Chapter 5
Green Composites from Sustainable Cellulose Nanofibrils

Gonzalo Martínez-Barrera, Irna Zukeyt Garduño-Jaimes, Enrique Vigueras-Santiago, Julián Cruz-Olivares, Nelly González-Rivas, and Osman Gencel

1 Cellulose and Nanocellulose

Cellulose is considered to be the most abundant renewable polymer on earth. It is obtained from diverse sources as plants and algae, as well as by bacterial, enzymatic and chemical processes. Natural fibers are essentially made of cellulose, hemicellulose and lignin. Pectin, pigments and extractables can be found in low amounts. For this reason, natural fibers are also referred to as cellulosic fibers or lignocellulosic fibers [1]. The chemical structures of natural fibers are sophisticated. Each fiber is a compound in which the rigid cellulose microfibrils are embedded in a soft matrix mainly composed of hemicellulose and lignin. The cellulose fiber properties depend on the chemical composition, microfibril angle, cell dimensions and defects; they differ either from different sections of the plant or from different plants. The

G. Martínez-Barrera (✉) · E. Vigueras-Santiago
Laboratorio de Investigación y Desarrollo de Materiales Avanzados (LIDMA), Facultad de Química, Universidad Autónoma del Estado de México, Km.12 de la carretera Toluca-Atlacomulco, 50200 San Cayetano, México
e-mail: gonzomartinez02@yahoo.com.mx

I. Z. Garduño-Jaimes
Posgrado en Materiales, Facultad de Química, Universidad Autónoma del Estado de México, Paseo Colon Esquina Paseo Tollocan S/N, 50180 Toluca, México

J. Cruz-Olivares
Facultad de Química, Universidad Autónoma del Estado de México, Paseo Colón Esq. Paseo Tollocan S/N, 50120 Toluca, Estado de México, México

N. González-Rivas
Centro Conjunto de Investigación en Química Sustentable UAEM-UNAM, Km. 14.5 Carretera Toluca-Atlacomulco, Campus UAEMex "El Rosedal" San Cayetano-Toluca, 50200 Toluca de Lerdo, Estado de México, México

O. Gencel
Department of Civil Engineering, Bartin University, Bartin 74100, Turkey

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Table 1 Types of nanocellulose and their properties [author]

Parameter	Cellulose nanocrystals (CNCs)	Cellulose nanofibrils (CNFs)	Bacterial cellulose (BC)
Diameter (nm)	3–50	2–10	1.5–4
Length (μm)	0.1–1	>2	1–9
Crystallinity index (CI)	85–100%	40–78%	84–90%
Degree of polymerization (DP)	140–6,000	200–10,000	300–10,000
Tensile strength (MPa)	7,500–7,700	2–2,000	200–2,000
Young's module (GPa)	130–250	13–180	15–138

mechanical qualities of natural fibers also depend on the cellulose type, because each one has its own degree of crystallinity [2].

In recent years, cellulose has been studied at nanostructural level. Cellulose nanoparticles can be obtained from lignocellulosic fibers, which are located around the world, mainly in tropical countries. They can be obtained from sisal fibers (from Agave sisalana leaves), which are easily cultivable in India, Brazil and Tanzania [3]. Three types of structures are known for nanocellulose: (a) cellulose nanocrystals (CNCs), also referred to as nanocrystalline cellulose (NCC) and cellulose nanowhiskers (CNWs); (b) cellulose nanofibrils (CNFs), also referred to as nanofibrillated cellulose (NFC); and (c) bacterial cellulose (BC) [4–6], as it is shown in Table 1.

The relevance of the nanocellulose investigations is related to its green nature, physical and chemical properties and applications. Moreover, its crystallinity, surface area and mechanical properties depend on the extraction and processing methods [7].

The cellulose nanocrystals (CNCs) are obtained mainly by acid hydrolysis. They have high resistance, large surface area, 3–50 nm diameters and 0.1–1 μm length [8]. Their properties make it an excellent material for manufacturing biopolymers, antimicrobial films, medical implants and automotive components [9]. Cellulose nanocrystals have been mixed with different polyvinyl alcohol (PVA) concentrations, which reducing the fibers diameter [10].

