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REVIEW PAPER

Cellulose-based electrospun nanofibers: a review

Jonas Kerwald · Celso Fidelis de Moura Junior · Emanuelle Dantas Freitas · João de Deus Pereira de Moraes Segundo · Rodrigo Silveira Vieira · Marisa Masumi Beppu

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Abstract Cellulosic materials have gained a lot of attention in the last decades because of their abundancy, renewability and excellent physicochemical properties. Meanwhile, research on nanofibers has also been increasing with the aim of producing or modifying materials that can have a wide range of applications, such as tissue engineering, drug delivery, protective clothing and wound dressing. In order to produce these fibers, electrospinning is shown to be a promising and extensively used technique. Electrospun cellulosic fibers maintain the optimal characteristics of cellulose while improving its surface area to volume ratio and mechanical properties, in addition to the possibility of surface tailoring of bulk materials. However, there are several limitations related to the utilization of cellulose and most of its derivatives with the electrospinning technique. Poor solubility in most common solvents and inability to melt are major drawbacks. Thus, this review describes mostly recent research in which cellulose and its derivatives have been the feedstock for fabrication of nanofibers by electrospinning, exploring processing details and potential applications.

Keywords Cellulose · Electrospinning · Nanofibers · Cellulose derivatives

J. Kerwald · C. F. de Moura Junior · E. D. Freitas · J. D. P. de Moraes Segundo · M. M. Beppu (✉)
School of Chemical Engineering, Department of Materials and Bioprocess Engineering, University of Campinas, Campinas, São Paulo 13083-852, Brazil
e-mail: beppu@feq.unicamp.br

R. S. Vieira
Department of Chemical Engineering, Federal University of Ceará, Fortaleza, Ceará 60455-760, Brazil

Introduction

Recently, natural polymers have gained massive attention due to their sustainability, eco-efficiency and renewability. Cellulose has been of great interest because of its abundancy, biocompatibility, biodegradability and regenerative properties (Frey 2008). Moreover, cellulose nanomaterials have advantageous surface area to volume ratio, high crystallinity, high mechanical strength and low density (Zhang et al. 2021). Cellulosic nanofibers, when compared to nanocrystals, present an even higher surface area to volume ratio (Bian et al. 2017) and excellent flexibility, elasticity and impact resistance (Lindström 2017). The fabrication of nanofibers (NFs) is generally classified into physical–chemical techniques and electrospinning and non-electrospinning techniques. Since the 2000s, electrospinning has been the most widely investigated method for the production of NFs (Gugulothu et al. 2019).

Electrospinning is a simple and effective method that utilizes high electrostatic forces to prepare NFs

with a high surface area to volume ratio (Xu et al. 2008). The electrospun fibers also present high porosity and flexibility, with the possibility of tuning pore structure. Such materials are ideal for applications in energy, healthcare and biotechnology. Nonetheless, electrospinning normally requires the use of hazardous solvents to dissolve the polymers and form the precursor solutions (da Silva et al. 2021). The problem is aggravated when there is cellulose in the polymeric solution because it does not dissolve in the majority of the most common solvents used in electrospinning (Prasanth et al. 2014). Therefore, the improvement and development of existing technologies are of utter importance in order to overcome these drawbacks (Khoshnevisan et al. 2018).

In the last decade, the number of publications involving electrospinning of cellulose has grown each year, as it can be seen in Fig. 1. The data was obtained through a search in ScienceDirect using the keywords "cellulose" AND "electrospinning", considering only research articles. This visualization shows that the subject has been attracting more attention as time passes and it may help to overcome major limitations of the electrospinning technique, especially when applied to cellulosic materials, hence increasing the technical and economic feasibility of cellulose NFs in its varied range of applications.

Cellulose and its derivatives

Cellulose is the most abundant natural biopolymer in the world. Because it is the main component of plant cell wall, it is mainly obtained from plants, but other sources such as algae (Gao et al. 2018), bacteria (Lotfy et al. 2021) and fungi (Thygesen et al. 2003) are also available. Its major sources are wood and cotton. Once obtained, depending on the treatment and chemical modification it suffers, cellulose has several different uses, such as in paper, fibers and clothes, cosmetic and pharmaceutical industries (Joseph et al. 2020). Extraction of cellulose depends on the biomass material (and their amount of hemicellulose and lignin). The most conventional method is isolation (also called pulping) by kraft, which involves the digesting of the material at elevated temperature and pressure in a solution of sodium sulfide and sodium hydroxide. Another highly applied method is the organosolv process, which uses an aqueous organic solvent (methanol, ethanol, acetone…) to solubilize lignin, which can subsequently be removed (Gauss et al. 2021).

Chemically, cellulose is a linear polysaccharide made up of repeating glucose units connected to each other by β-(1 → 4) linkages (French 2017). The three hydroxyl (-OH) groups present in every glucose residue are highly reactive and the substitution of the

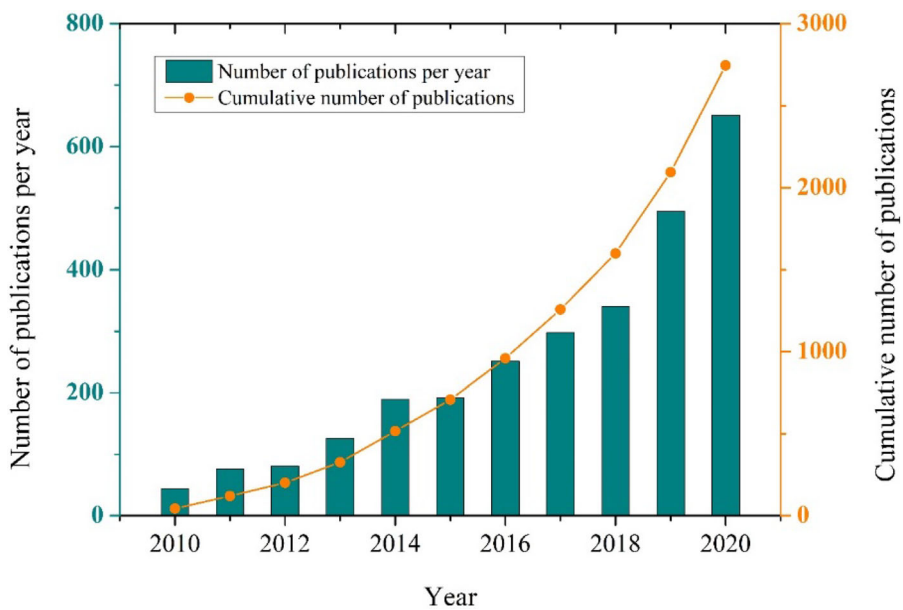


Fig. 1 Annual and cumulative number of publications in ScienceDirect platform in the last decade (keywords: cellulose AND electrospinning)

hydrogens by other radicals lead to the formation of cellulose derivatives, as it is displayed in Fig. 2, where R is the radical and n is the number of repeating units, which can range from 10,000 to 15,000 depending on the source of cellulose (Moon et al. 2011). The strong interacting hydroxyl groups bestow cellulose materials with a strong tendency to self-associate and form an extended network via intramolecular and intermolecular hydrogen bonds (Jorfi and Foster 2015).

Cellulose is a semicrystalline polymer comprised of crystalline (ordered) and amorphous (disordered) regions, which will vary according to the origin of cellulose and its isolation method. The native degree of crystallinity of the material is usually found to be in the range of 40–70% and depends on its origin and isolation method (Nechyporchuk et al. 2016), but these values vary greatly according to the determination method for the crystallinity of cellulose (Ahvenainen et al. 2016). Because of its abundancy, biodegradability, biocompatibility and nontoxicity, cellulose has received much attention in the last decades. Its interesting mechanical properties include high tensile strength, its Young elastic modulus, and its crystallinity (Tanpichai et al. 2012). It is insoluble in water and many organic and inorganic solvents. This is due to the strong interchain hydrogen bonds in the crystalline regions. However, because chains are more separated in amorphous regions and hydrogen bonding with other molecules is easier, cellulose can

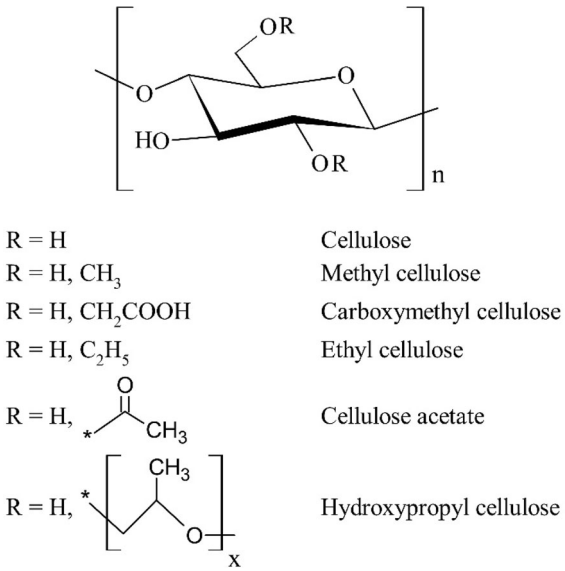


Fig. 2 Structure of cellulose and its main derivatives

absorb large quantities of water, being a hygroscopic material (Kalia et al. 2011). Despite being commonly overlooked in the literature, van der Walls attractive forces and hydrophobic interactions also play important roles in determining the properties and behavior of cellulose even in the most polar aqueous environments (Kaya et al. 2009; Glasser et al. 2012).

Chemical modifications can solve some of the drawbacks of natural cellulose fibers such as poor interfacial adhesion and high-water absorption, along with improving its existing properties and its processibility. Most of the modifications aim at improving cellulose processing with nonpolar matrices or changing its affinity with polar/nonpolar molecules (Abushammala and Mao 2019). Among the most common cellulose derivatives are cellulose acetate (CA), methyl cellulose (MC), carboxymethyl cellulose (CMC), ethyl cellulose (EC) and hydroxypropyl cellulose (HPC). Because of its good solubility in a wide variety of solvents and its possibility to melt, cellulose acetate has been the material of choice for several applications, including cigarette filters and high absorption diapers (Konwarh et al. 2013). Its fibers have been the focus of extensive research in recent years, especially regarding textile and biomedical uses (Chen et al. 2020c; Wei et al. 2020).

