

Life Cycle Assessment of Electrospun Cellulose-Based Nanocomposite Membrane Fabrication

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Abstract - Polymer nanocomposite membrane is an innovative and promising approach with a broad spectrum of potential applications in filtration processes. It is used to selectively separate molecules and ions. A comprehensive understanding of its environmental impacts, covering the life cycle of the used materials and the fabrication process, is crucial for its long-term sustainable success. This research aims to elaborate and implement a decision-making tool for greener membrane fabrication process. The environmental impacts of synthesizing one batch of Nanocomposite cellulose nanofibrils/cellulose acetate membrane using 50 gr polymer dope solution by electrospinning technique was determined based on a life cycle assessment methodology. The eco-sufficiency and sustainability of the electrospinning method were evaluated through a cradle-to-gate life cycle assessment (LCA) adopting the Cumulative Energy Demand (CED), and IMPACT2002+ impact assessment methods. According to CED assessment, the majority of energy consumed during electrospun membrane synthesis, amounting to 382 MJ, was consumed by the production of cellulose nanofibers. This is related to non-renewable fossil energy consumed in Ethanol production. As per IMPACT2002+ impact assessment, cellulose acetate and cellulose nanofiber manufacturing, and medium voltage electricity are the main contributors to the overall midpoint environmental effects.

Keywords: Life Cycle Assessment, Electrospinning, Membrane Technology, Polymer Nanocomposite, Cellulose

1. INTRODUCTION

It is expected Many advanced separation processes are made possible by membrane technology, which has found a wide range of applications in industry and in human life. Membrane technology is used in water treatment, wastewater treatment, food processing, drug delivery, biotechnology, and other industrial applications. It is also used in medical treatments such as dialysis and in the production of hydrogen fuel cells [1]–[3]. It is relatively inexpensive, environmentally friendly, and easy to process, which makes polymers the most popular membrane material in the industry [4]. Polymeric membranes offer advantages over traditional techniques such as distillation, adsorption, and absorption. In recent years, polymeric membranes have gained industrial attention because of their simplicity, cost-effectiveness, and small footprints [1], [5].

The electrospinning process is a simple, innovative, versatile, and a relatively low-cost way to produce nanofibers and nanocomposites with superior properties compared to conventional fibers, such as enhanced tensile strength, thermal stability, and chemical resistance [6]. A further advantage of electrospinning is its ability to create fibers with controllable diameters, higher porosity and surface to volume ratio which are desirable for water and wastewater treatment applications. Diameters of electrospun fibers usually range from 50 nm to 10 μ m [7]–[9]. It has been known that electrospinning mechanisms date back to the 19th century [10], but environmental effects associated with nanofibrous membrane fabrication are still under investigation. There has been a significant focus on making synthesis techniques more eco-friendly and greener [11], [12]. Figure 1- (a) depicts electrospinning process including heat treatment schematically. Electrospinning offers an incredibly flexible way to produce a wide range of fiber assemblies by fine-tuning polymer solution and electrospinning parameters. In this method, an electrospun nanofibrous membrane (ENM) is fabricated by overcoming surface tension with a polymer solution stretched by electrostatic forces. The electrostatic forces create a uniform nanofibrous membrane with a pore size that can be adjusted by varying the voltage and the concentration of the solution. In this technique, which is based on the electric field between polymer solution droplets in the needle existence and the collector, the conical-shaped droplets are stretched out and form nanofibers collecting on the collector [8], [13], [14].

