A Review on Cellulose-based Materials for Biomedicine

Hani Nasser Abdelhamid[1,2]*, Aji P. Mathew[1]
[1]Department of Materials and Environmental Chemistry, Stockholm University, SE-10691 Stockholm, Sweden
[2]Advanced Multifunctional Materials Laboratory, Department of Chemistry, Faculty of Science, Assiut University, Assiut 71515, Egypt
*Corresponding to Abdelhamid (hany.abdelhamid@aun.edu.eg); (hany.nasserabdelhamid@mmk.su.se)

Abstract

There are various biomaterials in nature, but none fulfills all the requirements. Cellulose, eco-friendly material-based biopolymers, have been advanced biomedicine to satisfy most market demand and circumvent many ecological concerns. This review aims to present an overview of the state of the art in cellulose's knowledge and technical biomedical applications. It included an extensive bibliography of recent research findings for fundamental and applied investigations. The chemical structure of cellulose allows modifications and simple conjugation with several materials, including nanoparticles, without tedious efforts. Cellulose-based materials were used for biomedicine applications such as antibacterial agents, antifouling, wound healing, drug delivery, tissue engineering, and bone regeneration. They advanced the applications to be cheap, biocompatible, biodegradable, easy for shaping and processing into different forms, with suitable chemical, mechanical and physical properties.

Keywords: Cellulose; Biomedical; Wound healing; Drug delivery; Antibacterials; Tissue engineering.

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1. Introduction

Cellulose is a natural linear-structural biopolymer of anhydroglucose monomer linked via β-(1–4) bond ($(C_6H_{10}O_5)_n$; n is the degree of polymerization; n equals 10000-5000 depending on the source used for cellulose extraction)[1–7]. It can be produced from plants, seaweeds, sugarcane bagasse, tunicate, marine algae, and bacteria [8–11]. The annual production of cellulose is more than several hundred billion tons. The market demand is continuously increasing over the years. Cellulose exhibits good mechanical, physical, and chemical properties such as high stability under acidic conditions, chirality, high tensile strength, good elastic modulus (130–150 GPa), low density or lightweight (density of 1.6 g/cm^3), high biodegradability, and abundant hydroxyl functional groups on their surfaces enabling chemical modification with high wettability. Thus, it was applied for several applications such as energy, environmental and health-based technologies [12–18].

Cellulose-based materials have advanced biomedical applications [19–34]. They were reported for antibacterial agents [35–40], wound dressing [41–47], drug delivery [18,48–51], tissue engineering [24,33,52], artificial blood vessels [53,54], and for the protection from UV radiation [55,56]. Cellulose can proceed into different forms such as hydrogels [57,58], aerogels [59], membrane [60], and three-dimensional (3D) scaffolds [61,62]. They exhibit several properties, making them interested in biomedical applications. They offered good binding properties [63]. They can be conjugated with organic [64] and inorganic-based materials. The surface chemistry of cellulose can be modified with several functional groups and compounds [65–68].

This review summarized the applications of cellulose-based materials for biomedicine. It covers the potential of cellulose and its composites with other materials for drug delivery, tissues engineering, wound healing, antifouling, and antimicrobial agents (**Figure 1**). Cellulose-based materials exhibited several advantages: high biocompatibility, transparency,

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production from renewable sources, cheap, high mechanical and physical strength, ease for shaping and processing, and require simple procedures for conjugation with other materials.

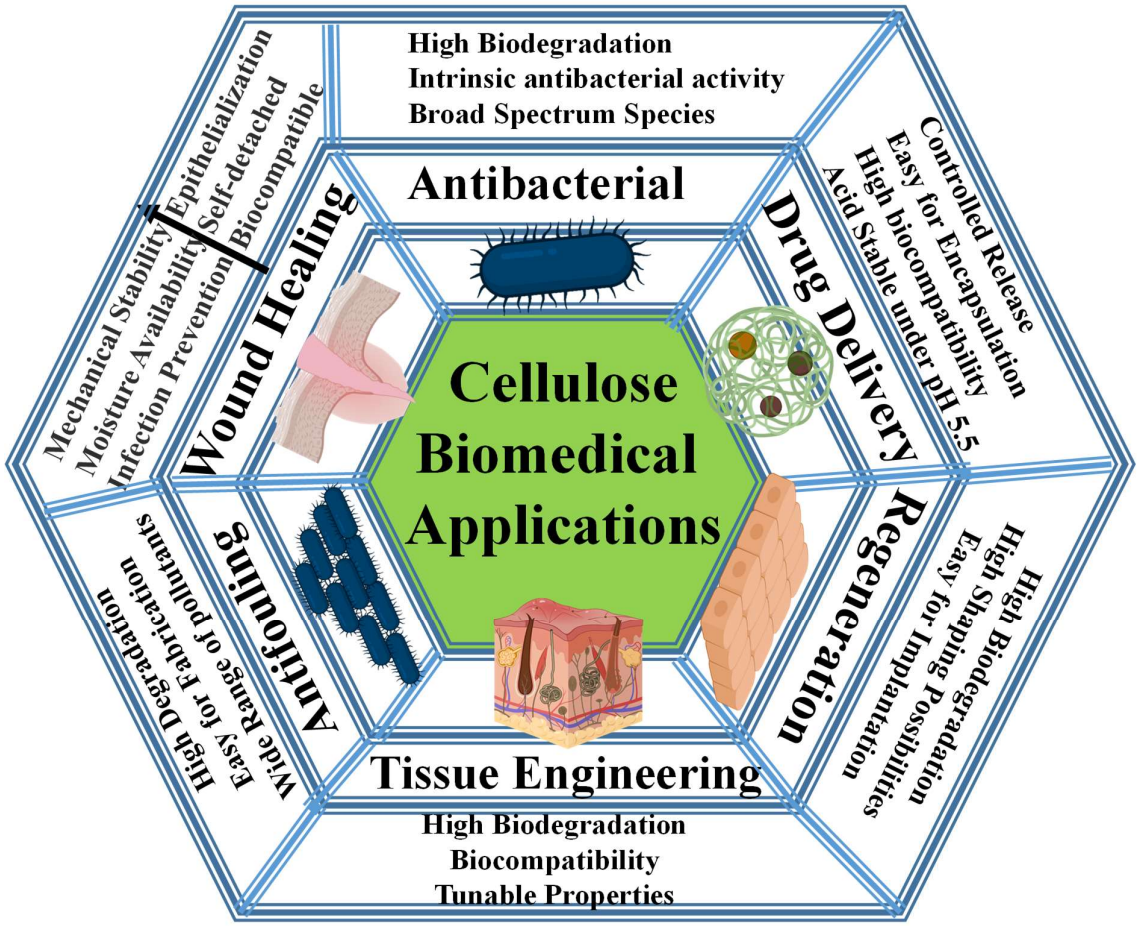

Figure 1 Overview for biomedicine applications covered in this review.

