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Chapter

Conversion of Cellulose into Cellulose Acetate and Evaluation of Biomedical and Wastewater Cleaner Application of Electrospun Cellulose Acetate Nanofibers

Rebika Baruah and Archana Moni Das

Abstract

Conversion of biomass into useful organic chemicals has great demand in the twenty-first century. Cellulose is the most natural abundant biopolymer. Production of cellulose acetate (CA) from cellulose has garnered tremendous global attention because of their myriad application. CA is produced in huge amounts globally. Annual production of the derivative crossed 1.5 billion pounds. CA has remarkable biomedical applications due to their capability of drug delivery and anti-inflammatory, anti-cancer, antioxidant, antimicrobial properties. This chapter highlighted the synthesis and chemical and physical properties of CA polymer and electrospun CA nanofibers (CA NF) and their tremendous applications in biomedical and wastewater treatment.

Keywords: cellulose, cellulose acetate, nanofibers, biomedical, wastewater treatment

1. Introduction

Electrospinning is one of the cost-effective procedures that were applied to synthesize multifunctional nanofiber from the origin of this century [1, 2]. Since the mid-1990s, researchers started to explore the big potential of nanofiber in the area of nanotechnology [3]. Due to the large surface area to volume ratio, about a thousand times higher than that of a human hair, nanofiber are tremendously applicable engineering material. Nanofibers (NFs) have diverse application in various fields such as catalysis, tissue scaffolds [4, 5], textiles, wastewater remediation, drug delivery, wound healing, food packaging, and electronics [6]. NFs are synthesized via electrospinning is grounded on the uniaxial unfolding of a visco-elastic mixture. The precepts of electrospinning and different factors like time, temperature, etc., affects the shape of the nanofiber assembly. Different techniques of electrospinning such as dry-spinning and melt-spinning build develop electrostatic forces to stretch the solution as it solidifies. In the electrospinning the drawing of the solution to make the fiber will proceed as long as there is enough solution to feed the electrospinning jet. This is the

advantages of electrospinning over the formal fiber spinning techniques. Synthesis of fiber is continuous without any interruption to the electrospinning jet [7].

Researchers of this era search out for economically feasible and commercially successful pathways to convert biomass into useful organic chemicals [8]. Cellulose is the most abundant natural biopolymer in the world. Cellulose is converted into different types of valuable cellulosic derivatives. Among derivatives, cellulose acetate gathers the attention of researchers due to its cost-effective, sustainable and eco-friendly nature [9]. According to the survey in 2005, the annual production of cellulose acetate is 1.5 billion pounds due its versatile applications in different fields [10]. Traditionally, cellulose is reacted with acetic acid and acetic anhydride in presence of sulfuric acid and synthesized cellulose acetate. Now days, some innovative and effective methods were developed to synthesize cellulose acetate, e.g., use of ionic liquids, green catalyst like nanoparticles, super acids like $\text{SO}_4^{2-}/\text{ZrO}_2$ in a solvent free ball milling reactor, etc. [11].

Cellulose acetate is applied as coating materials in textile industry, waste water treatment for filtration, pharmaceutical industry, stabilization of nanoparticles, paper industry, leather, dye, and food industries, and lamination, etc. [12]. Cellulose acetate/montmorillonite bionanocomposites were applied in the adsorption of organic dyes like Methylene blue, Methyl Orange, Orange G, Rhodamine B, Eosin Y, etc. They were degraded the harmful dyes by adsorption due to their large surface to volume ratio [13, 14]. Cellulose acetate based nanocomposites can be effectively used in the remediation of organic pollutants and save the world from the detrimental effects of water pollutants. They can successfully overcome the disadvantages of traditional method used in waste water treatment like ultra-filtration, electro-chemical adsorption, coagulation, photooxidation, ultrasound-assisted, flocculation-coagulation, etc. Bionanocomposites effectively remediates the pollutants through physical adsorption. Physical adsorption is an inexpensive, highly efficient method to remove harmful entities in waste water and make the water safe for drinking [15].

Cellulose acetate (CA) based bionanocomposites have profound biomedical applications, like coating of medicine, membrane, drug delivery, blood purification in chronic renal dysfunction, hemodialysis, etc. Their biodegradability, renewability, non-corrosivity, non-toxicity, sustainability, and biocompatibility make them a suitable and innovative alternative in the place of multi-drug resistant medicines [16]. As dialyzers cellulose and its derivatives are 1st generation of polymers, they play an important role in the removal of blood toxins by diffusion and convection. This chapter is designed to explore the wastewater remediation capability and biomedical applicability of cellulose acetate based bionanocomposites.

