Bioresource Technology 377 (2023) 128955

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech

Techno-economic analysis and life cycle assessment of cellulose nanocrystals production from wood pulp

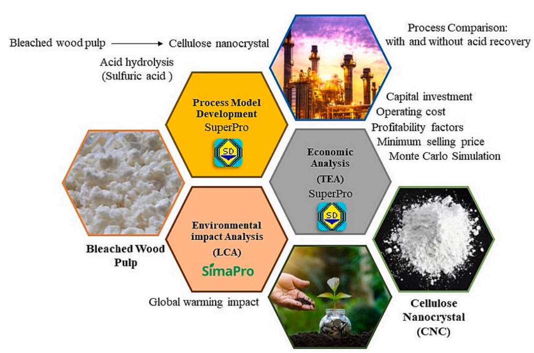
Naveenkumar Rajendran [a,b,*], Troy Runge [a], Richard D. Bergman [b], Prakash Nepal [b], Carl Houtman [b]

[a] *Biological Systems Engineering, University of Wisconsin, Madison, WI 53706, USA*
[b] *Forest Products Laboratory, USDA Forest Service, Madison, WI 53726, USA*

HIGHLIGHTS

- CNC production on a commercial scale from wood pulp was explored for two scenarios.
- Acid recovery process showed a better economic performance.
- The MSP of 1 kg of CNC with acid recovery was $4.69.
- Acid recovery process required more steam which increased fossil GHGs.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Wood pulp
Cellulose nanocrystal
Techno-economic analysis
Monto Carlo simulation
Environmental impacts

ABSTRACT

Cellulose nanocrystals (CNCs) are biobased materials with many desirable properties such as high aspect ratio, mechanical strength, crystalline nature, and biodegradability. This study developed a commercial-scale process model of CNC production from wood pulp using sulfuric acid treatment and evaluated its techno-economic and environmental performance with and without acid recovery. The results indicated that CNC produced with acid recovery process was financially more profitable with higher project net present values than without acid recovery process but required higher capital which resulted in a longer payback period and lower return on invested capital. The estimated minimum selling prices of CNC produced with and without acid recovery were $4.69/kg and $4.89/kg, respectively. The global warming (GW) impacts of 1 kg CNC production with and without acid recovery were 11.39 and 11.18 kg CO_2 eq, respectively, showing that higher steam use and consequently more fossil fuels were needed in the acid recovery process.

* Corresponding author at: Biological Systems Engineering, University of Wisconsin, Madison, WI 53706, USA.
E-mail address: nrajendiran@wisc.edu (N. Rajendran).

https://doi.org/10.1016/j.biortech.2023.128955
Received 30 January 2023; Received in revised form 17 March 2023; Accepted 21 March 2023
Available online 23 March 2023

1. Introduction

Increasing the utilization of renewable biobased products and processes across all economic sectors is recognized as a key strategy for sustainable economic, social, and environmental development. Bio-based products are synthesized from renewable biomass using physical, chemical, or biological techniques (Jakes et al., 2016; Van Schoubroeck et al., 2018). Bio-based materials and chemicals are known to substantially reduce the environmental impacts compared to their non-renewable counterparts while becoming relatively competitive on costs over time (Sahoo et al., 2019; Sahoo et al., 2021). In recent years, bio-based nanomaterials have gathered considerable interests due to their eco-friendliness and advanced physical and thermal properties (Chausali et al., 2022).

Cellulosic nanomaterials (CNMs) are considered a sustainable material of choice for the 21st century because they are widely available, renewable, and have many favourable properties allowing their usage in a wide range of applications including health care, biomedical, food industries, biopolymer production, film and package materials, construction, 3D printing, fuel cell application, chemicals industries, membrane and more (Wegner et al., 2013). CNMs have received significant interest from research, academia, and industry due to their unique structural features and physicochemical properties including biodegradability, renewability, biocompatibility, high aspect ratio, surface area, mechanical strength, and crystalline nature (Moon et al., 2016; Trache et al., 2017). Two types of CNMs are distinguished based on their shape, size, and methods by which they are extracted. While cellulose nanocrystals (CNCs) are produced from acid hydrolysis, and consist of rod-like crystalline particles that are 20–100 nm wide, >1 μm long, and have an aspect ratio of 5–30 (Foster et al., 2018), the cellulose nanofiber (CNFs) are produced using mechanical process and consists of fibril like particle types that are 4–100 nm wide, longer than 1000 nm in length, and has aspect ratio of 10–100 (Moon et al., 2016). Different methods are used for CNC production such as acid hydrolysis, ionic liquid-mediated method (Abushammala et al., 2015), deep eutectic solvent method (Sirviö et al., 2016), enzymatic hydrolysis, TEMPO-mediated oxidation (Sacui et al., 2014) and oxidation degradation (Xie et al., 2018). Acid hydrolysis using sulfuric acid and hydrochloric acid is a commonly used method to produce high-purity CNC (Beyene et al., 2018; Shankaran, 2018). The global cellulose nanomaterial production has increased considerably over the last few years, with an estimated nanocellulose market of more than $430 million in 2022, which is forecasted to grow at a compound annual growth rate of 24.7% from 2022 to 2028 (Stratistics Market Research Consulting, 2022). Currently, there are more than 150 companies in operation worldwide that produce CNC, CNF and bacterial nanocellulose (BNC); the largest among those being CNC (21), followed by CNF (125) and BNC (20) (Future Markets, 2023).

Wood biomass is a sustainable and economically feasible source for CNCs production and has been proven by many patents and research articles (Trache et al., 2017). CNCs are promising materials with a potential to use in a variety of applications as additives and composite materials, although the high cost of CNCs limits their application. The current commercial-level production process of CNCs utilizes acid hydrolysis technology, which requires high amounts of acid and water, produces large amounts of waste, and thus increases production costs and environmental impacts (Dong et al., 2016). Over the past few decades, numerous scientific articles have been published that focus primarily on production pathways, the characterization of CNC, and potential applications. However, very few studies have focused on the economic and environmental impacts of CNC production. An improved understanding of the economics and environmental impacts, and identification of a more sustainable and eco-friendly process of CNCs production through techno-economic analysis (TEA) and life-cycle analysis (LCA) would help in widespread commercialization (de Assis et al., 2017).

TEA is a tool to determine the financial and technical feasibility of a production process or technology by identifying various measures of profitability of the entire production process and the minimum selling prices (MSPs) of the products (Rajendran et al., 2022). de Assis et al. (2017) performed a TEA of CNC production from bleached wood pulp using sulfuric acid hydrolysis and estimated that an MSP of CNC produced from such a process was lower than $7.20/kg. They also reported that the process with acid recovery increased the MSP of CNC and that the feedstock cost and capital investment played a major role in the economics of CNC production (de Assis et al., 2017). Rosales-Calderon et al. (2021) performed an economic analysis of CNC production from bleached eucalyptus kraft pulp using acid and enzymatic hydrolysis. Their estimated MSP of CNC by acid hydrolysis and enzymatic hydrolysis were $10.03/kg and $65.74/kg, respectively. The main reason for their estimated higher MSP for enzymatic hydrolysis was the lower yield resulting from this process (Rosales-Calderon et al., 2021). These studies suggest that capital investment, operating cost and MSP of CNC production depend primarily on the hydrolysis technique, process flow, and feedstock.

LCA, commonly referred to as life-cycle assessment is a scientific method to estimate the life-cycle environmental impacts of a product, a process or a technology resulting from the raw material extraction to its transportation, further processing/manufacture, use, and final disposal (ISO, 2006a, 2006b). Zhang et al. (2022) performed an LCA of CNC production from cotton using different filtration processes. They found that the global warming (GW) impact of CNC produced from cotton pulp would range from 29.6 to 49.4 kg CO_2 eq/kg of CNC depending on the filtration process. Their experiment indicated that acid recovery would help to reduce acid consumption and GW impact (Zhang et al., 2022). Gu et al. (2015) performed LCA of CNC production from wood pulp-based pilot plant production data and found GW impact of 29.6 kg CO_2 eq/kg of CNC (Gu et al., 2015). These studies indicated that the usage of a larger number of chemicals and energy during the CNC production process would proportionately increase the environmental impacts.