2. Cellulose Nanoparticles

Cellulose was marketed into several types, including microfibrillated cellulose (MFC), microcrystalline cellulose (MCC), nanofibrillated cellulose (NFC), cellulose nanocrystals (CNCs), microfibrils, and bacterial cellulose (BC). It can be obtained in micro and nano-scale regimes (**Figure 2**). MCC is microsize cellulose particles with a length of up to 1 µm and width of higher than 1 µm. MFCs are commonly produced via the mechanical or chemical treatment of wood. Microfibrils are individual fibers with a length of >10 µm and width of 2–20 µm. Cellulose in nanoscale is called cellulose nanomaterials or nanocellulose

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(**Figure 2**). CNCs, 2,2,6,6-tetramethyl-piperidine-1-oxyl (TEMPO)-mediated oxidized cellulose nanofibers (TOCNF), and BC are the common cellulose nanoparticles. CNCs are common products of the acid hydrolysis of cellulose-based materials [69]. The size distribution of CNCs is in the range of 50–500 nm and 2–20 nm for length and width, respectively. BCs are produced via bacteria cells. They exhibit high crystallinity and good elastic and mechanical properties. The length of BC tends to be 200–3000 nm, and the width can be in the range of 10-75 nm (**Figure 2**). The extraction of cellulose at the nanoscale removes the defects associated with the hierarchical structure and offers new properties such as a large surface area.

Cellulose nanomaterials exhibit a degree of crystallinity of 50–90%. The common functional group of cellulose is hydroxyl groups. NFC can be further oxidized to carboxylic functional groups via TEMPO-mediated oxidation offering TOCNF. The cellulose surface can be modified using several methods, including adsorption or chemical modification via covalent bond formation [5]. The surface charge of cellulose is an essential parameter for the material's characterization. The cellulose colloids' high charge ensures high stability and prevents nanoparticles' aggregation [70].

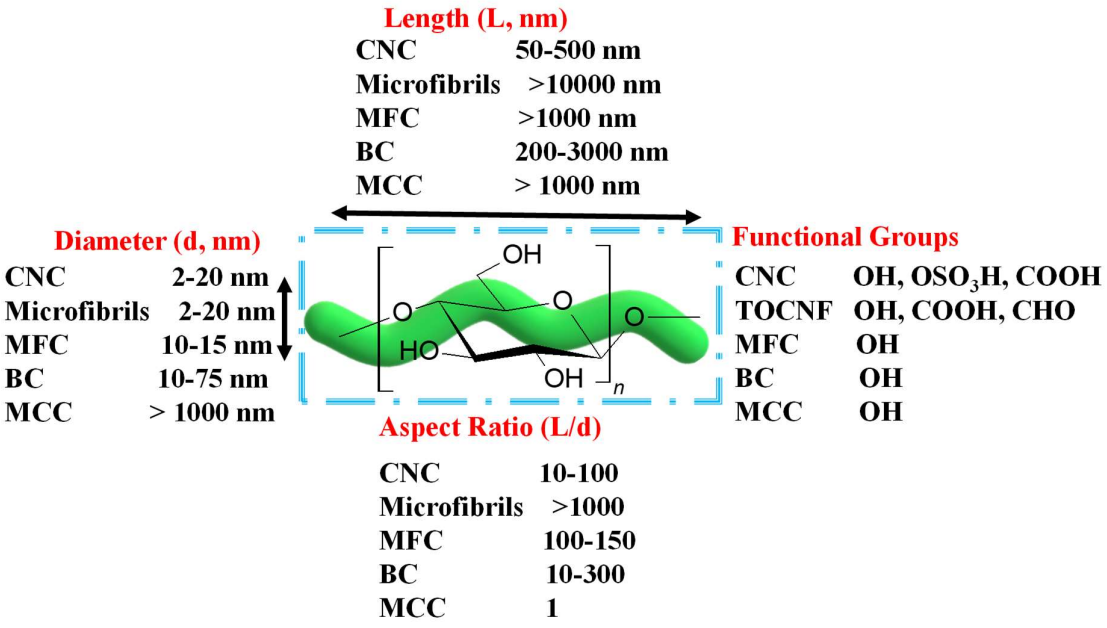

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Figure 2 Summary for cellulose nanoparticles with sizes and functional groups.

3. Applications of Cellulose Nanoparticles as Antibacterial Agents

Cellulose exhibits no intrinsic biocidal activity compared to other natural biopolymers such as cationic chitosan [71]. However, it can be used as an antibacterial agent via several methods, including surface modification and conjugation with antibacterial agents, including organic and inorganic materials (**Figure 3**) [37,72]. The following section summarized the applications of cellulose-based materials as antibacterial agents.

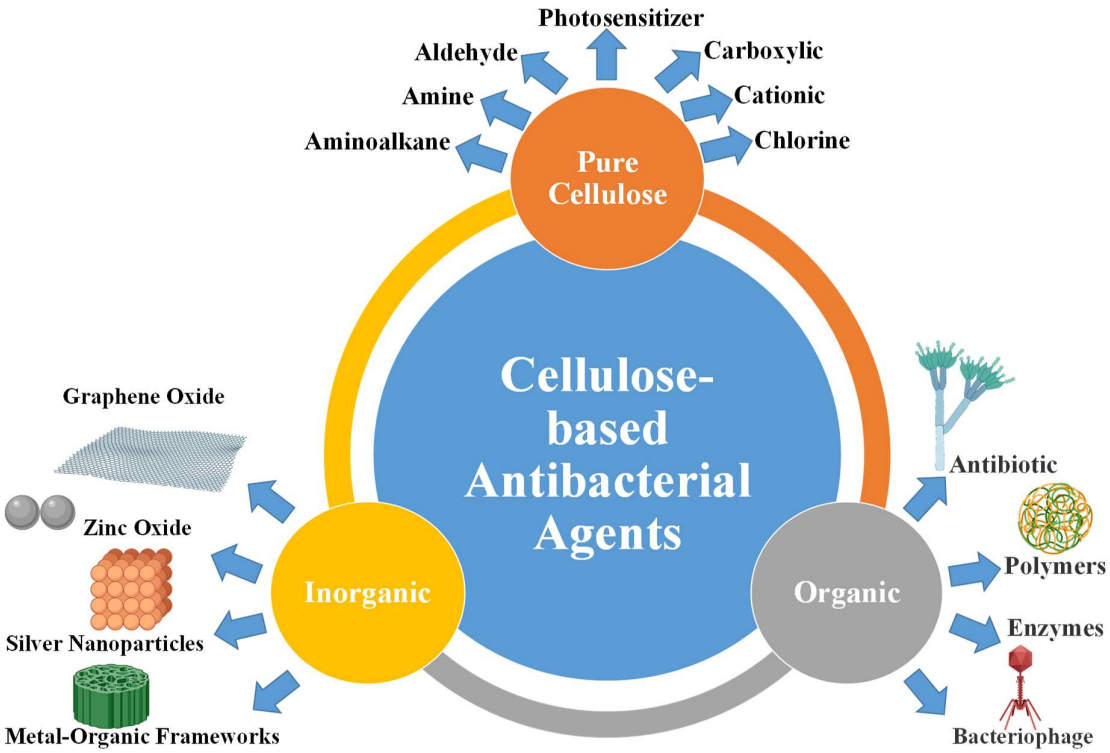

Figure 3 Cellulose-based materials as antibacterial agents.

3.1. Pure Cellulose with Modification for Antibacterial Agents

The antibacterial properties of pure cellulose can be improved via surface modification with several functional groups such as carboxylic groups, aldehyde, amine, alkylamine, and

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quaternary ammonium groups (**Figure 3**). The functional groups of cellulose can be modified with organic molecules that exhibit photosensitization properties. A summary of some of the antibacterial agents using cellulose-based materials is tabulated in **Table 1**.