Since membrane technology has developed applications across several markets, for sustaining the growth of the membrane industry, it is critical that we understand how much environmental impact each fabrication method will generate and select the more sustainable and greener membrane fabrication process accordingly. To better assess sustainability in membrane production, it is critical to quantitatively evaluate the impact of the entire process from different perspectives, i.e., global warming, human carcinogenic toxicity, human non-carcinogenic toxicity, fossil resource scarcity, and marine ecotoxicity by Life Cycle Assessment (LCA) approach [15][16][17]. LCA is an analytical and well-established environmental assessment tool to evaluate the cumulative environmental impacts of a product, process, or human activity to derive improvement actions, to develop the products and processes, and to help the decision makers [18]–[21]. Several LCA studies on polymer membranes are available in the literature, with many of them focused on membrane processes (i.e., filtration, desalination, water and wastewater treatment technologies) [22][23][24]. On the contrary, to the best of the authors' knowledge, few life cycle assessment studies have addressed the environmental aspects of the membrane fabrication process, and it is likely due to its complexity involving many different chemicals and polymers. The scope of this research focused primarily on the optimized enhancement of mechanical properties of the cellulose acetate nanocomposite membrane and secondary on the evaluation of the environmental impacts. In this study, we aimed to investigate the environmental impacts of electrospinning technique to synthesize cellulose-based nanocomposite fibrous membrane.

2. METHODOLOGY

Subsection 2.1 explains the preparation of nanocomposite cellulose acetate (CA) polymer solution and the synthesis of membrane via electrospinning method. A description of the two phases of life cycle assessment, i.e., the goal and scope definition and the life cycle inventory (LCI), follows in subsection 2.2.

2.1. Membrane production process

As part of the preparation of 50 gr of polymer solution composed of 15wt% CA (7.5 gr) as matrix polymer, and 0.25wt% 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO)-oxidized cellulose nanofiber (TOCNF) (0.125 gr) as reinforcing agent were dissolved homogeneously in 22.44 mL Dimethylformamide (DMF), and 26.79 mL acetone as the mixture solvent for 24 hours with a magnetic stirrer at 300 rpm, followed by ultrasonication at ambient temperature to achieve a good dispersal of nanofiller. Figure 1- (a) schematically illustrates the electrospinning process for fabricating nanocomposite fibrous membranes. To form the fibers, a BD plastic syringe with a capacity of 20 mL is connected to a spinneret with an inner diameter of 0.8 mm. The produced nanofiber samples are collected using a collector covered with aluminum foil to facilitate the peeling off the membrane from the collector. Moreover, the spinneret and the collector are two electrodes that are driven by a power supply (0-40 kV) in order to form nanofibrous membrane mats by electrostatic force.

2.2. Life cycle assessment methodology

For investigating the environmental impacts of the membrane fabrication process by electrospinning, LCA provides a standardized method (ISO 14040, ISO 14044). As a final product, the nanocomposite 0.25TOCNF/CA membrane is considered in order to quantify all emissions and resources consumed, as well as the associated environmental and health impacts. Defining the goal and scope, analyzing life cycle inventories, assessing environmental impacts, and interpreting the results are all critical components of an LCA study. This section covers the scope and goals, system boundaries, and LCI, while section 3.

