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Research Article

Environmental Impact Assessment and Process Upscaling for Sustainable Production of Cellulose Nanofibre, a Biopolymer Derived from Lignocellulosic Biomass

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Abstract: As an agricultural waste, rice straw has a requisite quality for conversion to nanocellulose; however, the environmental and technical feasibility of the processes available has not been assessed at a commercial scale. Therefore, in the present work commercialization and environmental impacts of the Cellulose Nanofibre (CNF) production from rice straw were evaluated by scaling up the laboratory-scale process to the pilot scale level. Life Cycle Assessment (LCA) of the laboratory scale process showed the maximum impact of electricity (161 kg CO_2 eq) and sodium hydroxide (7.82 kg CO_2 eq) on global warming potential; though, the overall impacts got reduced approximately three times (~51 kg CO_2 eq for electricity) with the scale-up of the process. When compared with the reported literature both the scales of production were found to have lesser environmental impacts. Further, the techno-economic feasibility of the process was evaluated at different plant capacities for the optimum Minimum Selling Price (MSP) of CNF. At the plant capacity of 15 kg/day, the MSP of CNF was calculated as $16,537.36/tonne dropped to $14,944.87 and $14,590.98/tonne with the increased batch capacity of 30 kg/day and 45 kg/day, respectively. Through sensitivity analysis, the MSP of CNF was observed to be highly sensitive to the cost of capital investment and chemicals. In addition, analysis of the uncertainty associated with the identified cost drivers reflects the scope for inherited risks, as determined by the Monte-Carlo simulation method. This study established an environmentally and economically sustainable process for the commercialisation of nanocellulose production from agricultural waste at a minimum cost of production.

***Keywords*:** cellulose nanofibre, life cycle assessment, process scale-up, rice straw, techno-economic analysis

1. Introduction

The commercialization of agricultural products and their byproducts has increased considerably as an outcome of a rise in population. These intensified agronomic activities led to tons of waste, mainly because of modern farming techniques[1]. The waste thus generated is dealt with by the standard practices of accumulation on the land or direct burning, which adversely affect human health and the environment. Agricultural waste is always an under-utilized resource for energy production and novel products[2,3]. To date, very little attention has been paid to the 'low carbon' route of valorization of waste. However, residues from agricultural waste represent a great source of carbohydrates,

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polymers, and other bioactive compounds[4]. A few reports on microcrystalline cellulose and novel lactic acid production using lingo-cellulosic biomass showed detailed life cycle assessment (LCA)[5]. Along with the co-production of value-added products such as 2,5-Furandicarboxylic acid (FDCA) and tetrahydrofurfuryl alcohol (THFA)[6].

Nanocellulose production from algae biomass, bacterial culture, and agricultural waste has gained much attention in recent years, but their industrial application remains deprived of factual value at the commercial level[7,8]. Cellulose nanofibre (CNF) production from natural resources presents an eco-friendly alternative to plastics, composite materials, resins, packaging materials, and others. The CNF can be reinforced with other biopolymers such as polyhydroxybutyrate, poly-lactic acid, starch, and others for various applications[9,10]. The global Cellulose nanocrystal (CNC) market value was estimated at USD 2.97 billion in 2021 and is expected to reach USD 7.83 billion by 2025[11]. As a developing country, India is looking for alternate resources and products to meet its basic needs and to achieve the United Nations Sustainable Development Goals (UN-SDG). Sustainable production of cellulose from agricultural waste requires the development of technologies that can provide green alternatives with improved productivity and a lower environmental profile. For environmental analysis, the life cycle assessment of agricultural products has been considered by many researchers[12], but still, a few have practiced this methodology for the products derived from agricultural waste[13,14]. The motto of the current research work is thus to perform the environmental and techno-economic analysis to examine the environmental and economic costs of producing cellulose nanofibre from waste rice straw.

The environmental feasibility of the laboratory scale and pilot scale process can be determined using the Life Cycle Assessment (LCA) method. In cradle to the grave method of LCA, evaluation of the environmental impacts of a product is based on all resource inputs and emissions from the entire process (ISO Standards 14040 & 14044 (2006); U.S. EPA (2006) Life Cycle Assessment Principles & Practice EPA/600/R-06/060)[15]. The environmental impacts can be studied in four phases, i.e., goal and scope of the study, life cycle inventory analysis, life cycle impact assessment (LCIA), and interpretation and reporting of results. LCA analysis of the processes was facilitated using different software such as SimaPro and GaBi Solution Education Software 2020. In the present work, life cycle inventory analysis was carried out using SimaPro 9.2 to identify the potential impacts of all the inputs and outputs of the process. The life cycle impact assessment of cellulose nanofibre production from rice straw was examined based on 18 categories of ReCiPe 2016 methodology. Further, design supported by techno-economic evaluation was projected for successful commercialization of the process. Economic assessment is critical for establishing the cost feasibility of the process and determining the cost reduction prospects for optimized planning[16]. A few have reviewed the economic feasibility of the integrated biorefinery model based on the economic values of bio-products, product generation, and cost-benefit analysis[17]. A study on bagasse valorization to value-added products and energy production reported LCA and techno-economic analysis of the process[6]. Reports showed the positive impact of fossil fuel-based energy sources on electricity production. The techno-economic analysis recommended the valorization of the process with the cogeneration of value-added products. Another study assessing the economic and environmental feasibility of cellulose nanocrystal production from sugarcane bagasse showed the major effect of pre-treatment steps, due to the various chemicals utilized during the processing of the feedstock[14]. The large-scale production of a product from agricultural waste prefers to be technically viable, economically feasible, and accepted socially as an environmentally friendly process, but it is the least explored field of research. Therefore, in this study, much emphasis has been given to the techno-economic assessment of commercial scale production of cellulose nanofibre along with its process designing for reduced environmental impacts.

Given quite a lot of potential of rice straw as a feedstock for nanocellulose production, its commercial-scale production was studied in the present report. Pilot-scale production of nano-cellulose from agricultural waste is a recent advancement in the area. Still, it lacks much information on its establishment as a sustainable industrial process performing the best productivity with the most negligible impact on the environment. To the best of our knowledge, study on the commercial feasibility of cellulose nanofibre production from rice straw has not been reported much, especially in the Indian context. However, India is among the largest producers of rice, producing ~120 million tonnes/year[18]. Therefore, the primary aim of the present study is to conduct the stochastic techno-economic assessment of the process at the pilot-scale level, to check its feasibility at a commercial scale, along with its viability as an environment-friendly procedure using LCA. Further, improvement in the process designing was performed based on the results obtained after analyzing the environmental impacts of the same process at a laboratory scale. A comprehensive model such as Monte Carlo simulation and sensitivity analysis was also studied to check the uncertainty and variability in the data. With these aims, an up-scaled robust process having a minimum risk of environmental impacts and invariability is

developed for its commercial applicability.

2. Methodology

2.1 *Materials*

Rice straw was collected from the local farms of Mohali, Punjab, India. Rice straw was washed with water and then dried to remove impurities or dirt. It was then grounded using a commercial grinder (PX-MFC 90). All chemicals like Sodium hydroxide flakes (NaOH, CDH-97%), sodium chlorite ($NaClO_2$, 80%), technical grade, Sigma Aldrich), acetic acid glacial (100%, Emparta grade, Merck), and sulphuric acid (H_2SO_4, 98%, Emparta grade, Merck) were used as without further purification[19].

2.2 *Rice straw pre-treatment and nanocellulose preparation*

Process parameters that were used in the preparation of α-cellulose from rice straw were considered as per the previous report by Sharma et al. 2018[19]. The extraction of cellulose from biomass is a process with two stages which involves pre-treatment and delignification. The rice straw was initially submerged in hot water and stirred for 1 hour to impurities, followed by drying overnight at 40 °C. 10 gm of the powdered rice straw was treated with 12 wt% NaOH at 121 °C for 1.5 h, maintaining a bath ratio of 1:10. The purpose of the procedure was to extract the cellulose and eliminate the lignin and hemicelluloses from rice straw. The obtained solution was washed with water to neutralize the pH. The sample was treated with acidified sodium chlorite (5 wt%) for delignification, with the pH adjusted with 1 M glacial acetic acid. thereafter, a white residue termed cellulose was produced and washed with distilled water to neutralize it followed by drying overnight at 50 °C. finally, the dried cellulose was homogenized using an ultra-high-pressure homogenizer (Microfluidics M-110P) followed by freeze-drying at -94 °C.

2.3 *Characterization*

The nanocellulose fibers so prepared were characterized using thermogravimetric analysis (TGA) and the expected functional group was determined by thermal stability and crystallinity[19].