This study focused on understanding the financial feasibility and life-cycle environmental impacts of a commercial scale CNC production from wood pulp by performing TEA and LCA. Process models of an industrial scale CNC production from wood pulp utilizing acid hydrolysis technique, with and without acid recovery, were developed to identify their economic costs and environmental impacts, followed by sensitivity analysis to understand the major factors and/or variables affecting the results and to identify and discuss ways to reduce the cost and the life-cycle environmental burden of the production process.

2. Materials and methodology

2.1. Techno-economic analysis

2.1.1. Process description

The process used to produce CNC from wood pulp involved the acid hydrolysis technique. The process was developed based on the SuperPro Designer software version 12 (Intelligen Inc., 2020) coupled with the information available from published peer-reviewed research articles based on a commercial scale production (de Assis et al., 2017). The base manufacturing plant capacity was assumed to be 100 MT/day of CNC with annual operating hours of 7,920. Similarly, the assumed plant construction period, start-up period and lifetime were 12 months, 4 months, and 20 years respectively (Naveenkumar and Baskar, 2021; Rajendran and Han, 2022a,b). In this study, the acid hydrolysis process was used for CNC production from wood pulp. Sulfuric acid (H_2SO_4) was used in hydrolysis due to its low cost, high yield, and stability. The process was analysed in two ways: with and without acid recovery.

2.1.2. Economic description

The base analysis assumed a 21% tax rate, based on the current

United States corporate income tax rate (IRS, 2017). The currency type was the US dollar ($), and the reference year was 2022. The discount rate assumed for the base analysis was 10%. The cost of equipment was calculated using SuperPro built-in cost model (Intelligen Inc., 2020), which was the basis for calculating capital investment (Rajendran and Han, 2022a,b). The equipment cost details are shown in supplementary materials. The capital investment includes equipment costs, plant construction and miscellaneous costs (see supplementary materials). The operating cost includes the cost of raw materials, labour, facility dependent, quality control and assurance, utilities, and miscellaneous. The facility-dependent cost includes the costs associated with equipment maintenance, depreciation, and local tax (Naveenkumar and Baskar, 2020). The raw materials costs were obtained from literature, local markets, and trading websites (see supplementary materials). Finally, various measures of profitability were calculated using these data and assumptions, including return on investment (ROI), payback period, internal rate of return (IRR), and net present value (NPV).

2.1.3. Minimum selling price estimation and sensitivity analysis

Analyses were conducted to estimate the MSP of CNC with and without acid recovery. MSP refers to the minimum price at which a product can be sold while covering the cost of production (Khan et al., 2022). By knowing the minimum price that can sustain the business, the MSP estimation helps find the CNC's market and economic sustainability. Because the estimated MSP and the profitability depend on assumed processes and economic factors, a sensitivity analysis was conducted to understand the effects of these assumptions on the results and the associated uncertainty. The sensitivity analyses were conducted by varying the price of raw materials, electricity, and utilities covering a range of minimum and maximum values. The uncertainty analysis was conducted only for the estimated MSPs utilizing the Risk software (Palisade Corporation, 2020). A Monte Carlo simulation with 10,000 iterations was obtained using a Project Evaluation and Review Techniques (PERT) distribution for the uncertainty analysis. The PERT distribution is commonly used in risk analysis to portray the uncertainty of the value of some quantity where one is depending on subjective estimates. The range of uncertainty on different input factors was specified by varying costs of the raw materials, consumables, and utilities by ±50% to find the lower and upper bound of the estimated MSP (Rajendran and Han, 2023).

2.2. Life-cycle environmental impact analysis

A LCA was performed to investigate the environmental impacts of CNC production using wood pulp with and without acid recovery. A cradle-to-gate LCA approach was followed based on the ISO 14040 and 14044 standards (ISO, 2006b, 2006a). The LCA analysis was facilitated by SimaPro 9.3 LCA software (PRé Consultants, 2021). The LCA followed four major steps including i) goal and scope definition, ii) life-cycle inventory (LCI) analysis, iii) impact analysis, and iv) interpretation.

2.2.1. Goal and scope

The first step in a typical LCA is to define the goal and scope, which establishes the context of assessment and identifies the objective of the study, the functional or declared unit, the system boundary, the data needed, the assumption, and the limitations (Itskos et al., 2016). The functional unit is a common reference unit that enables consistent comparison among all inputs and outputs while the declared unit refers to the production mass or volume of a given product; for this analysis, a declared unit of 1 kg of dry CNC was selected. The system boundary was cradle-to-gate, which primarily focuses on the CNC manufacturing process while including the extraction (i.e., harvesting) of raw material and its transportation to the production facility. An economic allocation was used instead of mass allocation because of the large quantities of other low-value co-products in the process. Key data needed to conduct LCA (e.g., mass and energy balances, material inputs and outputs, and fugitive emissions associated with CNC production) were based on the process model described in the TEA section (Section 2.1). Additional data required for LCA were supplemented from literature and available databases (Gu et al., 2015).

2.2.2. Life-cycle inventory analysis

The next step in an LCA is life-cycle inventory (LCI) analysis, which quantifies energy, water, and materials usage and environmental releases (e.g., CO_2 emissions, and wastewater discharge).

2.2.3. Impact analysis

In this stage, the potential impacts on humans and the environment are estimated based on the energy, water and materials consumption quantified in the LCI analysis stage. This study estimated the life-cycle environmental impacts of 1 kg of CNC production under 10 impact categories following the tool for the reduction and assessment of chemical and other environmental impacts (TRACI) 2.1 methodology (Bare, 2011). These impact categories included acidification, carcinogenics, ecotoxicity, fossil fuel depletion, eutrophication, global warming, ozone depletion, non-carcinogenics, respiratory, and smog (Bare, 2011).

2.2.4. Interpretation

Finally, the interpretation stage involves scrutinizing and discussing the results of the LCI analysis and impact assessment to identify a product, process or service that has the least environmental impacts. Here, this was achieved by conducting a sensitivity analysis and by identifying the key raw materials and processes of the CNC production that would result in the lowest or the highest impacts. Such information is helpful to manufacturers interested in CNC production from wood pulp, its improvement, and potential marketing. Similarly, the LCA results would be useful to policymakers or regulators to develop strategic planning and public policymaking related to the climate change mitigation potential of the CNC manufacturing industry (Itskos et al., 2016). Finally, such LCA results would be useful to the public, who are interested in understanding the environmental burden of CNC production from wood pulp.