In the last two decades, with the advances of nanotechnology, the obtention and application of cellulose in the form of NFs or nanocrystals have been extensively studied. While maintaining the formidable properties of the material, nanocellulose has increased stiffness and tensile strength (greater than cast iron). Whereas nanocrystals/nanowhiskers are known for their high crystallinity (54–88%) (Moon et al. 2011), nanofibers have higher surface area and higher length to diameter ratio (Lavoine et al. 2012). Research on applications of nanocellulose has grown exponentially in recent years, it is mainly used as the basis of functionalized materials and fiber-reinforced composites, especially in the field of biomedicine (Liu et al. 2016; Paulraj et al. 2018).

Preparation of cellulosic NFs can be carried out with a variety of physical–chemical methods, such as spinning and freeze-drying. One of the most common method for fabricating NFs in general is electrospinning. Even though it is a suitable technique to be used with cellulose, there are a lot of technical issues that need to be addressed, especially because native cellulose either does not dissolve in most of the

conventional solvents or is melted (Lundahl et al. 2017). There are reports of electrospinning of cellulose in its original form (Robles-García et al. 2018), or of its derivatives, such as cellulose acetate (Beikzadeh et al. 2020), carboxymethyl cellulose (Allafchian et al. 2020), hydroxypropyl cellulose (Shukla et al. 2005) and others (Ahmad et al. 2013).

Electrospinning

Electrospinning is a highly versatile technique for generating NFs from polymeric solutions that are used in a wide range of applications (Wen et al. 2020). It was in the 90 s that this technique became popular due to the growing interest in nanotechnology and the search for new materials, a fact evidenced by the significant increase in the number of publications on the subject (Xie et al. 2020). The electrospinning technique gained prominence for producing polymer fibers in micro (10–100 μm), submicron and nano (0.01–0.1 μm) scales, with characteristics of high surface area concerning the volume of interconnected NFs networks, adjustable surface properties and flexibility, in addition to good mechanical performance and high water permeability (Bavatharani et al. 2020). Due to these characteristics, electrospinning has been applied in several areas of science, such as the production of three-dimensional scaffolds in tissue engineering (Pattison et al. 2005), regeneration of skin tissue (Yu et al. 2019), controlled release of drugs (Qin et al. 2019), sensors and electronic devices (Zhang et al. 2017), membranes for water treatment (Liao et al. 2018), smart textile industry (Yu et al. 2021), among others.

In its most basic form, any electrospinning system (a basic setup of electrospinning is shown in Fig. 3) requires at least three main components (Huang et al. 2003; Wen et al. 2020):

(i) Some method of feeding the spin-dope (solution containing polymer);
(ii) A high voltage source to create an electric field;
(iii) A grounded metallic collector for the deposition of electrically spun fibers ejected from the supply.

Polymeric NFs are obtained from a liquid jet that is generated with the contribution of the external electric field with interactions of the electric charges in the polymer fluid (Bavatharani et al. 2020). The electric field is subjected to the end of the capillary tube containing the polymeric solution, inducing a charge on the liquid's surface and forming a drop. As the intensity of this electric field increases, there is an elongation of the spherical surface of the fluid at the tip of the capillary tube, in a conical shape, known as the Taylor cone. As the jet moves to the collecting surface, the solvents present in the solution evaporate. This causes the polymer chains to be closer and, consequently, solidification and formation of fibers that deposit on the collecting surface occur (Huang et al. 2003; Xue et al. 2017; Liao et al. 2018).

Although it is an easily applicable technique, several parameters can influence the production and performance of the NFs. These parameters can alter the diameter, morphology, pore size and other characteristics (Sun et al. 2014). According to Haider et al. (2018), a complete understanding of the effects of all the parameters that govern electrospinning is essential for a better knowledge of the technique.

Some parameters can influence the transformation of polymeric solutions into nanofibers by electrospinning. By varying the parameters of the polymer solution (viscosity, conductivity, surface tension), process (feed flow, voltage, distance between needle and collector) and environment (temperature and relative humidity) it is possible to control the formation of fibers as well as their morphology for a multitude of applications (Pelipenko et al. 2013; Sun et al. 2014; Hassiba et al. 2016; Khajavi and Abbasipour 2017). A summary of these parameters and the influence they have on the morphology of the fiber produced and on the electrospinning process is shown in Table 1.

Electrospinning of natural polymers

Electrospun fibers have great potential for application in the area of bioengineering. Because natural polymers present high biocompatibility and low immunogenicity, these materials are more suitable for biomedical applications when compared to synthetic polymers (Ramakrishna et al. 2006). The use of natural polymers in electrospinning systems have a strong relationship with their inherent capacity for binding cells, since they carry specific protein sequences (Bhardwaj and Kundu 2010). Collagen,

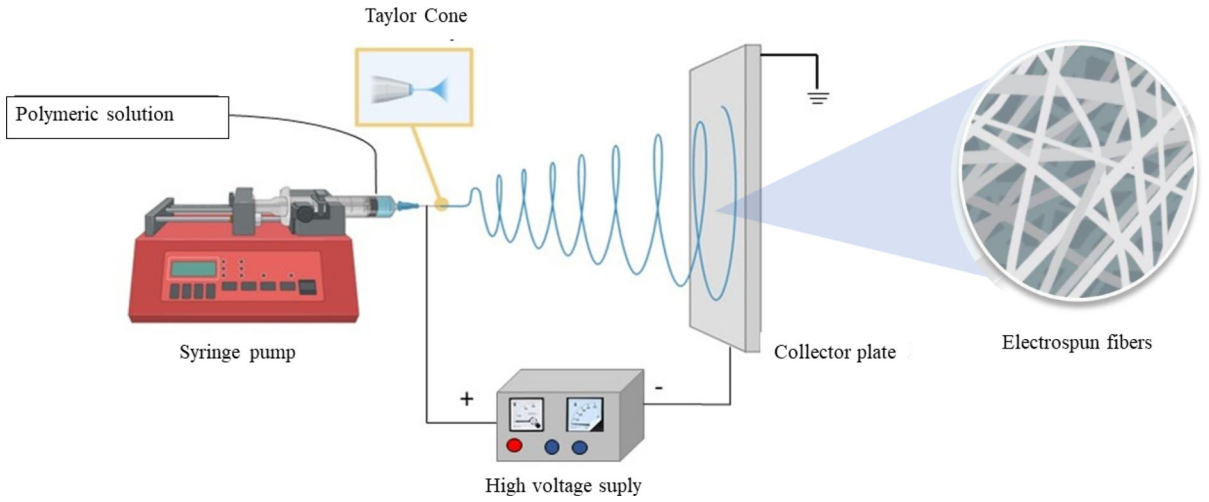


Fig. 3 Schematic drawing of a basic electrospinning system

alginate, chitosan, gelatin, casein, cellulose and its derivatives, silk fibroin, chitin and fibrinogen are examples of natural polymers that can be electrospun (Baker et al. 2012; Luo et al. 2018; Selvaraj et al. 2018; Sizeland et al. 2018; Dodero et al. 2020; Arumugam et al. 2021; Cui et al. 2021; Jing et al. 2021). In addition, these polymers can be used in blends with other polymers. Some examples are described below.

Collagen extracts from the skin of *Macruronus novaezelandiae* (hoki) were electrospun to obtain a fibrous material by Sizeland et al. (2018). Characteristics of intact alpha chains in collagen after being electrospun were observed and electrospinning can lead to the reform of collagen fibers with nanostructural properties similar to those found in native collagen tissues.

Dodero et al. (2021) prepared electrospun mats of alginates with different molecular characteristics crosslinked with strontium ions, which presented an interconnected porous arrangement capable of promoting the adhesion of both skin and bone cell lines, as well as providing enhanced mechanical properties. In addition, alginate solutions resulted in homogeneous membranes and without defects.

Beikzadeh et al. (2020) encapsulated the essential oil of lemon myrtle (LMEO) in cellulose acetate nanofibers by electrospinning and evaluated the antimicrobial, morphological and thermal properties of the fibers as well as the prolonged release of LMEO from the fibers. The results showed that the material is highly antibacterial against Gram-positive (*E. coli*) and Gram-negative (*S. aureus*) bacteria, even after one month of storage.

Zhang et al. (2008) evaluated scaffolds of electroplated silk fibroin for vascular cell responses in vitro. The authors characterized the potential of these NF silk matrices to support vascular smooth muscle cells and the functions of endothelial cells. The results suggested the potential of these scaffolds for the exploration of tissue engineering vascular grafts, since they support the viability of vascular cells, maintain the cellular phenotype and promote cellular reorganization.

Nawalakhe et al. (2013) developed a new compound for wound dressing with great adherence by electrospinning chitosan nanofibers onto 100% cotton gauze substrates treated with atmospheric plasma. The antibacterial properties of chitosan against *B. cereus* and *E.coli* were maintained after being electrospun. Chitosan nanofibers also improved absorbency of cotton gauzes by an average of 68–82%, which is likely to enhance absorption of blood and wound exudates.

Electrospinning of cellulose

Electrospinning of pure cellulose is a process that usually limited by its low solubility in common solvents and inability to melt due to its various inter- and intramolecular hydrogen bonds. However, cellulose is reported to be the first electrospun material in history, dating back to 1934 (Formhals 1934).