2.3.1 *Fourier-transform infrared spectroscopy (FTIR)*

The FTIR-ATR spectra of samples obtained at different stages were analyzed using an instrument (Agilent Technologies Cary 600) in the range of 400-4,500 (in cm^{-1}).

2.3.2 *Thermogravimetric analysis*

The thermogravimetric analysis of the obtained CNF was analyzed by a thermogravimetric analyzer (Mettler Toledo thermal analyzer) in a temperature range of 25-800 (in °C) at a heating rate of 10 °C/min in a nitrogen atmosphere.

2.3.3 *X-ray diffraction (XRD)*

Diffraction patterns and crystallinity of the solid fibers were carried out using an X-ray diffractometer (XPERT-PRO D8Bruker) equipped with Cu $K\alpha$ (λ = 0.154 nm) in the 2θ range 5-40°. The empirical method was used to obtain the crystallinity index, X of samples as shown in Eq. (1).

$$X_C = \frac{I_{cry} - I_{am}}{I_{xc}} \times 100 \quad (1)$$

where I_{cry} and I_{am} are the peak intensities of crystalline and amorphous materials, respectively. Scherrer Eq. (2) is used

to calculate the crystalline size.

$$\tau = \frac{\kappa\lambda}{\beta \cos\theta} \tag{2}$$

diffracting planes with Miller Indices of hkl, λ is the wavelength of the X-ray radiation (λ = 0.154 Å), and $\beta_{1/2}$ is the full width at half maximum (FWHM) of the diffraction peaks.

2.4 *Scale-up study of CNF fibers*

The CNF production was scaled up from laboratory scale to commercial scale production, with the inventory as mentioned in Table 1.

Table 1. Translation of laboratory to pilot scale process

Laboratory scale process	Scaled-up process
Rice straw particle reduction	
Mortar pestle, grinder, Milling	Super Mass Collider/Grinder
Reaction under heating	
Magnetic stirrer	Insulated batch reactor with in-tank stirring
Blending	
Homogenization	Rotor-stator type homogenizer
Filtration	
Membrane filtration, reverse osmosis, dialysis, Sieving, Centrifugation	Vacuum filter, basket centrifuge, bowl centrifuge
Drying	Vacuum drying
in oven/Vacuum drying	Tray drying
Transferring of liquids	
Manual	Pumping
Others	
Waste disposal	Recycling/ Co- and by-product separation Heat recovery through heat exchangers Water recovery

The pilot-scale CNF production started with a 100 L stainless-steel reactor. Based on that process, a simplified process flow diagram was designed, including all the essential raw materials, machinery, and equipment (See supporting information Figure S1). Initially, rice straw was pre-treated in the reactor using (12 wt%) weight percent NaOH with a bath ratio of 1:10 at 121 °C for 1.5 hours under constant agitation of 940 rpm. Here, the reaction mixture was cooled from 75 °C to 35 °C using cooling water. The reaction mixture was washed and neutralized in a basket centrifuge

with a capacity of 50 L at 150 rpm. thereafter, the moisture content was determined in the pre-treated sample before delignification using a moisture analyser (Aczet MB 200). The pre-treated sample had a dry weight of 0.99 kg (around 1 kg), which was less than its wet weight of 2.75 kg with a moisture content of 64%. Using acidified $NaClO_2$ (5 wt%) at a bath ratio of 1:10 (pH 3-5), the delignification of the pre-treated dry sample was carried out at 75 °C under constant stirring of 656 rpm. Approximately 540 g of the delignified sample known as α-cellulose was obtained after centrifugation. Later α-cellulose was homogenized using Ultra High-Pressure Homogenizer (Microfluidics M-110P) at 20,000 Psi followed by freeze drying at -94 °C for 48 hrs (0.0254 mbar vacuum pressure). Further, the data obtained from the above steps were used to perform an LCIA study to make a comparison between the environmental impacts of the production procedure.

2.5 *Calculations*

Some important equations have been taken care of for the ease of stoichiometric calculations.

Heat energy required for air drying the sample = Heat energy to raise temperature to 100 °C + latent heat to remove water (3)

Heat energy required in freeze drying = Latent heat of sublimation (4)

The net yield of CNF was calculated as the ratio between the weight of the freeze-dried CNF and the initial weight of biomass taken.

2.6 *Heating energy for pre-treatment*

The heating energy required for pre-treatment (Q_{react}) of the rice straw, mainly includes the heat energy needed to raise the reaction mixture temperature up to the desired level (Q_{heat}) and to maintain that level (Q_{loss}) during the reaction. To calculate this, the sum of Q_{heat} and Q_{loss} was divided by the efficiency of the heating device (η_{heat}) (Equation (5)). Thus, for the calculation of the required energy equation 5 was applied.

$$Q_{react} = \frac{Q_{heat} + Q_{loss}}{\eta_{heat}} = \frac{C_p \times m \times (T_r - T_o) + A \times \left(\frac{k_a}{s}\right) \times (T_r - T_{out}) \times t}{\eta_{heat}} \quad (5)$$

$$Q_{loss} = A \times \left(\frac{k_a}{s}\right) \times (T_r - T_{out}) \times t \quad (6)$$

where Q_{react} = heating energy required for the reaction; Q_{heat} = energy to reach the reaction temperature; Q_{loss} = energy to compensate the heat loss; η_{heat} = efficiency of the heating device; C_p = specific heat capacity; m_{mix} = mass of the reaction mixture; A = reactor surface area; k_a = thermal conductivity of the insulation material; s = thickness of insulation; T_o = starting temperature; T_r = reaction temperature; T_{out} = temperature outside of reactor; t = time of reaction.

2.7 *The heat energy to reach the reaction temperature (Q_{heat})*

The specific heat capacity (C_p) of the solution is the amount of heat energy required to raise the temperature of one kilogram of the substance by one degree Celsius. If the solution is a mixture having different solvents then its specific heat capacity can be evaluated by averaging the mole fraction of the pure component[20].

The specific heat capacity of the solution can be calculated according to the equation:

$$Q_{heat} = C_p \times m \times (T_r - T_o) \quad (7)$$

Generally, the reactor is not filled completely and leaves some space for air or other gases involved in the reaction. However, the contribution of the mass of the gases can be neglected due to its extremely low mass compared to the components of the reaction mixture.

2.8 *The heat energy to compensate for the losses (Q_{loss})*

The heat loss occurring through the surface of the reactor should be balanced to maintain the required temperature in the reactor. The heat losses vary according to the jacket covering the reactor or the heating coil inside the reactor. The insulation part was only considered for the simplification of calculations, and conduction losses were neglected. To calculate the heat loss based on the procedure described above, the following equation was applied.

$$Q_{loss} = A \times \left(\frac{k_a}{s}\right) \times (T_r - T_{out}) \times t \quad (8)$$

For the pre-treatment stage in the reactor of 1,001 capacity, the following values were inserted:
A = 1.078 m^2, k_a = 0.035 W/(m*K), s = 0.08 m;
T_r = 394.15 K, T_{out} = 313.15 K, T_o = 301.15 K, t = 5,400 s.
This results in an energy requirement equal to the heat loss of:
Q_{react} = 256,317.68, J = 0.071 kWh.

2.9 *Cooling phase*

The reaction mixture was cooled by replacing steam with hot water to reduce the temperature to a level of 75 °C, and for further reduction till 35 °C cooling tower water was used.

2.10 *Delignification of rice straw*

Delignification of the pre-treated dried sample (~1 kg) was performed using acidified $NaClO_2$ (5 wt %) at a bath ratio of 1:10 (pH 3-5). The reaction mixture was kept at 75 °C under constant stirring of 656 rpm. Steam was supplied to maintain 75 °C temperature for 1.0 hr. Thus, the energy required for delignification would be:

$$Q_{react} = \frac{C_p \times m \times (T_r - T_o) + A \times \left(\frac{k_a}{s}\right) \times (T_r - T_{out}) \times t}{\eta_{heat}} \quad (9)$$

where, T_r = 348.15 K, T_{out} = 303.15 K, T_o = 306.25 K, t = 3,600 s, Q_{react} = 94,850.306, J = 0.026 kWh.