3. Results and discussion

3.1. Process flow and mass balance

Producing 100 MT/day of CNC required 208 MT of wet wood pulp (5% of moisture content). Initially, the pulp was shredded to reduce particle size for improved hydrolysis. The shredded pulp was transferred to the reactor for the hydrolysis process using a screw conveyor. In the hydrolysis process, 981 MT of 64% sulfuric acid was used for 1hr at 45 °C. After the hydrolysis process, water (2% acid) from the second washing was added to stop the hydrolysis reaction and then transferred to the decanter to separate the CNC slurry from the acid solution. The 11.5% acid mixture was transferred to the diafiltration process to separate the sugar and acid solution. The filtered acid solution was transferred to the evaporator to remove the 64% sulfuric acid solution, which was recycled back into the process. The sugar solution contained acid which was removed using 96 MT/day of calcium oxide (CaO) with gypsum ($CaSO_4$) being produced in the process. The 1,198 MT of 7.8% sugar solution and 448 MT of 50% wet gypsum were separated using a microfilter. The CNC slurry was washed twice, and water was reused. Then 0.26 MT/day of sodium chlorite ($NaClO_2$) was added to the slurry to maintain the CNC brightness. The washed slurry still contained a small amount of acid, which was neutralized using 25% NaOH. The 2% CNC slurry was obtained and filtered using diafilter and ultrafilter to concentrate up to 8%. The final slurry was dried using a spray dryer to obtain 100 MT/day of dry CNC powder. The CNC production process was investigated with and without acid recovery. The quantities of 64%

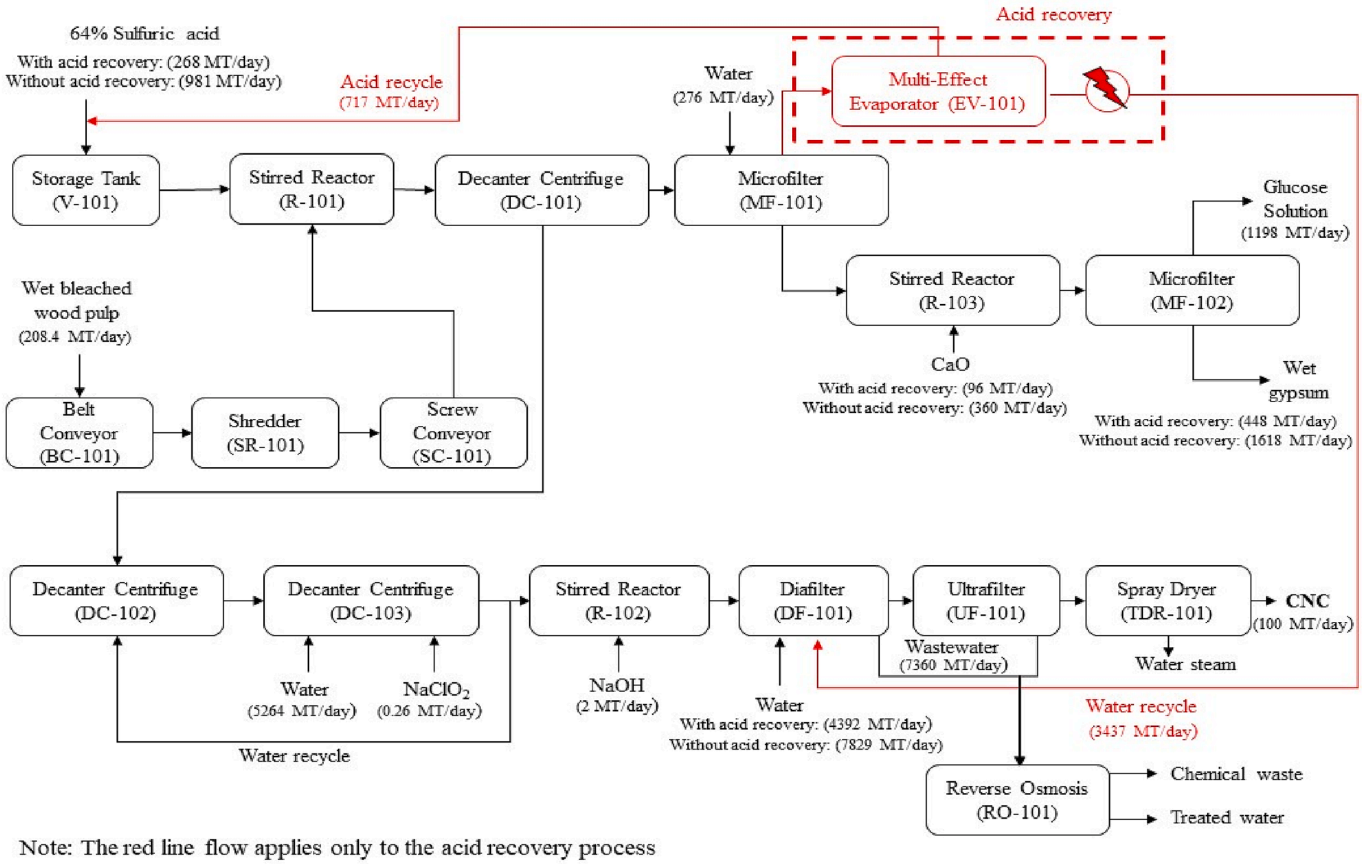


Fig. 1. Process flow of CNC production from wood pulp with and without acid recovery.

Table 1
Economic summary of wood pulp CNC production with and without acid recovery process.

Description	With Acid Recovery	Without Acid Recovery
Total Capital Investment ($)	285,853,000.00	227,738,000.00
Operating Cost ($/yr)	144,352,000.00	149,796,000.00
Savings ($/yr)	60,748.00	0
Main Revenue ($/yr)	198,009,000.00	198,008,000.00
Other Revenues ($/yr)	3,953,068.00	0
Total Revenues ($/yr)	201,962,000.00	198,008,000.00
Cost Basis Annual Rate (kg/yr)	33,001,527.00	33,001,314.00
Net Unit Production Cost ($/kg)	4.37	4.54
Unit Production Revenue ($/kg)	6.12	6.00
Gross Margin (%)	28.56	24.35
Return On Investment (%)	24.73	25.38
Payback Time (yr)	4.04	3.94
IRR (After Taxes) (%)	29.42	30.40
NPV (at 10.0% Interest) ($)	307,477,000.00	257,826,000.00

sulfuric acid used in the with and without acid recovery processes were 268 MT/day and 981 MT/day, respectively. In without acid recovery process, 360 MT/day CaO was used to neutralize the acid waste. The glucose solution was not obtained in without acid recovery process. The wastewater, once treated at the plant noted above was disposed of without further treatment. The process flow is shown in Fig. 1.

3.2. *Economic analysis*

3.2.1. *Capital investment*

The estimated total capital investment for wood CNC production with and without acid recovery was estimated to be $285.85 million and $227.74 million, respectively (Table 1). The capital investment for both processes was higher because of the utilization of a large amount of acid and water, which increased the size of the equipment. The capital investment for with acid recovery process was higher than without the acid recovery process because additional unit operations were used in the acid recovery process. In our process model, facility, construction, and installation costs were estimated based on equipment purchase cost so an increment of equipment cost would increase the overall capital investment. The detailed capital investment for both processes is shown in Fig. 2(a). The capital investment for both with and without acid recovery process consisted of direct cost (with acid recovery – 50.28%, without acid recovery – 49.54%), indirect cost (with acid recovery – 30.17%, without acid recovery – 29.72%), other costs (with acid recovery – 12.07%, without acid recovery – 11.89%) and miscellaneous cost (with acid recovery – 7.49%, without acid recovery – 8.84%). The equipment, engineering and construction costs played a major role in capital investment. In equipment cost, reverse osmosis filters accounted for a major part of both processes (with acid recovery – 31.98 %, without acid recovery – 40.33%) due to the utilization of a large amount of water generating a large amount of wastewater. In with acid recovery process, three decanter centrifuges (22.92%), microfilter – MF-101 (15.45%), multi-effect evaporator – EV-101 (10.89%) and diafilter – DF-101 (4.55%) accounted for 53.82% of total equipment cost. In without acid recovery process, three centrifuges (29.04%) and microfilter – MF-102 (7.18%) accounted for 36.22% of the total equipment cost. In terms of equipment cost, separation and filtration equipment accounted for most of the cost due to the high flow rate and large equipment size. Advanced process development would help to reduce unit operations, equipment size and overall capital investment.