Table 1 Effect of electrospinning parameters on fiber morphology. (Adaptation with permission from reference Sun et al. (2014). Copyright 2014 Elsevier)

	Parameter	Effect on fiber morphology	Account	References
Solution parameters	↑ Polymer concentration/ viscosity	Fiber diameter increase Formation of continuous and longer fibers	Jet elongation becomes more difficult and slower. Increase in intermolecular bonds	Fong et al. (1999) and Baykara and Taylan (2021)
	↑ Conductivity	Fiber diameter decrease Bead-free fibers	The increase in conductivity causes bending instability and greater repulsion during electrospinning, with the fibers being subjected to more stretching	Uyar and Besenbacher (2008) and Wang and Wang (2021)
	↑ Surface tension	Formation of beaded fibers	Jet instability causing electrospray	Ren et al. (2017)
	↓ Flow rate	Fiber diameter increase Formation of beaded fibers	Higher mass flow results in thicker fibers and, above a critical value, beaded fibers are observed due to unstable jet formation	Fong et al. (1999) and Topuz et al. (2021)
Process parameters	↑ Applied voltage	Fiber diameter increase (in most cases) Formation of beaded fibers	There is an increasing electrostatic repulsion of charges on the surface of the droplet, facilitating the formation of fibers	Topuz et al. (2021)
	↑ Distance needle-collector	Fiber diameter decrease	Higher distances mean that the solvent will be allowed to vaporize before reaching the metal plate, forming thinner fibers	Fong et al. (1999)
Environmental Parameters	↑ Relative humidity	Decrease in fiber diameter Uneven diameter distribution	Longer jet elongation time due to slow solidification	Tripatanasuwan et al. (2007)
	Temperature	Has no direct effect on fiber morphology	Effect on parameters such as solution density, vapor diffusivity, viscosity, relaxation time, etc	Thompson et al. (2007) and De Vrieze et al. (2009)

Throughout this time, the great challenge for researchers was to discover different solvents that met the dielectric requirements to be used in cellulose electrospinning processes (Prasanth et al. 2014; He et al. 2015). The destruction of highly ordered crystalline regions through the dissolution of cellulose and the reduction of its intermolecular interactions in the dissolved state is fundamental to the electrospinning process (Ohkawa et al. 2009). Frey (2008) reviewed some of the results presented throughout the twentieth century and the beginning of the twenty-first century. Therefore, this review will focus on important results reported more recently.

An efficient solvent for electrospinning requires semi-conductivity with moderate charge capacity, high volatility for rapid solidification of polymeric fibers, and the ability to dissolve polymers with less intermolecular interaction (Ohkawa et al. 2009). Among the various solvent systems used for dissolution of cellulose for electrospinning, the mixture of lithium chloride and N,N-dimethylacetamide (LiCl/DMAc) is one of the most studied. Although being widely used, the solubility of cellulose in this solvent depends on factors such as its source, molecular weight and degree of crystallinity (Sadeghifar et al. 2019). However, its low toxicity and ability to dissolve

different types of cellulose with negligible degradation, make it a promising solvent system (Chen et al. 2015).

Chen et al. (2015) highlighted the importance of carrying out an activation on cellulose before its dissolution in LiCl/DMAc. Among the procedures employed, water presented a good activation result, but it impaired the electrospinning process. The best result was obtained by pre-activation with monohydric alcohols, which enabled the dissociation of hydrogen bonds from cellulose and allowed the production of well-distributed ultrafine cellulose fibers, with a diameter in the range of 500 nm–2 μm. Li et al. (2011) used cellulose from cotton, which was also pretreated, to weaken the polymer chains and later dissolved it in LiCl/DMAc. The high degree of polymerization of cotton cellulose (12,000) conferred high viscosity to the solutions, even in low concentrations, and the increase in concentration from 1 wt% to only 1.3 wt% impaired the normal flow. In the optimized condition, fibers with mean diameters of 150 nm were obtained.

He et al. (2015) studied the key factors of the electrospinning process, such as the intrinsic properties of the solution, using cotton cellulose and LiCl/DMAc as a solvent. The authors observed that a well-defined range of cellulose concentration is essential for electrospinning, since low concentrations lead to low viscosities and do not allow the formation of continuous fibers in the collector, and high concentrations cause high viscosity and surface tension, impairing the process of electrospinning. Besides, a relationship was observed between the applied voltage and the diameter distribution of the fibers. An increase in voltage contributes to the radial force to be stronger than the cohesive force of the polymer jet. Thus, the single jet can be divided into two or more jets, leading to a decrease in the diameter of the fibers and to a more uniform diameter distribution.

Another solvent commonly used in the dissolution of cellulose is trifluoroacetic acid (TFA), a very strong organofluorine compound that dissolves cellulose by attacking glycosidic linkages and that can be easily evaporated from the solution due to its low boiling point (72.4 °C) (Kiper et al. 2020). Ohkawa et al. (2009) proceeded to successfully electrospinning cellulose obtained from cotton and wood pulp by dissolving it in TFA. The authors obtained spinning concentrations in the range of 4.0–5.0 wt% and mean fiber diameter of 40 nm. Smaller concentrations provided low viscous solutions and their electrostatic deposition generated mainly bead, instead of uniform fibers. Higher concentrations resulted in failure of the jet emission due to the high viscosity. The produced nanofiber matrix exhibited an almost amorphous state, implying the disruption of crystalline regions of cellulose by TFA.

Montaño-Leyva et al. (2011) proposed the production of electrospun nanofibers from durum wheat straw cellulose dissolved in TFA. Tests with the addition of water or acetic acid to TFA, aiming to decrease the volatility of this solvent, generated beads instead of fibers, possibly due to incomplete evaporation of TFA. NFs with high thermal stability, high crystallinity, and extremely high contact surface were obtained, making them suitable to reinforce biomaterials. Robles-García et al. (2018) used agave bagasse, an agro-industrial residue, to obtain cellulose for electrospinning. Nanofibers with an average diameter of 54.57 nm were obtained and there was a loss of crystallinity of cellulose because of the use of TFA as a solvent.

Kiper et al. (2020) compared the use of LiCl/DMAc and TFA as solvents for the electrospinning process of cellulose obtained from recycling toilet paper. Dissolving such cellulose samples in LiCl/DMAc was not simple, generating very viscous solutions and difficulty in electrospinning. In TFA, however, the dissolution occurred more easily and generated significantly less viscous solutions, suited to be electrospun. The advantage of TFA over LiCl/DMAc was its ability to also dissolve hemicellulose and lignin, which are usually present when the cellulose source is a paper-derived material. Even though the generated fibers are ultrafine with nanoscale diameter, their morphologies are mostly in the shape of bead-on-string, which in general are regarded as poor quality fibers, but some specific applications such as drug delivery or separation processes can take advantage of these characteristics.

Another aqueous solvent widely used to dissolve cellulose for electrospinning is N-methylmorpholine-N-oxide (NMMO), capable of dissolving it by penetrating into the amorphous regions and causing the molecular chains in these regions to disentangle, followed by the scattering of the crystalline regions with greater penetration of the solvent and consequent dissolution of the polymer (Peng et al. 2017). Tomaszewski et al. (2012) studied the production of

cellulose nanofibers using NMMO/H_2O as a solvent. The authors verified the need for a hydrothermal pretreatment of cellulose to obtain an adequate solution for electrospinning, which resulted in a reduction of the degree of polymerization from 1400 to 370. Solutions with a concentration of 3 wt% and 6 wt% provided fibers with an average diameter of 0.66 μm and 10.6 μm, respectively, with irregular shapes and frequent joints of thin and thick fibers. Absar et al. (2015) found that the solution resulting from the dissolution of cellulose in NMMO showed a paste-like viscosity, impairing its electrospinning because of high instability in jet. NFs with a diameter of 70–90 nm were obtained, but they were discontinuous and had many surface defects.

The most common solvents cited previously can be replaced by ionic liquids, considered a potential green solvent for dissolving cellulose, with high thermal and chemical stability and a simple recycling process (Ahn et al. 2012). Ionic liquids are molten salts composed of organic cations and inorganic anions, with the cations having a melting point below the boiling point of water, and making it possible to vary the combinations of cations and anions to suit a specific application. In the case of cellulose dissolution, ionic liquids are of great interest due to their highly polar character, low vapor pressure, and loosely coordinating bulky ions that interact with hydroxyl groups from cellulose (Phadagi et al. 2021). However, its high melting point combined with its low vapor pressure requires application of wet electrospinning, as common dry electrospinning is not capable of producing the desired NFs. By doing this, the electrospun fibers slowly solidify in a coagulation bath, in which the ionic liquid is replaced by a co-solvent, which is later removed by desiccation or freeze-drying (Quan et al. 2010; Ciuzas et al. 2021).

Quan et al. (2010) evaluated the electrospinning of cellulose dissolved in a hydrophilic ionic liquid, 1-butyl-3-methylimidazolium chloride (BmimCl), and also the influence of dimethyl sulfoxide (DMSO) as an additive in solutions with a higher concentration of cellulose. Optimal cellulose fibers were obtained at a concentration of 4.0 wt%. Lower concentrations led to the formation of massive blocks or beads while higher concentrations (higher viscosity) generated larger fibers. The addition of DMSO in the 4.0 wt% solution generated smooth fibers of finer diameters. Ahn et al. (2012) also used an ionic liquid (1-ethyl-3-methylimidazolium acetate—EmimAc) to dissolve cellulose, but with the aid of co-solvents such as dimethylformamide (DMF) and N, N-dimethylacetamide (DMAc), to study the influence of the type and concentration of the co-solvent on spinnability and properties of the resulting fibers. Regardless of the co-solvent used, spinnability was improved by the presence and increased concentration of the co-solvent, due to the reduction in viscosity and the partial polarity of the co-solvent.

Jing et al. (2020) obtained NFs from bacterial cellulose using 1-allyl-3-methylimidazolium chloride and N, N-dimethylformamide (AmimCl/DMF) as the solvente system. When comparing common bacterial cellulose (BC) membranes and electrospun membranes (EBC), the authors observed that electrospinning provided better nanoporosity and fiber arrangement (Fig. 4), in addition to improving water holding capacity.

Table 2 presents a summary of solvent systems used for cellulose electrospinning.

It is common for the electrospinning process to generate NFs with poor mechanical strength, which is one of the major limitations of electrospinning pure cellulose. One of the alternatives to eliminate this issue is the insertion of rigid nanoparticles into the solutions for electrospinning (Zhang et al. 2019). Among them, cellulose nanocrystals (CNCs) can be mentioned, which are crystalline subunits of cellulosic fibers obtained through acid or enzymatic hydrolysis that remove the amorphous region and can be considered an effective reinforcement for polymeric nanocomposites (Habibi et al. 2010). He et al. (2014) produced electrospun nanofibers from cellulose/CNCs nanocomposites, both originated from cotton, using LiCl/DMAc as the solvent system. The authors observed that the addition of CNCs to the system containing cellulose NFs resulted in a well dispersed medium and provided a more uniform morphology, better tensile properties, and improved thermal stability of the resulting fibers. Also, no toxic effects on fibroblast cells were observed, demonstrating their biocompatibility.