2. Cellulose acetate (CA) electrospun nanofibers

Cellulose acetate can be synthesized from cellulose via a green and economic method and it has tremendous application in food technology for packaging and biomedical fields as CA is an easily obtainable substantial derived from cellulose at low cost and it is for regenerative medicine, tissue engineering, bone regeneration, and stem cell research [17, 18]. CA is more easily electrospun as compared to cellulose. Conversion of nanocellulose from cellulose acetate is easier than that from cellulose. Electrospun cellulose acetate nanofiber (CA NF) is completely novel candidate in the field of biomedical and food industry. Global industrial demand of Ca fiber is about 1.05 M metric tons by 2017 [19, 20]. Environment friendly CA NF has great

demand in the industrial world due to their biodegradability, higher tensile strength, enough angularity, and more modulus [21–23]. Electrospun cellulose acetate nanofibers have commendable applications in different fields and introduction of new functional groups into the CA NF increases the utilization of this NF to greater extent [24, 25]. CA NF possesses many beneficial characteristics like biocompatible, biodegradable, water resistance, etc. Therefore CANF can be safely used in the preparation of bandage loaded with nanocomposites for biomedical uses [26].

2.1 Choice of solvent for cellulose acetate e-spun nanofabrication

Choice of solvent is an important step to synthesize electrospun CA NF. It affects the morphology, size, and shape of the NF [27]. Researchers used different types of solvents like chloroform, N, N-dimethylformamide, dichloromethane, formic acid, and methanol (MeOH) and pyridine and they also used mixture of solvents like acetone, dimethyl acetamide (DMAc), chloroform-MeOH, and DCM-MeOH [28]. Formhals synthesized electrospun CA NF in acetone as the solvent [29]. Viscosity, surface tension, and conductivity of solvent system play a vital role in the development of smooth NF. Acetone, DMF, trifluoro-ethanol and mixture of acetone and water are the best solvent systems for preparation of e-spinning CA NF [30, 31]. CA NF with poly(ethyleneglycol), poly(ethylene oxide) and hydroxyapatite becomes an effective bionanocomposites in the mentioned solvent systems [32]. Haas et al. reported the effect of solvent system in the restraining of the coordination between CA NF [33]. Binary solvent system of less volatile solvents produced ribbon like structure with good packaging property [34]. Humidity and temperature of solvents have adverse effects on the shape and size of CA NF [34]. Along with solvent system, the field strength, distance between tip to collector, feed rate of solution affects the properties of CA NF like structure, diameter of fiber, etc. [35]. By using Box-Behnken pattern method the affected property of the CA NF can be modified [36]. Researchers concern for the preparation of e-spinning CA NF by blending with natural polymer and metal nanoparticles like AgNPs, etc. [37].

3. Properties of cellulose acetate

3.1 Thermal properties of cellulose acetate

Thermal properties like glass transition temperature (T_g), melting point (T_m) and thermal decomposition (T_d) of CA is dependent on degree of substitution (DS) and average molecular weight [38]. Plasticizers are used with CA to prepare thermoplastic to overcome the disadvantages of decomposition [39, 40]. Liu et al. studied the effects of degree of substitution on the thermal properties of CA NF in the aqueous hydrolysis of CA. He stated that DS have negative effects on T_g . When DS decreased from 2.45 to 1.77 T_g increased from 198°C to 205°C [41]. Elevation of T_g was due to the increasing number of free hydroxyl groups at low DS. On mixing of CA with copolymers nanofiber also increases the T_g [42]. Intermolecular hydrogen bonds and dipole-dipole interactions between the monomers of CA polymer have significant impact on the properties of CA NF. Number of both of hydrogen and acetyl groups affects the density and strength of hydrogen bond and dipole-dipole interactions of CA [41]. FTIR spectra of CA confirmed the presence of hydroxyl (OH) stretching band at 3322 cm^{-1} and the carbonyl (CO) stretching band at 1751 cm^{-1} . On increasing degree

of acetylation, the reduction of hydroxyl groups and increasing of carbonyl group were achieved [41]. Electrospun CA NF that possessed DS value of 2.4 or 2.5 can be effectively utilized in the biomedical application [41]. Inner structure of CA NF is more stable than original CA. CA NF possessed higher thermal stability and broad range of thermal degradation as compared to CA [42]. Presence of acetyl groups in presence of hydroxyl groups in the CA NF is the reason for the higher thermal stability of NF [42].