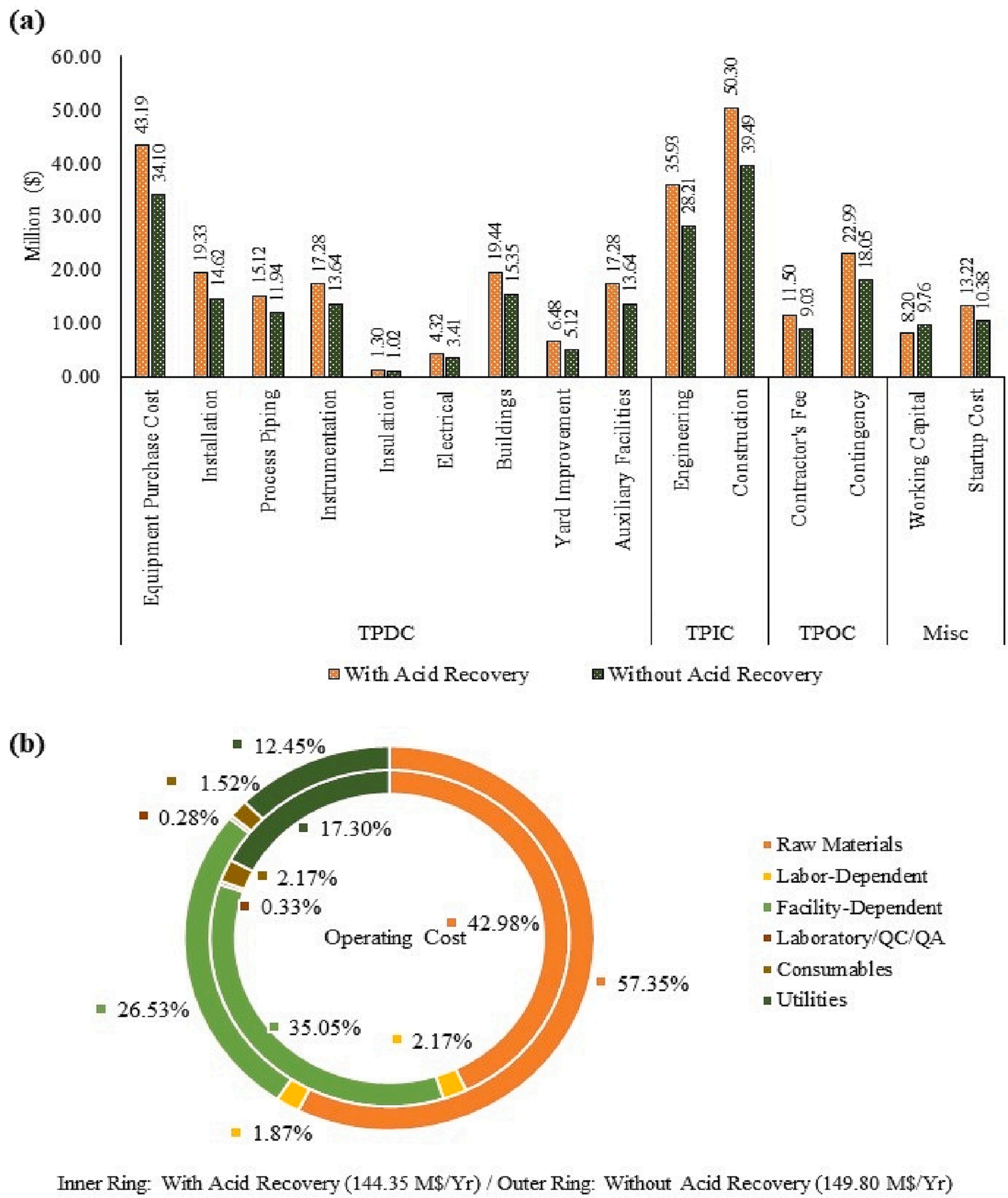


Fig. 2. Total capital investment a) and annual operating cost b) of CNC production from wood pulp with and without acid recovery. Note: TPDC = Total Project Direct Cost, TPIC = Total Project Indirect Cost, TPOC = Total Project Other Cost).

Table 2
Input values of sensitivity analysis.

Input Materials	Minimum Value	Base Value	Maximum value
Raw Materials			
Wood pulp ($/kg)	0.38	0.76	1.14
CaO ($/kg)	0.07	0.14	0.21
H_2SO_4 ($/kg)	0.04	0.07	0.11
Consumable			
Membrane ($/m^2)	200.00	400.00	600.00
Utilities			
Electricity ($/kWh)	0.05	0.10	0.15
Steam ($/kg)	6.00	12.00	18.00
Chilled Water ($/kg)	0.20	0.40	0.60

3.2.2. Annual operating cost

The second major cost was operating cost, which includes costs associated with raw materials, labour-dependent, facility-dependent, labour, quality control and assurance, consumables, and utilities. The annual operating costs of with and without acid recovery processes were $144.35 million and $149.80 million, respectively (Table 1). In both processes of CNC production, the costs of raw materials, consumables, utilities, and facility-dependent constituted a major share of the operating costs. The detailed operating costs for the two processes are shown in Fig. 2(b). Wood pulp cost accounted for the major part, 80.35% (with acid recovery) and 58.03% (without acid recovery) of total raw material cost. In with acid recovery, CaO (7.01%) and H_2SO_4 (6.44%) accounted for 13.45% of total raw material cost. In without acid recovery, CaO