In addition to the insertion of rigid nanoparticles to obtain NFs with improved properties, cellulose can also be used with other polymers to produce polymeric composite/blend nanofibers. Awal et al. (2011) evaluated the effect of the main electrospinning parameters on the diameter and morphology of green

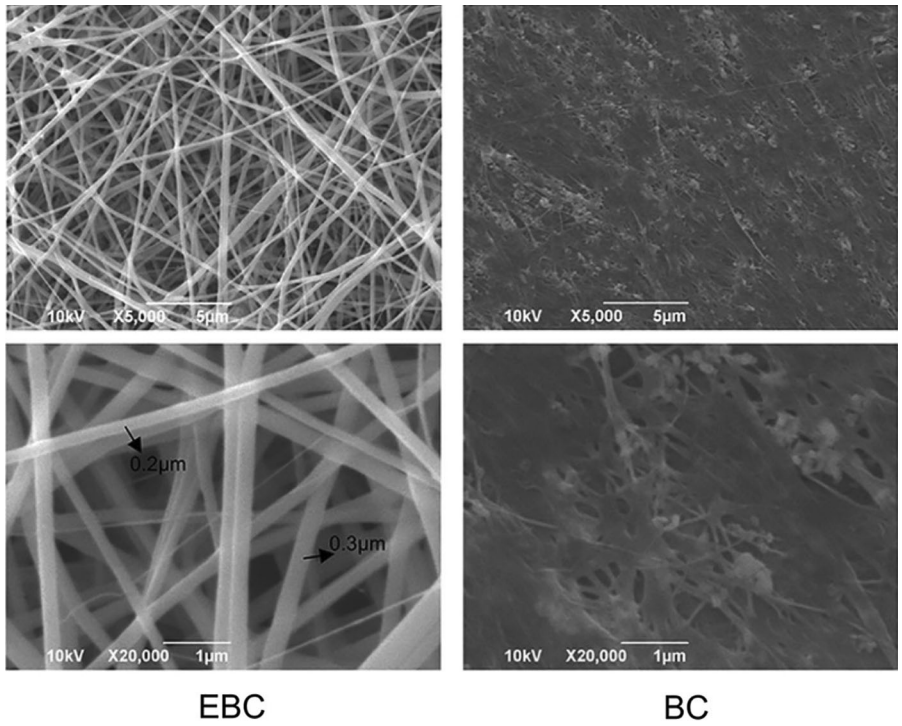


Fig. 4 Microstructure of common bacterial cellulose membranes (BC) and electrospun membranes (EBC) obtained by Scanning Electron Microscopy (SEM). (Reproduction with permission from reference Jing et al. (2020). Copyright 2020 Elsevier)

composite fibers developed from wood pulp cellulose and nylon 6.6, using formic acid as a solvent. The morphology of the fibers was affected by the processing conditions, while the diameter was more significantly affected by the concentration of the solution, although the applied voltage and the distance from the needle to the collector also presented some influence.

Several other authors have evaluated the electrospinning of polymeric composites or blends containing cellulose. Among the polymers used for this purpose, some can be mentioned: poly (2-hydroxy ethyl methacrylate) (pHEMA) (Rao et al. 2014) and poly (ethylene terephthalate) (PET) (de Oliveira Santos et al. 2017), both in TFA solvent, poly (ε-caprolactone) (PCL) in chloroform and TFA as a co-solvent (Ramamoorthy and Rajiv 2015), silk in a TFA and acetic acid co-solvent system (Begum et al. 2018), poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) in chloroform and DMF co-solvent system (Benini et al. 2017) and poly(vinyl alcohol) (PVA) in water (Chahal et al. 2013). In general, the blends and composites cited previously resulted in biocompatible NFs with a bead-free and smooth morphology, with a tendency to reduce porosity and to reduce or to change the crystalline structure of the polymers after the addition of cellulose.

Applications of electrospun cellulose

Due to the high biocompatibility of cellulose, the use of its derivatives in the biomedical area is widely studied. The in vivo application of cellulose-based biomaterials has shown only negligible inflammatory reactions (Zhang et al. 2019). Ohkawa et al. (2009) developed electrospun cellulose fibers and evaluated their use as a drug carrier for epidermic administration, using acetylsalicylic acid (ASA) and nicotine as model drugs. For this study, the drugs were loaded into the pre-spun cellulose solution, followed by electrospinning. The electrospun fibers loaded with nicotine, a liquid at room temperature, proved to be more homogeneous than those loaded with ASA, which is crystalline at room temperature. Also, drug dissolution results showed that the high surface area of the electrospun materials promoted greater drug release,

Table 2 Solvent systems and optimal parameters used in cellulose electrospinning and characteristics of formed fibers

Solvent system	Cellulose concentration (%)	Average fiber size	Tip-to-collector distance (cm)	High voltage (kV)	Feed flow rate (mL/min)	Comments	References
9 wt% LiCl/DMAc	–	500 nm–2 μm	10–15	18–21	0.03	Well-distributed ultrafine cellulose fibers	Chen et al. (2015)
8.5 wt% LiCl/DMAc	1.15 wt%	150 nm	–	–	0.04	Dry, smooth, uniform and well separated nanofibers	Li et al. (2011)
8% LiCl/DMAc	1–2%	220–280 nm	10	20	0.03	Distinct and uniform fiber texture	He et al. (2015)
TFA	4.5 wt%	40 nm	15	15	–	Electrospun nanofibers with an amorphous character	Ohkawa et al. (2009)
	4–5.5%	270 nm	7	15	0.025	Nanofibers remained highly crystalline although the crystallinity of the cellulose was reduced after electrospinning	Montaño-Leyva et al. (2011)
	11%	54.57 nm	5	17	0.0083	Nanofibers demonstrated a fibrillar shape	Robles-García et al. (2018)
70 wt% NMMO/H_2O	3 wt%	0.66–10.6 μm	5	11	–	Fibers with irregular shapes	Tomaszewski et al. (2012)
	6 wt%		5	30	–		
NMMO/H_2O	2 wt%	70–90 nm	10	22	0.03–0.1 mL/h	Interconnected nanofibers	Absar et al. (2015)
BmimCl/DMSO	4 wt%	500–800 nm	15	15	–	Well-defined ultrafine fiber	Quan et al. (2010)

proving that they can be used as drug carriers for epidermic administration.

Other authors made use of nanofibers for drug delivery using cellulose in composites or as a starting material for CNCs, used as a reinforcement component in various polymeric solutions. Rao et al. (2014) and Ramamoorthy and Rajiv (2015) produced composite nanofibers from cellulose and poly (2-hydroxyethyl methacrylate) (pHEMA) and poly (ε-caprolactone) (PCL), respectively. In the former, cellulose was obtained from bamboo and evaluated as nanocomposite fibers for incorporation of the anticancer drug paclitaxel, with promising results for its applicability as an anticancer scaffold for, e.g., the treatment of skin cancer. In the last study, wheat cellulose was used in composites to incorporate L-carvone essential oil, known to be antibacterial, antifungal, antioxidant, and insecticidal. The entrapment of L-carvone was effective, with little change in thermal stability and therefore showing the suitability of these NFs in antimicrobial textile applications.

Hivechi et al. (2019) and Chen et al. (2020b) used CNCs as reinforcement materials for PCL and poly (N, N-dimethylaminoethyl methacrylate) (PDMAEMA), respectively. In the case of CNC-*g*-PDMAEMA, the combination with poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) was used to produce the polymeric NFs. Hivechi et al. (2019) found that the incorporation of CNCs improved the biodegradability of PCL and generated optimal mechanical behavior, under the condition of 1% CNC in PCL solution. The antibiotic tetracycline was incorporated into the PCL and CNC NFs system, resulting in controlled and slower release of the drug by increasing CNC concentration. The same drug was used as a model drug in the study by Chen et al. (2020b), who achieved long-term sustained release behavior. The addition of CNC-*g*-PDMAEMA to PHBV improved thermal

properties, crystallization, hydrophilicity and a more uniform diameters of the fibers.

In the tissue engineering field, cellulose can also be used in its pure form, as a reinforcement material or in composites with other materials. He et al. (2015), using pure cotton cellulose in a LiCl/DMAc solvent system, developed electrospun non-woven NFs for potential application in tissue engineering. Almost amorphous and highly aligned nanofibers were obtained, with promising results for application as a scaffold. Experiments with human dental follicle cells showed that not only did the cells attach to the surface of the scaffolds, they were also able to proliferate throughout the inner part of the scaffold. A similar application was evaluated by Ao et al. (2017), but using a scaffold electrospun from cotton cellulose and nano-hydroxyapatite (nano-HA) composites. The presence of nano-HA contributed to the increase of the average diameter of the fiber and the tensile strength of the electrospun NFs. In vitro tests have shown that nano-HA did not generate cytotoxicity in cellulose scaffolds and allowed cells to proliferate properly, especially by allowing the obtention of a structure that is similar to the natural extracellular matrix.

The reinforcement of CNCs was evaluated by its graft in poly (ethylene glycol) (PEG) and later incorporation in poly (lactic acid) (PLA) to produce electrospun scaffolds. Initially, it was observed that CNC-g-PEG showed better dispersion in the DMF solvent than the unmodified CNC. When comparing PLA, PLA/CNC, and PLA/CNC-g-PEG scaffolds, it was found that all of them had homogeneous and randomly oriented fibers, with no visible beads, as it can be seen in Fig. 5. Also, the addition of CNCs or CNC-g-PEG did not alter the wettability of the original material, indicating that the fillers remained inside of the fiber matrix instead of the surface, which would generate a hydrophilic behavior. The PLA/CNC-g-PEG nanofibers showed improved tensile strength when compared to PLA/CNC, while both were non-cytotoxic and capable of attaching and growing human mesenchymal stem cells (Zhang et al. 2015).

Apart from the biomedical application, cellulose electrospinning can also find use in the energy sector, specifically in next-generation batteries, based on the Li–S chemistry (lithium-sulfur batteries—LSBs). It is common for LSBs to have problems of low conductivity and degeneration of its performance, associated with the presence of sulfuric cathodes, requiring the introduction of an interlayer, such as activated carbon NFs based on electrospun cellulose, which guarantees a high surface area and high conductivity (Park et al. 2021). It is also possible to apply cellulose nanofibers in oil–water separation processes, for example, to solve environmental issues or those of the oil industry itself. Cellulose NFs are capable of separation efficiencies above 98%, with good reusability and degradability (Shu et al. 2020).