3.2 Mechanical properties of electrospun cellulose acetate

Mechanical properties of CANF are analyzed by measuring the tensile strength, young modulus, and elasticity of fiber. Mechanical properties are dependent on the morphological structure and packing of nano fiber. Mechanical properties help to determine the suitability of application of CA NF in biomedical fields [43, 44]. Golizadeh et al. reported the tensile strength and Young's modulus of CA NF approximately 2.5 MPa and nearly 125 MPa [44]. In their study the tensile strength and Young's modulus were increased by chemical and heating treatment of Ca NF. Bonding of CANF at crossover points occurred on heating due to the cross linking of nanofibers and it increased the tensile strength of CANF [44].

Van der Waals interaction and fiber-fiber fusion in CANF also affect the mechanical properties of NF [45]. Increase in the cross-linking between fiber improves the mechanical properties of fiber like, shear resistance, tensile strength, and compressive [46]. Mixing of another polymer with CANF also improves the mechanical strength of nano fiber. The mechanical properties of CA could be enhanced by mixing with another polymer. E. g., Poly hydroxybutyrate (PHB)/CANF scaffold possessed high tensile strength and Young's modulus at 7.86 ± 0.67 MPa and 854.2 ± 187.6 MPa whereas the pure CANF has low tensile strength and Young's modulus was 1.56 ± 0.19 MPa and 41.6 ± 12.3 MPa, respectively. Improvement of mechanical properties of CANF was due to the intermolecular hydrogen bond in CANF and intermolecular interactions of C=O groups in PHB [46]. Increase in the bonding of CANF and adjustment in the degree of molecular orientation of CA also improves the mechanical properties of nano fiber [46].

3.3 Biodegradability of cellulose acetate

Cellulose acetate nano fiber is a biodegradable polymer. Microorganisms degrade CANF in both aerobic and anaerobic condition by using cellulase and acetyl esterase enzymes [20]. Degree of polymerization, degree of substitution, and location affect the rate of degradation of CANF [21]. Acetyl-esterase is an essential enzyme for the initial step of biodegradation of CANF and cellulase is important in second step in which hydrolysis of the backbone of cellulose take place [22]. In the biodegradation, acetyl group from the C2- and C3-positions of the CA convert to acetic acid in first step and in hydrolysis free glycosyl residues release into the metabolic cycle [21]. Puls et al. reported the biodegradation of CANF with the help of cellobiohydrolase I (CBHI) and *cellobiohydrolase* II (CBHII) as cellulolytic enzymes. They catalyze the splitting of β -1,4-glycosidic bonds at the reducing and non-reducing end side of the cellulose chain.

CANF from bacterial cellulose loaded with cellulose is degraded by the *in vitro* degradation. *In vitro* biodegradation converts CANF to simulated body fluid within 6 months. Modulation of the content of cellulose modifies the rate of degradation [44].

Although many studies about the biodegradation CANF are done in microorganisms but biodegradation of CANF has not been studied yet. Therefore insertion of CANF loaded composites in to the human body is a challengeable research till now.

3.4 Biocompatibility of cellulose acetate

Biocompatibility is defined as the behavior of implanted biomaterials in living tissues without causing any local or systemic adverse effect in their surroundings. Biomaterials also possess an effective host response in a specific application [46].

Biocompatibility test is carried out under the guidance of the International Organization for Standardization (ISO) 10993 series. The aim of this test to protects the humans from future biological risks of medical devices or materials [47]. Positive results of this test provide the final approval of implantation of biomaterials in humans. Biocompatibility tests include cytotoxicity, sensitization, irritation, systemic toxicity, genotoxicity, and hemocompatibility tests [47]. CANF possessed excellent biocompatibility [48]. Liang et al. reported the biocompatibility of poly (ethylene terephthalate) (CDA-g-PET) grafted CANF and showed that the viability of fibroblast on CDA-g-PET grafted CANF significant increase with incubation time.

Biocompatibility of other polymers can be improved by addition of CA to them. E. g., electrospun CANF grafted PHB showed better biocompatibility than pure HPOB film [48]. Biocompatibility of CANF/carboxymethylcellulose (CMC) scaffolds showed better results than that of pure CMC scaffolds [48].

4. Application of cellulose acetate nanofiber

4.1 Waste water treatment

More than one billion people in the world are suffered from lack of safe drinking water [49]. Remediation of waste water is a great challenge for researchers till now [49]. Many industries like textiles, dye, pharmaceutical, etc. discharged their waste water to water bodies and cause water pollution. Water pollutants like, harmful dyes, heavy metals such as, Fe, etc., and pathogens have detrimental effects on the aquatic lives and terrestrials organisms [50]. Traditional methods like chemical precipitation, membrane separation, electrocoagulation, anodic oxidation, TiO₂ photocatalysis, biodegradation, adsorption, and ion exchange are applied in the purification of waste water. Adsorption is the effective one that overcomes the all the disadvantages belongs to other methods and it possesses easily handling, reversibility, recyclability and cost-effective nature [51].