2.11 *Life cycle assessment*

2.11.1 *Goal and scope*

The goal of the LCA was to generate data on the environmental impact of CNF production at a commercial scale compared to the laboratory scale process. The cradle-to-gate life cycle assessment starts with the waste of rice straw to produce CNF. The laboratory-scale production process already described in our previous publication was modeled for larger-scale production, to draw a comparison of their environmental impact[19,21]. The system boundaries consider all the unit operations involved in the process, including alkaline treatment and delignification. Chemical and energy consumption are evaluated for these unit processes at the laboratory and pilot scale. During these assessments,

building construction and commissioning and decommissioning of infrastructure and equipment are not included. The starting material rice straw derived from waste was considered a free input, carrying no load into the LCA. Further, the hemicellulose and lignin produced as by-products during the pre-treatment and delignification step can be used in other processes and, therefore, considered valuable products.

2.11.2 *Life cycle inventory (LCI) analysis*

Under the LCI analysis, all the inputs and outputs from the process are quantified within the system boundaries. The inputs to the LCI analysis include natural resources and materials/fuels (raw materials, chemicals, energy requirements, etc.), and outputs include output to the technosphere (products and by-products) and output to nature (emissions to air, water, soil; non-material emissions, economic issues, social issues, waste to treatment, etc.). Ecoinvent database was used for life cycle inventory analysis in the present study for inputs such as chemicals (NaOH, NaOCl, Glacial acetic acid), raw material, water from unspecified natural resources, and energy consumption. Emissions to water resulting from the process were not included as it was recycled and reused, and no release in the environment was observed. Some important steps are followed during LCI assay, i.e., Designing a process flow diagram within the system boundaries, formulating a data collection methodology to meet the goal and scope of the study, collecting and validating the functional units assumed for each unit process, and finally reporting the results.

2.11.3 *Life Cycle Impact Assessment (LCIA)*

For the life cycle impact assessment of the process, different methodologies present in the SimaPro (version 9.2) software were exploited. Out of the methods available in the software, such as European, North American, Global, Single issue, Water footprints, and others, the commonly used global methodology, i.e., ReCiPe 2016 (with the hierarchic perspective), was implemented in the present study. With the ReCiPe 2016 methodology, 18 midpoint and 3 endpoint indicators were calculated[22]. The three endpoint indicators, including ecosystem quality, human health, and resource depletion, were associated with high levels of uncertainty. Therefore, 18 midpoint indicators discussing the global warming potential, agricultural land occupation, ozone formation, human carcinogenic toxicity, fossil depletion, etc., were explored.

2.11.4 *Techno-economic analysis*

The techno-economic analysis was carried out to systematically evaluate the effect of significant cost categories on sustainability and the large-scale application of the process. Through the analysis, the total cost of production, including the capital cost, utility cost, raw materials cost, and the competitiveness of the process in the market, was investigated. The equipment cost of the process was adapted from the National Renewable Energy Laboratory (NREL) report and the economic analyzer[23,24]. To estimate the impact of the uncertainties in input variables on the life cycle cost of the process, sensitivity analysis was also carried out using different methods.

2.11.5 *Capital expenses*

The total capital expenses of the process can be calculated by evaluating the cost of the process equipment and utilities. The prices for the equipment used in the present study have been sourced, and a cost correlation was applied where equipment of different capacities was utilized. The chemical engineering plant cost indices (CEPCIs) were used for cost appreciation[25].

2.11.6 *Operational cost*

The variable operational expenditures were categorized as fixed and variable. Fixed operating expenses recur annually regardless of the product of that year. These expenses include annual maintenance charges, taxes, insurance, and labor costs. Regarding the capital cost, the fixed operating expenses were calculated. The variable operational cost comprises raw materials and utilities such as boiler feedwater, electricity, steam, and high-speed diesel. All materials required were scaled to account for plant-wide inefficiencies to meet the specified plant capacity.

2.11.7 *Total cost*

The total cost of production was the addition of the fixed capital cost and variable operational cost. Depreciation @ 10% on plant and machinery was considered.

3. Results and discussion

Production was scaled up based on previously optimized parameters for preparing CNF at the lab scale. For each procedure, the stage efficiency was determined. Purified cellulose content appears to have significantly increased from 46.5% to 91.2%, whereas hemicellulose content decreased from 31.5% to 5.34% followed by lignin content (21.5% to 1.32%) (Table 2). The table showed that after chemical treatment, nearly all of the hemicellulose and lignin content were removed from the fibers.

Table 2. Chemical composition analysis of rice straw components at each stage of the treatment

Material	Scale	α-cellulose content (in %)	Hemicellulose content (in %)	Lignin content (in %)
Untreated rice straw		46.5	31.5	21.5
Alkaline cellulose fibers	Lab scale	72.3	17.1	10.6
	Pilot scale	79.02	12.07	8.91
Sodium chlorite-treated fibers	Lab scale	82.64	10.99	4.97
	Pilot scale	87.31	7.63	5.06
Cellulose nanofibers	Lab scale	91.2	5.34	1.32
	Pilot scale	92.36	4.5	2.64

3.1 *Characterization Attenuated Total Reflectance Fourier Transform Infrared Spectroscopy (ATR-FTIR)*

FT-IR spectrum of the chemically treated rice straw fibers is shown in Figure 1a. The smaller peak at 1,732 cm^{-1} in the untreated rice straw attributed to the characteristics of esters in lignin or hemicelluloses, which was removed after chemical treatment indicating near cleavage of the ester bonds. The spectra in the region of 3,400 cm^{-1} showed the broadened absorption band characteristic of the OH stretching. A minor peak at 2,897 cm^{-1} was identified attributed to the aliphatic saturated C-H stretching vibration in cellulose and hemicelluloses. In response to the OH bending of the absorbed water, the peak intensity at 1,640 cm^{-1} has greatly enhanced in CNF. The peak at 1,266 cm^{-1} was sharply reduced after chemical treatment, revealing the removal of hemicelluloses. The partial acetylation of -OH groups in both polysaccharides and remaining lignin caused C-O stretching that gave rise to the peak at 1,060 cm^{-1}. The glycosidic bonds between the glucose units in cellulose present in chemically treated fibers are responsible for the gradual increase in peak at 897 cm^{-1}. Further, the lignin was removed from the extracted CNF which was later confirmed by chemical composition analysis (Figure 1a).

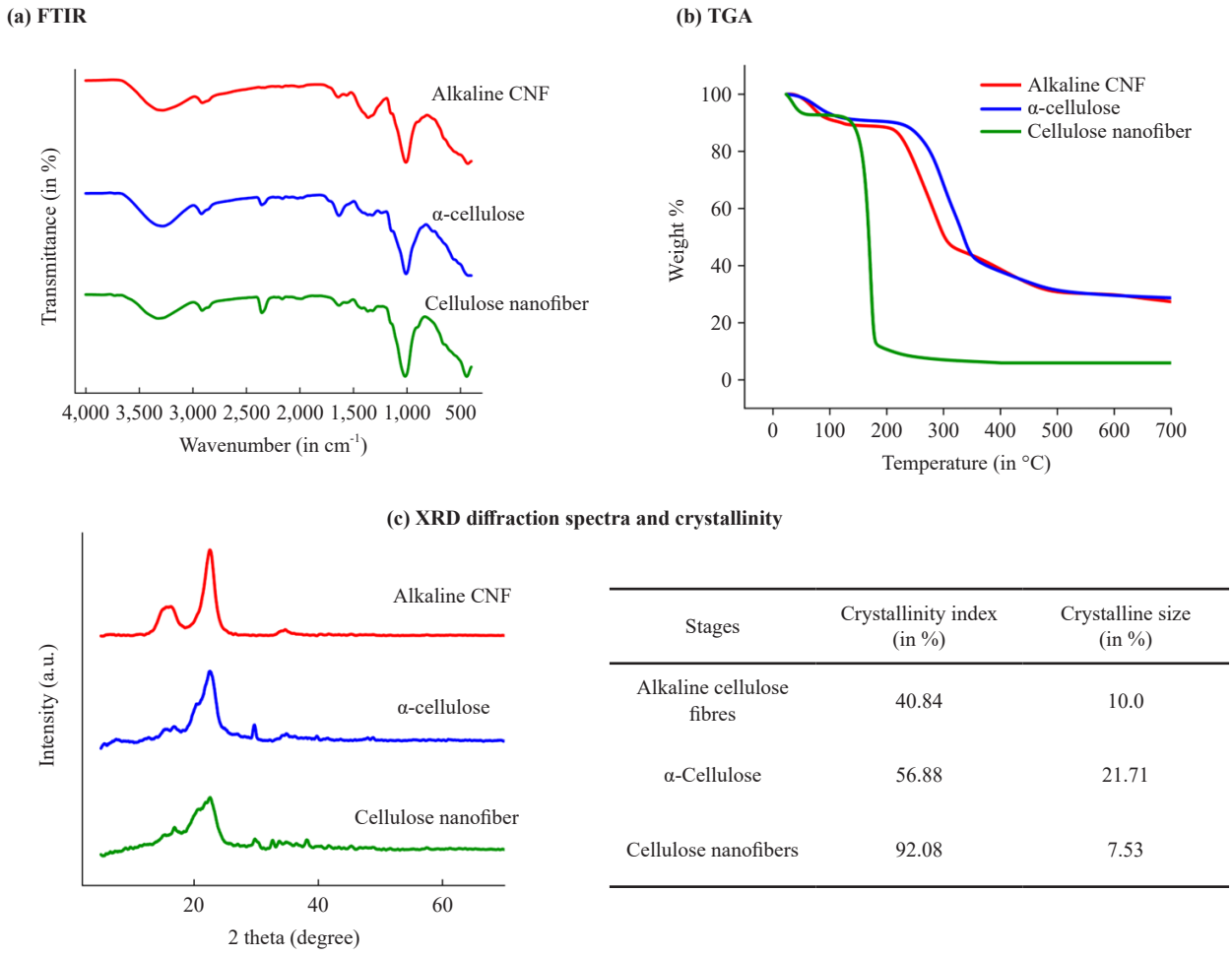
3.2 *Thermal gravimetric analysis (TGA)*