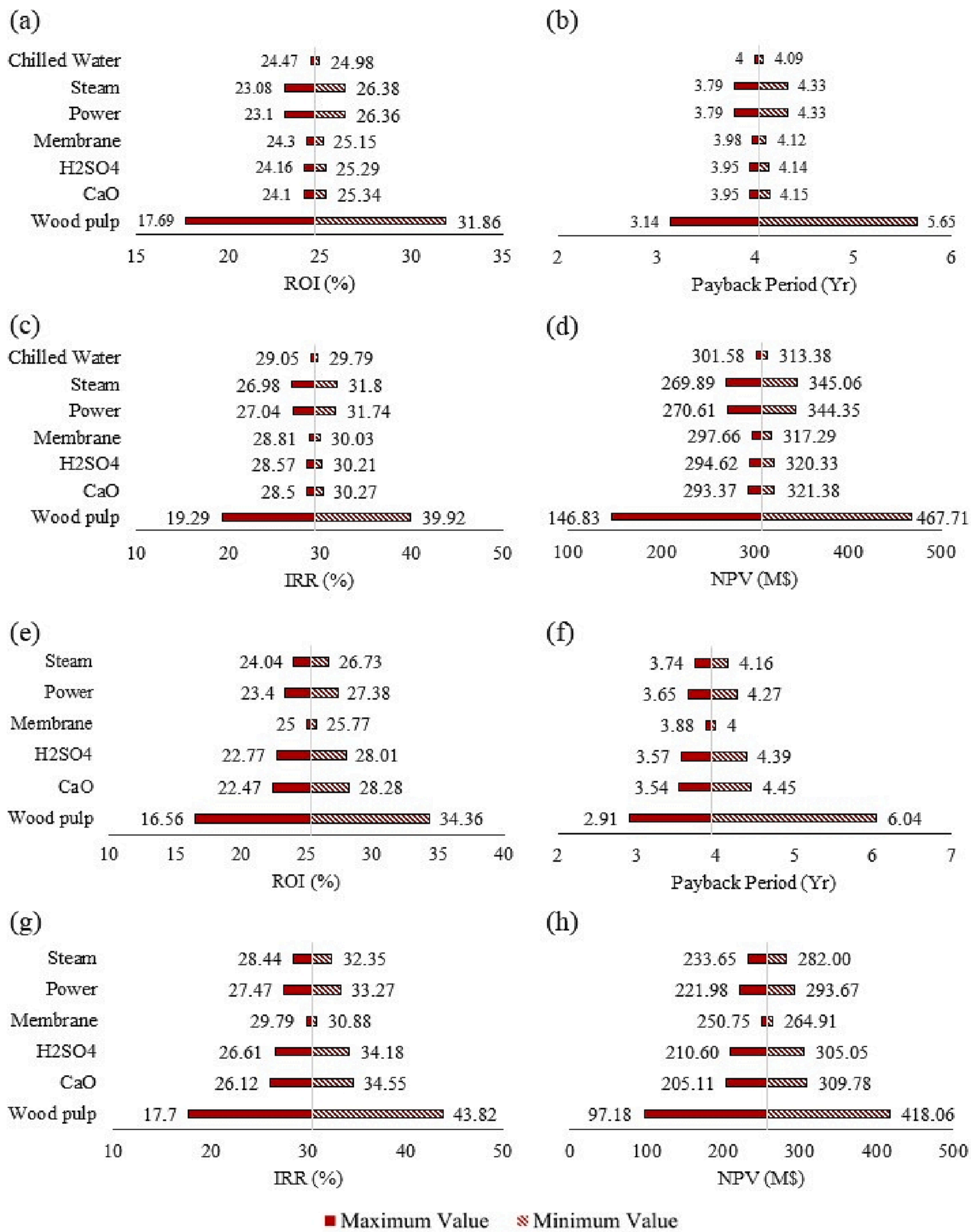


Fig. 3. Sensitivity analysis of economic factors (a-d) with acid recovery, (e-h) without acid recovery.

(18.93%) and H_2SO_4 (17.08%) accounted for 36.01% of total raw material cost. The cost of consumables (filter membrane) was $3.14 million in with acid recovery process and $2.78 million in without acid recovery process, representing 2.17% and 1.52% of total operating costs, respectively. Similarly, the utilities accounted for 17.30% ($24.97 million) and 12.45% ($18.65 million) of total operating costs in with and without acid recovery processes, respectively. In without acid recovery process, material (mainly H_2SO_4 and CaO) costs were high, and in acid recovery process, facility-dependent and utilities costs were high.

3.2.3. Economic summary and sensitivity analysis

The detailed economic summary of the base scenarios is shown in Table 1, which indicates that the overall economic feasibility of the with acid recovery process was better than the without acid recovery process of wood CNC production. In both scenarios, CNC was the main source of revenue. Whereas the with acid recovery process included glucose solution as additional source of revenue. The total annual revenues of with and without acid recovery process were $201.96 million and $198.01 million, respectively.

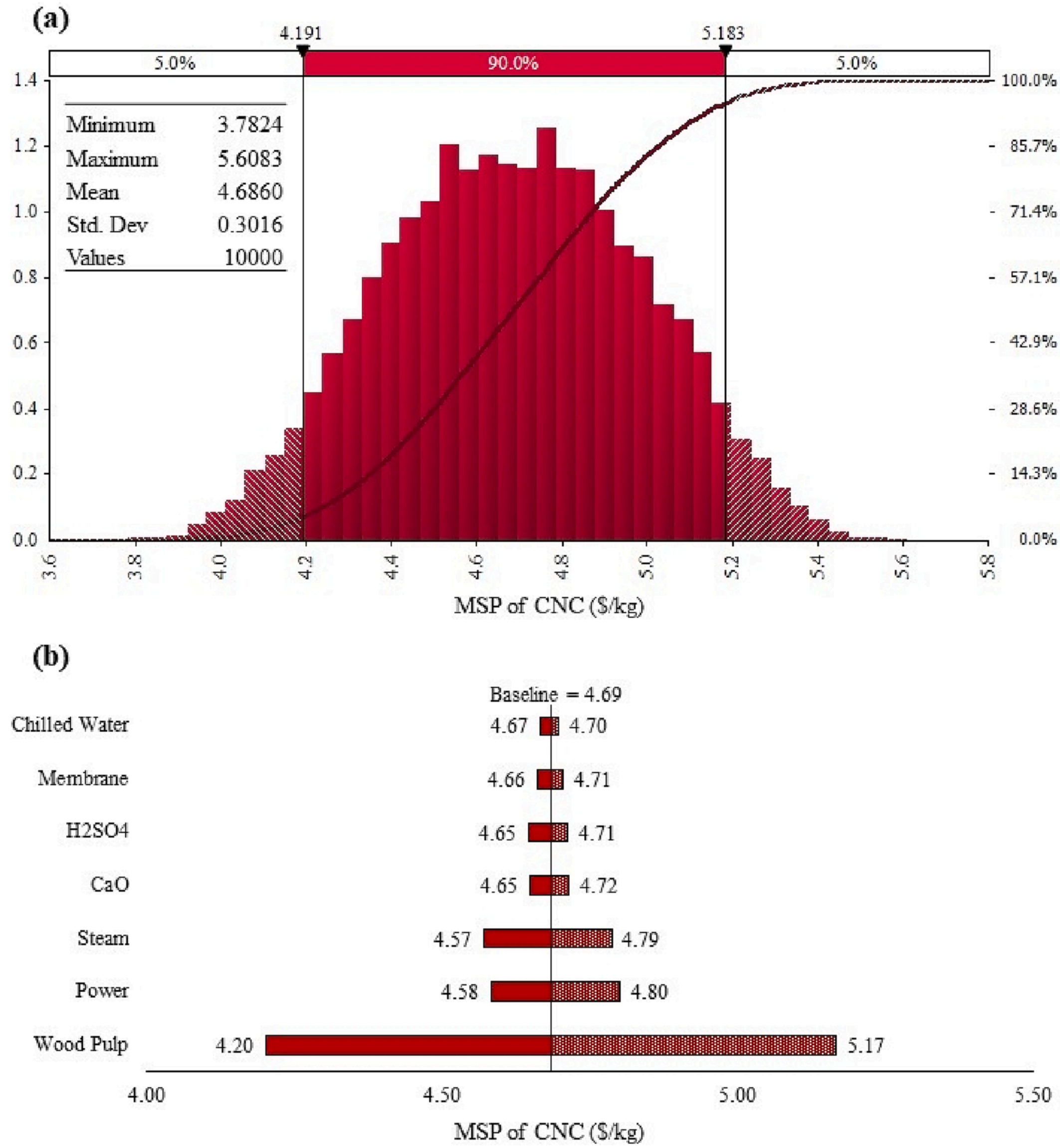


Fig. 4. Uncertainty analysis of MSP of wood pulp CNC produced with acid recovery, a) probability distribution and lower and upper bounds, and b) impacts of input prices.

The robustness of calculated financial outcomes such as ROI, payback period, IRR and NPV were evaluated by conducting a sensitivity analysis, where the costs of input raw materials (wood pulp, CaO, H_2SO_4), consumables (membrane), and utilities (steam, cooling water) were modified by ± 50% (Table 2). The results of the sensitivity analysis are shown in Fig. 3. In the process that includes acid recovery, the cost of the wood pulp, electricity and steam had a major influence on the calculated financial results. The evaporation technique used in this study for the acid recovery process also required a large amount of heat transfer agents (steam), which also increased the overall operating cost of this process. Advanced alternative technology will be required for acid separation for this process to become more financially feasible. In without acid recovery process, wood pulp, CaO and H_2SO_4 had major impacts on the estimated measures of profitability. The large amount of acid waste produced in this process required a large amount of CaO for neutralization.

3.2.4. *Uncertainty analysis of minimum selling price*

The calculated MSPs of wood CNC produced with and without acid recovery for the base scenario were $4.69/kg and $4.89/kg respectively, which were notably lower to those estimated in previous studies by Bondancia et al. (2020) and de Assis et al. (2017), who reported MSPs of CNC of $6.9/kg and $7.2/kg, respectively. In addition, the estimated MSP of wood CNC produced through acid hydrolysis by Rosales-Calderon et al. (2021) was $10.03/kg, which was substantially higher than the MSP estimated in this study, probably due to different source, capacity, cost, and process. The MSPs in this study were calculated using the base costs of input raw materials, consumables, electricity, and utilities. To understand the uncertainties in the estimated MSPs, these costs were varied by ± 50% and their effects on the calculated MSP of CNC were investigated. The 90% confidence intervals of the estimated MSP of CNC were $4.19 to $5.18/kg and $4.37 to $5.41/kg in with and without acid recovery process, respectively (Figs. 4 and 5). The wood pulp price showed a major impact on the estimated MSP of CNC in both processes. However, a large amount of steam used in the acid recovery process to concentrate the acid also had a substantial impact on the calculated MSP of CNC with acid recovery. The without acid recovery process required large amounts of acid and CaO which increased the raw material cost as well as the MSP of CNC.

