH~~ALH~~ | ATH~~OTH~~ | ALH~~OLH~~ |
| Yield | 9~~9~~ | 16~~16~~ | 9~~9~~ | 16~~16~~ |
| Cost | -~~-~~ | -~~-~~ | -~~-~~ | -~~-~~ |
| Temperature/time | 4~~3~~ | 4~~3~~ | 3~~4~~ | 3~~4~~ |
| Safety | -~~10~~ | -~~10~~ | 10~~-~~ | 10~~-~~ |
| Technical setup | 4~~1~~ | 4~~1~~ | 1~~4~~ | 1~~4~~ |
| Workup ~~,~~and purification | 2~~3~~ | 2~~3~~ | 3~~2~~ | 3~~2~~ |
| EcoScale score | 82~~74~~ | 74~~67~~ | 74~~82~~ | 67~~74~~ |

**Table 5.** R~~The r~~esults of LCIA (Midpoint method, Endpoint method ~~,~~and IPCC 2013 GWP 100a/V1.03 method)

| **Impact category (Endpoint Method ~~,~~and midpoint)** | **Unit** | **OTH** | **OLH** | **ATH** | **ALH** |
|---|---|---|---|---|---|
| Climate change | kg $CO_2$ eq. | 7.89 | 9.96 | 4.08 | 2.85 |
| Terrestrial acidification | kg $SO_2$ eq. | 0.07 | 0.09 | 0.03 | 0.02 |
| Human toxicity | kg 1,4-DB eq. | 61.37 | 85.23 | 41.44 | 23.52 |
| Freshwater ecotoxicity | kg 1,4-DB eq. | 0.03 | 0.04 | 0.01 | 0.01 |
| ~~Marin~~Marine ecotoxicity | kg 1,4-DB eq. | 18.66 | 24.87 | 13.40 | 8.90 |
| Ionising radiation | kBq U235 eq. | 0.36 | 0.45 | 0.25 | 0.23 |
| Metal depletion | kg Fe eq. | 0.40 | 0.48 | 0.15 | 0.15 |
| Fossil depletion | kg oil eq. | 3.42 | 4.56 | 2.13 | 1.64 |
| Impact category (IPCC 2013 GWP, 100a Method) | | | | | |
| GWP | kg $CO_2$ eq | 8.92 | 11.2 | 4.17 | 3.17 |

**Table 6.** Share of each inputs on human health, ecosystem, and resources

| Input | | ATH | | | ALH | | | OTH | | | OLH | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Unit | Human health | Ecosystem | Recourses | Human health | Ecosystem | Recourses | Human health | Ecosystem | Recourses | Human health | Ecosystem | Recourses |
| Acetic acid | % | 56.55 | 51.54 | 52.40 | 68.3 | 71.2 | 79.9 | 25.40 | 27.51 | 12.43 | 31.24 | 33.08 | 16.61 |
| Chemicals inorganic | % | 8.98 | 12.18 | 8.87 | 16.2 | 12.0 | 7.9 | 0.49 | 0.80 | 0.26 | 0.41 | 0.65 | 0.23 |
| Sodium hydroxide | % | 1.61 | 2.18 | 1.90 | 2.3 | 2.1 | 1.0 | – | – | – | – | – | – |
| Electricity | % | 1.30 | 1.61 | 1.47 | 13.2 | 14.7 | 11.2 | 18.26 | 22.35 | 10.81 | 22.46 | 26.87 | 14.45 |
| Chemicals organic | % | 31.55 | 32.49 | 35.37 | – | – | – | 0.52 | 0.76 | 0.31 | - | - | - |
| Ethanol | % | - | - | - | – | – | – | 55.33 | 48.58 | 76.20 | 45.90 | 39.40 | 68.71 |
| Total | % | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 |