Cellulose is also biosynthesized by some bacteria, which is called bacterial cellulose (BC), the bacteria that generate it are mainly those from the genders Gluconacetobacter, Sarcina and Agrobacterium [11]. By Gluconacetobacter (formerly called Acetobacter) is possibly to produce cellulose at commercial levels [12]. The main applications of BC are in biomedicine, explosives, membranes, magnetic materials and even in the food industry [13, 14].

Nanocellulose-based materials are non-toxic, carbon neutral, sustainable and recyclable. Investigations of novel, efficient and environmentally safe treatments continue as main objective. Despite all difficulties for nanocellulose production, it is easily found on the market. However, many established and proposed methods have emerged with respect to its large-scale production [15].

2 Cellulose Nanofibrils (CNFs)

Cellulose nanofibrils (CNFs), also referred to as nanofibrillated cellulose (NFC), consist of long, flexible, entangled nanometer-sized cellulose fibrils (2–10 nm diameter and >2 μm length). It exhibits amorphous and crystalline domains, with high aspect ratio and specific surface area. Currently, cellulose nanofibrils are considered high-performance, abundant, renewable, biodegradable and biocompatible materials, as well as with great potential for several industrial applications [6, 16, 17].

The first CNFs were obtained in the early eighties by Herrick and Turbak, who disintegrated cellulose by mechanical treatments in a high-pressure homogenizer. The cellulose was brought to a low aqueous consistency several times during the mechanical process. The final product had highly viscous gel consistency [18].

Various pretreatments and processes for obtaining CNFs have been developed. The objective is to eliminate some disadvantages for its production as improving the quality and reducing the large amounts of energy required; thus, the production cost is noticeably affected. Another important disadvantage after producing CNFs is the produced chemical wastes; so many efforts are focused for obtaining sustainable processes.

The most common mechanical procedures for producing CNFs are high-pressure homogenization (HPH), microfluidization, grinding, high-intensity ultrasound and ball grinding (Table 2). However, the scaling of mechanical processes becomes quite complex due to its high energy consumption. In the case of homogenizer treatment,

Table 2 Mechanical processes for producing CNFs [author]

High-pressure homogenization (HPH)	The velocity of the fibrous suspension is stopped by a homogenization valve, which causes low pressure, but high turbulence, temperature, impact and shear forces. Pressures of 30–150 MPa and energy ranging from 12,000–70,000 kWh/ton are used. The fibrillation degree depends on the applied pressure and the number of the passes
Microfluidization	The fibers are placed in an inlet tank and are propelled to interaction chamber by an intensifier pump, which provides high pressure. Inside, the pulp circulates through microchannels. High pressures (>100 MPa) are required and 500–2,550 kWh/ton energy; the fibrillation degree depends on the chamber size and the number of the passes
Grinding or ball process	It is based on impact and friction forces. Pressures of 30–50 MPa are used. However, it consumes high energy, due to the large number of passes, namely 16–30 times
High-intensity ultrasound	Hydrodynamic forces produce vibrations in the cellulose fibers, generating millions of microbubbles, which break the cell structure forming cellulose fibrils. It is a process that does not allow large-scale production

the electrical consumption reaches 70,000 kWh/ton; however, pilot-scale plants have been developed in some companies and research centers for such purposes [17].

The mechanical processes are normally accompanied by an enzyme or chemical pretreatment; otherwise, the energy costs rise to a great extent to achieve the cellulose defibrillation. The most widely used pretreatment process involves enzymatic hydrolysis, alkaline acids and ionic liquids.

In the case of enzymes, those with selective hydrolysis capacity as laccase can degrade or modify the lignin and hemicellulose content without altering the cellulose content [19]. A single enzyme is not capable for fiber degradation, as cellulose contains many organic compounds; thus, a set of cellobiohydrolase-type enzymes is required for the breakdown of crystalline cellulose and endoglucanases for disordered cellulose [20].