Electrospinning of cellulose acetate

Cellulose acetate (CA), a semi-synthetic polymer, is produced from the esterification reaction of cellulose with acetic anhydride in the presence of sulfuric acid as a catalyst (Nicosia et al. 2016; Rosdi et al. 2018). Despite being extensively investigated for its low cost and easy processability, factors such as a maximum operating temperature of approximately 40 °C and a narrow pH range (3–6) lead to the reduction of its potential applications (Guglielmi and Andreottola 2010). Due to its properties such as good thermal stability, chemical resistance, biodegradability, and biocompatibility, electrospun CA has been gaining a lot of attention.

Similarly to what has been described previously for pure cellulose, the selection of the ideal solvent is one of the starting points for the production of electrospun nanofibers of CA, due to the strong influence that the solvent has on the diameters of the fibers. The choice of the appropriate solvent system has been commonly based on trial and error, similar systems or solubility models (Konwarh et al. 2013). Studies have been developed to evaluate the effect of several solvent systems, either containing only one solvent or binary and even ternary systems (Liu and Tang 2007; Tungprapa et al. 2007; Rodríguez et al. 2012; Naragund and Panda 2020). Solvents such as chloroform, acetone, N,N-dimethylformamide (DMF), formic acid, dichloromethane (DCM), methanol (MeOH), trifluoroacetic acid (TFA), and pyridine are used in simple solvent systems (Mikaeili and Gouma 2018; Angel et al. 2020; Li and Yang 2020). In mixed systems, binary mixtures of acetone/DMAc, chloroform/MeOH, DCM/MeOH and acetic acid/water can be used (Pang et al. 2016; Lee et al. 2018; Dizge et al. 2019; Wang et al. 2020), and ternary mixtures as is the

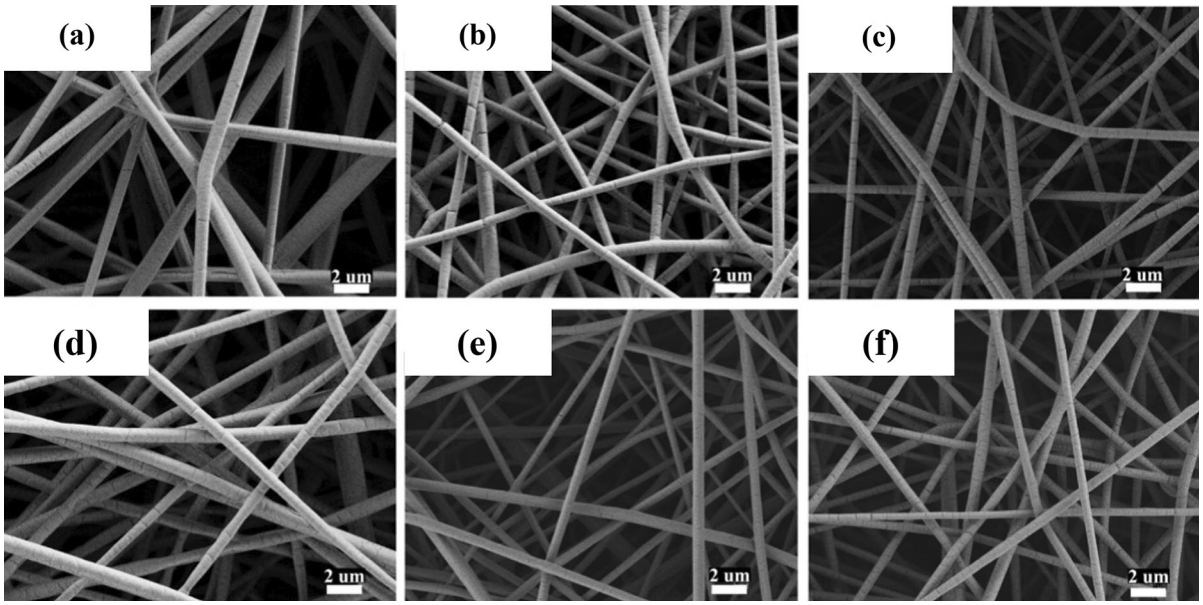


Fig. 5 SEM images of electrospun nanofibers: **a** PLA, **b** PLA/CNC (1%), **c** PLA/CNC (5%), **d** PLA/CNC-g-PEG (1%), **e** PLA/CNC-g-PEG (5%), and **f** PLA/CNC-g-PEG (10%). (Reproduction with permission from reference Zhang et al. (2015). Copyright 2015 Elsevier)

case of acetone/DMF/trifluoroethanol (Ma and Ramakrishna 2008).

However, in some cases of single solvent systems, the dielectric constant and the boiling point of the solvent are low, making fiber formation difficult, as in the case of acetone (Liu and Hsieh 2002; Rodríguez et al. 2012). As a result, mixed solvent systems are more efficient in the formation of CA electrospun fibers (Tungprapa et al. 2007). In most cases, binary solvent mixtures consist of solvents with a high boiling point (DMAc, acetic acid or water) and solvents with a low boiling point, such as acetone and MeOH (Celebioglu and Uyar 2011). According to the results presented by Majumder et al. (2019), the DMAc/acetone binary mixture presents itself as the ideal solvent system in obtaining electrospun bead-free CA fibers with uniform diameter distribution.

Despite not being an ideal solvent for CA, water has been tested in solvent systems with acetone for promoting a decrease in the general evaporation rate, allowing the formation of fibers during the electrospinning process (Frey 2008). However, the fibers obtained by this type of system have a ribbon format, usually attributed to the viscosity of the solution, requiring an optimization in the concentration of the polymer necessary to maintain the formation of fibers instead of tape-shaped materials (Olaru and Olaru 2010; Nista et al. 2012; Majumder et al. 2019). When evaluated in an acetic acid/water system, it is possible to obtain fibers that have nanoheterogeneous morphology in the single fiber unit, with an average fiber diameter of approximately 2 μm, and without beads or granules at a concentration of CA of 21%, a 3:1 ratio of the solvents, a voltage of 21 kV and a flow of 0.8 mL/h (de Almeida et al. 2020).

More recently, Naragund and Panda (2020) evaluated the mixed system containing methyl-ethyl ketone (MEK)/DMAc in different proportions and observed that when these solvents were evaluated separately they produced only beads and not fibers. When evaluated in a mixed solvent system, the authors observed the formation of cylindrical nanofibers with increasing polymer concentration, as it can be seen in the SEM micrographs in Fig. 6. Additionally, the authors were able to observe that higher concentrations of CA were needed to form the cylindrical fibers when the solvent system had high surface tensions due to high concentrations of DMAc.

Due to the high temperatures, in the range of 80–130 °C, the direct electrospinning of cellulose in hydrated NMMO solution ends up producing relatively less uniform NFs due to the fusion of the NMMO hydrate. To solve this problem,

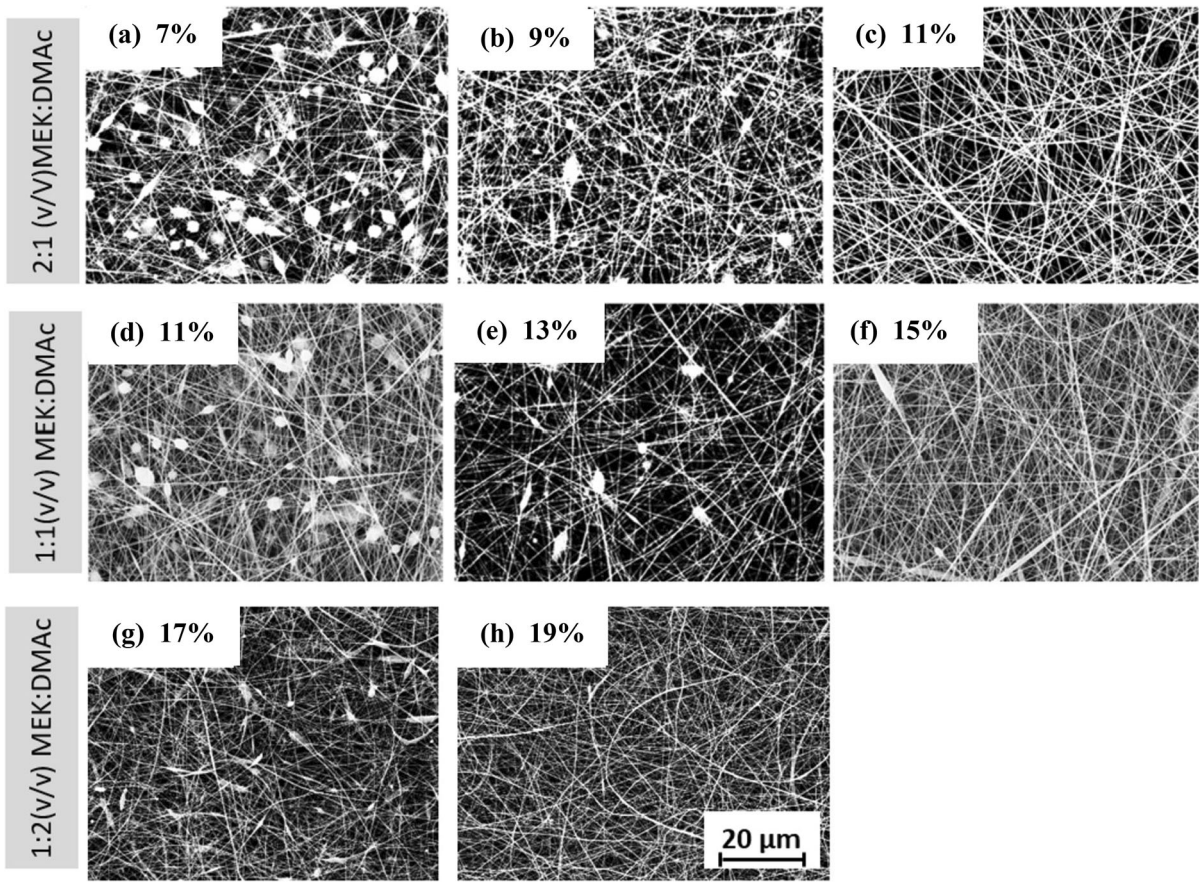


Fig. 6 SEM micrographs showing effect of polymer concentration and solvent ratio on morphology of nanofibers. (Reproduction with permission from reference Naragund and Panda (2020). Copyright 2020 Elsevier)

electrospinning the CA followed by its deacetylation is an alternative (Han et al. 2008).