| Input | | OTH | | | OLH | | | ATH | | | ALH | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Unit | Human health | Ecosystem | Recourses | Human health | Ecosystem | Recourses | Human health | Ecosystem | Recourses | Human health | Ecosystem | Recourses |
| Acetic acid | % | 25.4 | 27.5 | 12.4 | 31.2 | 33.1 | 16.6 | 56.5 | 51.5 | 52.4 | 68.3 | 71.2 | 79.9 |
| Chemicals inorganic | % | 0.49 | 0.80 | 0.26 | 0.41 | 0.65 | 0.23 | 8.98 | 12.2 | 8.87 | 16.2 | 12.0 | 7.90 |
| Sodium hydroxide | % | - | - | - | - | - | - | 1.61 | 2.18 | 1.90 | 2.30 | 2.10 | 1.01 |
| Electricity | % | 18.3 | 22.3 | 10.8 | 22.5 | 26.9 | 14.4 | 1.30 | 1.61 | 1.47 | 13.2 | 14.7 | 11.2 |
| Chemicals organic | % | 0.52 | 0.76 | 0.31 | - | - | - | 31.5 | 32.5 | 35.4 | - | - | - |
| Ethanol | % | 55.3 | 48.6 | 76.2 | 45.9 | 39.4 | 68.7 | - | - | - | - | - | - |
| Total | % | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 |

**Table 7.** Share of each chemicals, and energy on GWP

| Input | GWP, 100a (kg $CO_2$ eq) | | | |
|---|---|---|---|---|
| | ~~ATH~~OTH | ~~ALH~~OLH | ~~OTH~~ATH | ~~OLH~~ALH |
| Acetic acid | 2.362.09 | 3.52.09 | 2.092.36 | 2.093.5 |
| Chemicals inorganic | 0.060.46 | 0.060.46 | 0.460.06 | 0.460.06 |
| Sodium hydroxide | -0.09 | -0.07 | 0.09- | 0.07- |
| Electricity | 2.531.74 | 3.750.57 | 1.742.53 | 0.573.75 |
| Chemicals organic | 0.070.07 | -- | 0.070.07 | -- |
| Ethanol | 3.9- | 3.9- | -3.9 | -3.9 |
| Total | 8.931.47 | 11.23.17 | 1.478.93 | 3.1711.2 |