According to Figure 1b of the TGA curve, the thermal degradation of CNF was higher than that of cellulose and

alkali-treated cellulose, respectively. The partial/complete removal of hemicelluloses, lignin, and pectin from the fibres and the increased crystallinity of cellulose can be attributed to the higher temperatures for thermal degradation of the purified CNF. Initial weight loss can be attributed to water loss or loss of low molecular weight compounds in the range of 35-150 °C. The second stage of degradation can be attributed to cellulose and hemicelluloses pyrolysis from 260 °C to 310 °C. The aforementioned results were consistent with results obtained from XRD and FTIR analysis. The significant second decomposition peak for alkaline cellulose fibre at about 321.86 °C was attributed to lignin degradation. In CNF, cellulose breakdown was indicated by a strong peak at 437.58 °C. All of these results show a considerable improvement in the thermal stability of rice straw fibres.

3.3 *X-Ray diffraction (XRD)*

X-ray diffraction analysis for extracted nano cellulose at various stages of extraction is shown in Figure 1c. According to the XRD data, there were three distinct peaks in the rice straw CNF that were extracted at the diffraction angles (2) = 15°, 22.5°, and 34°, which correspond to the crystalline cellulose. However, the crystallinity and crystallite size of cellulose are influenced by chemical and mechanical processes. It is inferred that crystallinity increases with increasing cellulose content in the processed sample (Figure 1c). Further, treatment with strong alkali treatment (NaOH, 12%), patly releases the amorphous and crystalline regions of cellulose.



Stages	Crystallinity index (in %)	Crystalline size (in %)
Alkaline cellulose fibres	40.84	10.0
α-Cellulose	56.88	21.71
Cellulose nanofibers	92.08	7.53

Figure 1. Characterization study of extracted nanocellulose at different stages (a) FTIR (b) TGA (c) XRD Life cycle assessment

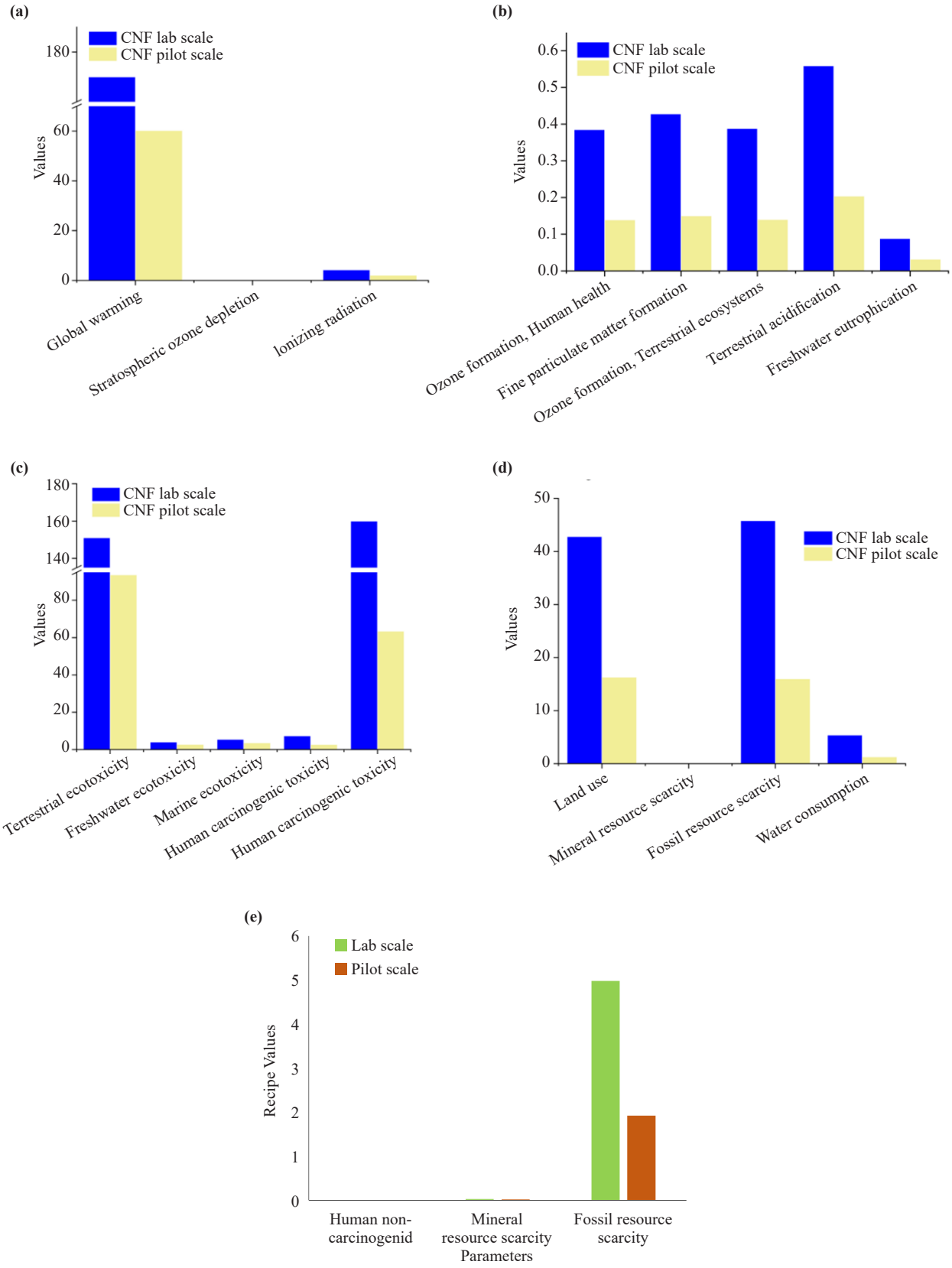


Figure 2. Influence of the CNF production process at laboratory and pilot scale on environmental impact categories using ReciPe 2016 (midpoint analysis) (a-d). (e) Comparing the LCIA major affected parameter of CNF production at laboratory and pilot scale using ReciPe (2016) endpoint method

To develop a green and sustainable process, it is vital to ensure its environmental impacts[26]. The current research work presents a cradle-to-gate approach for the life cycle assessment of the process at the laboratory as well as on a pilot scale, incorporating the previous impacts of the production of the electricity and chemicals used in the process. ReciPe 2016 methodology (incorporating the CML-IA approach of problem-based analysis and Eco-indicator 99 approach of damage-based analysis) was employed to investigate and compare the various impact categories observed during the production of 1 kg of nanocellulose fibers, at both scales. With the Hierarchical perspective (H), both the midpoint (problem-oriented) and endpoint (damage-oriented) impact categories were studied. The categorization factors such as Global warming, Stratospheric ozone depletion, Ionizing radiation, Ozone formation, Terrestrial ecotoxicity, Freshwater eutrophication, Marine eutrophication, Terrestrial ecotoxicity, Freshwater ecotoxicity, Marine ecotoxicity, Human carcinogenic toxicity, Human non-carcinogenic toxicity, Mineral resource scarcity, Fossil resource scarcity, and water consumption, were represented for global-scale evaluation of the impacts, as shown in Figure 2.