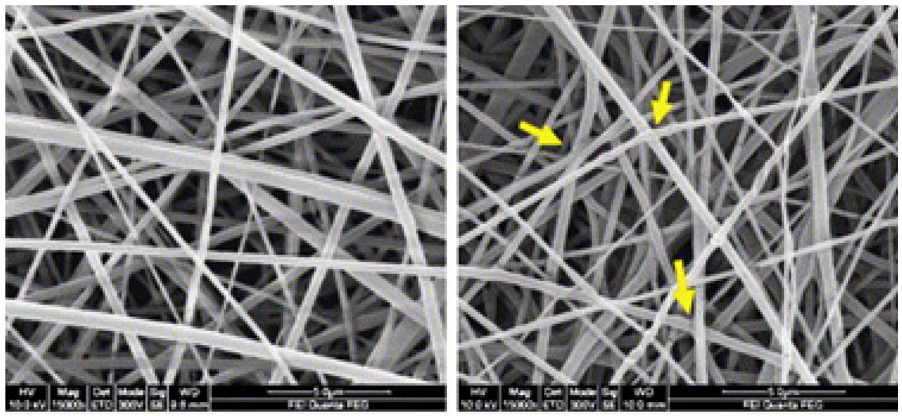
Ma and Ramakrishna (2008) evaluated the feasibility of applying cellulose NFs membranes as an affinity membrane. The authors prepared the cellulose NFs from the electrospinning of cellulose acetate, using a solvent mixture containing acetone/DMF/trifluoroethylene (3:1:1). Cellulose NFs were prepared by treating CA NFs with NaOH in water/ethanol. When evaluating a ternary solvent system, the formation of an electrospun CA nanofiber fabric is observed, which presents a very weak mechanical resistance, such as very fluffy cotton, requiring a heat treatment at a temperature close to the melting point of the material for the nanofibers to merge, as can be seen in the SEM images in Fig. 7.

Table 3 presents some solvent systems developed for CA electrospinning, highlighting the size of the fibers and some process parameters used.

Electrospinning of CA with other materials

Due to the limited solubility in most organic solvents, little molecular flexibility, and the ability to form hydrogen bonds in the three-dimensional structure, electrospinning of biopolymers has proved to be a great challenge. To improve the electro-spinnability of CA, the mixture of CA with different polymers, either natural or synthetic, promotes an improvement in fiber properties such as tensile strength, as shown by Baek et al. (2011). As a result, several studies were carried out to evaluate the electrospinning of cellulose acetate with other materials such as synthetic and natural polymers, nanomaterials, and, more recently, with gums, as described below.

ElMessinry and Fadel (2019) prepared NFs from PVC/CA blends and dissolved them in a THF/DMF solution to improve the mechanical resistance of the nanofiber mat. An improvement in the properties of



(a) (b)

Fig. 7 SEM images of the surface of: (a) untreated CA nanofibers and (b) CA nanofibers with heat treatment. (Reproduction with permission from reference Ma et al. (2005). Copyright 2005 Elsevier)

electrospun fiber carpets has been reported at a concentration of 8% CA. Such blend promoted a 350% increase in the breaking strength of the PVC/CA nanofiber blanket and a break length of 210%.

The use of nanofibers of CA encapsulated with sodium polyacrylate (SPA) as absorbents were evaluated by Yadav et al. (2016). In the study, a solvent system composed of acetone/DMAc was used, in a volumetric ratio of 2:1 (v/v), a voltage of 10–12 kV and a distance of the needle-collector of 10 cm. In comparison to commercial absorbents, there was a reduction in the diameter of the fibers by two orders of magnitude for the SPA/CA and long and continuous changes, resembling pure CA fibers.

Chen et al. (2013) prepared, via coaxial electrospinning, ultrafine phase change fibers (PCFs) from poly(ethylene glycol)/CA blends in a mixed solvent system composed of acetone/DMAc (2:1, w/w). The fibers presented a typical core-sheath structure, where PEG was completely encapsulated as a thread by the CA sheath, and both morphology and thermal properties are affected relatively by the concentration of PEG fibers.

Biodegradable and biocompatible polymeric systems composed of a hydrophobic polymer (PLA) mixed with a hydrophilic polymer (CA and/or PEO) were prepared by Elsayed and coworkers to increase cell interaction with fibroblasts (Elsayed et al. 2020). The synthesized fibers showed smooth, dense, and uniform bead-free morphology, with an average diameter ranging between 53 and 161 nm. However, it was observed that the incorporation of CA in high concentrations in the matrix with PLA resulted in fibers with smaller diameters, around 53–58 nm, due to the secondary interactions between the polymeric chains of PLA and CA, which consequently reduced the viscosity of the solution (Thompson et al. 2007).

The preparation of electrospun nanofiber scaffolds from a poly (hydroxybutyrate)/CA mixture using chloroform /DMF as a co-solvent system was evaluated by Zhijiang et al. (2016). The scaffolds made up of PHB/CA have a porous structure and the nanofibers are cylindrical, uniform in diameter, bead-free and have random orientation. The obtained fibers were of nano and submicron size, with a diameter varying between 80 and 680 nm, comparable to collagen fibers (50–500 nm).

In some applications, such as the encapsulation of probiotics, the use of electrically spun CA fibers is limited due to the solvents commonly used, which reduce cell viability. With that in mind, a double-angled nozzle electrospinning system to manufacture PVA/CA composite fibers was proposed by Çanga and Dudak (2021). Smooth, bead-free fibers were produced, with an average diameter of 754 nm, being larger than pure PVA and pure CA fibers.

More recently, electrospinning of the mixture of a natural hydrocolloid with CA was evaluated. Çanga and Dudak (2019) used gum Arabic (GA) as a water-soluble component with CA hybrid fibers. The results reported the obtainment of bead-free CA-GA hybrid fibers with smooth surface, with their diameters ranging from 527 to 670 nm. There was an increase

Table 3 Solvent systems used in CA electrospinning and characteristics of formed fibers

Solvent system	CA concentration (%)	Collector	Tip to collector distance (cm)	High Voltage (kV)	Feed flow rate (mL/h)	Average fiber size (nm)	Comments	References
Acetone/DMF / Water (3:2:1)	5	Aluminum foil	10	16	–	–	Messy and coarse fibers with many beads or clumps due to the viscosity of the solution	Chen et al. (2020a)
Single solvent system		Aluminum foil					There was no fiber formation	Naragund and Panda (2020)
MEK	8 and 10		15	15	0.5–1.0	–		
DMAc	15 and 20					–		
Mixed solvent system MEK/DMAc (2:1) (1:1) (1:2)	7–19		10	19	2.0	40–500	An increase in the average diameter of the fibers with the augmentation in the concentration of CA	
DMF/DCM (9:1) (5:1) (1:1)	8	Metal plate	15	15	1.0	700–800	1:1 ratio can produce granule-free fiber membranes with superior mechanical properties	Li and Yang (2020)
Acetone/Water (8:2)	10	Glass plate	0.02	–	–	3000–4000	Uniform fibers with smooth surfaces	Hong and Kang (2020) and Hong et al. (2020)
Acetone/DMAc (2:1)	17	Aluminum rotating drum collector	15	17.5	0.4	616–624	Smooth, uniform and bead-free nanofibers were obtained At higher rotations (6000 rpm), there was greater fiber alignment	Patrojanasophon et al. (2020)
Acetone/DMAc (2:1)	13.5	Aluminum rotating drum collector	10	14	2	600	Smooth fibers and uniform fiber entanglements	Das and Gebru (2017)

Table 3 continued

Solvent system	CA concentration (%)	Collector	Tip to collector distance (cm)	High Voltage (kV)	Feed flow rate (mL/h)	Average fiber size (nm)	Comments	References
Acetone/DMAc (2:1)	17	Aluminum rotating drum collector	15	17.5	0.4	500–730	Smooth, uniform and bead-free nanofibers were obtained and fibers with smaller diameters and better aligned at higher collector drum rotations	Tidjarat et al. (2014)
Acetone/DMAc (1:1) (2:1) (3:1)	16	Aluminum sheet	–	–	–	160–320	Smooth fibers	Tungprapa et al. (2007)
Chloroform/MeOH (9:1) (4:1) (7:3) (3:2) (1:1)	5					520–1090	Smooth fibers	
DCM/MeOH (9:1) (4:1) (7:3) (3:2) (1:1)	5					670–1060	Smooth fibers	
Acetone/DMF/Trifluorethanol (3:1:1)	16	Aluminum plate	15	25	4.0	200–800	Fibers with ~ 80% porosity and distributed	Ma and Ramakrishna (2008)
Acetone/DMAc/Ethanol (4:1:1)	10	Aluminum foil	15	8–15	1.0	100–200	Continuous and fine fibers	Wu et al. (2010)

in fiber diameter with higher concentrations of GA due to the increased viscosity of polymer solutions.

Another alternative to enhance the properties of electrospun CA fibers is the use of blends with other natural polymers, such as chitin (Goetz et al. 2016; Pereira et al. 2020), chitosan (Phan et al. 2019), silk fibroin (Arumugam et al. 2021) and collagen (Lukanina et al. 2018), gelatin (Farahani et al. 2020). The results show that CA matrices loaded with other natural polymers have better mechanical properties,

mostly smooth fibers without beads in nanometric scales and high stability in water, resulting to be materials of great interest for biometric applications or as filter media.

Applications of electrospun CA

Electrospun CA nanofibers have been studied for several applications due to their properties, such as biocompatibility, biodegradability, and non-toxicity. Immobilization of bioactive substances, drug delivery, battery separator, water treatment, dressings, tissue engineering and female intimate absorbent are some examples of the several possible applications of CA electrospun NFs.