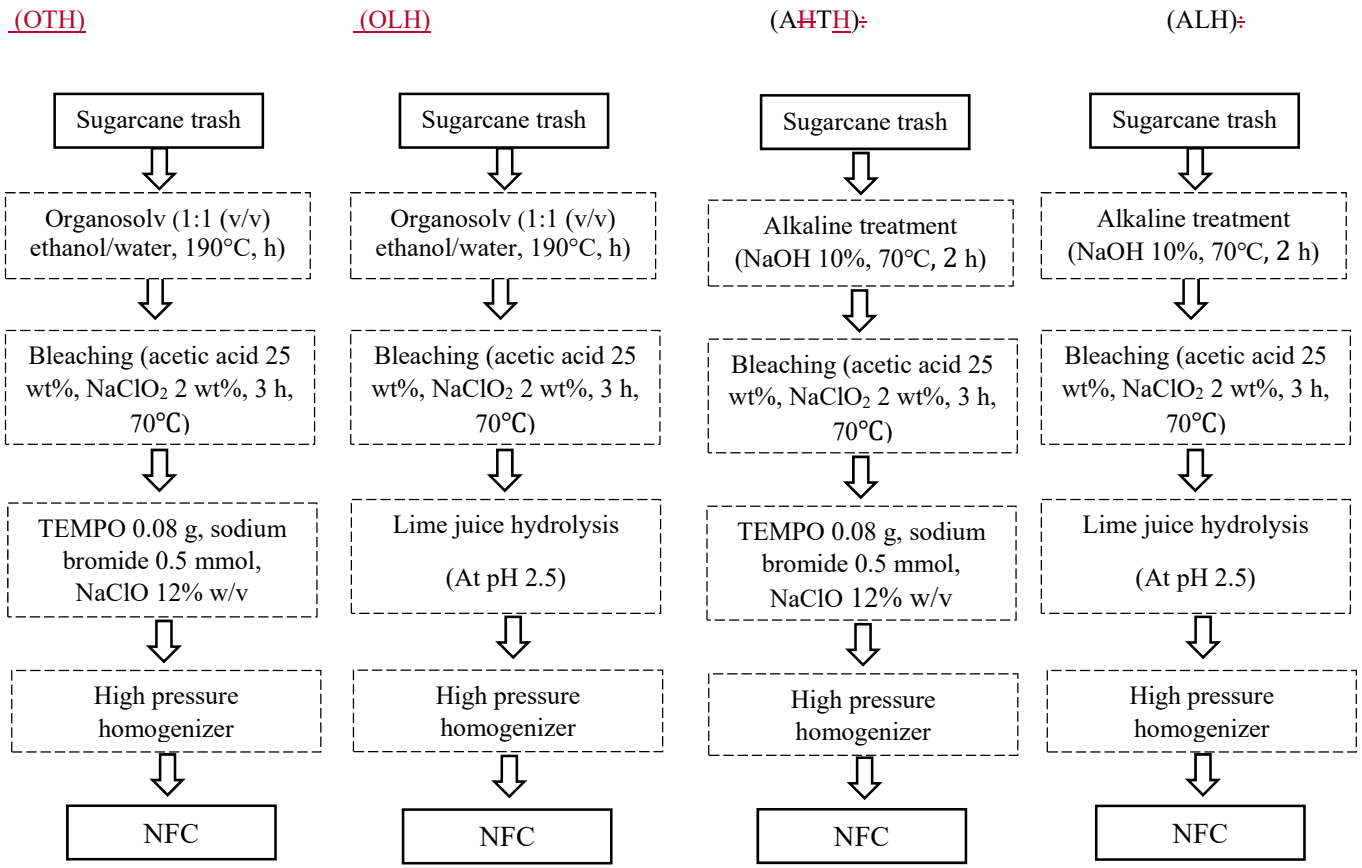



**Figure 1.** SThe summarize of the four baseline scenarios

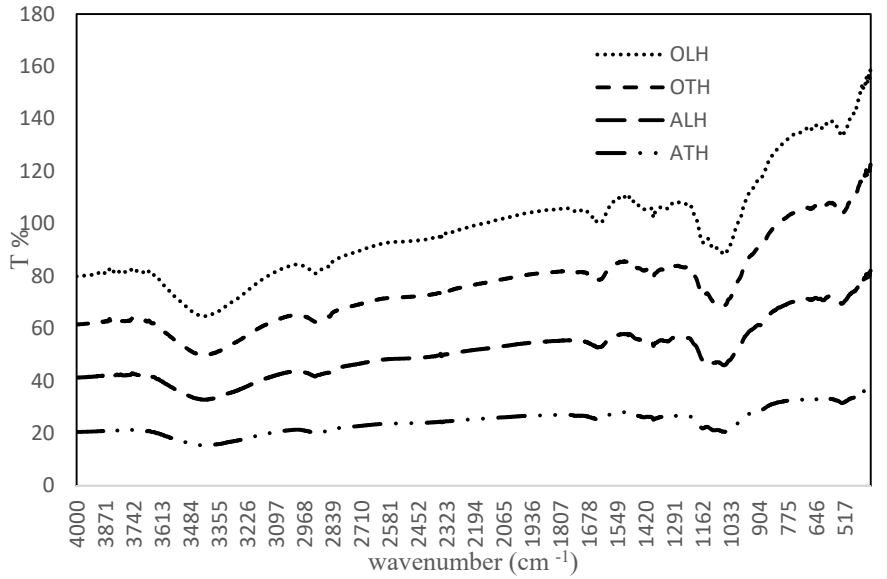



**Figure. 2.** FT-IR spectrum of the