Antibacterial activity of 2,3-dialdehyde nanofibrillated cellulose (DANFC) was investigated against *Staphylococcus aureus* (*S. aureus*) and methicillin-resistant *Staphylococcus aureus* (MRSA) [73]. Dialdehyde formation was performed via the oxidation cleavage of C_2 and C_3 bonds in the D-glucose monomer of cellulose using a chemical reagent such as sodium periodate ($NaIO_4$). The antimicrobial activity of DANFC is enhanced by increasing the time of oxidation. The antibacterial effect of DACNF is due to the aldehyde groups that cause a drop in the pH value (5.7–6.2) [73]. The antibacterial activity for dialdehyde microcrystalline cellulose (DAMC) was also reported [74]. DAMC with aldehyde contents of 5.14 mmol/g showed the most potent antibacterial activity against *S. aureus*, *Bacillus subtilis* (*B. subtilis*), *E. coli*, and *Salmonella typhimurium* (*S. Typhimurium*). It exhibited minimum inhibitory concentration (MIC) values of 15, 15, 15, and 30 mg/mL for *S. aureus*, *B. subtilis*, *E. coli*, and *S. Typhimurium*, respectively [74]. Cellulose was extracted from ginger residual, denoted as GNFs (ginger nanofibers), via acid hydrolysis and high-pressure homogenization [75]. GNFs were tested for antibacterial activity [75]. The MIC values of GNF was 14±2, 13±1, 18±0, and 31±0 µg/mL for *B. cereus, E. coli, S. aureus, and S. Typhimurium,* respectively [75].

Cellulose with carboxylic groups shows high antibacterial activity (**Table 1**). A gel of TOCNF (0.2-0.8 wt.% in water) inhibited the growth of a wound with the infection of pathogen *P. aeruginosa* [76]. The physical, chemical, antibacterial activity of carboxylate CNF can be modified via treatment such as autoclaves [77]. The autoclave treatment at 121 °C for 20 min reduced the gels' viscosity, increased the ultraviolet-visible absorbance maxima at 250 nm, and increased the aldehyde content. Autoclaved carboxylate CNF showed high antibacterial activity with minimal toxicity toward L929 mouse fibroblasts and reconstructed human

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epidermis (RhE)[77]. The antibacterial activity of carboxylate CNF with different oxidation levels was evaluated against *P.aeruginosa* and *S.aureus* [45,77]. Oxygenated CNF exhibited higher antibacterial activity than non-oxygenated CNF dispersion [45,77].

The antibacterial of pure cellulose nanoparticles could be due to several mechanisms, including; decrease the mobility of bacteria cells [76], surrounding and entrapping the bacteria via the formation of a network [45], and reduced pH value due to the increase of aldehydes groups in CNFs [73,77]. However, further investigations should be carried out to understand the key parameters affecting the antibacterial activity of pure cellulose nanomaterials.

Pure cellulose with suitable functional groups offers good antibacterial activity. However, it is crucial to consider the presence of foreign species such as endotoxins or lipopolysaccharides that can cause inflammation leading to antibacterial activity [78]. Modified TEMPO-mediated oxidation method using sodium hydroxide as a pre-treatment produced CNF showed an endotoxin level of 45 endotoxin units (EU) per g of cellulose [79]. This value may cause no toxicity at low concentrations. However, it can be critical for high concentration [80]. The presence of endotoxin should be considered during the evaluation of the antibacterial activity of cellulose-based materials.

3.2. Photoactive cellulose for antibacterial agents

Photo-based treatments using light radiation are promising for antibiotic resistance bacteria (**Table 1**) [81–84]. They required the presence of photosensitizer molecules that absorb the light radiation and convert it to thermal energy (photothermal therapy) or generation reactive species (i.e., photodynamic treatment) such as reactive oxygen species (ROS). Pure cellulose lacks photosensitizer's properties. Thus, it is usually modified with small molecules via covalent and non-covalent interactions to absorb light. Most of these photosensitizers are

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bacteria inactive materials. However, they are effective for bacteria inactivation using cheap light sources such as light-emitted diode (LED) lamps [85].

Photodynamical inactivation (PDI) against bacterial using CNC [86] and hairy aminated nanocrystalline cellulose (ANCC)-based material was reported [87]. Reactive oxygen species can be generated under light via the modification of cellulose with molecules such as anthraquinone vat dyes [88], 3,3′,4,4′-benzophenone tetracarboxylic acid [89], ketoprofen [90], hypocrellin [91], xanthene [68], BODIPY (**Dipy**rromethene**bo**ron difluoride) [81,92], chlorin-e6 [93], phthalocyanines [94,95], protoporphyrin-IX [96–98], and porphyrin [86,87,99–104]. CNC was chemically modified with cationic porphyrin, denoted as CNC-Por, via Cu(I)-catalyzed Huisgen-Meldal-Sharpless 1,3-dipolar cycloaddition (**Figure 4a**). The reaction occurs between the azide and alkyne groups on the cellulosic and porphyrinic molecules, respectively (**Figure 4a**). PDI of CNC-Por against *Mycobacterium smegmatis, S. aureus* and *E. coli* was investigated under white light radiation (400–700 nm, 60 mW/cm^2) [86]. CNC-Por exhibited high PDI against *M. smegmatis, and S. aureus* and insignificant activity against *E. coli* after illumination for 15 min. However after 60 min, PDI activity of the material against all bacteria was higher than 99% (99.9999% for *S. aureus*) [86]. ANCC was modified with a natural photosensitizer; Rose bengal (RB) via covalent bond (**Figure 4b**) [87]. RB-ANCC showed PDI over 80% for pathogens *Listeria monocytogenes* and *S. Typhimurium* under illumination using normal light irradiation. Interestedly, ANCC improved the PDI of free RB against *S. Typhimurium* [87].

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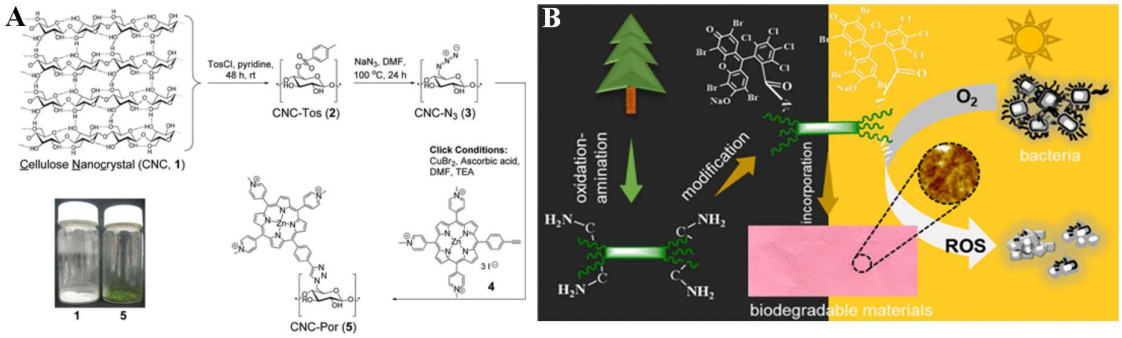

Figure 4 A) Synthesis of CNC-Porphyrine; 1) CNC preparation via acid hydrolysis, 2) surface tosylation of CNC, CNC-Tos, 3) synthesis of Azide Bearing CNC-N_3, 4) Click Reaction of CNC-N_3 with Porphyrin, Figure reprinted with permission from Ref. [86], Copyright belongs to ACS (2011); B) Chemical modification of ANCC with Rose Bengal as photosensitizers, Figure reprinted with permission from Ref. [87]. Copyright belongs to ACS (2021).