Pretreatment with alkaline acids is the most widely used method for removal of lignin, hemicellulose and pectin. In a study, sodium hydroxide and hydrochloric acid were used to remove cellulose, and then, it was submitted to mechanical treatment to obtain CNFs, and the results showed improvement on the cellulose yielding from 43 to 84% [21].

In the case of ionic liquids, they are organic salts, which have a high melting point and work at very low vapor pressure, prior to a mechanical treatment. Moreover, they have been used as solvents to dissolve cellulose and exhibit good results because an equivalent number of cations accompany anions and the electro-neutrality is maintained. The interactions of the ions (cation or anion) differ from each other but lead to self-organizing behavior [22].

Anions with high hydrogen bond basicity can solvate cellulose by binding to their hydroxyl groups, when the total hydrogen bond basicity of the solvent or the solvent mix has certain values [23]. The ionic liquids are salts with melting point less than 100 °C and better physicochemical properties compared to conventional organic solvents. They can dissolve polar, non-polar, organic, inorganic compounds and polymer materials. Recently, alternative solvents have been used for polymer production, such as supercritical carbon dioxide and water. In the CNFs production, high propagation rates and decreased termination rates have been observed in free radical polymerizations. Moreover, in the production of electrospinning CNFs can happen moderate reaction conditions, reuse of the catalytic system without decreasing of its activity and higher yields [24].

Catalytic oxidation by TEMPO is a process to obtain CNFs with a basic pH, very small diameters and transparent gels. Other characteristics also studied are quality, microfibrils angles and residual chemical composition (lignin and hemicellulose content). In this process, the production costs are reduced due to an energy efficient methodology, based first on the TEMPO process with catalytic oxidation and then on subsequent mechanical destructing. Moreover, this process is widely used to obtain nanopaper as a final product, with a high degree of transparency [25].

In recent years, a lot of investigations are concerning to use agro-industrial wastes as raw material for obtaining NFCs. The products are rich in natural fibers with structural arrangements and have high tensile strength. The chemical structure of such wastes are essentials for the nanofibers production, mainly the lignin and hemicellulose quantities, for example, the lignin content in the pulp can produce nanocellulose with diameters up to six times larger, and the hemicellulose content can influence on the diameter distribution [17].

The CNFs' properties depend on the chemical structure, surface chemistry, crystallinity and polymerization degree. The cellulose type and the used process are determinant on the morphological characteristics, which are studied by electron scanning microscopy (SEM), electron transmission (TEM) and atomic force (AFM) [26].

3 Green Composites

Research in sustainable technologies and highly resistant smart materials is an actual and future topic, supporting the need to balance economic growth with social and environmental concerns. One of the most actual research is concerning to nanocomposites, mainly those based on nanocellulose, which is an environmentally friendly material, and used in medical, automotive, electronics, packaging and construction industries as well as in the wastewater treatment, besides being an excellent substitute of synthetic materials.

Nanocomposites' production involves a lot of parameters, for example, process improvements, industrial scale production, energy consumption and matrix blending. Green composites include nanofibril-based polymer nanocomposites, as well as bacterial cellulose, which have been used in biomedical and technological applications. They show improvements on the mechanical, optical and barrier properties.

The global demand for textile fibers is supposed to increase as the population increases, and the quality of life improves. It is a fact that the CNFs' market is constantly increasing, due to the lack of cotton production and its high demand around the world. Cotton production cannot increase significantly; therefore, it is expected that, in few years, the CNFs' production will cover from 33 to 37% of the total market. Nevertheless, it would cause an increase in demand by 2030 year and would represent the production of 15 million tons CNFs by year. Such forecasts are due to some properties of the CNFs as absorption and humidity, among others [27]. The large-scale production of nanocellulose is already a fact. Both research laboratories and industries around the world promote its research; however, there are many challenges to overcome for its manufacture.

In the investigations about CNFs have been studied natural and synthetic polymer matrices. However, the scope of the polymers is very broad. Thus, it is essential to conduct more investigations. The objective is to obtain materials with good optical, mechanical and barrier qualities for their use in biomedical and technological applications. The nanocomposites based on CNFs show great advances, but it is necessary to develop properties, such as high tensile resistance, low density, high barrier properties (for sound, oxygen or other gases), optical transparency, biodegradability, renewability, among others [28].