The climate change impact of the CNF production process at the pilot and laboratory scale has been presented in Figure 3. In the analysis of Global Warming Potential (GWP), the laboratory scale process showed a higher potential of 170 kg CO_2 eq. per kg of CNF, whereas the pilot scale showed much less impact of 60.2 kg CO_2 eq. During the lab-scale process, energy consumption (electricity) alone contributed more than 90% of the total climate change impact. However, with the scale-up of the process, a 60% contribution of electricity and a 36.8% contribution of NaOH were observed. The lab scale process findings help identify the limitations of each step from an environmental perspective. Thus, after better insight into the process, its industrial scale application was carried out, reflecting a 7.5 times reduced impact on the environment. Significant damage to aquatic ecosystems detected in both scales was mainly due to the electricity utilization at various stages of the process. However, the chemicals used in the process, including NaOH, $NaClO_2$, and acetic acid showed less influence. These observations identified electricity consumption as the key contributor to environmental sustainability.

The release of huge amounts of CO_2, heavy metals, organic compounds, and wastewater into the atmosphere during the production of electricity and chemicals utilized during the process contributes to the increased GWP at the laboratory (161) and pilot scale (36.8). In corroboration of the above-said results, studies on MCC (Micro Crystalline Cellulose) production also reported the key role of energy (electricity) and raw materials in GWP impact[27,28]. Freshwater and marine eutrophication potential at the pilot scale were three times less than that observed during laboratory-scale production of CNF. Among techno-spheres input, rice straw showed an almost negligible effect on all the impact categories due to its amicability towards the environment.

Katakojwala and Mohan (2020)[5], recently compared LCA results for MCC production from sugarcane bagasse through three different methodologies. The climate change impacts for MCC1, MCC2, and MCC3 methods were reported as 261.35, 203.755, and 146.677 kg CO_2 eq., respectively. While carrying out the production of MCCs, electricity and deionized water consumed for various stages, along with sodium hydroxide utilization for pre-treatment were shown as the significant contributors. These results indicated a 10 times higher impact on GWP when compared with the process developed in the present study[5]. Leao et al. (2017)[14] studied gate-to-gate processes for cellulose nanocrystal production from sugarcane bagasse, considering the environmental impacts of value-added products. The GWP for the processes was reported in 13.7 to 3,950 kg CO_2 eq. The values obtained were very high, compared with the cradle-to-gate study of the present work where each input and output of the process was considered[14]. Another study on nanofibrilated cellulose production represented total impact scores of 807 kg CO_2 eq based on the ReciPe 2016 method. The impacts on human health and aquatic ecotoxicity were 20 to 30 times higher than obtained with the pilot scale study of the present work (Turk). In the report by Figueiredo et al. (2012), the GWP for 1 kg of CNCs from coconut fibre was reported as 1.08×10^3 kg CO_{2eq}[29]. However, Husgafvel et al. (2016)[30] reported GWP for eight different methods within 1.217×10^3 to 3.634×10^6 kg CO_2 eq. Another study by[31] found the contribution of 1 kg production of CNF to GWP equivalent to 0.7-1.3 $\times 10^3$ kg CO_2 eq. The report identified the use of sulfate-bleached pulp and heat energy from wood residues as the main contributors to the impacts[31]. The present report showed a radically lesser effect of the nanocellulose fiber production from rice straw compared to the reported literature.

The comparison between laboratory scale and pilot scale production of CNF was also demonstrated using endpoint indicators from the ReCiPe (E/A) method (Figure 2e). The results indicated a dramatically decreased impact in almost all the categories with the scale-up of the production procedure. Even when the highest impacted category (fossil resource scarcity) was compared, the commercial scale showed an 82% reduced impact than the laboratory scale

procedure. The effect of GWP, on human health at a laboratory scale was 0.00205, whereas, with the scale-up of the process, it was reduced by a factor of 10 to 0.000723. Considering the credit from the heat and water recycling recovery at the pilot scale, the results shift more towards a sustainable process. A report on predicting the environmental impact of future nanocellulose production at an industrial scale also showed a many-fold reduction in all the impact scenarios with the scale-up of the production[32]. These findings play an important role in recommending lab-scale process improvements concerning industrial-scale production.

3.4 *LCIA representation through Sankey diagrams*

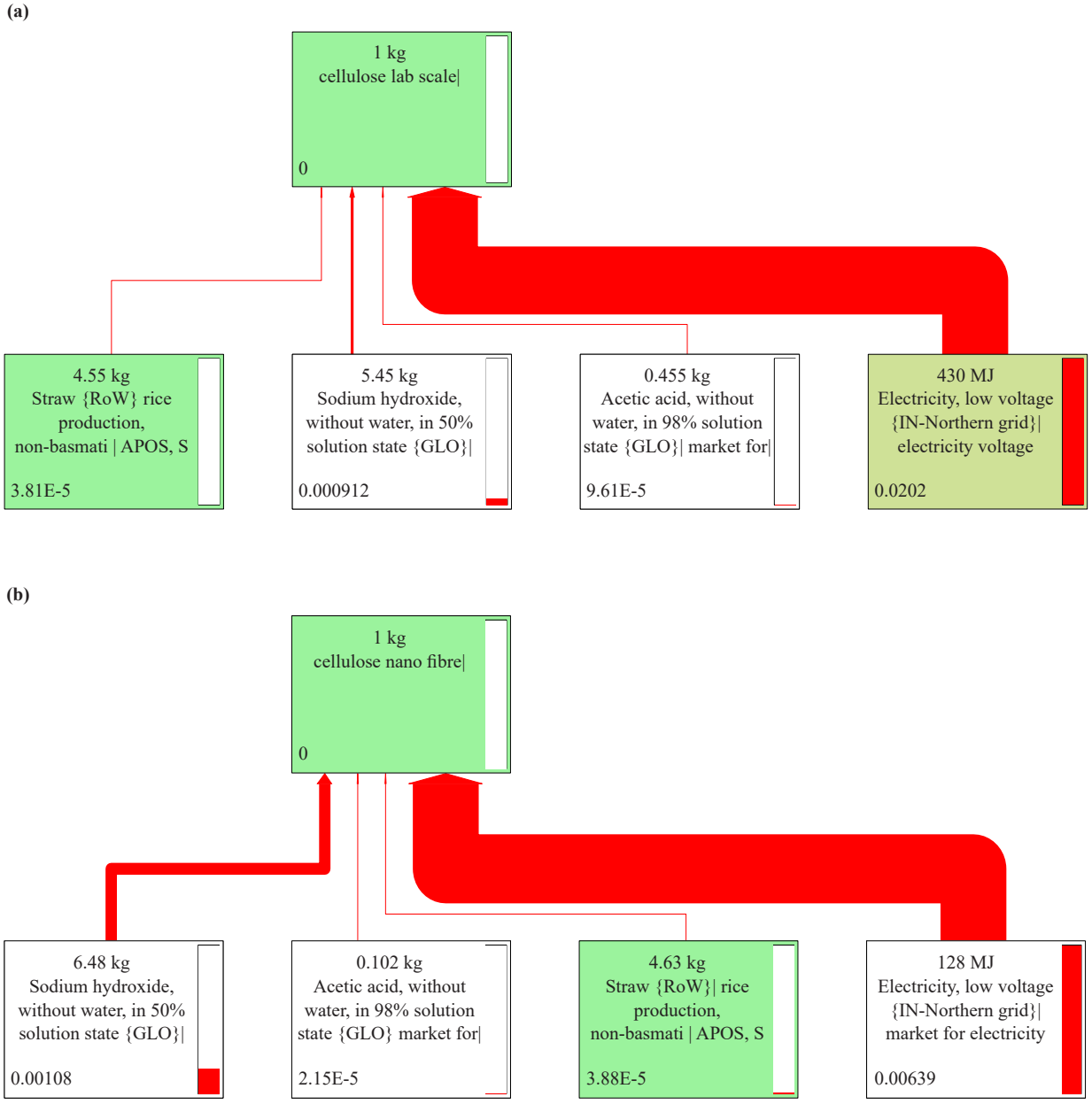


Figure 3. Network diagram for CNF production at (a) laboratory scale and (b) pilot scale for LCIA data

The LCIA results were mainly reported as independent and relative scores of each input to the process. The scores

can also be represented as Sankey diagrams. These diagrams were complex looped systems in the LCA studies having many repeated units, presented linearly by merging the processes, as shown in Figure 3[33]. In the diagram, the width of the lines represents amounts of resources and the value displayed in the box refers to the contribution of the single unit in the process. As shown in Figure 3, electricity has a significant contribution to GWP for the laboratory scale process. In contrast, in the pilot scale, the change in the width of the lines represents a lesser effect of electricity and sodium hydroxide on the environment. The network diagrams again displayed the reduction in the environmental impacts with the scale-up of the process of the CNF production from rice straw.