Por(+)-the paper was illuminated using visible light of wavelength and power of 400-700 nm and 65 ±5 mW/cm2, respectively, for 30 min. Cationic porphyrin (Por(+)) conjugated cellulose was performed as paper for scalable antimicrobial treatment using PDI [81]. The antibacterial and antiviral efficacies were investigated against bacteria (S. aureus, vancomycin-resistant Enterococcus faecium (VER), Acinetobacter baumannii, P.aeruginosa, and Klebsiella pneumoniae) and viruses (dengue-1 virus, influenza A, and human adenovirus-5). The inactivation efficiencies for all investigated species, e.g., bacteria and virus, were higher than 99.9% [81].

PDI exhibit several advantages, such as high antibacterial efficiency up to 99.999% (**Table 1**). The method can be applied for the treatment of antibiotic resistance bacteria. Photosensitizer-conjugated cellulose fibers can be used to inactivate viruses such as dengue-1 virus, influenza A, and human adenovirus-5 with efficiencies of 99.995%, 99.5% 99%, respectively [81]. Cellulose chemistry offers the fabrication of the materials as paper [81], fibers [100], or textiles [105] that enable scalable and straightforward uses for the antibacterial treatment. It provides immediate covalent modification using advanced methods such as photo-strain-triggered click ligation [106]. It may open a new venue for photoactive textiles [107].

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3.3. Cationic Cellulose for Antibacterial Agents

Similar to chitosan (CTS), cationic cellulose exhibits intrinsic antibacterial activity. The mechanism of the antibacterial action of cationic biopolymers depends on the high binding affinity between the positive charge of these polymers and the negative charge of the bacteria cells. The surface for bacteria cells, i.e., Gram-positive and Gram-negative, is negative due to hostile phosphate groups in peptidoglycan and phospholipids. Following this principle, creating a positive charge on cellulose enables high antibacterial activity (**Table 1**). Cationic CNCs can also be used as immune-modulators [108].

Cationic cellulose can be achieved via the modification with quaternary ammonium compounds such as poly(isopropanol dimethylammonium) chloride (PIDMAC)[109], quaternized poly(2-(dimethylamino ethyl) methacrylate) (PDMAEMA)[110], cetyltrimethylammonium bromide (CTAB) [111], 3-chloro-2-hydroxypropyl-tri-methyl ammonium chloride 3-chloro-2-hydroxypropyl-trimethyl ammonium chloride (CHPTAC) [112], pyridinium/N-chloramine [113], benzalkonium chloride [114], and quinolinium silane salt [115]. Cellulose grafted with DMAEMA can be prepared via polymerization using a reversible addition-fragmentation chain transfer (RAFT) [110]. Cationic cellulose can also be prepared via direct covalent bonding of quaternary ammonium moieties without the need for a linker [116]. The silane group of 3-(trimethoxy silyl)-propyl-dimethyl octadecyl ammonium chloride reacted with the hydroxyl groups in cellulose, forming Si–O–Si bonds [116]. Cellulose-QA showed complete inactivation of *E. coli* and *P. aeruginosa* after one h and 10-fold inactivation of B. cereus [116].

Cellulose can be modified with quaternary ammonium and porphyrin moieties via esterification [98]. Protoporphyrin IX (PpIX) moiety offered white-light radiation photosensitization (**Figure 5**). While, quaternary ammonium moieties offer dual-functional;

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antibacterial agents and prevent aggregation of porphyrins that cause quenching of the generation of reactive oxygen species (ROS). Photodynamic treatment of bacteria using quaternary ammonium-porphyrin modified cellulose showed effective antibacterial activity against antibiotic-resistant *E. coli* and *S. aureus* strains (**Figure 5**). The antibacterial activity of the materials are due to the intrinsic bioactivity of quaternary ammonium moieties and the ROS generation [98]. The treatment using the material required relatively low concentrations of porphyrin and can be applied under a low dosage of white-light irradiation (2.4 J/cm) [98].

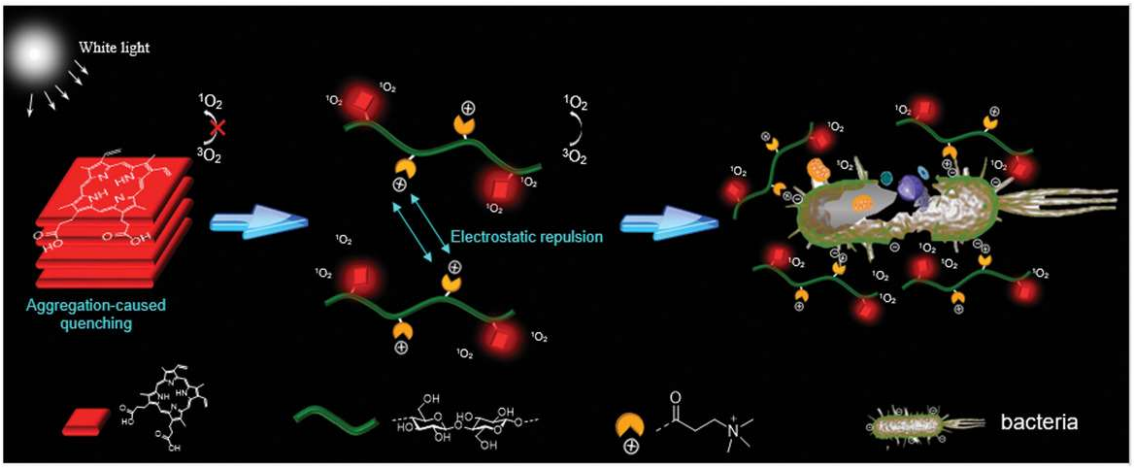

Figure 5 Antibacterial mechanism for porphyrin and quaternary ammonium-modified cellulose under light radiations. Figure reprinted with permission from Ref. [98]. Copyright belongs to John Wiley & Sons (2019).

The antibacterial activity of cationic cellulose can be due to several mechanisms such as destabilization of the bacterial intercellular membranes due to Ca^{2+} or Mg^{2+} ion exchange, membrane disruption due to the release of potassium ions, formation of ROS, increase of amine groups [117], or increase lipophilicity using amino-alkyl [118].

3.4. Organic-modified cellulose as antibacterial agents

The antibacterial activity of cellulose can be enhanced via chemical modification with organic bioactive molecules, including antibiotic, antimicrobial peptides, N-halamines, aminoalkyl groups, bacteriophage, and polymers (**Figure 3**).

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3.4.1. Antibiotic-modified Cellulose

Antibiotics are widely used for bacteria treatments. Cellulose was grafted with antibiotic including β-lactam antibiotic benzyl penicillin [119], ciprofloxacin [120], tetracycline hydrochloride [121], silver sulfadiazine (Ag SD) [122], 3-pentadactylphenol [123], Allicin [124], and amoxicillin [125]. Antibiotics such as penicillin can be covalently modified with cellulose via ester bond formation (**Figure 6**)[119]. The covalent modification of cellulose ensures high durable properties with good antibacterial activity. The materials can proceed into film simply using thermal treatment without destroying the formed bond (**Figure 5**).