Table 3 Applications of green composites based on CNFs [author]

Biomedical industry	• In moisturizing creams with ingredients capable of penetrating the skin and carrying bioactive agents • As matrix for tissue engineering (bone, cartilage, vascular, nerves and skin) • In the development of multifunctional matrices that allow the controlled release of therapeutic agents
Automotive and space industry	• As substitute of steel in the manufacture of bonnets and other auto parts. For reducing fuel consumption and carbon dioxide emissions • In aircraft fuselage manufacturing
Food industry	• Used as films in food wraps that produce strong barrier effect against gases penetration (mainly oxygen) are effective for maintaining food freshness • As food additive to reinforce the body feeling or food chewiness
Paper industry	• Filters that collect dust, small particles or deodorant substances that absorb odor-bearing microparticles • Manufacture of sheets or foils with deodorizing and antibacterial functions • Gel ink pens in which cellulose nanofibers are used as a thickener

Nowadays, there is a great deal of information related to the processing and properties of nanocellulose, which include several topics such as processing strategies, chemical modification of surfaces, biocompatibility, toxicity, characterization and possible applications. In addition, obtaining cellulose nanofibrils has been studied too; the reports are focused on optimizing chemical–mechanical treatments for the extraction of nanofibrillar cellulose and the final properties. Moreover, in recent years CNFs have been used in different sectors (Table 3).

Production of CNFs hydrogels is a novel topic; they have been recently used for the segregation of pollutants in water, non-polar hydrocarbons, solvents and oils as well as in chloroform adsorption.

In the coming years, the use of CNFs is expected to grow considerably, especially in tissue engineering, in the development of implants where surgeons replace the damaged tissue resulting from trauma, as well as in orthopedic reconstruction, where congenital deformity or pathological deterioration is replaced with autogenous grafts, made with CNF-reinforced composites.

Cellulose nanofibers are extensively used as reinforcing agents (up to 2.5 wt%) for manufacturing strong, highly flexible polymer gels, which are obtained by in situ polymerization, or heat treatment. They are designed for biomedical applications [29].

CNFs obtained from vegetables as well as synthetic nanofibers have been used as reinforcements of polymer materials. In a study, water soluble polymer matrices were used, due to their polar hydrogen bridges, which can bond with cellulose surface by hydroxyl groups, and thus generate a single and more stable phase. Polyvinyl alcohol

(PVA) matrices were dissolved in water at low percentages of CNFs. According to the results, an increase of approximately 100% in tensile strength was obtained when CNFs are added [30].

Another application of CNFs hydrogels is for elimination of heavy metals in water treatments to obtain purified water, due to their elevated specific surface area, nanoscale size, low toxicity, hydrophilicity and bioadsorption ability. Hydrogels produced with chemically cross-linked CNFs, alginate and polyvinyl alcohol (PVA) exhibited high swelling and adsorption capacity, as well as high-storage modulus [31]. The main objective is to produce CNFs with polymer networks or with nanomaterials nets. For example, aerogels are novel materials that are being applied in catalysis, filtration, graft, damping and liquid storage [29].

High-performance biomaterials are produced adding nanocellulose fibers, which produce high uniformity and few defects [32]. Addition of nanocellulose to biodegradable polymers produces improvement on the mechanical properties and accelerates the biodegradation rate. Moreover, nanocellulose is used in water soluble polymer solutions to modify their viscosity and mechanical properties.

In the biomedical area, the biodegradable CNFs are not toxic to humans and biocompatible. It is used for personal hygiene products, cosmetics, biomedicines, burned skin treatment. They hold the stabilization of medical suspensions and decrease the sedimentation and phase separation of heavy ingredients. Moreover, after its chemical modification, it serves as a scaffold for enzymes and other medications [33].