different Scenariosmethods

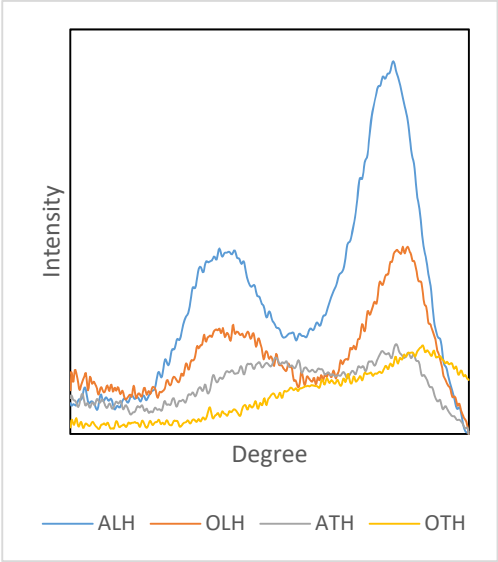
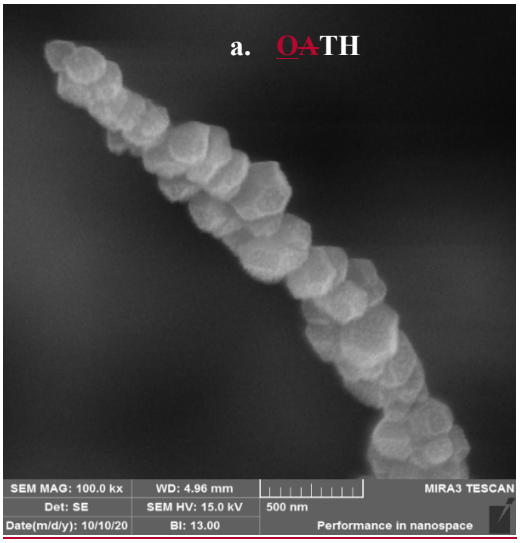
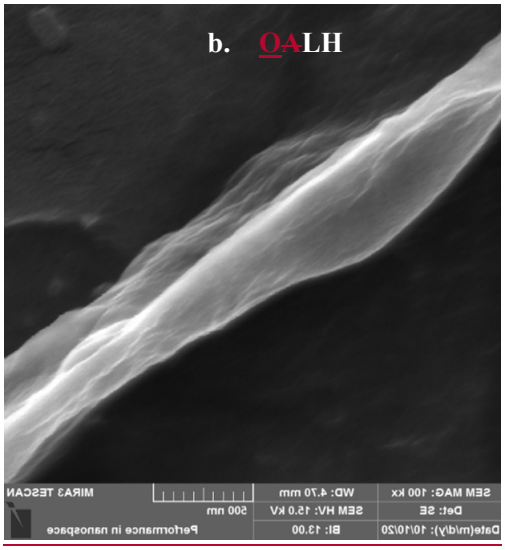
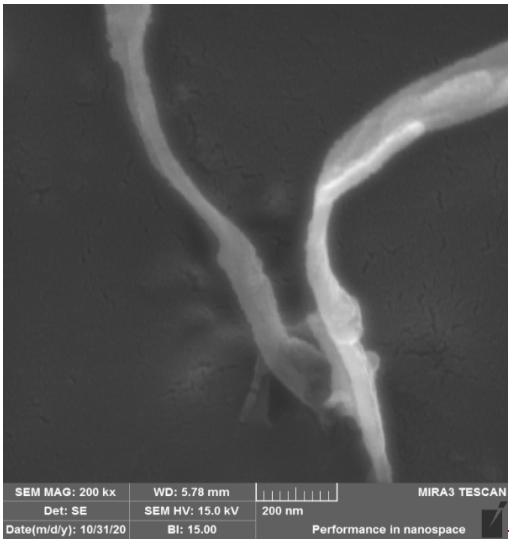