3.5 *Techno-economic analysis of the pilot scale study*

3.5.1 *Total cost investment*

The cost investment for the production of 15 kg/day of cellulose nanofibre was evaluated for economic analysis of the process and to determine the minimum selling price of the product. The total cost includes fixed (equipment, installation, piping, electrical systems, building, and utility facilities) and variable costs (raw material, electricity, water, chemicals), as presented in Table 3. Detailed energy and mass flow balances were studied to estimate the consumption of raw materials, chemicals, water, and electricity[34].

Table 3. Summary of fixed capital investment (FCI) estimation and utility consumption for the CNF production (15 kg/day)

Units	Cost (in Rs.)	Data source
	Plant & Machinery cost (Rs. in lakhs)	
Equipment	129	Stoichiometric analysis based on the study
Piping	38.7	
Electrical	25.8	
Instrumentation	12.9	
Total	206.4	
Depreciation	10 years	
	Total cost per kg of CNF	
Cost of Goods	621.13	Elementary calculations based on the study
Depreciation	573.33	
Other overhead charges	20	
TOTAL	~1,215	

The total cost of production for CNF from rice straw was calculated using process development studies reported by the National Renewable Energy Laboratory (NREL) and Aspen Process Economic Analyzer[24,35]. The cost for other subsystems, such as utilities and storage, was developed based on the internal cost database and costs mentioned in NREL[23]. For the 15 kg/day batch, the minimum selling price (MSP) of CNF was calculated as Rs. 1,215/kg, whereas, with increased batch capacity i.e., 30 kg/day and 45 kg/day, the MSP got reduced to Rs. 1,098/kg and Rs. 1,072/kg, respectively. The reduced cash flow calculations for CNF were based on numerous economic factors and assumptions.

The costs for all the raw materials and equipment utilized in the process were indexed using the Chemical Engineering's Plant Cost Index (CEPCI) for the year 2021.

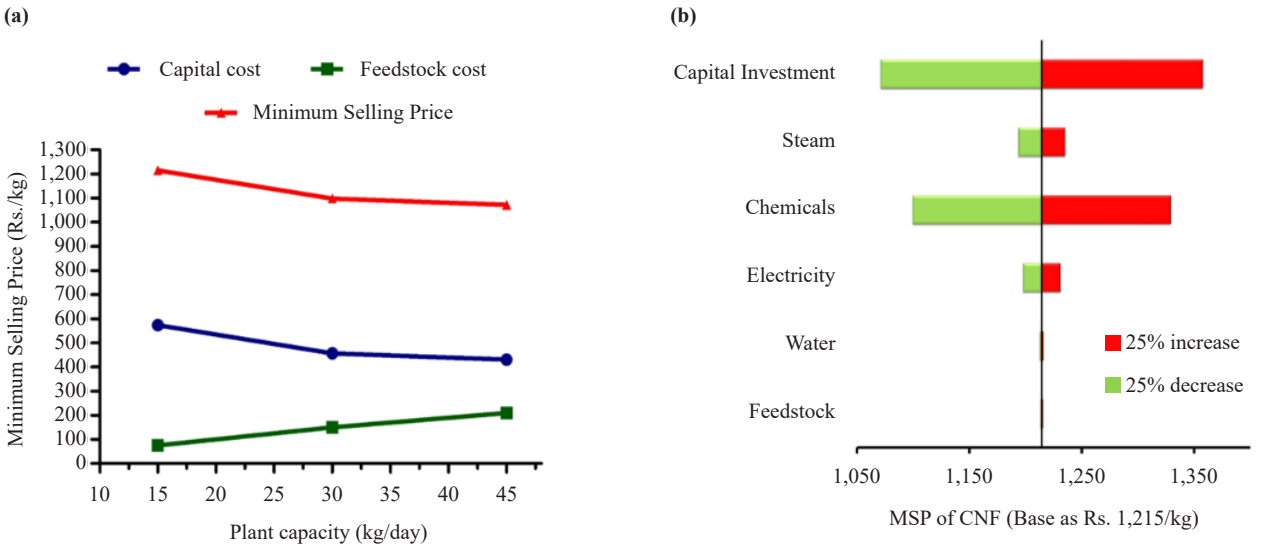
Phase-wise distribution of the production cost specified that chemicals utilized during the pre-treatment and delignification stages contributed 55% and 27.67% of the total variable cost, respectively. These findings indicated that the recycling of chemicals and solvents might substantially impact the total finances. The fixed capital cost accounted for ~47% of the total cost, and chemicals and electricity accounted for the rest of 30%. The fixed capital cost for instruments and utilities was mainly dominated by the pre-treatment stage, ~56% of the fixed capital cost. Depreciation cost was included for @10 yrs, to show its effect on fixed-cost investment. With the variations in the plant capacity of 15 kg/day, 30 kg/day, and 45 kg/day, the minimum selling price for CNF was evaluated as $16,537.36/tonne, $14,944.87/tonne, and $14,590.98/tonne, respectively (The cost conversion rate was taken as 1 $ = 73.47 Rupees).

The primary cost drivers were identified as chemicals and capital cost, followed by electricity in all cases. A reduction in capital cost and minimum selling price can be demonstrated with an increase in the plant capacity; however, it led to an increase in feedstock cost, as shown in Figure 4a. On the contrary, in our study, the feedstock used was a waste (rice straw) that was available at a meager cost, thus not influencing the overall investment cost to a larger extent. However, it fulfills the objective of waste valorization and sustainable development. The plant capacity at which an optimal MSP can be obtained represents the best option for investment, i.e., 30 kg/day for the present work.

3.5.2 *Sensitivity analysis*

Sensitivity analysis was performed to assess the impact of variations in each input on the scenario of the lowest MSP. For the analysis, the input cost of all the parameters, including fixed and variable, were assorted by ± 25% independently, and their impact on the MSP was recorded. Inputs evaluated were: the cost of chemicals, feedstock, electricity, water, steam, and capital investment. The sensitivity of CNF on MSP was presented as a Tornado diagram, as shown in Figure 4b.

The results showed the highest sensitivity of MSP of CNF towards the capital investment, followed by the cost of chemicals, Figure 4b. The feedstock being a natural resource, represents the least impact on MSP even with the variation of ± 25%. Although, cellulose obtained from other sources reported in the literature such as wood ($450-500/t), grasses ($45-170/t), and jute fiber ($390-780/t), showed relevant impact during sensitivity analysis[36]. The sensitivity analysis identified the chemical cost as one of the foremost cost drivers among all other variable parameters. Therefore, the recycling of chemicals and solvents should be promoted to reduce the impact on total finances. Regarding the rest of the cost drivers, such as steam price and electricity, examinations related to the heat exchanger networks and the use of renewable energy resources should be incorporated into the study. Further to improve the commercialization of the product, the valorization of other by-products such as hemicellulose and lignin should be considered.



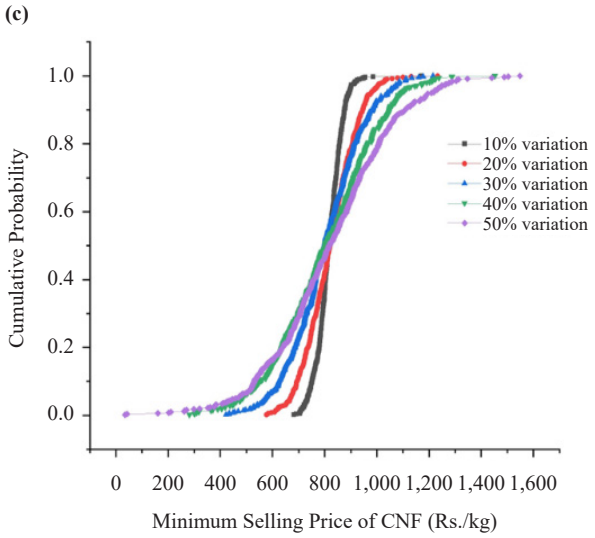


Figure 4. (a) Variation of minimum selling price according to plant capacity and contribution of feedstock and capital cost (b) Sensitivity tornado chart of the economic parameters in the proposed process (c) Cumulative distribution function to the MSP of CNF obtained using Monte-Carlo simulation by varying the range of influential parameters

3.5.3 *Sensitivity analysis*

The influential factors identified through sensitivity analysis were selected to perform the Monte-Carlo simulation to understand the chances for an event to happen and to enumerate the risks linked with the process under unpredictable environments. Earlier, for the techno-economic analysis, it was assumed invariability in each parameter despite the inherited uncertainties of the system. Through sensitivity analysis, it was not feasible to characterize an assimilated effect of each of the parameters on the economics of the process. Therefore, an uncertainty analysis was performed using a cumulative distribution function based on the influential inputs (capital investment, chemicals, steam, and electricity) identified in the sensitivity analysis. Figure 4c represents cumulative probability concerning the MSP of CNF obtained using Monte-Carlo simulation. The influential parameters varied over the range of 10 to 50%. The impact of the increased variations in the range of the parameter could be seen with an increase in uncertainty. A shift in the curves in both directions was observed as the variation percentage increased from 10% to 50%. The results showed that the cumulative probability of the MSP of CNF was 0.49. It indicates a 49% probability of having a selling price lesser than the MSP of CNF and a 51% probability of having an increased cost. Uncertainty analysis was carried out to quantify the inherited risk in the immature technologies and to develop insight into the decision-makers of the innovative processes.