Currently, the care and preservation of natural resources require the use of technologies based on biodegradable materials. For example, an ecological sport vehicle, called nanocellulose vehicle (NCV), was made with cellulose nanofibers from plants and agricultural waste, which are a fifth lighter than steel but five times more resistant. Various automotive parts were developed, such as doors, roof and hood. The cellulose nanofibers reduced the NCV body weight up to 50% compared to a traditional car. In addition, this novel technology helps to reduce the carbon emissions related to automobile manufacturing [34].

The paper industry has developed novel researches and applications based on the use of CNFs, becoming one of their main activities. As it is known, fibers decrease their properties during recycling processes, mainly due to drying. To resolve such problems, mechanical refining techniques have been used for their rehydration, but they cause other irreversible structural damages [35].

Production of electrospinning CNFs depend on the polymer concentration, tip-collector distance, viscosity, solution flow and applied voltage. Moreover, require of environmentally friendly solvents. Several investigations are related to the collector shape, which directly affects the diameter and resistance of the CNFs.

One of the advantages of the electrospinning technique is its ability to mix CNFs with synthetic polymer matrices. In this case, prior chemical modification is often necessary to ensure good compatibility with the matrix. Poly (vinyl alcohol), poly (lactic acid), polyurethanes, conductive polymers and acrylic resins are a few examples of used synthetic polymers. Electrospun CNFs have small pore sizes and a large surface area compared to commercial fabrics. They have controllable dissolution properties and are used as tissue engineering scaffolds and drug delivery systems [32].

Inside the production, 3D printing manufacture has been added. Now, it is possible to create three-dimensional scaffold-like structures. These nanomaterials can respond to the temperature and color changes and are known as intelligent materials. Due to their swelling, they become soft and elastic. In the case of nanocomposites produced with CNFs and impenetrable network (IPN) systems, the 3D structures show the matrix and an interwoven network [36].

In the future, it is planned to program the shape and behavior of 3D-printed objects, but adding a fourth dimension related to their temporal changes. This new scientific revolution is called 4D printing and is based on novel printing materials called "intelligent," they undergo a controlled structural change when they are submitted to external stimulus. Their shape and properties change with temperature, humidity, light and time. For example, it could be possible design smart fabrics that react to our thermal sensation, that is to say, if we feel heat they can release this energy and on the contrary, when we feel cold they can retain body heat. Thus, 4D printing is a new technological paradigm in areas such as textile, membranes, medicine, robotics and energy production.

4 Experimental Studies

In the investigation group belonging to Laboratory of Research and Development of Advanced Materials (LIDMA) at Autonomous University of the State of Mexico (UAEM), some experimental studies related to the production of CNFs, involving raw materials, production methods, characterization and applications are conducted.

In the first experimental stage, electrodynamic methods were used for to obtain bacterial cellulose (BC) by using environmentally friendly solvents. According to the literature, BC has been applied as a skin transitory substitute in the treatment of wounds, burns and ulcers, as well as in dental implants or acoustic transducers, among others, due to its high mechanical resistance acquired after chemical treatments. With elasticity modulus of 16–18 GPa, tensile stress of 260 MPa, 2.1% deformation and high purity and degree of crystallinity [37, 38].

The BC synthesis was carried out at 28–32 °C and using the *Gluconacetobacter xylinus*. The produced BC was crystalline, free of lignin and hemicelluloses, with 4–6 pH. According to the literature, highest cellulose yield can achieve with controlled quantities of glucose; high concentration of this can inhibit the cell growth and production as well as decrease pH, due to accumulation of (keto)gluconic acids [39]. The carbon source is a fundamental factor in the production of BC, whereby two types of carbon sources were used in the static bioreactors: sugar and beet molasses.

Gluconacetobacter xylinum was obtained from apple cider vinegar. Hestrin and Schramm were used as culture medium, whose composition was glucose (20 g/L), peptone (5 g/L), yeast extract (15 g/L), disodium phosphate (2.7 g/L) and acid citric (1.15 g/L). The pH was adjusted to 5.5 using 0.1 N sodium hydroxide, while the culture medium was autoclaved at 15 psi and 121 °C by 15 min. Then, 9 mL of culture medium and 1 mL of apple cider vinegar were placed in test tubes. The mixtures were homogenized with a shaker and incubated by 6 days at 32 °C. The variables were the chemical structure, pH, volume loss, the cellulose sizes, cellulose production and the bioreactor size.