**Figure 3.** X-ray diffraction patterns of CNF samples


a. OATH
SEM MAG: 100.0 kx
WD: 4.96 mm
MIRA3 TESCAN
Det: SE
SEM HV: 15.0 kV
500 nm
Date(m/d/y): 10/10/20
BI: 13.00
Performance in nanospace


b. OALH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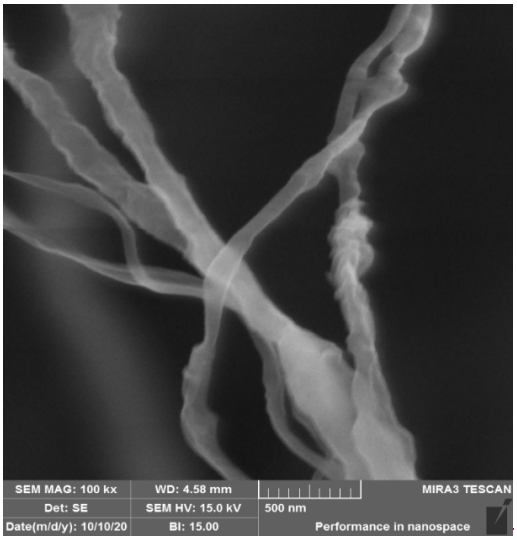
WD: 5.78 mm
MIRA3 TESCAN
Det: SE
SEM HV: 15.0 kV
200 nm
Date(m/d/y): 10/31/20
BI: 15.00
Performance in nanospace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WD: 4.58 mm
MIRA3 TESCAN
Det: SE
SEM HV: 15.0 kV
500 nm
Date(m/d/y): 10/10/20
BI: 15.00
Performance in nanospace