4. Conclusion

The present study has developed a robust methodology for producing nanocellulose from rice straw in an environmentally and economically sustainable manner. A life cycle assessment was performed to identify the environmental impact of all the inputs and outputs of the process at the laboratory and the pilot scale. Electricity consumed during the production of CNF was identified as a key contributor to LCIA at the laboratory scale. However, reduced global impacts were observed with the scale-up of the process, mainly because of the industrial heating systems utilizing natural gas or other fossil fuels as their source of energy. A difference of three times lesser GWP was observed during pilot-scale production due to less dependency on electricity. The cost of CNF production from rice straw was calculated as $14,590.98/tonne at a plant capacity of 45 kg/day, and it was found to be much less when compared with other methods of production. Further, to quantify the inherited risks in the developed technology, sensitivity and uncertainty analyses were performed using Monte-Carlo simulation. Out of all the parameters contributing to the total cost of production, the capital cost of investment and chemicals cost were identified as the most influencing factors. A

deeper insight into decision-making was attained through uncertainty analysis by incorporating the risk associated with present uncertainties. The outcomes of this study will help in the commercial-scale production of nanocellulose from agricultural wastes in an environmentally and economically sustainable manner.

Conflict of interest

The authors declare no competing financial interest.

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Appendix A. Process scale-up, techno-economic feasibility, and environmental impact assessment of cellulose nanofibre production from laboratory to pilot scale

A1. *Procedure for the extraction of Cellulose from Rice straw on a lab scale*

A1.1 *Pre-treatment of rice straw*

Process parameters used in the preparation of α-cellulose from rice straw have been already optimized in our previous work[19]. 10 gm of the grounded fine rice straw powder was treated with 12 wt% NaOH at 121 °C for 1.5 h, maintaining a bath ratio of 1:10. After the treatment, the solution was neutralized by washing with water until the solution's pH became neutral.

A1.2 *Delignification*

For delignification, acidified sodium chlorite (5 wt%) was added (pH was adjusted with 1 M glacial acetic acid) to the sample. The reaction was carried out at 75 °C for 1 h with continuous stirring on the magnetic stirrer. The obtained delignified residue is known as α-cellulose.

A1.3 *Mechanical treatment*

α-cellulose was washed with distilled water and kept overnight for drying in a hot air oven at 50 °C. Dried sample was homogenized using ultra high-pressure homogenizer (Microfluidics M-110P) followed by freeze-drying at -94 °C.

Table S1. Translation of laboratory to pilot scale process

Laboratory scale process	Scaled-up process
Pestle mortar	Super Mass collider/Grinder
Other particle size reduction	
Other particle size reduction	
Reaction under heating (Magnetic stirrer)	Insulated batch reactor with in-tank stirring
Blending	Rotor-stator type homogenizer
Homogenization	
Filtration (e.g. membrane, reverse osmosis, dialysis)	Filtration (e.g. pressure filter, vacuum filter)
Sieving	
Centrifugation/cyclonic separation	Centrifugation (Basket centrifuge, bowl centrifuge, disk stack centrifuge etc.)
Other separation techniques	
Drying in oven/Vacuum drying	Vacuum drying/Tray drying
Transferring of liquids (Manual)	Pumping
Waste disposal	Recycling/Co- and by-product separation Heat recovery through heat exchangers Water recovery

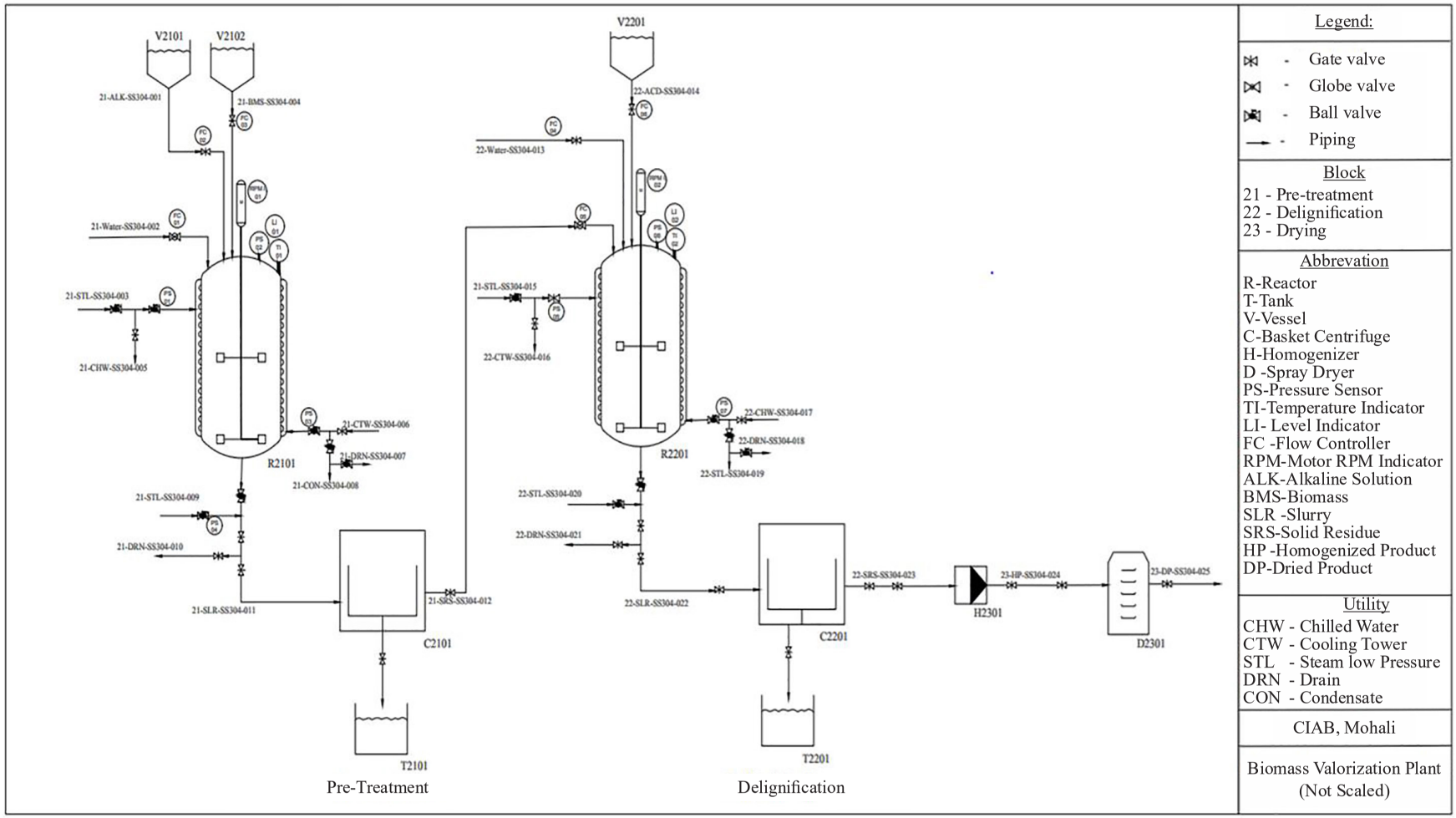


Figure S1. Cellulose nanofibre production from rice straw on a Pilot scale

A2. *Scaleup and process calculation from lab to pilot plant*

A2.1 *Heating energy for pre-treatment*

The heating energy required for pre-treatment (Q_{react}) of the rice straw, mainly includes the heat energy needed to raise the reaction mixture temperature upto the desired level (Q_{heat}) and to maintain at that level (Q_{loss}) during the reaction. To calculate this, the sum of Q_{heat} and Q_{loss} was divided by the efficiency of the heating device (η_{heat}) (Equation A1). Thus, for the calculation of the required energy equation A1 was applied.

$$Q_{react} = \frac{Q_{heat} + Q_{loss}}{\eta_{heat}} = \frac{C_p \times m \times (T_r - T_o) + A \times \left(\frac{k_a}{s}\right) \times (T_r - T_{out}) \times t}{\eta_{heat}} \quad \text{(A1)}$$

$$Q_{loss} = A \times \left(\frac{k_a}{s}\right) \times (T_r - T_{out}) \times t \quad \text{(A2)}$$

where Q_{react} = heating energy required for the reaction; Q_{heat} = energy to reach the reaction temperature; Q_{loss} = energy to compensate the heat loss; η_{heat} = efficiency of the heating device; C_p = specific heat capacity; *mmix* = mass of the reaction mixture; A = reactor surface area; k_a = thermal conductivity of the insulation material; s = thickness of insulation; T_o = starting temperature; T_r = reaction temperature; T_{out} = temperature outside of reactor; t = time of reaction.