Bacterial cellulose (BC) growth was performed in bioreactors, which were sterilized in an autoclave at 15 psi and 121 °C by 15 min. The bioreactors contained the test tube solution (1 mL), potassium sorbate (0.13%), sucrose (12.66%), yeast extract (1.26%), calcium chloride (0.76%), potassium phosphate (0.37%), distilled water (84.9%) and the corresponding percentage of each substrate. The bioreactors were covered with a cotton cloth to allow aeration in the culture medium, in order to promote contact with oxygen and achieve the bacteria growth. Then, the bioreactors were placed in an incubator with circulation at 30 °C. Produced BC is shown in Fig. 1.

The electrospinning process was used, due to its advantages to produce nanofibers. In recent years, numerous types of materials, including synthetic and natural polymers have been electrospun to obtain continuous fibers. They produce a highly porous spun bond nonwoven membrane, with fibers size ranging from nanometers to few microns [36]. Moreover, cellulose derivatives with greater solubility have been synthesized, as cellulose acetate, cellulose triacetate and methyl cellulose. Cellulose acetate nanofibers (247–265 nm diameters) have been used for protection of vitamins A and E, while cellulose acetate is mixed with water/acetic acid for producing pyranose 2-oxidase fibers, with 200–400 nm diameters. In the case of cellulose triacetate (4.7 k dielectric constant), this is used for produce CNFs with low surface area and without lumps or beads [40, 41].

Fig. 1 Bacterial cellulose (BC) obtained after 28 days of culture [author]

The parameters involved in the CNFs production are determinant on the applications; some of them are shown in Table 4.

The electrospinning equipment (shown in Fig. 2) consist of an infusion pump, which controls the flow rate, a high-voltage source (10,000 V as minimum) and the collecting system.

The configuration can include more variants, for example, type of XY movements, rotary nozzles, rotocollectors, arrangements with auxiliary electrodes, cryogenic collectors and coaxial nozzles. The electrospinning equipment was designed with CAD software and has movement on the three axes. Moreover, a collector with

Table 4 Types of CNFs produced and its applications [author]

Fiber type	Parameter	Applications
Flattened or tapes	It is attributed to the emergence of a polymer layer on the surface of the fiber, due to the uneven solvent evaporation. The atmospheric pressure tends to collapse the round shape of the fiber. It can be related to the solvent type and the addition of salts to the solution	• Biosensors systems, due to its electrochemical activity and the ability to transfer electrons
Helical	It occurs due to the jet deformation during the impact with the collecting plate. Solution concentration promotes this behavior. The incidence angle of the jet influences on its maintenance	• Drug delivery systems • Electromechanical and electromagnetic microsystems • Advanced optical components
Ramified	It is related to the small jets on the surface, produced by the initial jet. The instability between electrical forces and surface tension produces instability in the jet	• Drug delivery systems
Hollow	It is obtained by coaxial electrospinning processes or by chemical processes applied to electrospun fibers	• Nanoelectronic and optoelectronic devices • Energy conversion • Drug release • Environmental protection • Sensors
Fibers with beads	They are due to the surface tension and the viscoelastic properties of the solution. They depend on the outlet flow, distance between capillary and collector and voltage as well as from molecular weight or viscosity of the solution	• Tissue engineering
Fibers with pores	It is the consequence of the relative humidity and vapor pressure of the solvent	• Tissue engineering • Catalysis • Sensors