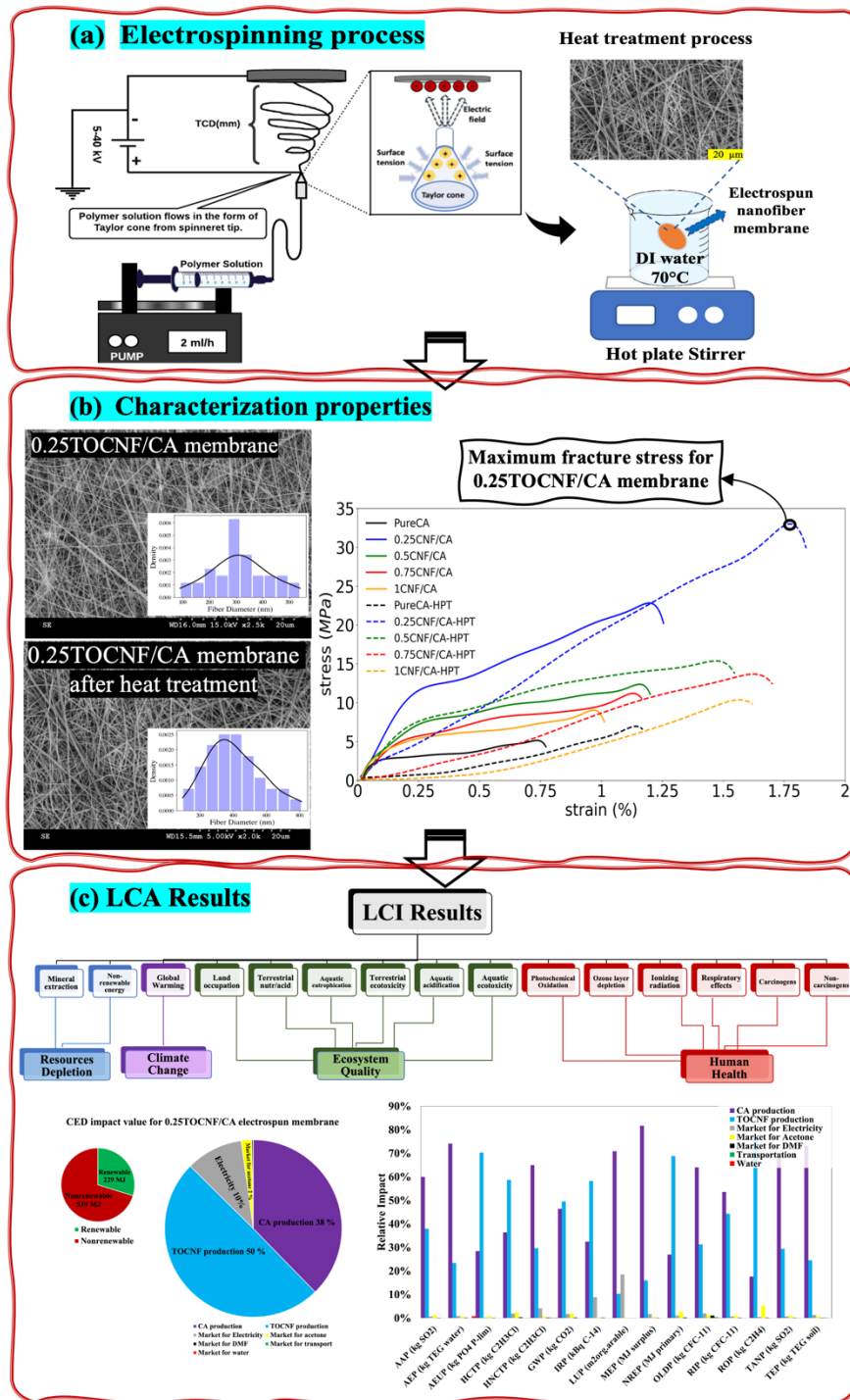


Figure 1 Comprehensive illustration of Process- Properties- Environmental impacts of 0.25TOCNF/CA ENM sample (a) Electrospinning method (b) Mechanical properties (c) LCA results of the membrane production process.

2.2.1. Goal and Scope definition

This study assessed the environmental impact of 0.25TOCNF/CA electrospun nanofibrous membrane production process. The functional unit (FU) of analysis was one batch of electrospun membrane samples prepared using 50 gr of 0.25TOCNF/CA polymer solution. We evaluated all inputs (materials and energy requirements) and outputs (emissions) on a per-FU basis. Table 1 shows an overview of the materials and energy requirements for electrospinning a batch of 0.25TOCNF/CA nanocomposite membrane sample. Nanocomposite membranes are the focus of the LCA in this work, making it a "cradle-to-gate" analysis. From the extraction of raw materials to the manufacturing of the final product, the system boundary encompasses all processes including all raw materials, energy, utilities (e.g., electricity and water), chemicals, and emissions during each stage. In this study, the environmental impact of the manufacturing of machinery and the equipment used to fabricate nanocomposite membranes was not taken into account. Emissions to water were considered in terms of environmental impacts.

Table 1 Life cycle inventory to produce one batch 0.25TOCNF/CA nanofibrous membrane using 50 gr polymer solution by electrospinning method.