Utilization of bio-based products in the adsorption of organic pollutant in the remediation of waste water is a fruitful idea due to their natural availability, biodegradability, sustainable, and biocompatibility [52]. Cellulose is the most preferred candidate in this category but the production of submicron cellulose fibers is difficult due to the insolubility of cellulose in conventional solvents [53]. To overcome these obstacles cellulose derivatives are synthesized to access their applicability in different fields. Cellulose acetate is the most preferable one due to its environment friendly and economic nature. CA possesses versatile application in many industries like textiles, cigarette filters, surface coating, inks, motion picture film, microfilm, and audio tape as a carrier [54]. CA is soluble in non-polar solvents and therefore it can be easily electrospun to form CANF membrane with high mechanical strength [55].

CANF with ribbon-like porous structure successfully prepared to adsorb Cu^{2+} ions were studied. CANF could remove the Cu^{2+} ions up to 88.6% [55].

Fabrication of metal nanoparticles and another biopolymer on the CANF as inorganic filler overcome the drawback of lack of active site on the Ca backbone [56]. CA acts as soft matrix in the accommodation of inorganic fillers on it to functionalize it. E.g., chitosan/CANF composites effectively removed copper from waste water [57]. CANF/HAp bionanocomposites successfully removed harmful bisphenol [58]. These bionanocomposites have size of 92 nm and a new chromatographic method was developed for the quantification of bisphenol A in samples of baby food [58].

4.2 Biomedical application

Incorporation of inorganic nanofillers with CANF makes the bionanocomposites tremendous applicant in the field of biomedical. Due the biodegradability, renewability, non-corrosiveness, non-toxicity and biocompatibility nature cellulose acetate based bionanocomposites have tremendous applications in medicinal fields, like coating of medicine, drug delivery, tissue engineering, bandage preparation, etc. [59].

4.2.1 Antimicrobial activity

Infectious diseases are become threatened to various industries like textile, food, health care and packaging industry. Due to the uses of limitless antibiotics without proper knowledge, microorganism become multi-drug resistant and cause persistent infections. Developing a sterile and cost-effective medicine is an urgent need for twenty-first century that can control the infection caused by multi-drug resistant microbes. This kind of clinically approved medicine will decrease the infection of microorganism in medical instruments, filters, implants, tissue scaffolds, wound dressing materials, etc. [59]. Inorganic antimicrobial nanoparticles like Ag, Cu, and ZnO, and organic materials like N-halamine and chlorhexidine can be incorporated to CANF and increased the antimicrobial property of CANF [59]. Types of incorporated polymers, shape, size, dispersion, and load ratio of nanoparticles adversely affects the antimicrobial activities of CANF based bionanocomposites. Therefore, appropriate inorganic fillers should be incorporated to CANF which have positive effects on the biomedical activity of CANF materials [60].

4.2.2 Packing biomedical and pharmaceutical products

Contamination of food and pharmaceuticals by surroundings is very common and causes undesirable side effects. Extra care is required in the packaging of these products to safe them microbial contamination. de Moraes et al. developed grapheme oxide (GO)/CA packaging film for pharmaceutical, biomedical, and food products. GO/CA films possessed UV light absorption potential, visible light transferable and non-catalytic property towards polymer degradation [61]. CANF based packaging materials provide homogenous and smooth film [61].

4.2.3 Controlled drug release

Drug delivery systems are developed in such way that they can deliver drug at a specific target, i.e., either tissues or intracellular sites within short period of

time. Delivering drug to the site action has advantages of control delivery of drugs, requirement of low concentration of drugs, reduction of dose of drugs, effective accumulation of drug to the location of action and overcome the disadvantages of side effects, instance decomposition of drugs, etc. Different materials are employed in the drug delivery like clay, biopolymer, LDH, etc. Cellulose based drug delivery systems are very effective due to their natural abundance, renewability, biocompatibility, biodegradability, and unique biological activities. Madaeni et al. reported the drug delivery ability of polyethylene glycol (PEG)/CANF bionanocomposites. The composites showed fast rate of drug deliver with zero order kinetics. Hydrophilicity of the composite membrane minimized the agglomeration of the nanocomposites and addition of PEG improved the structure and porosity of composite membrane [62].