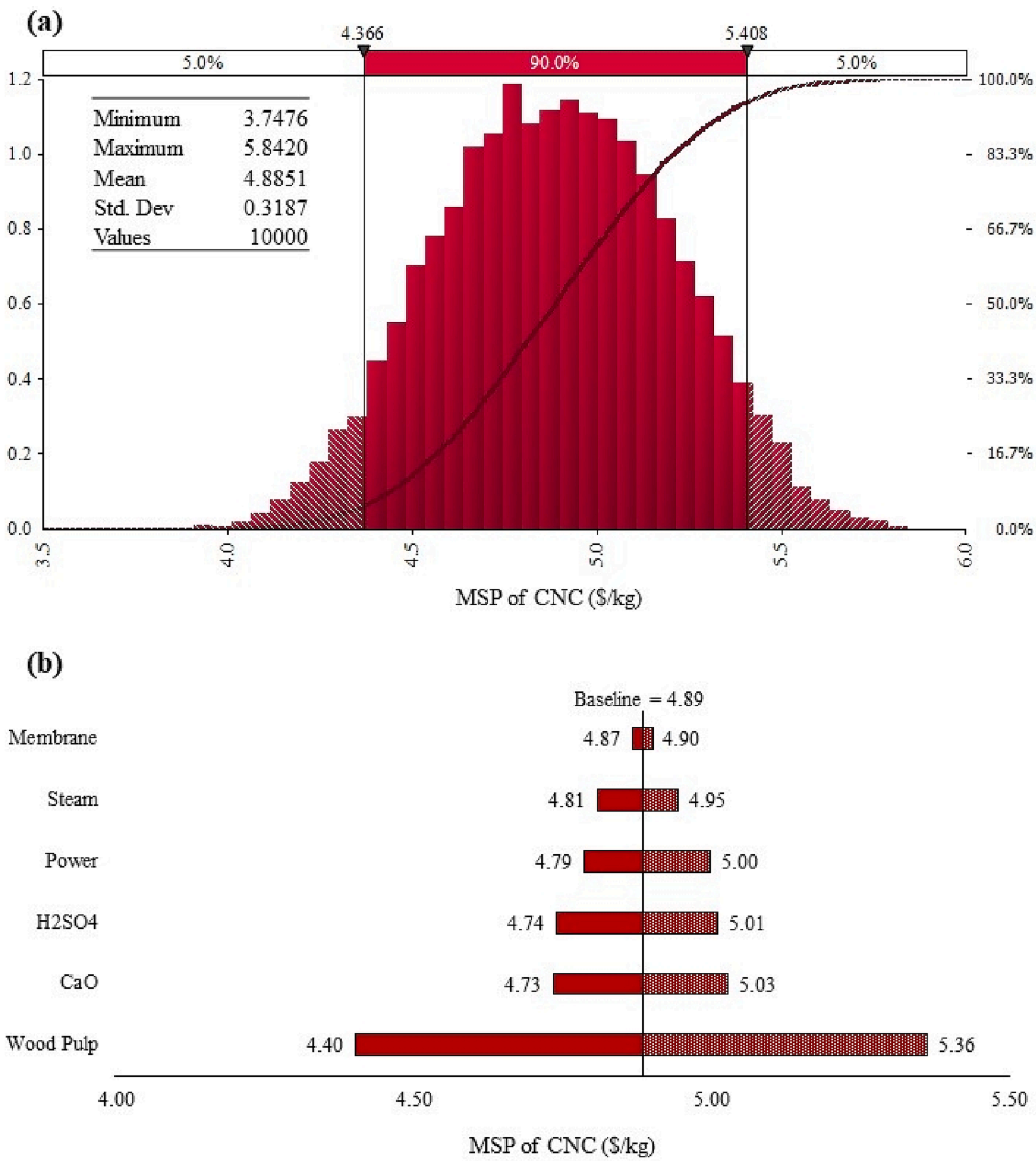


Fig. 5. Uncertainty analysis of MSP of wood pulp CNC produced without acid recovery, a) probability distribution and lower and upper bounds, and b) impacts of input prices.

3.3. *Environmental impact analysis*

The volume of input materials and energy utilities of production of 1 kg of CNC and LCI are shown in Table 3. These data were obtained from the SuperPro designer model for a specified CNC plant production capacity of 100 MT/day. The CNC production process utilized bleached pulp, H_2SO_4, CaO, $NaClO_2$, NaOH, and water. The estimated cradle-to-gate GW impacts of 1 kg of CNC produced with and without acid recovery processes were 11.39 kg CO_2 eq and 11.18 kg CO_2 eq, respectively (Fig. 6). Sodium chlorite was not available in the SimaPro database, so sodium hypochlorite was used as a proxy in this study. Steam was a major contributor to the GW impact in both processes. A large amount of steam was needed during the acid recovery process. The without acid recovery process required a large amount of CaO for the neutralization process, which also contributed to increase GW impact. The detailed contribution analysis by inputs for their environmental impacts is shown in supplementary materials. The development of an alternative acid recovery process that would reduce steam usage would reduce the environmental impact of CNC produced with acid recovery. Also, sourcing the fuel to generate steam from biomass instead of fossil fuels could lower the overall impacts and especially GW.

3.4. *Critical review and future aspects*

The process models developed in this study for both with and without acid recovery processes consumed a large amount of acid and water, which increased the equipment size, and water washing steps. The acid recovery process further increased the capital investment due to the need for additional equipment for acid recovery, but the operating cost was lower than without acid recovery due to acid recovery and low

Table 3
Input materials and energy consumption per kg of dry CNC production from wood pulp and the associated LCI unit processes.

Material	Unit	1 kg of CNC		LCI Unit Process
		With acid recovery	Without acid recovery	
Bleached wood pulp	kg	1.98	1.98	Pulp, kraft market, bleached, average production, at mill/kg/RNA
CaO	kg	0.96	3.60	Quicklime, at plant/US
H_2SO_4	kg	1.73	6.35	Sulphuric acid, liquid, at plant/US- US-EI U
$NaClO_2$	kg	<0.01	<0.01	Sodium hypochlorite, 15% in H_2O, at plant/US- US-EI U (proxy)
NaOH	kg	0.02	0.02	Sodium hydroxide, production mix, at plant/RNA
Water	kg	95.88	130.62	Water, deionised, at plant/US* US-EI U
Steam	kg	29.49	18.97	Steam, for chemical processes, at plant/US- US-EI U
Electricity	kW-h	3.47	3.38	Electricity, medium voltage, certified electricity, at grid/US* US-EI U

utilization of CaO in the acid waste neutralization step. The without acid recovery process required a large amount of acid and CaO which increased the raw material cost as well as the operating cost. This trade-off between capital and operating expenses can also be seen in the financial metrics with higher calculated NPV for the acid recovery scenario but higher ROI and lower Payback time for the no-acid recovery scenario. Based on these results acid recovery is warranted for projects especially if material costs are expected to rise or if lower energy costs can be procured. Advanced cost-effective and eco-friendly techniques are required for acid recovery to reduce the operating cost and GW impact. Future development of a process that reduces acid and water usage would help to reduce the equipment size and overall capital and operating cost.