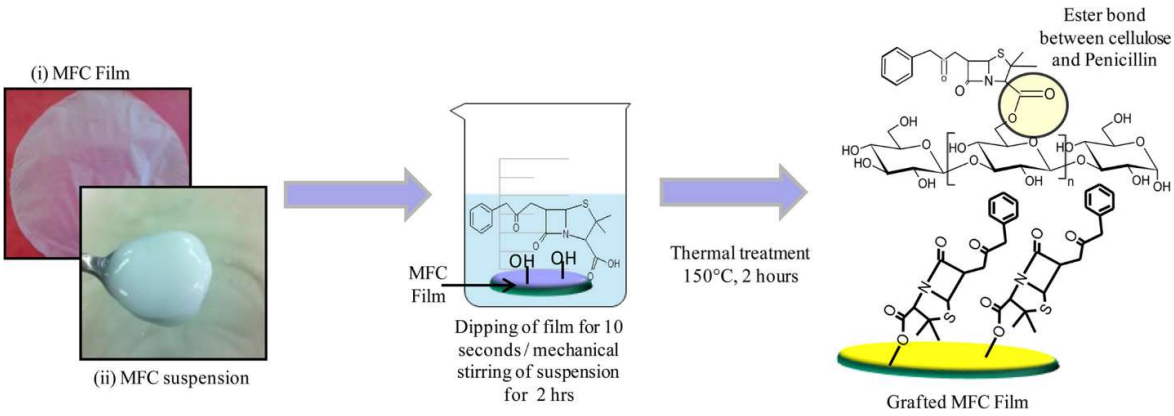

Figure 5 Schematic representation for suspension and film of MFC and the chemical modification with Benzyl Penicillin via esterification. Figure reprinted with permission from Ref. [119]. Copyright belongs to ACS (2015).

The cationic cellulose filter paper was reported for water treatment with antibacterial activity [109]. The cellulose filter paper was coated with PIDMAC as a cationic polyelectrolyte binder (CPE). It was further loaded with amphiphilic block copolymer micelles containing triclosan (antibacterial and antifungal agent). The micelles interacted with CPE via polystyrene-block-polyacrylic acid (PS-b-PAA) as the block copolymer. The materials contain two antibacterial

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agents, i.e., triclosan (hydrophobic) and ammonium compound [109]. Thus, it showed high antibacterial activity [109].

Cellulose is promising support for antibacterial agents, including antibiotics [126]. The use of conventional antibacterial agents such as antibiotics ensures high antibacterial activity for cellulose-based materials. However, the release of antibiotics into water causes environmental concerns and raises the risk of high antibiotic-resistant bacteria.

3.4.2. Aminoalkyl-modified Cellulose

Cellulose modified with aminoalkyl functional groups exhibit antibacterial activity [118,127–130]. The modification takes place via the reaction with silanol groups as coupling agents. The process involved the formation of a covalent bond between silanol groups (Si-OH) and hydroxyl groups (OH) of cellulose, i.e., the formation of Si–O–C bond. BC membrane modified with aminoalkyl groups using APMS ((3-aminopropyl)trimethoxysilane) exhibit antibacterial activity against *S. aureus* and *E. coli* [118]. The antibacterial activity of APMS-modified cellulose is due to the polycationic nature of the membrane and the alkyl chains in APMS. The long chains of the alkyl groups increase the lipophilicity that ensures high interactions with the cytoplasmic membrane of the investigated bacteria cells [118]. The alkyl groups with a length of up to 10 carbons exhibited antibacterial activity against a broad spectrum of antibacterial and antifungal activity [131]. The cellulose membrane with aminoalkyl groups exhibited non-toxic properties to human adipose-derived mesenchymal stem cells (hAMSC) [118] and human embryonic kidney 293 cells (HEK-293) [131].

Besides the chain length, the increase of amine groups ensures high antibacterial activity [117]. Cellulose was modified aminoalkyl containing different amine groups of 1, 2, 3 using APMS, 2-aminoethyl 3-aminopropyl trimethoxysilane (DAMS), and 3-2-(2-aminoethyl amino) ethyl aminopropyl-trimethoxysilane (TAMS), respectively. Cellulose-TAMS showed the highest

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antibacterial activity against Gram-positive bacteria [117]. The increase of amine groups offers high antibacterial activity.

3.4.3. N-halamine@Cellulose

N-halamines, halogen atoms linked to the nitrogen-containing compounds, were used to modify cellulose [132–134]. The functional groups of *N*-halamine, such as epoxy groups or organosiloxane, offer the grafting onto cellulose via the reaction with the hydroxyl groups [135]. The materials exhibit durable antibacterial activity [135]. N-halamine of s-triazine-based quaternized molecule was used to modify cellulose. The material showed a 6-log reduction in *S. aureus* and *E. coli* after treatment for 1–5 min [134]. The oxidant chlorine in *N*-halamine molecules showed 50% retention of their activity even after 50 cycles of washing and 30 days of storage. The remaining 50% of the material's activity could be regenerated after exposure to a bleach solution [134]. N-halamines modified cellulose can be restored via simple methods such as treating with a diluted bleach solution [136].

3.4.4. Antimicrobial peptides-modified Cellulose

Antimicrobial peptides are attractive compared to small organic molecules used as antibiotics [137,138]. Cellulose was modified with several peptides such as gentamicin [139]. Bacterial cellulose (BC) was chemically modified with RGDC peptides (R, G, D, and C refer to arginine, glycine, aspartic acid, and cysteine, respectively) and gentamicin via covalent bonds[139]. The process involves the cross-coupling of RGDC peptide to BC using a coupling agent such as 3-aminopropyltriethoxysilane (APTES). Gentamicin was then attached to the surface of the RGDC-BC membrane [139].

3.4.5. Polymers-modified Cellulose

Cellulose was modified with polymers to enhance antibacterial activity. The polymers can be directly blended (anchored or grafted) to cellulose using a simple procedure (post-synthetic

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procedure) or via in-situ polymerization of the monomers in the presence of cellulose (in-situ procedure). Cellulose was modified with several polymers including; 1) biopolymers such as chitosan [140], and 2) synthetic polymers such as polypyrrole [141], polypropylene [123,142], polyethyleneimine [143], polyhexamethylene guanidine hydrochloride (PHMG-Cl) [144], polyvinyl [145], 2-aminoethyl methacrylate [146], and poly(3-hydroxy-acetylthioalkanoate-co-3-hydroxy alkanoates) [147].

Fibers of dialdehyde cellulose (DAC)/chitosan (CTS) composite were prepared via stirring and filtration [140]. The antimicrobial activity of DAC/CTS against *E. coli* and *S. aureus* was investigated. Data analysis showed antibacterial activity of 90.2% and 95.1% against *E. coli* and *S. aureus*, respectively [140]. The antibacterial activity is due to the aldehyde groups of cellulose and the intrinsic antibacterial activity of CTS [140].

Polyrhodanine (PR) was prepared in the presence of CNC [148]. The synthesis procedure involves the polymerization of rhodanine on the surface of CNC using ferric chloride ($FeCl_3$) as the initiator and oxidant. The negatively charged surface of CNC assisted the polymerization and led to the formation of core-sheath nanoparticles of CNC@PR. Using the plate colony counting method, the antibacterial activities were evaluated against *E. coli* and *B.subtilis*. CNC@PR showed good antibacterial activity with good MICs [148].

Cellulose was modified with polymers such as guanidine polymer [149]. In-situ polymerization of 2-aminoethyl methacrylate (AEM) into BC network was reported with and without cross-linker such as *N*,*N*-methylenbis(acrylamide) (MBA)[146]. The polymerization takes place on the BC network via a radical-based reaction offering BC/poly-AEM/MBA. The synthesized polymer filled the pore of the BC network. BC/poly-AEM and BC/poly-AEM/MBA materials were evaluated for antimicrobial activity against *E. coli*. BC/poly-AEM exhibited higher antibacterial activity than cross-linked BC/poly-AEM/MBA material. The authors explained

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that the cross-linking reduced the diffusion of the bacteria into the BC network leading to low contact of the *E. coli* with the ammonium groups [146]. Thus, cross-linking of the network reduced the antibacterial activity.