4.2.3.1 Non-steroidal anti-inflammatory and antioxidant drug

Inflammation and swelling symptoms are decreased by the use of non-steroidal anti-inflammatory medicines (NSAIDs). NSAIDs have some side effects like gastrointestinal problems due to their poor solubility in the water. To overcome this problem local or targeted delivery of NSAIDs are developed by using electrospun biopolymer coated drug delivery systems [63]. CANF can effectively deliver NSAIDs like naproxen, indomethacin, ibuprofen, and sulindac, etc. CANF transdermally deliver drugs like naproxen with sustainable release [64].

4.2.3.2 Anticancer and antioxidant agents

Electrospun CANF/polyethylene oxide (PEO) effectively delivered potential anticancer drugs like cisplatin. Incorporation of cisplatin in CANF/PEO formed the drug formulation with the repeating dumbbell-shaped structures [65]. Electrospun CANF is utilized in the local delivery of silymarin through the skin in various liver diseases. Silymarin was well incorporated in CANF and released gradually to the targeted site within 20 days [66].

4.2.3.3 Antimicrobial agents

Antimicrobial drugs are incorporated with electrospun CANF and utilized to control the growth of microbes [67]. CANF/poly (vinylpyrrolidone) (PVP) was incorporated with amoxicillin and formulates as (CA/PVP-Amoxi/CA) [68]. Tetracycline was also incorporated with CANF and applied in periodontal reconstitution as an adjuvant.

CANF membrane acts as a prospective degradable drug release device in the drug release study [69]. CANF was incorporated with Bis *N*-Chloro-2,2,6,6-tetramethyl-4-piperidyl sebacate (CI-BTMP) and applied in the controlling of bacterial infection [70]. Ag/TiO₂/CANF was applied as an antibacterial for future bone tissue regeneration. Ag/TiO₂/CANF possessed potential antibacterial activity against gram-negative bacteria (*E. coli* and *P. aeruginosa*) and gram-positive bacteria (*S. aureus*). CANF composites showed significantly improved cell viability and the proliferation of fibroblasts [71, 72].

Khan et al. reported the delivery of silver sulfadiazine (SSD), a topical wound dressing antibiotic via Ag NPs/CANF bionanocomposites [73]. Ag NPs/CANF bionanocomposites possessed remarkable antibacterial properties against gram-negative *E. coli*.

4.2.3.4 Wound dressing

Development and designing of wound dressing materials are vital keeping in mind wound pathogenesis. Wound dressing materials should be non-toxic, antimicrobial, maintain moisture, non-adherent, wound care, prevent bacterial infection, and endorse wound healing. Recently, different biological materials have been used for wound dressing. Among them, polymers that considered as soft biomaterials; such as cellulose derivatives and their nanocomposites have been widely used as wound dressing materials [74].

4.2.3.5 Bone repairing and tissue engineering

In the field of bone repairing and tissue engineering, it is most viable to choose a biocompatible and degradable material that lack of cytotoxic effects and could be replaced by the regenerated tissue biomaterial. In addition, considerable attention should be given to provide a sufficient environment for tissue to promote cell integration, differentiation, and proliferation. All these could be achieved by implantation of the appropriate three-dimensional porous scaffolds seeded by the bioactive molecules [75]. Accordingly, due to the fact that cellulose acetate was not only naturally abundant but also has good hydrolytic stability, good mechanical properties, excellent compatibility, low toxicity and relatively low cost [76], many attempts have been conducted to investigate the possibility of fabricating CANC in tissue and bone engineering [77].

5. Conclusion and future prospective

Cellulose acetate nanofiber can be successfully synthesized via electro spinning. CANF possesses remarkable waste water remediation and biomedical properties. CANF shows potential drug delivery application as locally also transdermally. Biodegradability, biocompatibility, and sustainability of electrospun CANF will be cost effective green material in environmental remediation and biomedical application. Due to the lack of cellulose enzyme in human body, biodegradability of CANF is a challengeable topic till now. Incorporation of drugs in e-spinning CANF with degree of substitution of 2.5° is an effective localized drug delivery system without the risk of second surgery. Excellent solubility, hydrophobic nature, mechanical strength, and thermal property are due to the presence of acetyl groups present in the backbone chain of CA and cross linking between the fibers. CA with 2.5° of substitution has commonly been used in the drug delivery systems due to inherited properties. Therefore, CANF electrospun bionanocomposites will be an eco-friendly and cost effective materials in waste water treatment and localized drug delivery systems. These purposeful properties of CANF based bionanocomposites make them strong materials for industrial applications to provide human clean and safe drinking water and effective drug delivery systems in wound dressing, cancer treatment, liver disease, etc., without the risk of secondary infection [78–80].

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Conflict of interest

The authors declare no conflict of interest.

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