Environmental impacts for the two scenarios when compared showed neither system to be significantly different for most categories. However, the with acid recovery scenario had a significantly higher GW impact, due to the increase in energy required to recover the sulfuric acid. Using steam and electrical power from lower carbon-intensive sources or finding improve separation efficiency operations would be key in further reducing the environmental impacts of recovering the acid and should continue to be investigated.

Finally, it should be noted that the TEA and LCA results were based on data from the USA only, and therefore, they may not be generalized for different geographical areas where the cost of input materials, energy and labour, and tax rates differ. However, this study has provided the necessary information and methodological framework needed to conduct TEA and LCA of CNC production in different geographies.

4. Conclusions

The analysis found higher capital investment for with-acid recovery process due to the need for extra unit operations but resulted in lower operating cost due to recovery of acid and glucose solution and reduction in CaO use. Results of advanced separation processes should be further explored to further reduce the equipment capital and operating costs. However, the with acid recovery scenario had a significantly higher GW impact, due to higher energy required to recover the acid, suggesting the importance of utilizing lower carbon-intensive energy sources or finding improved separation efficiency operations in reducing overall environmental impacts of recovering the acid.

Data Availability Statement: Some authors of this paper are employees of the United States Department of Agriculture, U.S. Forest Service. The findings and conclusions in this report are those of the author(s) and should not be construed to represent any official USDA or U.S. Government determination or policy. This research was supported in part by the U.S. Department of Agriculture, Forest Service. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. government.

CRediT authorship contribution statement

Naveenkumar Rajendran: Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Visualization. **Troy**

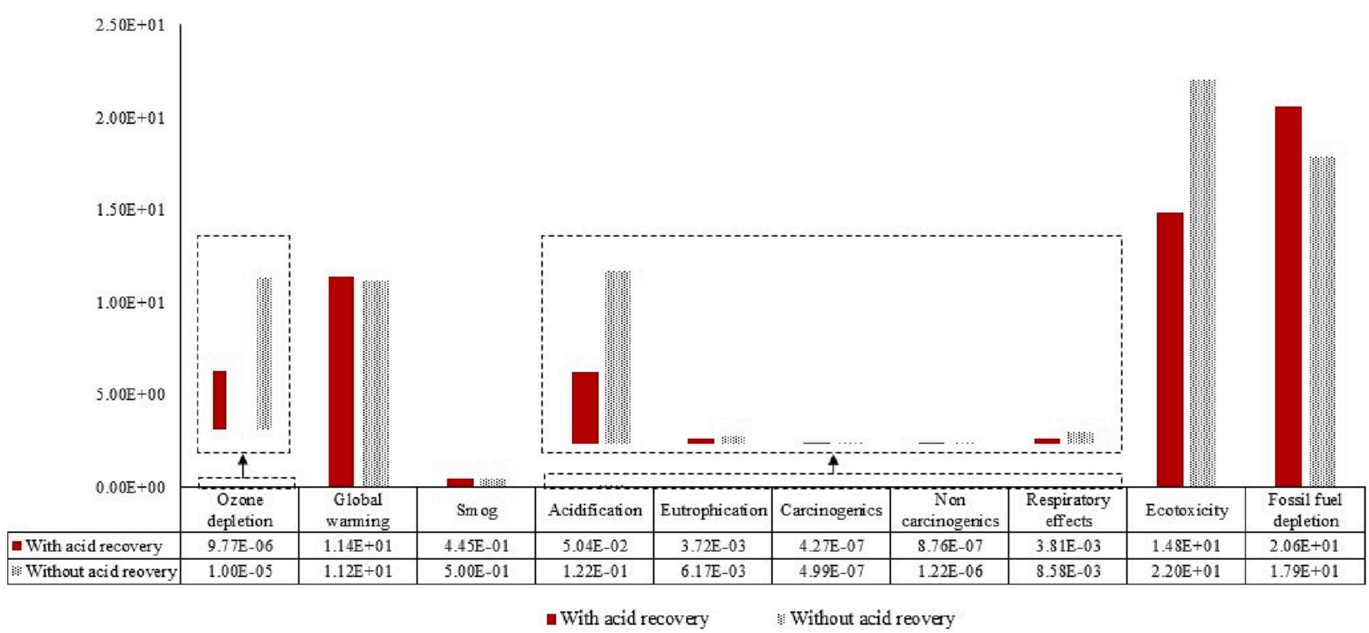


Fig. 6. Environmental impact of CNC production from wood pulp.

Runge: Formal analysis, Resources, Writing – review & editing, Visualization, Funding acquisition. **Richard D. Bergman:** Formal analysis, Resources, Writing – review & editing, Visualization, Funding acquisition. **Prakash Nepal:** Validation, Writing – review & editing, Visualization. **Carl Houtman:** Formal analysis, Resources, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

This project was made possible by financial and technical assistance provided by the U.S. Endowment for Forestry and Communities, Inc., under projects 20-00098 and 21-00288.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.biortech.2023.128955.

References

Abushammala, H., Krossing, I., Laborie, M.P., 2015. Ionic liquid-mediated technology to produce cellulose nanocrystals directly from wood. Carbohydr. Polym. 134, 609–616. https://doi.org/10.1016/j.carbpol.2015.07.079.

Bare, J., 2011. TRACI 2.0: the tool for the reduction and assessment of chemical and other environmental impacts 2.0. Clean Techn. Environ. Policy 13, 687–696. https://doi.org/10.1007/s10098-010-0338-9.

Beyene, D., Chae, M., Dai, J., Danumah, C., Tosto, F., Demesa, A., Bressler, D., 2018. Characterization of cellulase-treated fibers and resulting cellulose nanocrystals generated through acid hydrolysis. Materials 11 (8), 1272.

Bondancia, T.J., De Aguiar, J., Batista, G., Cruz, A.J.G., Marconcini, J.M., Mattoso, L.H. C., Farinas, C.S., 2020. Production of Nanocellulose Using Citric Acid in a Biorefinery Concept: Effect of the Hydrolysis Reaction Time and Techno-Economic Analysis. Ind. Eng. Chem. Res. 59, 11505–11516. https://doi.org/10.1021/acs.iecr.0c01359.

Chausali, N., Saxena, J., Prasad, R., 2022. Recent trends in nanotechnology applications of bio-based packaging. J. Agric. Food Res. 7, 100257 https://doi.org/10.1016/j.jafr.2021.100257.

Palisade Corporation, 2020. @Risk Getting Started Guide. https://help.palisade.com/v8/en/Guides/@RISK-Getting-Started-Guide.pdf. Accessed Nov 11, 2022.

de Assis, C.A., Houtman, C., Phillips, R., Bilek, E.M.T., Rojas, O.J., Pal, L., Peresin, M.S., Jameel, H., Gonzalez, R., 2017. Conversion Economics of Forest Biomaterials: Risk and Financial Analysis of CNC Manufacturing. Biofuels Bioprod. Biorefin. 11, 682–700. https://doi.org/10.1002/bbb.1782.

Dong, S., Bortner, M.J., Roman, M., 2016. Analysis of the sulfuric acid hydrolysis of wood pulp for cellulose nanocrystal production: A central composite design study. Ind. Crop. Prod. 93, 76–87. https://doi.org/10.1016/j.indcrop.2016.01.048.

Foster, E.J., Moon, R.J., Agarwal, U.P., Bortner, M.J., Bras, J., Camarero-Espinosa, S., Chan, K.J., Clift, M.J.D., Cranston, E.D., Eichhorn, S.J., Fox, D.M., Hamad, W.Y., Heux, L., Jean, B., Korey, M., Nieh, W., Ong, K.J., Reid, M.S., Renneckar, S., Roberts, R., Shatkin, J.A., Simonsen, J., Stinson-Bagby, K., Wanasekara, N., Youngblood, J., 2018. Current characterization methods for cellulose nanomaterials. Chem. Soc. Rev. 47, 2609–2679. https://doi.org/10.1039/C6CS00895J.