A2.2 *Heat energy to reach the reaction temperature (Q_{heat})*

The specific heat capacity (C_p) of the solution is the amount of heat energy required to raise the temperature of one kilogram of the substance by one degree celsius. If the solution is a mixture having different solvents then its specific heat capacity can be evaluated by averaging the mole fraction of the pure component[20].

The specific heat capacity of the solution can be calculated according to the equation:

$$Q_{heat} = C_p \times m \times (T_r - T_o) \quad \text{(A3)}$$

Generally, the reactor is not totally filled, leaving some space for air or other gases involved in the reaction. However, the contribution of the mass of the gases can be neglected due to its extremely low mass compared to the components of the reaction mixture.

A2.3 *Heat energy to compensate the losses (Q_{loss})*

The heat loss occurring through the surface of the reactor should be balanced to maintain the required temperature in the reactor. The heat losses vary according to the jacket covering around the reactor or the heating coil inside the reactor. The insulation part was only considered for simplification of calculations, and conduction losses were neglected. To calculate the heat loss based on the procedure described above, the following equation was applied.

$$Q_{loss} = A \times \left(\frac{k_a}{s}\right) \times (T_r - T_{out}) \times t \quad \text{(A4)}$$

For the pre-treatment stage in reactor of 100 l capacity, the following values were inserted:

A = 1.078 m^2, k_a = 0.035 W/(m*K), s = 0.08 m;

T_r = 394.15 K, T_{out} = 313.15 K, T_o = 301.15 K, t = 5,400 s.

This results in an energy requirement equal to the heat loss of:
Q_{react} = 256,317.68, J = 0.071 kWh.

A2.4 *Cooling phase*

The reaction mixture was cooled by replacing steam with hot water to reduce the temperature to a level of 75 °C, and for further reduction till 35 °C cooling tower water was used.

A2.5 *Washing*

For washing and neutralizing the reaction mixture, it was fed into a basket centrifuge of 50 l capacity at 150 rpm. After centrifugation, dry weight of the pre-treated sample to be used for delignification was estimated by calculating the moisture content of the wet sample collected. Wet weight of the pretreated sample was found to be 2.75 kg and moisture content estimated using moisture analyzer (Aczet MB 200) was found to be 64%, thus the dry weight of the pre-treated sample would be 0.99 kg.

A2.6 *Delignification of rice straw*

Delignification of pre-treated dried sample (~1 kg) was performed using acidified $NaClO_2$ (5 wt %) at a bath ratio of 1:10 (pH of the solution was maintained between 3-5 using glacial acetic acid) in the reactor, fig. S1. The reaction mixture was kept at 75 °C under constant stirring of 656 rpm. Steam was supplied to maintain 75 °C temperature for 1.0 hr. Thus, the energy required for delignification would be:

$$Q_{react} = \frac{C_p \times m \times (T_r - T_o) + A \times \left(\frac{k_a}{s}\right) \times (T_r - T_{out}) \times t}{\eta_{heat}} \quad \text{(A5)}$$

where, T_r = 348.15 K, T_{out} = 303.15 K, T_o = 306.25 K, t = 3,600 s, Q_{react} = 94,850.306, J = 0.026 kWh.

A2.7 *Cooling phase*

Cooling water was used to lower down the temperature of the reaction mixture from 75 °C to 35 °C.

A2.8 *Washing*

For washing and neutralizing the reaction mixture, the mixture collected from the reactor was fed into a basket centrifuge running at an rpm of 1,500. After centrifugation, ~540 g of the delignified sample was collected.

A2.9 *Homogenization and freeze-drying*

The obtained delignified residue known as α-cellulose was homogenized using Ultra High-Pressure Homogenizer (Microfluidics M-110P) at 20,000 Psi followed by freeze drying at -94 °C for 48 hrs (0.0254 mbar vacuum pressure).

Table S2. Influence of the CNF production process at laboratory and pilot scale on environmental impact categories using ReciPe 2016 (midpoint analysis)

Sr. No.	Impact Category	Unit	CNF lab scale	CNF pilot scale
1	Global warming	kg CO_2eq	170	60.2
2	Stratospheric ozone depletion	kg CFC11eq	5.56E-5	2.55E-5
3	Ionizing radiation	kBq CO-69eq	4.26	2.08
4	Ozone formation, Human health	kg NOxeq	0.385	0.139
5	Fine particulate matter formation	kg PM2.5eq	0.428	0.150
6	Ozone formation, Terrestrial	kg NOxeq	0.388	0.140
7	Terrestrial acidification	kg SO_2eq	0.559	0.204
8	Freshwater eutrophication	kg Peq	0.0883	0.0316
9	Marine eutrophication	kg Neq	0.00609	0.0026
10	Terrestrial ecotoxicity	kg 1,4-DCB	151	93.7
11	Freshwater ecotoxicity	kg 1,4-DCB	4.04	2.77
12	Marine ecotoxicity	kg 1,4-DCB	5.44	3.57
13	Human carcinogenic toxicity	kg 1,4-DCB	7.3	2.73
14	Human non-carcinogenic	kg 1,4-DCB	160	63.4
15	Land use	M2a cropeq	42.8	16.3
16	Mineral resource scarcity	kg Cueq	0.0913	0.0663
17	Fossil resource scarcity	kg oileq	45.8	16
18	Water consumption	m^3	5.39	1.27

Table S3. Comparing the influence of the CNF production process at laboratory and pilot scale on environmental impact categories using ReciPe 2016 (endpoint analysis)

Sr. No.	Impact Category	Unit	NCF laboratory scale	NCF pilot scale
1	Global warming, Human health	DALY	0.00205	0.000723
2	Global warming, Terrestrial	species.yr	4.1E-6	1.45E-6
3	Global warming, Freshwater	species.yr	1.12E-10	3.94E-11
4	Stratospheric ozone depletion	DALY	1.1E-7	4.75E-8
5	Ionizing radiation	DALY	9.04E-8	4.44E-8
6	Ozone formation, Human health	DALY	3.5E-7	1.27E-7
7	Fine particulate matter formation	DALY	0.000269	9.49E-5
8	Ozone formation, Terrestrial	species.yr	5E-8	1.81E-8
9	Terrestrial acidification	species.yr	1.18E-7	4.34E-8
10	Freshwater eutrophication	species.yr	5.91E-8	2.21E-8
11	Marine eutrophication	species.yr	1.03E-12	4.43E-12
12	Terrestrial ecotoxicity	species.yr	1.85E-9	1.15E-9
13	Freshwater ecotoxicity	species.yr	2.8E-9	1.93E-9
14	Marine ecotoxicity	species.yr	3.26E-6	1.42E-6
15	Human carcinogenic toxicity	DALY	0.00171	6.43E-5
16	Human non-carcinogenic	DALY	0.00645	0.00269
17	Land use	species.yr	3.8E-7	1.45E-7
18	Mineral resource scarcity	USD2013	0.0211	0.0154
19	Fossil resource scarcity	USD2013	4.98	1.92
20	Water consumption, human	DALY	9.02E-7	6.86E-7
21	Water consumption, Terrestrial	species.yr	3.34E-8	9.03E-9
22	Water consumption, Aquatic	species.yr	9.3E-12	1.80E-12

Table S4. Summary of fixed capital investment (FCI) estimation and utilities consumption for the CNF production (15 kg/day)

Units	Cost (in Rs.)
Plant & Machinery cost (Rs. in lakhs)	
Equipment	129
Piping	38.7
Electrical	25.8
Instrumentation	12.9
Total	206.4
Depreciation	10 years
Total cost per kg of CNF	
Cost of Goods	621.13
Depreciation	573.33
Other overhead charges	20
TOTAL	~1,215