Post-synthetic modification of BC with octenidine dihydrochloride was reported [150]. The material showed significant antimicrobial activity against *S. aureus* even after six months of storage. It exhibited minimal cytotoxic effects against human keratinocytes [150].

3.4.6. Bacteriophage-modified cellulose

Cellulose can be modified with a virus form called a bacteriophage (or phage for simple description) to induce DNA or RNA inside the cells [151–154] or bacteriophage endolysins (enzyme causes hydrolysis)[155,156]. The phage can be immobilized into cellulose via non-specific interactions (adsorption) [157], or covalent [158].

Several enzymes, such as lysozyme (muramidase), can be used as antibacterial agents. The mechanism of most enzymes is the cleavage of the bond in the cell membrane, such as the peptidoglycan layer of bacteria leading to cell lysis [159]. Enzymes such as lysozyme cause hydrolysis of the 1,4-β-linkage between N-acetylmuramic acid and N-acetylglucosamine [159]. CNC was used to immobilize Hen Egg White Lysozyme (HEWL) and T4 lysozyme (T4L, **Figure 6**) [158]. The immobilization of lysozyme into CNC causes no decrease in the enzyme's enzymatic activity for lytic and hydrolytic. The process involves covalent coupling via carbodiimide-activated CNC via carboxylate groups and to glutaraldehyde-activated aminated CNC (Am-CNC, **Figure 6**). Am-CNC-HEWL and Am-CNC-T4L showed lytic activity of 86.3% and 78.3%, respectively. The enzyme after immobilization exhibited high bactericidal activity compared to the free enzymes. They also showed high stability during storage at 4°C and 22°C (**Figure 6**) [158]. A thin film of CNC–lysozyme composite was prepared using the evaporation-induced self-assembly method [160]. The technique offers a

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film with HEWL enzyme loading of 10 wt.%. However, they showed insignificant antibacterial activity against Gram-positive bacteria. The optimization of enzyme-modified cellulose is critical to achieving high antibacterial activity.

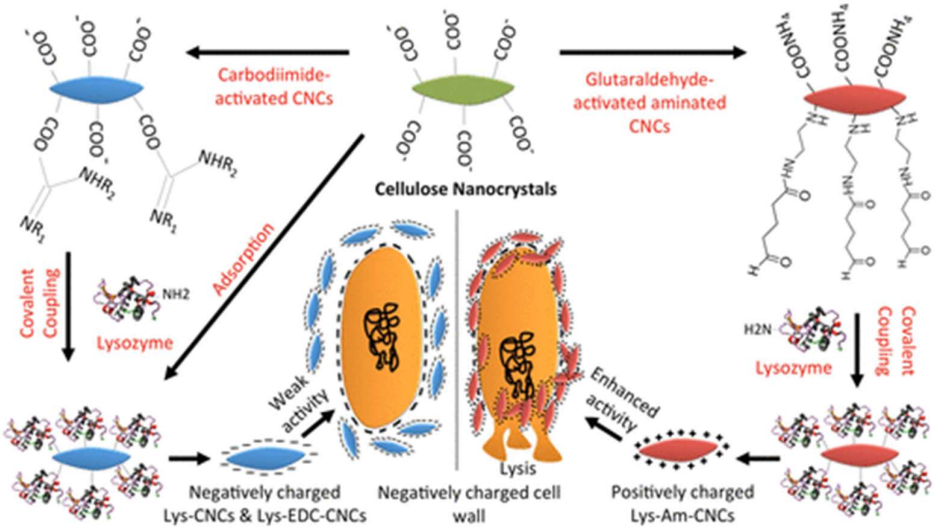

Figure 6 Scheme illustration for the immobilization of Lysozymes into CNCs for antibacterial activity. Figure reprinted with permission from Ref. [158]. Copyright belongs to ACS (2017).

Enzyme-based antibacterial agents offer several advantages, such as selectivity. For example, lysozyme exhibit high activity against Gram-positive bacteria compared to Gram-negative bacteria. This selectivity is because of the peptidoglycan layer only in Gram-positive bacteria cells [20]. The antibacterial agents should be active against broad-spectrum bacteria.

Bacteriophages exhibit high antibacterial activity. However, they are usually negatively charged at their head. Thus, they are difficult to interact directly with the negative charge of cellulose. They interacted easily with the positively charged surface via electrostatic interactions. The presence of functional groups such as amine and carboxylic acid on the phage surface offers the possibility for functionalization via covalent bonds. The latter method enables a highly durable and robust phage attachment to cellulose materials.

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Cellulose-organic composite with antibacterial activity can be synthesized via several methods. The simple attachments of both components via grafting or non-covalent interactions require no tedious efforts or expensive equipment. However, they lack high stability or long-term activity due to leaching. Thus, strong bonds such as covalent bonds are required to ensure high strength and long-term activity [161].

3.5. Cellulose-Inorganic nanoparticles for antibacterial agents

Cellulose was modified with inorganic nanoparticles such as carbon nanomaterials, metal oxides, and metallic nanoparticles, and metal-organic frameworks (MOFs) (**Figure 3**)[162–166]. Most of these materials exhibit intrinsic antibacterial activity leading to high performance against Gram-positive and Gram-negative strains.

Carbon nanomaterials exhibit high antibacterial activity. They were conjugated with cellulose. A composite of BC and graphene oxide (GO) was reported as an antimicrobial agent against *E. coli* and *S. aureus* [167]. Electrostatic modification improved the antimicrobial activity of GO/BC nanocomposites [168]. The antibacterial activity of carbon nanosheets such as GO is mainly due to the sharp edge of the sheets that sever as knife to cut the cell's membrane causing rupture of the outer envelope [169–172].

Metal oxide such as zinc oxide (ZnO) nanoparticles exhibits high antibacterial activity. A composite of cellulose acetate (CA) and hydrophobic polysulfone (PSf) polymer was used to modify with 0.1 wt.% of ZnO NPs [173]. The membranes containing ZnO NPs showed good antibacterial activity against *E. coli* [173]. ZnO nanoparticles were prepared via an in-situ procedure [174]. Zn^{2+} ions were adsorbed into cellulose before precipitation using ammonium hydroxide [174]. The surface functional groups of cellulose, such as hydroxyl groups, enable the adsorption of Zn^{2+} ions via electrostatic and offer control of the morphology and particle

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size. The antibacterial activity of ZnO-based nanomaterials depends on the ZnO contents in composites [175].

Other metal oxide nanoparticles such as TiO_2 nanoparticles [176,177], faujasite [178], and montmorillonite (MMT) [179] were modified with cellulose for antibacterial activity. Cellulose acetate/TiO_2 nanoparticles exhibited high antibacterial activity [177]. A membrane of Faujasite-cellulose composite was used to purify water contaminated with bacteria (*E. coli*, *Enterococci*, and *Clostridium*). It showed high removal efficiency, offering <100 colonies/100 mL [178]. The presence of MMT into the cellulose membrane enabled the modification with several metal ions such as Na, Ca, and Cu [179]. BC/Cu-MMT composites showed the highest antibacterial activity against the investigated bacteria [179].