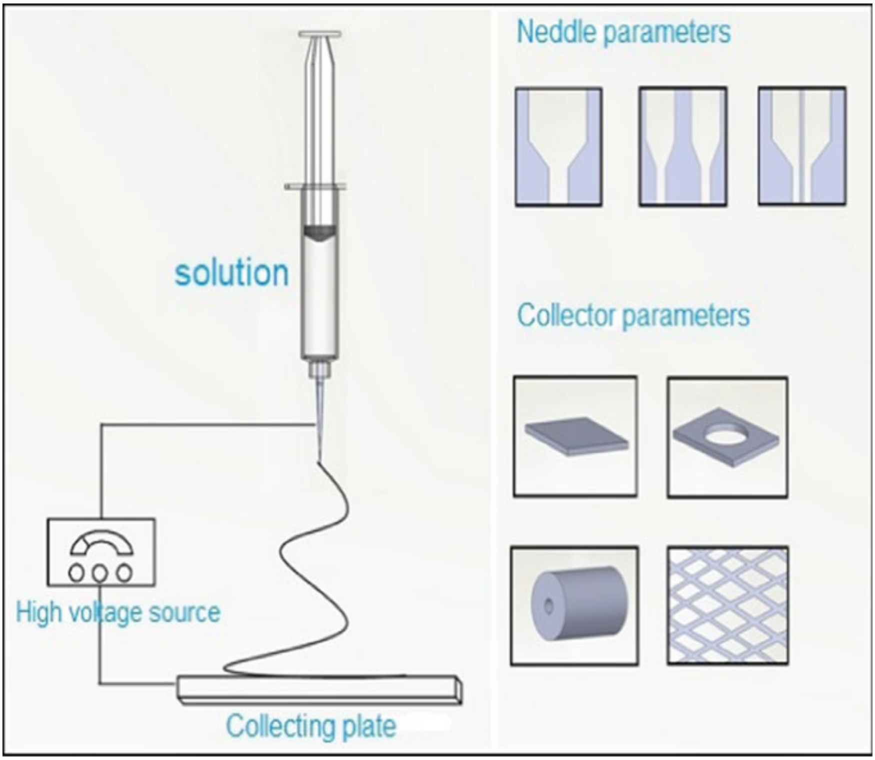

Fig. 2 Electrospinning process and their parameters for producing CNFs [author]

speed was also designed to determine the accuracy deposition of CNFs. The objective was to have several needles connected to the dosing pump and a collector in motion, as it is shown in Fig. 3.

The electrospinning design gives the possibility of using a single injector with specific movement, in order to reproduce the patterns designed in the CAD software. The parameters shown in Table 5 were taken into account for production of electrospinning CNFs.

In the second experimental stage of the electrospinning process, the cellulose triacetate was replaced with eutectic mixtures having low melting point. In the process, sustainable and biodegradable solvents with low toxicity and ecological footprint were used. As it is known, molecular weight and solvent type are the most important factors for producing electrospinning cellulose. A homogeneous distribution of solvents produces a homogeneous material; otherwise, the needle capillary will be obstructed.

A possible application of the produced CNFs was as an interchangeable mask filter, as it is shown in Fig. 4. The mask was designed with CAD software and 3D printing in a molten deposition printer, by using a flexible and low-density filament.