The drug delivery by electrospun fibers plays an important role for biomedical applications, due to high flexibility, improved control over the kinetics of drug release, simultaneous delivery of different therapies and promotion of local therapeutic effects (Khoshnevisan et al. 2018). The manufacture of electrospun NFs from biopolymers has gained prominence in the delivery of different therapies in drug distribution systems because of their increased surface-to-mass ratio, ease of operation, and excellent cost–benefit ratio. Using a new approach via modified triaxial electrospinning, Liu et al. (2019) elaborated scaffolds with the adjusted zero-order release of ferulic acid (FA) from nanoscale formulations of CA. In the study, two un-electrospinnable liquids were used as the external and intermediate working fluids, with only the core solution being individually electrospinnable into fibers. High-quality core–shell NCs were generated with a thin CA coating and a FA-loaded core. The same approach was used by Yang et al. (2019), in which they manufactured core–shell nanostructures comprised of a drug-protein coated with a thin layer of CA for the controlled release of ibuprofen. The reported results showed that the NFs had linear and cylindrical morphologies with a diameter ranging from 0.66 to 0.87 μm. The presence of the CA coating extinguished the initial release of ibuprofen, allowing the drug to be manipulated at a percentage of 90% during a tunable interval of 23.5 to 43.9 h.

Arumugam et al. (2021) recently investigated the biocompatible potential of electrospun NFs composed of SF/CA/Au–Ag for anticancer applications. As a result, the composite nanofiber exhibited excellent biological activity compared to other fibrous materials. This simple and economical approach to manufacture composite nanofibers with enhanced anticancer capability is very promising for biomedical applications.

Dos Santos et al. (2021) prepared scaffolds of CA NFs loaded with annatto (*Bixa Orellana* L.), a natural dye from tropical plants of South America, and evaluated their potential for wound healing. The authors reported that NFs promoted fibroblast cells proliferation and that they spread and penetrated the structure after 48 h. Also, the bioactive molecules were able to modulate the inflammatory process in vitro, indicating the potential of this material for applications in wound healing. The use of CA NFs as a biocompatible and antimicrobial dressing was also evaluated by Ullah et al. (2020), but using electrospun CA and Manuka honey (MK) NFs. The authors reported that the inclusion of MK in the CA-MK mats shows high efficacy in preventing bacterial growth (*E. coli* and *S. aureus*) on the wound surface, in addition to possessing good antioxidant capabilities, high porosity and adequate values of water vapor transport rate for breathability of the wound. Khan et al. (2019) developed new high-profile antibacterial dressings from the electrospinning of CA and silver sulfadiazine (SSD) nanoparticles. It was observed that the addition of SSD does not alter the morphology and diameter of the CA fibers, in addition to being uniformly distributed throughout the fiber. The NFs showed antimicrobial activity against several bacteria and can be used as a promising and differentiated product for applications in dressings.

The performance of electrically spun fibrous filter used for microfiltration of high-flow water has recently been investigated. Zhou et al. (2016) manufactured and characterized non-woven fibrous CA membranes with varying diameter and areal-weight for application as a filter medium. The results showed that the rate of rejection of CA fibrous membranes to polystyrene (PS) particles with a diameter of 2 μm was 99.8%, in addition to an increase in filtering capacity as there is an increase in the areal-weight of the membrane. Goetz et al. (2016) coated electrically spun CA membranes with chitin nanocrystals to obtain water filtration membranes with customized surface characteristics. A significant impact was observed on the surface properties of the membranes with the chitin nanocrystal coating, producing a superhydrophilic

membrane, also showing a significant reduction in biofouling and biofilm formation.

Recently, Bhute and Kondawar (2019) prepared a new polymer electrolyte membrane of hybrid poly (vinylidene fluoride) PVF/CA/$AgTiO_2$ polymer by electrospinning to apply in a lithium-ion battery separator. To manufacture the membranes, Ag-TiO_2 nanoparticles were incorporated into the PVF/CA electrospinning solution. The membranes presented excellent thermal stability, high porosity and super-electrolytic compatibility. They also presented high ionic conductivity and good electrochemical performance, making CA-based composite polymer electrolyte membranes potential candidates for lithium-ion batteries.

The application of electrospun cellulose acetate fibers as pre-cursor materials for carbonization aiming their application as supercapacitors has been reported in recent literature (Deng et al. 2013; Li et al. 2018; Cao et al. 2020).

In order to solve the problem of poor thermal stability of cellulose, which results in the morphological collapse of carbon nanofibers (CNFs) based on pure biomass, Cao et al. (2020) proposed a strategy to simulate the covalent bond between lignin and cellulose acetate for the preparation of CNFs in order to improve their extensibility and stability. The CNFs were prepared from electrospinning and the fibers exhibited independent filamentous morphology, uniform diameter, large surface area, and excellent storage capacity. The specific capacitance of 320.3 F/g was obtained using CNFs, prepared with 10% epichlorohydrin, like a supercapacitor of the CNFs.

The effects of multi-walled carbon nanotubes (MWNT) on the stabilization and carbonization of cellulose nanofibers were evaluated by Deng et al. (2013). For this, the authors produced cellulose nanofibers from a solution of CA followed by deacetylation to regenerate into cellulose and followed by the incorporation of MWNT. The results showed that the incorporation of MWNT reduced the activation energy of the oxidative stabilization of cellulose nanofibers. Furthermore, they promoted an increase in crystallite size, structural order, and electrical conductivity of activated CNFs.

Electrospinning of other cellulose derivatives

Cellulose acetate butyrate

Along with CA, cellulose acetate butyrate (CAB) and cellulose acetate propionate (CAP) also belong to the cellulose ester class (Edgar et al. 2001). Unlike CA, these polymers have been underexplored as electrospun fibers, especially CAP. However, the excellent solubility and structural stability of these polymers make them good candidates for producing electrospun fibers with unique characteristics (Huang et al. 2011).

When compared to cellulose acetate, CAB has improved dimensional stability and has higher chemical and moisture resistances. Additionally, it is inherently hydrophobic because of its butyryl chains (Huang et al. 2018). Recently, Tanvir et al. (2020) produced superhydrophobic electrospun CAB fibers that exhibited four times higher oil adsorbent capacity (60 g/g) than commercial polypropylene. The authors also reported that the surface porosity of CAB fibers considerably increased the oil sorption capacity, with the hydrophobicity of the porous fibers being superior than that of non-porous CAB fibers.

Nevertheless, the hydrophobicity of the CAB limits its application in the biomedical field because it compromises the adhesion and spreading of cells on fibrous CAB scaffolds (Huang et al. 2013; Tan et al. 2020). An alternative that has been performed is the combination of CAB with other polymers for blend formation, aiming to enhance its hydrophilicity. Recently, Tan el al. (Tan et al. 2020) manufactured scaffold of fibrous structure via electrospinning using CAB and polyethylene glycol (PEG). The polymers were dissolved in acetone and dimethylacetamide mixture and electrospun on a grounded plate collector, resulting in a fibrous CAB scaffold and fibrous CAB/PEG scaffold. Both scaffolds presented excellent biocompatibility in normal human dermal fibroblasts, but the cell adhesion on the CAB/PEG scaffold was greater than on the CAB scaffold. The authors also reported higher crystallinity, enhanced tensile strength and improved biodegradation of the CAB/PEG composite nanofibers.

Huang et al. (2013) compared conventional needle electrospinning and disc electrospinning of CAB. Fibers produced by both methods displayed similar surface morphology and the same chemical composition, but the disc-electrospun fibers had higher

crystallinity and wider diameter distribution. Additionally, by using disc as a spinneret, the authors were able to achieve a productivity 150 times higher than that of needle electrospinning. By cultivating fibroblast and Schwann cells on electrospun nanofiber webs, the ones produced by disc electrospinning showed enhanced cellular growth.

In biomedical applications, such as tissue engineering, sterilization of NFs is very important. However, sterilization methods can significantly affect the properties of fibrous scaffolds and, when using thermosensitive polymers, most of the methods cannot even be applied. To address this issue, Elashnikov et al. (2019) studied the effect of UV and 70% ethanol sterilization methods on CAB NFs. Regarding fiber morphology, UV sterilization had no influence whereas ethanol led to reduced porosity. Wettability was increased with the ethanol method. In vitro tests with SH-SY5Y cells showed that cell attachment, growth and migration is higher on the ethanol-sterilized CAB nanofibers. A summary of studies involving electrospinning of CAB and other cellulose derivatives is shown in Table 4.

Methyl cellulose and ethyl cellulose

Regarding the electrospinning of cellulose ethers, methylcellulose has a scarce number of studies. Frenot et al. (2007) investigated the spinnability of MC dissolved in a water and ethanol mixture in a proportion of 1:1 (w/w), using a voltage of 40 kV. The authors reported unsuccessful electro-spinnability, however, the reason for this was superficially explored. The SEM images revealed incomplete drying regions and coarse fibers, showing the low volatility of the solvent mixture during the electrospinning process used.

Unlike MC, fibers of ethyl cellulose (EC) were manufactured successively and extensively via electrospinning, even though its processing is challenging because of the formation of beads (Zaitoon and Lim 2020). EC is used as an emulsifier, thickening agent and dietary fiber in the food industry. Electrospun EC fibers, when used as a starting material or scaffold, have been reported to be potentially applied in metal adsorption (Tabatabaeefar et al. 2020), antimicrobial systems (Park et al. 2015), drug delivery (Jash and Lim 2018) and food packaging (Niu et al. 2020).

The electro-spinnability of EC fibers can be improved by the addition of a second polymer, such as gelatin (Liu et al. 2018a, b). Zaitoon and Lim (Zaitoon and Lim 2020) investigated the influence of poly(ethylene oxide) (PEO) on the electro-spinnability of EC fibers. The authors observed that electrospinning of 10% (w/w) EC resulted in the formation of interweaved particulates and fibers. When adding 1% PEO to the electrospinning solution, the spinnability was enhanced due to changes in solution viscosity, surface tension and conductivity. Such changes may be originated due to the interactions between ether groups of PEO with hydroxyl groups of EC, which were observed in FTIR spectra. These parameters contribute significantly to the jet stability during the electrospinning process, affecting the spinnability.

Li et al. (2014) combined the encapsulation technology with electrospinning for producing complex structure fibers containing quercetin, an important bioactive flavonoid with anti-inflammatory and anti-hypertensive properties (Anand David et al. 2016). Electrospun core–shell fibers were prepared, with the core phase containing a mixture of EC and quercetin, and the shell phase containing a mixture of poly (vinyl pyrrolidone) (PVP) and quercetin. The material offered tunable biphasic release of quercetin in physiological saline media at human body temperature.