Gu, H., Reiner, R., Bergman, R., Rudie, A., 2015. LCA Study for Pilot Scale Production of Cellulose Nano Crystals (CNC) from Wood Pulp. In: Proceedings from the LCA XV Conference - A bright green future. 6-8 October 2015. Vancouver, British Columbia, Canada. p. 33-42.

Intelligen, Inc., 2020. SuperPro Designer. User Guide. https://www.intelligen.com/wp-content/uploads/2020/05/SuperPro_ManualForPrinting_v11.pdf. Accessed Oct. 12, 2022.

IRS, 2017. Tax Cuts and Jobs Act of 2017 (TCJA) - Internal Revenue Service.

ISO, 2006a. Environmental Management: Life Cycle Assessments: Requirements and Guidelines. International Standardization Organization.

ISO, 2006b. Environmental Management: Life Cycle Assessment; Principles and Framework. International Organization for Standardization.

Future Markets. The Global Market for Nanocellulose 2023-2033 (Cellulose Nanofibers, Cellulose Nanocrystals and Bacterial Nanocellulose). https://www.futuremarketsinc.com/the-global-market-for-nanocellulose-2023/.

Itskos, G., Nikolopoulos, N., Kourkoumpas, D.-S., Koutsianos, A., Violidakis, I., Drosatos, P., Grammelis, P., 2016. Energy and the Environment. In: Environment and Development. Elsevier, pp. 363–452. https://doi.org/10.1016/B978-0-444-62733-9.00006-X.

Jakes, J.E., Arzola, X., Bergman, R., Ciesielski, P., Hunt, C.G., Rahbar, N., Tshabalala, M., Wiedenhoeft, A.C., Zelinka, S.L., 2016. Not Just Lumber—Using Wood in the Sustainable Future of Materials, Chemicals, and Fuels. JOM 68, 2395–2404. https://doi.org/10.1007/s11837-016-2026-7.

Khan, M.N., Lacroix, M., Wessels, C., van Dael, M., 2022. Converting wastewater cellulose to valuable products: A techno-economic assessment. J. Clean. Prod. 365, 132812 https://doi.org/10.1016/j.jclepro.2022.132812.

Moon, R.J., Schueneman, G.T., Simonsen, J., 2016. Overview of Cellulose Nanomaterials, Their Capabilities and Applications. JOM 68, 2383–2394. https://doi.org/10.1007/s11837-016-2018-7.

Naveenkumar, R., Baskar, G., 2020. Optimization and techno-economic analysis of biodiesel production from Calophyllum inophyllum oil using heterogeneous nanocatalyst. Bioresour. Technol. 315, 123852.

Naveenkumar, R., Baskar, G., 2021. Process optimization, green chemistry balance and technoeconomic analysis of biodiesel production from castor oil using heterogeneous nanocatalyst. Bioresour. Technol. 320, 124347.

PRé Consultants, 2021. SimaPro 9.3: Full update instructions, https://simapro.com/wp-content/uploads/2021/12/FullUpdateInstructionsToSimaPro930.pdf. Accessed Dec 11, 2022.

Rajendran, N., Han, J., 2022a. Techno-economic analysis of food waste valorization for integrated production of polyhydroxyalkanoates and biofuels. Bioresour. Technol. 348, 126796 https://doi.org/10.1016/j.biortech.2022.126796.

Rajendran, N., Han, J., 2022b. Integrated Polylactic Acid and Biodiesel Production from Food Waste: Process Synthesis and Economics. Bioresour. Technol. 343, 126119.

Rajendran, N., Han, J., 2023. Techno-economic analysis and life cycle assessment of poly (butylene succinate) production using food waste. Waste Manag. 156, 168–176. https://doi.org/10.1016/j.wasman.2022.11.037.

Rajendran, N., Kang, D., Han, J., Gurunathan, B., 2022. Process optimization, economic and environmental analysis of biodiesel production from food waste using a citrus fruit peel biochar catalyst. J. Clean. Prod. 365, 132712 https://doi.org/10.1016/j.jclepro.2022.132712.

Rosales-Calderon, O., Pereira, B., Arantes, V., 2021. Economic assessment of the conversion of bleached eucalyptus Kraft pulp into cellulose nanocrystals in a stand-alone facility via acid and enzymatic hydrolysis. Biofuels Bioprod. Biorefin. 15, 1775–1788. https://doi.org/10.1002/bbb.2277.

Sacui, I.A., Nieuwendaal, R.C., Burnett, D.J., Stranick, S.J., Jorfi, M., Weder, C., Foster, E. J., Olsson, R.T., Gilman, J.W., 2014. Comparison of the properties of cellulose nanocrystals and cellulose nanofibrils isolated from bacteria, tunicate, and wood processed using acid, enzymatic, mechanical, and oxidative methods. In: ACS Applied Materials and Interfaces. American Chemical Society, pp. 6127–6138. https://doi.org/10.1021/am500359f.

Sahoo, K., Bilek, E., Bergman, R., Mani, S., 2019. Techno-economic analysis of producing solid biofuels and biochar from forest residues using portable systems. Appl. Energy 235, 578–590. https://doi.org/10.1016/j.apenergy.2018.10.076.

Sahoo, K., Upadhyay, A., Runge, T., Bergman, R., Puettmann, M., Bilek, E., 2021. Life-cycle assessment and techno-economic analysis of biochar produced from forest residues using portable systems. Int. J. Life Cycle Assess. 26, 189–213. https://doi.org/10.1007/s11367-020-01830-9.

Shankaran, D.R., 2018. Cellulose Nanocrystals for Health Care Applications. In: Applications of Nanomaterials. Elsevier, pp. 415–459. https://doi.org/10.1016/B978-0-08-101971-9.00015-6.

Sirviö, J.A., Visanko, M., Liimatainen, H., 2016. Acidic Deep Eutectic Solvents As Hydrolytic Media for Cellulose Nanocrystal Production. Biomacromolecules 17, 3025–3032. https://doi.org/10.1021/acs.biomac.6b00910.

Stratistics Market Research Consulting, 2022. Nanocellulose Market Forecasts to 2028 – Global Analysis By Type (Cellulose Nanofibrils (CNF), Micro-Fibrillated Cellulose (MFC), Nanocrystalline Cellulose, Bacterial Cellulose (BC) and Other Types), Raw Material (Non-Wood and Wood), Application and By Geography. https://www.marketresearch.com/Stratistics-Market-Research-Consulting-v4058/Nanocellulose-Forecasts-Global-Type-Cellulose-33096424/. Accessed Feb 25, 2022.

Trache, D., Hussin, M.H., Haafiz, M.K.M., Thakur, V.K., 2017. Recent progress in cellulose nanocrystals: Sources and production. Nanoscale 9 (5), 1763–1786.

Van Schoubroeck, S., Van Dael, M., Van Passel, S., Malina, R., 2018. A review of sustainability indicators for biobased chemicals. Renew. Sustain. Energy Rev. 94, 115–126. https://doi.org/10.1016/j.rser.2018.06.007.

Wegner, T.H., Ireland, S., Jones, E., 2013. Cellulose Nanomaterials: The Sustainable Material of Choice for the 21st Century. In: Postek, M.T., Moon, R.J., Rudie, A.W., Bilodeau, M.A. (Eds.), Production and Applications of Cellulose Nanomaterials. TAPPI PRESS.

Xie, H., Du, H., Yang, X., Si, C., 2018. Recent Strategies in Preparation of Cellulose Nanocrystals and Cellulose Nanofibrils Derived from Raw Cellulose Materials. Int. J. Polym. Sci. 2018, 1–25.

Zhang, L., Jia, X., Ai, Y., Huang, R., Qi, W., He, Z., Klemeš, J.J., Su, R., 2022. Greener production of cellulose nanocrystals: An optimised design and life cycle assessment. J. Clean. Prod. 345, 131073.