Inputs	Units	Amount	Description	Source
Deionized Water	kg	3	Market for water, deionized- Cutoff, U-RoW	Ecoinvent 3.7
Tap Water	kg	5	Market for tap water- Cutoff, U-CA-QC	Ecoinvent 3.7
Cellulose Acetate	kg	0.0075	Cellulose acetate production	[25]– [27]
TOCNF	kg	0.00013	TEMPO-oxidized Cellulose nanofiber production	[28], [29]
Acetone	kg	0.212	Market for acetone, liquid- Cutoff, U - RoW	Ecoinvent 3.7
DMF	kg	0.0212	Market for N, N-dimethylformamide- Cutoff, U-GLO	Ecoinvent 3.7
Electricity	kWh	18.5	Market for electricity, medium voltage- Cutoff, U-CA-QC	Ecoinvent 3.7
Transportation	kg*km	138.91	Market group for transport, freight, lorry, unspecified Cutoff, U - GLO	Ecoinvent 3.7
Outputs	Units	Amount	Description	
Wastewater	kg	8	Emission to water	
0.25CNF/CA-ENM	number	1	Final product	

2.2.2. Life cycle Inventory

This LCA utilizes a comprehensive approach, combining data from the Ecoinvent database (Version 3.7), experimental measurements, literature findings, and estimations to construct its life cycle inventory. Table 1 and Table 2 further provide insights on the Life Cycle Inventory based on FUs. The background upstream manufacturing data for electricity, transportation, water, and chemicals are provided using the inventory Ecoinvent database (Version 3.7). Moreover, information on the synthesis process of CA and TOCNF was gathered from literature [25]–[29] and their environmental footprint was evaluated based on Ecoinvent database v3.7 (Table 2). The Ecoinvent database did not include two key reactants in the TOCNF production process. Due to limited information available on TEMPO's environmental impact, it was not included in the inventory. Furthermore, NaBr is not listed in any of the OpenLCA databases, and NaCl was substituted for it to estimate impacts due to their similar production processes and environmental impacts [23]. We also estimated a value from experience for some process data such as the volume of wastewater produced during the washing process. This analysis was conducted with the following assumptions: 1) While membrane fabrication is done by the NIPS method, nitrogen pressure is constant throughout the entire spinning process at 1 bar. 1) The analysis did not take into consideration the wastewater treatment system. 2) The analysis did not factor in air emissions caused by solvent volatility during membrane synthesis.

Table 2 Synthesis data inventory to produce 1.14 gr CA and 10 gr TEMPO-CNF

Cellulose acetate			
Inputs	Units	Amount	Description
Deionized Water	kg	6.22	Market for water, deionized- Cutoff, U-RoW
Acetic Acid	kg	0.063	Market for acetic acid, without water, in 98% solution station Cutoff, U-GLO
Cellulose Fiber	kg	0.001	Market for cellulose fiber-Cutoff, U-RoW
Acetic Anhydride	kg	0.0043	Market for acetic anhydride- Cutoff, U- GLO
Sodium Bicarbonate	kg	1.32	Market for sodium bicarbonate- Cutoff, U - GLO
Sulfuric Acid	kg	0.0004	Market for sulfuric acid- Cutoff, U-RoW
Electricity	kWh	4.7	Market for electricity, medium voltage- Cutoff, U-CA-QC
Outputs	Units	Amount	Description
Cellulose Acetate	kg	0.00114	Final product
Wastewater	kg	6.216	Emission to water
TEMPO-Oxidized CNF			
Inputs	Units	Amount	Description
Deionized Water	kg	22	Market for water, deionized- Cutoff, U-RoW
Ethanol	kg	504.96	Market for ethanol, without water, in 99.7% solution state, from ethylene-Cutoff, U-RoW
Kraft Paper	kg	0.040	Market for kraft paper- Cutoff, U-RoW
Piperidine	kg	0.24	Market for piperidine- Cutoff, U- GLO
Sodium Chloride	kg	0.0055	Market for sodium chloride, powder-Cutoff, U-GLO
Sodium Hydroxide	kg	0.43	Market for sodium hydroxide, without water, in 50% solution state-Cutoff, U-GLO
Electricity	kWh	331.22	Markey for electricity, medium voltage-Cutoff, U- CA-QC
Sodium Hypochlorite	kg	0.097	Market for sodium hypochlorite, without water, in 15% solution state- Cutoff, U-RoW
Outputs	Unit	Amount	Description
Wastewater	kg	22	Emission to water
TEMPO-CNF	kg	0.01	Final product

3. RESULTS

Among the benefits of LCA is the ability to evaluate different scenarios based on different assumptions. By accounting for these scenarios, LCA helps stakeholders understand the impact of their decisions on the environment. An electrospinning approach was used to synthesize 0.25TOCNF/CA nanocomposite membranes and an LCA approach was utilized to assess the environmental impacts of the process. After briefly reviewing the mechanical reinforcement of 0.25TOCNF/CA electrospun cellulose-based membrane sample, this section discusses the environmental impacts of the synthesis method.