Carboxymethyl cellulose

Carboxymethyl cellulose is a water-soluble anionic ether derivative of cellulose, which is normally applied as its sodium salt (NaCMC). However, it is not soluble in organic solvents and it is difficult to be processed by thermoplastic methods because of the strong interchain hydrogen bonds (Lin et al. 2013). Electrospinning of pure CMC is hindered because of its rigid structure, which does not allow chain entanglement and its high gel formation tendency drastically increases the viscosity of the solution, making it difficult to form CMC fibers during electrospinning process (El-Newehy et al. 2016; Hashmi et al. 2020). Due to this reason, other hydrophilic polymers are used as spinning agents to enhance the CMC spinnability and processability, such as PEO (Shi et al. 2016; Crabbe-Mann et al. 2018; Maver et al. 2020), PVA (El-Newehy et al. 2016;

Table 4 Summary of electrospinning of common cellulose derivatives (not including pure cellulose and cellulose acetate) fibers containing briefly relevant information and main goal of each study

Cellulose derivative	Electrospinning technique	Solvent system	Relevant information	Main goal and/or application	References
CAB	Traditional	Dichloromethane and acetone	Porous surface fibers (pores of 65–80 nm)	Application as a motor oil adsorbent with reusability potential	Tanvir et al. (2020)
	Traditional	Acetone	Sterilization with UV light or 70% ethanol	Human neuroblastoma cultivation on fibrous scaffold	Elashnikov et al. (2019)
	Traditional	2-Methoxyethanol	Growth of ZnO nanocrystals on CAB fibers	Photocatalytic activity by degradation of Rhodamine B	Pascariu et al. (2018)
	Traditional	DMAc and acetone	Lipase immobilization on modified CAB nanofiber membrane	Applications on lipase-catalyzed reactions	Chen et al. (2014)
	Traditional (Disc nozzle)	DMF and acetone	Comparison between disc-spinning and needle-spinning process	Cellular growth of fibroblast and Schwann cells on fibrous scaffold	Huang et al. (2013)
EC	Coaxial	Ethanol	Organic solvents (methanol, ethanol and DMF) were used as sheath fluids	Optimization of fabrication process and of EC fibers	Yu et al. (2014)
	Coaxial	DMAc and ethanol	Core solution of EC and quercetin and shell solution of PVP and quercetin	Biphasic quercetin release from EC nanofibers	Li et al. (2014)
	Traditional	THF and DMAc	Electrospun EC fibers with porous surface	Investigation of the effect of THF/DMAc solvent system on EC fibers	Park et al. (2007)
	Traditional	THF and DMAc	EC fibers containing antibiotic agent (streptomycin sulfate salt)	Streptomycin sulfate-loaded EC fibers and its antibacterial activity on *S. aureus*	Park et al. (2015)
	Glass needle	Ethanol and water	Use of glass and steel needle to produce EC fibers	Relationship between needle material and electrospun EC fibers	Ahmad et al. (2013)
	Traditional	Ethanol and water	Study varying the viscosity from 613 to 1652.5 mPa s	Electro-spinnability of EC using safe solvents	Crabbe-Mann et al. (2018)
	Traditional (spinning mandrel)	Ethanol	Polyoxyethylene sorbitan monooleate (PSM) surfactant added into EC solution	EC/PSM fibers as adsorbent for removing lead (II) ion from aqueous solution	Tabatabaeefar et al. (2020)
CMC	Traditional	Water	CMC was combined with alginate and PEO containing lidocaine	Lidocaine release from CMC/PEO/alginate nanofiber mat controlled by crosslink degree	Baek et al. (2020)
	Traditional	Water	CMC blended with PVA containing magnetic nanoparticles	Synthesis of magnetic nanofibrous materials with soft ferro-magnetic response	Durán-Guerrero et al. (2018)
	Traditional	Water	Tri-component composite nanofibers containing CMC, PVA and PVP	Investigation of spinnability and processability of CMC/PVA/PVP fibers	Hashmi et al. (2020)
	Needless	Water	CMC/PEO blend fibers with medicinal plant extracts	Plant extract-CMC/PEO fiber and its antibacterial activity on *S. aureus*	Maver et al. (2020)

Table 4 continued

Cellulose derivative	Electrospinning technique	Solvent system	Relevant information	Main goal and/or application	References
HPC	Traditional	Ethanol	HPC fibers in nano and submicron scales	Nano and submicron semiconductor SnO_2 fibers deposited on MEMS device	Shukla et al. (2005)
	Traditional	Water	HPC was blended with PVA and PVP containing diclofenac sodium	Production of HPC/PVA and HPC/PVP fibers. In vitro release of diclofenac sodium	El-Newehy et al. (2018)
	Traditional	Water and acetic acid	HPC blended with chitosan and PEG containing graphene	Nanofibrous mat with potential application as wound dressing	Lin et al. (2020)
	Traditional	DMF	HPC blended with polyurethane (PU) containing donepezil hydrochloride	HPC/PU nanofiber mat as transdermal drug delivery system	Gencturk et al. (2017)

Durán-Guerrero et al. 2018; Hashmi et al. 2021), and PEG (Kurečič et al. 2019).

Electrospun CMC fibers have been applied to drug delivery because of their biodegradability and the formation of covalent bonds between their carboxyl groups and amine groups present in most drugs. Allafchian et al. (2020) achieved 10 h of flufemic acid release time when loaded in CMC/PVA nanofibers with 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide as a crosslinker. El-Newehy et al. (2016) obtained a more sustained release of diclofenac sodium when using CMC/PVA nanofibers, in vitro studies showed that approximately 90% of the drug is released in 24 h. Esmaeili and Haseli (2017) fabricated thermoplastic CMC/PEO NFs and obtained a sustained release (after an initial burst release of approximately 26%) of tetracycline hydrochloride during 72 h.

Recently, CMC has been combined with more than one polymer (Baek et al. 2020; Hashmi et al. 2020, 2021). Hashimi et al. (2021) produced electrospun fibers of CMC blended with PVA and PVP in different polymer proportions (w/w). The fibrous mat of tri-component blend presented hydrophobicity, differently from the original polymers that are hydrophilic due to the hydroxyl groups present in CMC, PVA, and PVP. The material was evaluated in food packaging of fruits and vegetables. In comparison to conventional plastic packages, the fibrous mat presented lower accumulation of moisture. Also, the good air permeability resulted in fruits and vegetables fresh for a longer time.

Hydroxypropyl cellulose and hydroxypropyl methylcellulose

Electrospinning of hydroxypropyl cellulose has been suitable due to its solubility in water and organic solvents (El-Newehy et al. 2018), including methanol, which is widely used in solvent electrospinning (Zhang et al. 2017). The pioneering study on the electro-spinnability of HPC effectively showed the obtention of its fibers (Shukla et al. 2005). HPC solution of 15 wt.% was fixed to two different solvents, anhydrous ethanol, and 2-propanol, which were fundamental for the homogeneity of the fibers formed. When using 2-propanol as a solvent, the formation of beads was eliminated, unlike when using ethanol.

HPC has also been applied in electrospinning along with other polymers, such as PVA and PVP, known for their electro-spinnability (El-Newehy et al. 2018). Aiming drug delivery applications, when incorporating a model drug, it was found that the diameter of the fibers remained similar, with no changes in their morphology, indicating that the drug was adequately embedded within the fibers. In addition, the presence of PVA and PVP demonstrated better tensile strength in the blend with HPC, with this characteristic increasing due to the increase in the percentages of PVA and PVP. Gencturk et al. (2017) produced polyurethane/HPC electrospun NFs that showed great potential in transdermal drug delivery systems. In vitro studies demonstrated that the material exhibited Korsmeyer-Peppas drug release kinetic mechanism,

which means that the release rate is controlled by diffusion. Also, the fibrous mat was well-tolerated by human skin.

Lin et al. (2020) also produced electrospun membranes for wound dressing applications from blends containing HPC, in this case using chitosan and polyethylene oxide, with embedded graphene to improve antibacterial properties. A non-toxic solvent system (aqueous solution of acetic acid) was used, and it was shown to be difficult to obtain the membranes until reaching the optimum process conditions. The obtained hydrophilic membranes presented a high capacity to prevent adhesion of bacteria and reduce bacterial growth, becoming suitable for the intended application.

Hydroxypropyl methylcellulose (HPMC) is a cellulose derivative which, similarly to EC, has been used as a food additive and as thickening agent, emulsifier and stabilizer in the food industry (Aydogdu et al. 2018). Its electrospun nanofibers can be used in order to improve drug dissolution. Balogh et al. (2016) evaluated alternating and direct current electrospinning of HPCM/PEO. Fibers produced with alternating current (AC) were several times thinner than the ones produced with conventionally used direct current, mainly because of the multiple times higher feeding rates that are possible with AC. The authors were able to produce amorphous fibers with large surface area so that the dissolution of drug spironolactone in water was significantly increased. Paaver et al. (2015) fabricated supersaturated controlled-release solid dispersion polymeric nanofiber matrices of HPMC for the amorphization of poorly-water soluble drug piroxicam. The NFs were able to maintain the drug in amorphous state during 3 months when stored in low temperature and relative humidity condition.

Conclusions and future prospects

Electrospun nanofibers have been the target of an increasing number of studies because of their numerical advantages and wide range of potential applications. Large surface-to-volume ratio, high porosity, ease of processing and innumerous possibilities of surface modification are some of the properties responsible for making NFs interesting materials for applications in biomedicine (drug delivery, tissue engineering, wound dressing), food packaging, environmental engineering (wastewater treatment), energy, etc. Cellulosic NFs are of even higher interest because of its abundance, biodegradability and biocompatibility.

However, several challenges have been presented and need to be taken into account in cellulosic electrospinning systems. Depending on the derivative chosen, poor solubility, low thermal stability, formation of beaded fibers, need of complex solvent systems and uneven fiber diameter distribution are some of the major limitations that one may face.

Knowledge about electrospinning of cellulose and its derivatives is still limited and, therefore, extensive research and technology development are fundamental in the near future. The combination of the amazing materials that electrospinning can produce with the marvelous properties of cellulose can bring symbolic technological advances and improve the quality of life worldwide.

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