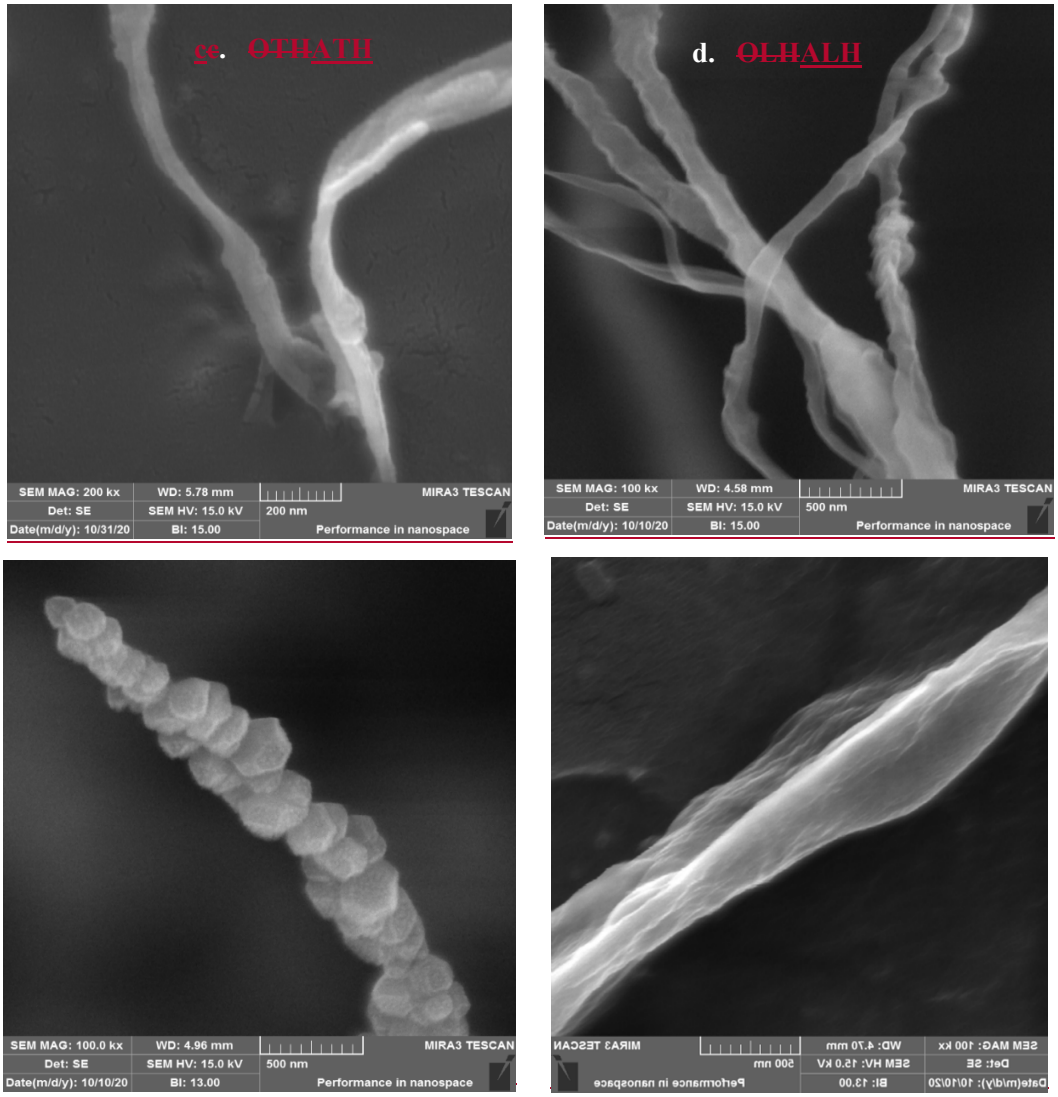



**Figure 4.** Morphology of CNF for all scenarios (SEM