3.1. Electrospun 0.25TOCNF/CA nano-composite membrane characterization

This study aims to determine the environmental impacts of the fabrication process of 0.25TOCNF/CA membrane that has previously been synthesized and mechanically strengthened [14], [30], [31]. The study was about the mechanical

reinforcement of cellulose acetate nanofibrous membranes using cellulose nanomaterials, i.e., TEMPO-Oxidized cellulose nanofibers (TOCNFs) and cellulose nanocrystals (CNCs) and the implementation of heat treatment process. Study results showed that TOCNF exhibited a better reinforcing capability than CNC nanofillers. As a result of heat treatment, the composite 0.25TOCNF/CA nanofibrous membrane sample reached maximum tensile strength of 33.31 MPa and elongation of 1.8% after reaching the breakpoint. A SEM and tensile analysis of the ultimate 0.25TOCNF/CA ENM sample are shown in Figure 1- (b). The fiber diameter distribution and SEM micrographs of 0.25TOCNF/CA nanocomposite membranes as synthesized and after heating are shown in the left panel. SEM images reveal an increase in fiber diameter after HPT at constant electrospinning conditions, due to the physical connection between the fibers and a slight melting in the surface of the fibers in the 0.25TOCNF/CA sample. The results of mechanical strength analysis of TOCNF/CA membrane samples are presented in Figure 1- (b). The solid line represents the stress-strain curve of the TOCNF/CA membrane samples before heat treatment, while the dashed line shows the stress-strain curve of the same samples after they were subjected to heat treatment. According to the results, heat treatment strengthened the samples' mechanical properties. Heat-treated 0.25TOCNF/CA membrane samples achieve the maximum ultimate tensile strength (UTS) and fracture strain (dashed blue curve) as compared to unheated samples. The improved mechanical strength and integrity of the membrane sample after heating is attributed to strong fiber connections caused by optimized TOCNF concentration and mean fiber diameter.