Silver nanoparticles (Ag NPs) are very active antibacterial agents [180]. They were widely modified with cellulose nanoparticles for antibacterial activity [181–189]. Ag NPs were synthesized directly into electrospinning fiber of cellulose acetate (CA) [190]. The procedure involved the adsorption of silver ions that were reduced via photon into Ag NPs. The prepared materials exhibited high antibacterial activity against *S. aureus*, *E. coli*, *K. pneumoniae*, and *P. aeruginosa* [190]. Nanofiltration (NF) membrane consisting of CNC/silver (CNC/Ag) using polyamide was fabricated using the interfacial polymerization (IP) method [191]. The membrane with 0.01 wt.% CNC/Ag showed antibacterial activity of 99.4% against *E. coli* viability [191]. The high antibacterial activity of silver-based nanomaterials can be explained in several mechanisms such as ionization and the release of silver ions [189]. Silver nanoparticles offered several advantages. It can be synthesized into cellulose textile via an in-situ procedure [192]. It can be conjugated with other materials such as magnetic nanoparticles (MNPs) [193] and gold nanoparticles [194]. Furthermore, cellulose offers biodegradable support for Ag NPs [195].

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The conjugation of cellulose with inorganic nanomaterials ensures high antibacterial activity. However, they can be toxic for mammalian cells or environmentally unfriendly due to toxic heavy metal ions released into drinking water.

4. Cellulose-based Materials for Antifouling

Pure cellulose nanoparticles with residual lignin [196,197] or carboxylic functional groups [198] offer bioactivity and antibacterial and antifouling properties. Thus, they are widely used as a coating for membranes for antifouling applications [198]. A membrane of TOCNF, poly(vinyl alcohol) (PVA), and polyethersulfone (PES) was fabricated for antifouling application. TOCNF/PVA@PES membrane exhibited high antifouling against *E. coli* [198]. Micro/nanocellulose membrane grafted with zwitterionic poly(cysteine methacrylate) (PCysMA) showed excellent antibacterial and antifouling properties. It showed a reduction efficiency of 85% in the biofilm formation for *S. aureus* [197]. Cellulose nanoparticles-based membranes exhibit high antifouling performance with high reflux compared to commercial membranes such as Millipore GS9035 [199].

CNC/silver/polyamide membrane using polyamide containing CNC/Ag (0.01 wt.%) showed high antifouling activity of 92.6% using humic acid with the antibacterial activity of 99.4% against *E. coli* viability [191]. A membrane of CMC/GO/magnesium oxide (MgO) nanoparticle (CMC/GO/MgO) was prepared for photocatalytic antifouling [200]. It can be used as an antifouling membrane due to the generation of electrons and ROS. Thus, it can be used to oxidize organic pollutants [200]. Photocatalytic disinfection using photoactive substances is based on generating free radicals under the light.

Cellulose-inorganic hybrids exhibited high antibacterial performance. CNC/silver/polyamide membrane offered high antifouling with flux recovery of 92.6% using humic acid [191]. A

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composite nanofiber of CA and polysulfone (PSf) with 0.1 wt.% ZnO exhibited high antibacterial activity against *E. coli* [173].

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Table 1 Antibacterial applications for cellulose-based materials.

Materials	Preparation Methods	Form	Microorganism	Methods	Time	Efficiency	Mechanism	Ref.
DANFC	1. Mechanical grinding 2. Enzyme treatment 3. $NaIO_4$ oxidation 4. Dialysis 5. Freezy-drying	Mat	S.aureus and MERSA	Zone inhibition Plat counting method	24h	100%	Drop-in pH value	[73]
TOCNF	1.TEMPO-oxidation 2. Oxygenation	Suspension	P. aeruginosa *and* S. aureus	Plate counting method	24h	71%	The formation of a network surrounding the bacteria	[45]
TOCNF	1.TEMPO-oxidation 2. Autoclaving (121 °C, 20 min)	Gels	*P. aeruginosa or S. aureus*	Plate counting method	24h	71%	Increase of aldehydes, drop in pH value	[77]
CNC-Porphyrin	1. HBr acid hydrolysis of Whatman #1 filter paper 2.Cu(I)-catalyzed Huisgen–Meldal–Sharpless 1,3-dipolar cycloaddition	Suspension	*Mycobacterium smegmatis, S.aureus, and E.coli*	Plate counting method	60 min	>99%	Generation of ROS, Photodynamic	[86]
Porphyrin-cellulose paper	Cu(I)-catalyzed Huisgen–Meldal–Sharpless cycloaddition	Paper	S.aureus, *VER,* Enterococcus faecium, Acinetobacter baumannii, P. aeruginosa, *and* Klebsiella pneumoniae	Plate counting method	30 min	>99.9%	Generation of ROS, Photodynamic	[81]

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NFC-Porphyrin	Cyanuric chloride coupling	Paper	MRSA, VER, *E. faecium*, *A. baumannii* and *K. pneumoniae*	Plate counting method	30 min	99.999%	Generation of ROS, Photodynamic	[104]
CHPTAC-Cellulose triacetate	1.Immersion precipitation technique 2. Alkaline hydrolysis 3. Esterification	Membrane	*E. coli* *S. aureus*	Plate counting method	24h	78.7–89.0% 64.7–76.6%	Cationic charge	[112]
BC-aminoalkyl	Stirring for 5h at 25 oC	Membrane	S. aureus *and* E. coli	Dynamic shake flask method	24h	>99.9%	Increase lipophylicity	[118]
DAC/CTS	1.$NaIO_4$ oxidation 2. Stirring 3. Filteration	Fibers	S. aureus *and* E. coli	Plate counting method	24h	95.1% 90.2%	Drop-in pH, cationic CTS	[140]
T7 phage-Cellulose acetate	Electrospinning of cellulose acetate	Membrane	*E. coli*	Plaque forming units (PFU)	24h	6 log(PFU/mL)	Release of phage and hydrolysis	[157]
Am-CNC-HEWL Am-CNC-T4L	1.Ammonium persulfate oxidation 2. Modification 3. Coupling	Suspension	M. lysodeikticus, Corynebacterium *sp.*, E. coli, *and* Ps. mendocina	Time-kill study with Alamar Blue assay	24h	100%	Lytic activity	[158]
CNC-Lysozyme	1. Sulfuric acid hydrolysis 2. Evaporation-induced self-assembly	Thin film	*E. coli and S. aureus*	Diffusion assays	24h	0%	Lytic activity	[160]
ZnO-BC	In-situ synthesis	Sheets	*E. coli and S. aureus*	Inhibition zone	24h	5.7 ± 0.29 mm 2.9 ± 0.75 mm	Formation of ROS	[174]
ZnO-BC	Ex-situ synthesis	Film	*E. coli*	Inhibition zone	24h	34-41 mm	Formation of ROS	[175]
TiO_2-BC	Ex-situ synthesis	Pieces	*E. coli*	Optical density (OD_{600})	12h	60%	Formation of ROS	[176]

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Faujasite-cellulose	Hydrothermal	Membrane	*E.coli*, *Enterococci* and Clostridium	Standard FS ISO9308 and ISO17025		80%		[178]
SSD/BC	Impregnation and ultrasonication	Commercial membrane	P. aeruginosa, E. coli and S. aureus	Zone-inhibition	24h	6.5 mm		[201]

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Inorganic nanoparticles improved the antibacterial activity of cellulose-based materials. The direct contact with the metallic ions causes damage to the cell membrane due to the generation of reactive oxygen species (ROS) that can permeate the cell wall. The formed ROS damage the phospholipid layer of the cell membrane can also cause cell disruption via the oxidation of thiol groups of amino acids present in the bacterial cells.