results)

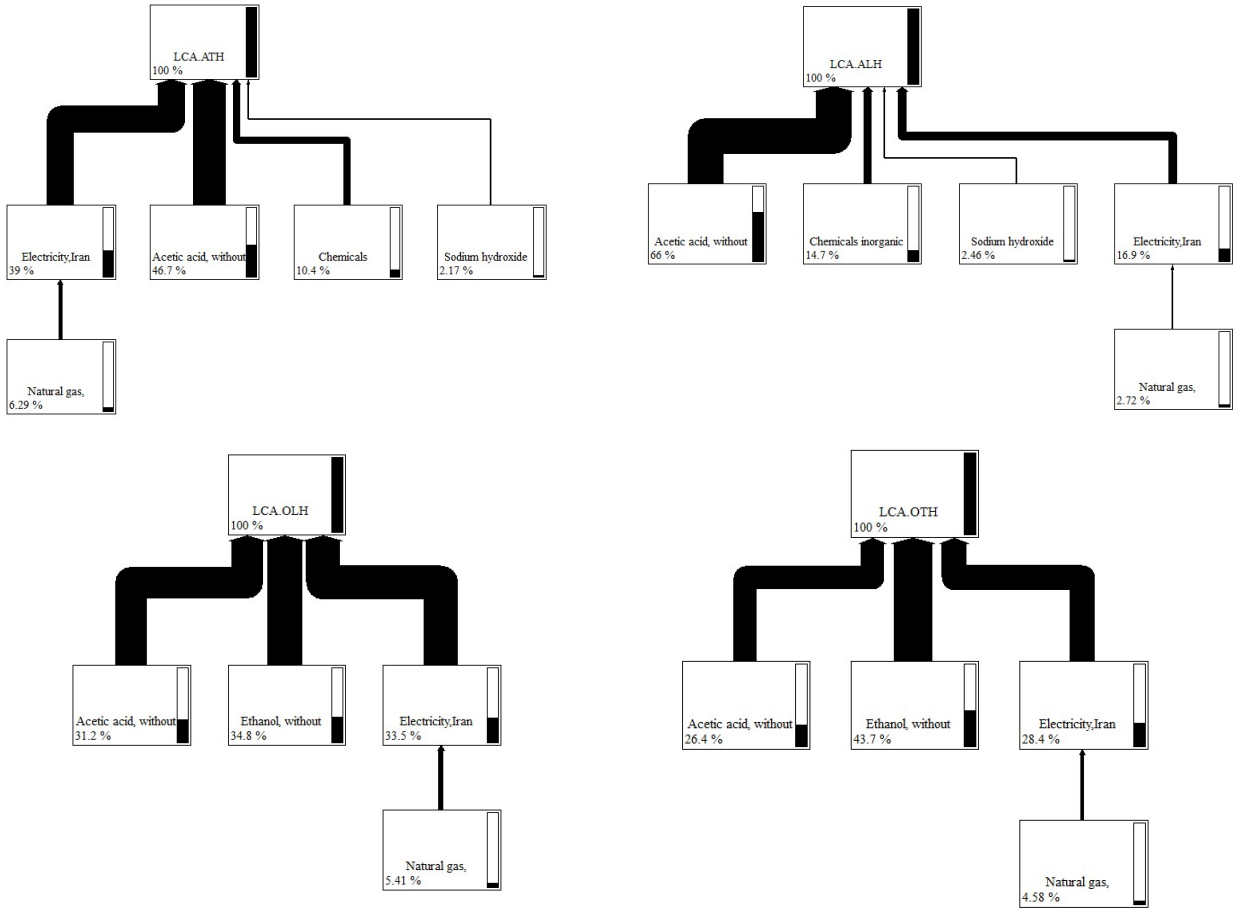
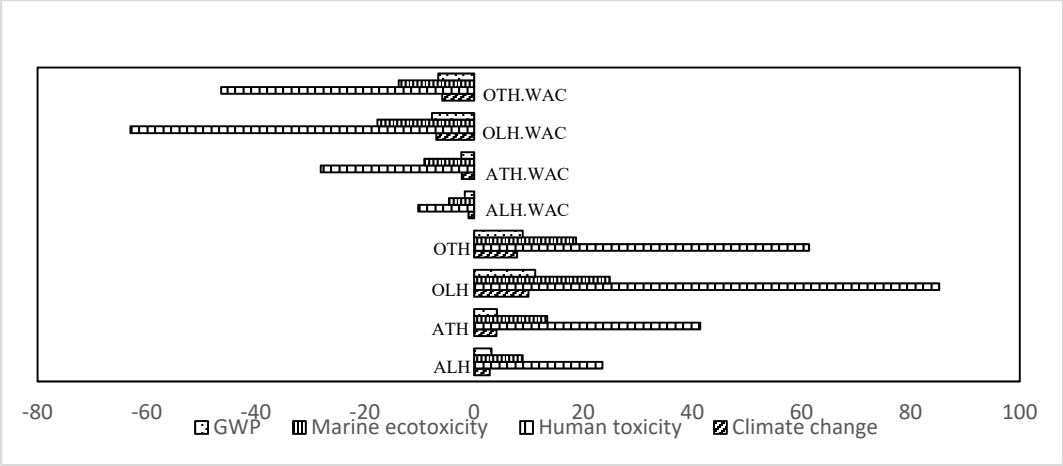