3.2. Environmental impacts of manufacturing process of electrospun 0.25TOCNF/CA nanofibrous membrane

Life Cycle Impact Assessment (LCIA): Using the right impact assessment method is crucial to the success of an LCA. Using characterization factors, these methods translate inventory amounts into environmental impacts. In this study, an impact assessment based on two methods was conducted. As part of the analysis, the IMPACT2002+ method was used to assess environmental impacts, which is a more appropriate method compared to other methods that were not developed in North America [32], [33]. Based on the 15 midpoint impact categories shown in Figure 1- (c), IMPACT2002+ provided an endpoint damage assessment. The fifteen potential impact categories assessed are aquatic acidification potential (AAP; kgSO₂ eq.), aquatic ecotoxicity potential (AEP; kg TEG water), aquatic eutrophication potential (AEUP; kg PO₄ P-lim), global warming potential (GWP; kg CO₂ eq.), ionizing radiation potential (IRP; kBq C-14 eq.), mineral extraction potential (MEP; MJ surplus), human carcinogenic toxicity potential (HCTP; kg C₂H₃Cl eq.), human non-carcinogenic toxicity potential (HNCTP; kg C₂H₃Cl eq.), land use potential (LUP; m²org.arable), non-renewable energy potential (NREP; MJ primary), ozone layer depletion potential (OLDP; kg CFC-11 eq.), respiratory inorganics potential (RIP; kg PM_{2.5} eq.), respiratory organics potential (ROP, kg C₂H₄ eq.), terrestrial acid/nutri potential (TANP; kg SO₂ eq.), terrestrial ecotoxicity potential (TEP; kg TEG soil). In terms of damage categories (endpoint impacts), midpoint impacts can be divided into four categories: human health (HH), ecosystem quality (EQ), climate change (CC), and resource depletion (RD). As energy consumption is the most common way of quantifying the environmental impact, we used cumulative energy demand (CED; MJ) to measure the energy consumption within the membrane production process. The pie chart in Figure 1- (c) illustrates the energy required to produce 0.25TOCNF/CA ENM sample based on material, electricity, water, and transportation requirements. It can be seen from the pie chart that TOCNF, CA, and electricity require more energy (50%, 38%, and 10% of total energy demand, respectively) than transportation, water, and solvent processes. The results showed that a total of 768 MJ of energy was consumed during electrospinning of 50 gr of 0.25TOCNF/CA polymeric solution, of which 382 MJ came from TOCNF production, 290 MJ came from CA production, and 80 MJ came from electricity. Energy demands for TOCNF production are mainly caused by the use of non-renewable fossil fuels in the ethanol production process. With CA production, a higher percentage of energy is derived from non-renewable fossil energy and renewable water energy that are used in the sodium bicarbonate production process and hydroelectricity power. In Quebec, 94% of electricity is generated by hydroelectric resources, so renewable water sources account for a large part of the cumulative energy demand. On the left side of Figure 1- (c), the smaller pie chart shows the breakdown of the production process' energy demand by renewable and nonrenewable sources. It is estimated that 70% of the energy used to produce 0.25TOCNF/CA ENM samples comes from nonrenewable sources, which account for 539 MJ. According to Figure 1- (c), the environmental impacts associated with the production of 0.25 TOCNF/CA ENM and the relative impacts of CA, TOCNF, acetone, and DMF production processes, as

well as electricity, water, and transportation requirements, are grouped into fifteen mid-point impact categories using the IMPACT2002+ assessment method. Across the whole electrospun membrane fabrication process, the CA and TOCNF production processes contributed >74% and 23%, respectively, to the midpoint category for aquatic ecotoxicity (AEP). Soda (>49%) and DI water (>10%) used for CA production and ethanol (>21%) used for TOCNF production processes were the largest contributors to the AEP midpoint category. Embedded aquatic toxicity impacts were primarily caused by heat production and lime used in soda production, and by ethylene and heat used for ethanol production in TOCNF production. A far greater burden is imposed on global warming (GWP) impact categories by sodium bicarbonate, electricity, and acetic acid requirements in CA production, and by ethanol consumption in TOCNF production. There are a number of factors that can damage human health, including carcinogens (HCTP), non-carcinogens (HNCTP), ionizing radiation (IRP), ozone layer depletion (OLDP), and respiratory effects (RIP and ROP). There were large embedded carcinogenic and noncarcinogenic toxicity potentials in the use of soda, acetic acid, ethanol, acetone, and electricity in CA and TOCNF production processes. The TOCNF production process, resulting in ethylene consumption, contributed the most to the carcinogenic potential, as shown in Figure 1- (c). CA production, on the other hand, posed the greatest risk for noncarcinogenic effects because of soda production. Additionally, 76% of risks associated with respiratory health were attributed to ethanol consumption. Moreover, 15% of the ionizing radiation was caused by sodium bicarbonate and 79% by electricity voltage transformation.

4. CONCLUSION

In conclusion, this study revealed the environmental impacts of the electrospinning process of 0.25TOCNF/CA nanofibrous membrane samples. By utilizing the LCA approach, it was possible to assess the environmental impacts of each step of the process. Results showed that TOCNF and CA consumption had a higher environmental impact in terms of midpoint categories related to aquatic ecotoxicity and global warming. Sodium bicarbonate and electricity consumption were two of the primary contributors to global warming and ionizing radiation potentials, respectively. Furthermore, the environmental burden caused by the production of TOCNF and CA were mainly attributed to the consumption of ethylene, ethanol, and acetic acid. This study provides valuable information in terms of environmental impacts of a novel fibrous membrane synthesis technique, i.e., electrospinning. It also provides a better understanding of potential damages to human health, global warming, and ecosystem quality. In the future work, further research is needed to examine the environmental impacts of different nanofibrous membrane production processes and a comparative LCA study should be conducted.

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