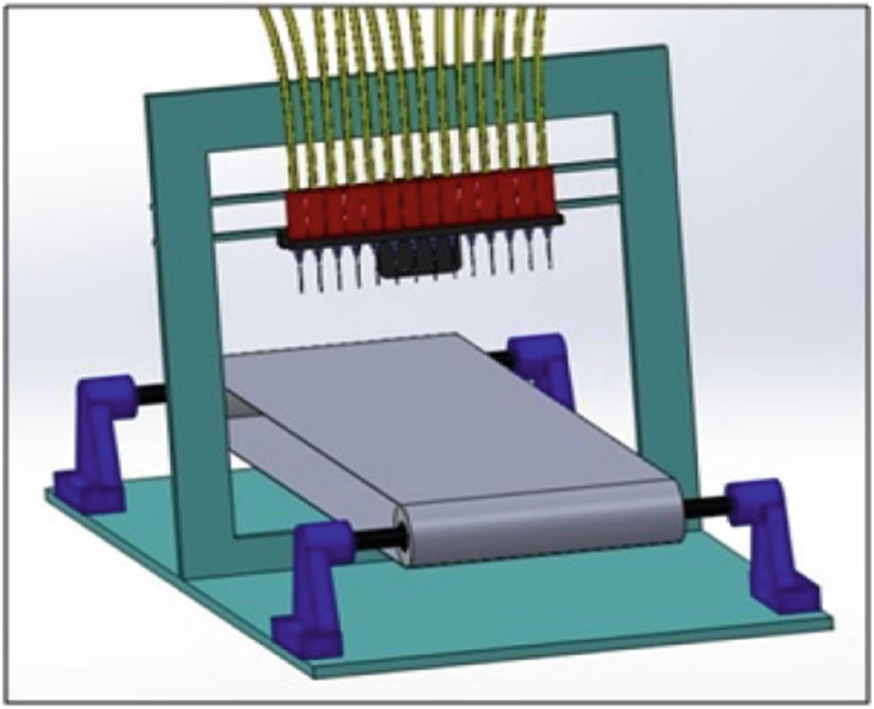

Fig. 3 Solidworks design of a multiple electrospinning machine with dynamic collector [author]

Table 5 Parameters in the electrospinning process [author]

	Parameters
Dissolution	• Concentration of the polymer solution • Surface tension • Solution conductivity • Dielectric effect of the solvent
Process	• Voltage • Outflow • Collector distance
Environmental	• Room temperature • Humidity

Undoubtedly, research and developments of CNFs will continue. The future nanocomposites are innumerable, and their success will depend on many factors including their novel methods and applications.

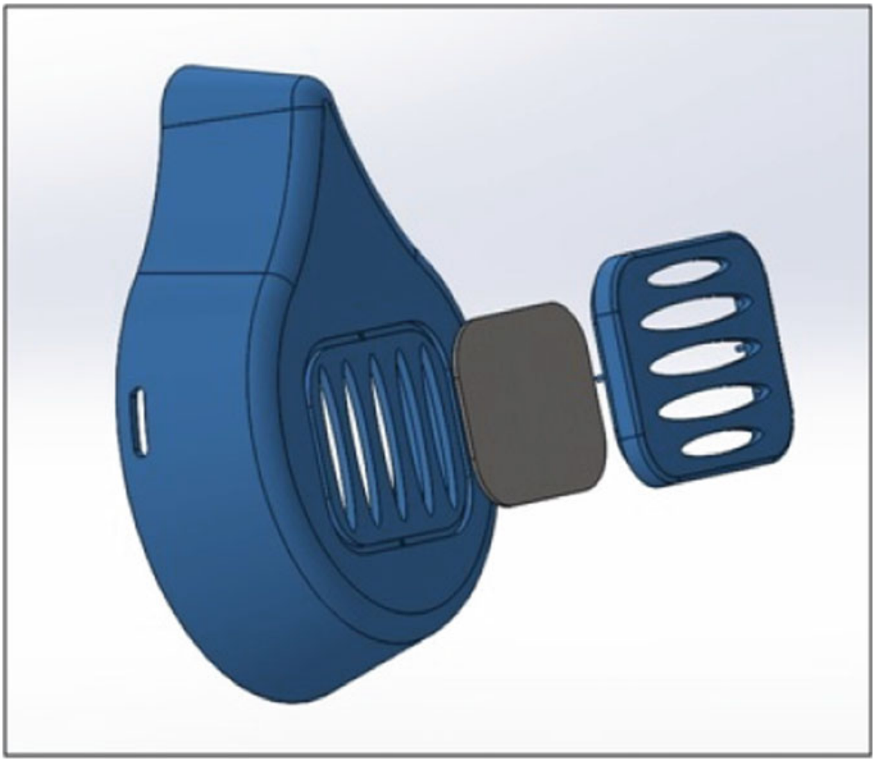

Fig. 4 Prototype mask including filter produced with NFC [author]

5 Conclusions

The CNFs' era continues to evolve; however, there are still many activities to do respect to the current production processes, to the use of available resources and mainly to the related to environment care. The electrospinning technique gives a very encouraging image for the improvement of the CNFs' properties as well as for producing green composites, which have a wide range of applications in many industrial sectors. Such materials will allow in the near future improving some needs of the human being.

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