**Figure 5.** Snaky diagram for GWP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OLH.WAC
ATH.WAC
ALH.WAC
OTH
OLH
ATH
ALH
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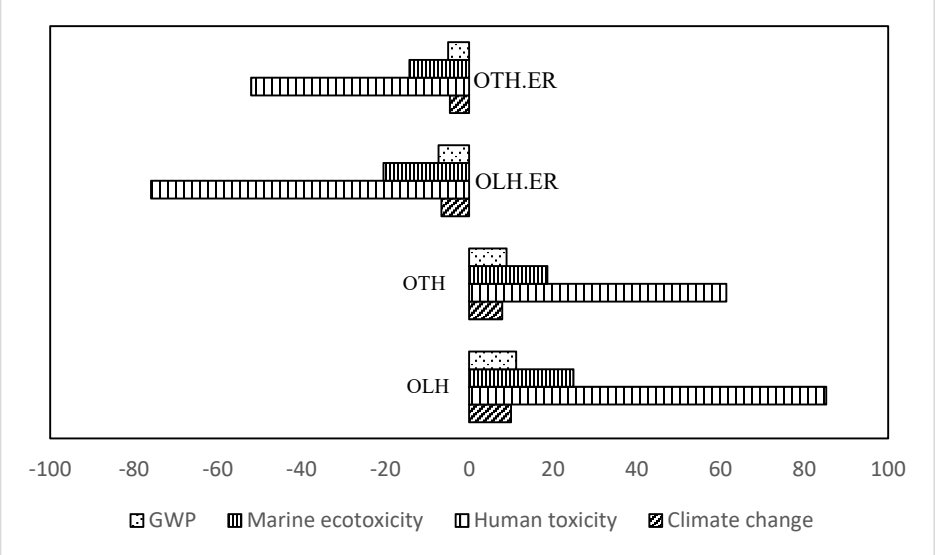
-60
-40
-20
0
20
40
60
80
100
GWP
Marine ecotoxicity
Human toxicity
Climate change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OLH.ER
OTH
OLH
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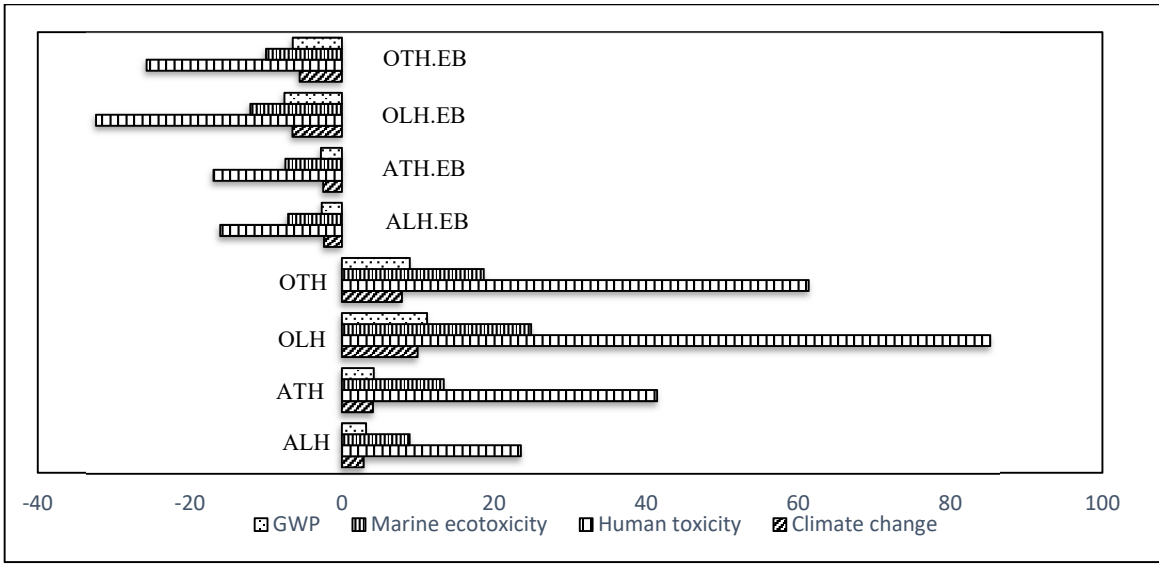
-40
-20
0
20
40
60
80
100
GWP
Marine ecotoxicity
Human toxicity
Climate change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OLH.EB
ATH.EB
ALH.EB
OTH
OLH
ATH
ALH
-40
-20
0
20
40
60
80
100
GWP
Marine ecotoxicity
Human toxicity
Climate change

**Figure 6.** a) R~~The r~~esults of sensitivity analysis by removing acetic acid, b) sensitivity analysis by recovery of ethanol, c) sensitivity analysis by electricity obtaining from biomass.