6. Cellulose Nanoparticles for Wound Dressing

During burn treatment, special care should be performed to avoid microbial infection in burn treatment [45]. Wound healing requires fast re-epithelialization within 10–14 days to prevent complications. Chronic wounds may undergo critical physiological change or initiate tumor growth leading to high levels of destroying tissues or organs. This situation is vital for diabetic patients who can suffer from chronic ulcers. Thus, the dressing becomes an essential active component during the healing process and is no longer considered a supplement. The dressing should offer a warm and moist environment to provide a fast and effective healing process. It should also be biocompatible, easy to detach, thermal insulator, and provide antimicrobial activity to prevent infections.

Natural polymers such as cellulose were widely investigated for wound dressing because of their high biocompatibility and biodegradable properties (**Table 2**). Bacterial cellulose (BC) has been intensively used for wound healing [47,202]. Several methods were reported to modify BC to improve the material's properties, such as biocompatibility, uptake/release of water or drug molecules, and ensure high antimicrobial activity. Cellulose can be fabricated into different forms such as films, foams, hydrocolloids, hydrogels, and nonwovens [203]. It can also support wounds with drugs, growth factors, and antimicrobial agents. Traditional gauze of cotton is usually modified with sodium periodate via oxidation, i.e., the formation of dialdehyde [204]. The oxidation process decreases elastase and offers the potential for a

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chronic wound. It can also improve the antibacterial activity and offers high protection due to the drop in pH value caused by the aldehyde functional group in cellulose [73]. BC-based wound dressings reduce the closure time (e.g., recovery rate) of the wound [205,206] without significant inflammation (**Table 2**) [207].

The wound dressing of pure cellulose such as NFC was reported [43]. The antimicrobial properties of dressing materials were investigated against bacterial pathogens such as *S. aureus* and *P. aeruginosa* [43]. NFC-based wound dressing showed no antibacterial activity against the tested species. The BC-based film exhibits good wound healing (**Figure 7**) [207]. The performance is lower than traditional wound dressing (gauze). However, it may be improved. The sides of BC film (i.e., bottom and top) exhibit different performances (**Figure 7**) [207]. The antibacterial activity of cellulose can be improved via the formation of carboxylated CNF [45]. Autoclaved CNFs exhibited a high antibacterial effect of wound infection bacteria [77].

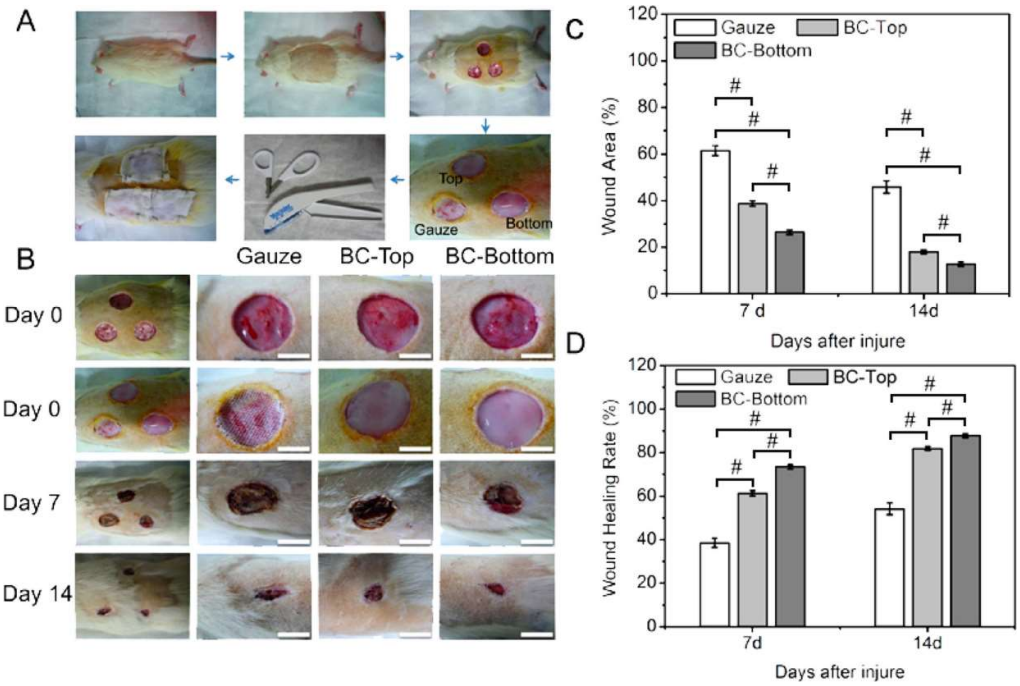

Figure 7 Wound healing treatment using BC-based dressing, A) description for the operation of skin injury model and the dynamic healing on a rat, B) the progress (0-14 days) of healing for skin injury model on Wistar rat using gauze and BC-based dressing of two sides; top and bottom (All the scale bars equal 10 mm), C) the wound area progression after the injury and

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D) wound healing rate. Figure reprinted with permission from Ref. [207]. Copyright belongs to the American Chemical Society (ACS, 2015).

Cellulose nanofibrils were prepared via the defibrillation of wood via mechanical method [41]. The fibrils have proceeded as membrane via filtration and drying under mild pressure. The prepared cellulose membrane was used as a wound dressing. It offered strong adhesion to the wound and promoted epithelialization after 4 days without observation of allergic or inflammation during the treatment [41]. A cellulose hydrogel was prepared from the alkali treatment and TEMPO-mediated oxidation of ginger fibers (T-GNF) [208]. It was designed via a simple vacuum-assisted filtration using only ginger nanofibers without cross-linking treatments. The preparation method required low energy and few components for the production procedure. The antibacterial and wound healing of the material were investigated [208]. The material exhibited a closure rate for wound healing without significant antibacterial activity against *E. coli* and *S. aureus* [208]. 3D printing of NC hydrogel scaffolds was reported for wound healing [209]. The process included a double cross-linking procedure via two steps; i) in situ Ca^{2+} crosslinking during 3D printing and ii) chemical crosslinking with 1,4-butanediol diglycidyl ether (BDDE) after printing. The scaffolds were used to support the proliferation of fibroblast cells with an attachment percentage of 84-86.5% [209]. Cellulose exhibits high performance for wound healing and is capable of proceeding via several procedures.

Modulating the wound's pH value enhanced the wound treatment [210]. Intact skin exhibit a slightly acidic pH value of 4-6 due to the secretion of keratinocytes for organic acids to regulate bacterial flora and prevent infection. While the infected wound exhibit a pH value of 7-7.5. The neutral pH value of the wound is an ideal condition for the growth of bacteria cells. The drop in the pH of the wound using aldehyde modified cellulose enable high protection against bacterial infection [73]. Dialdehyde of microcrystalline cellulose (DAMC) [74], and

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nanofibrillated cellulose (DANFC) [73] with aldehyde content of 6.5 mmol/g, and 1.5 mmol/g, respectively showed high antibacterial activity.

The antibacterial activity of cellulose-based wound dressing can be improved via loading antibiotics. Chloramphenicol (CAP)-loaded dialdehyde (DABC) and non-oxidized BC membranes were investigated in terms of their antimicrobial efficiency against *E. coli*, *S. aureus*, and *Streptococcus pneumoniae* (*S. pneumoniae*)[211]. The CAP drug loading capacity using DABC was low (0.1 mg/cm^2) compared to the BC-based membrane that showed a loading capacity of 5 mg/cm^2. However, both membranes, i.e., oxidized and non-oxidized BC, showed high antimicrobial activity against the tested bacteria. CAP/DABC showed high adhesion and proliferation of fibroblasts cell line L929 compared to non-oxidized BC. This study highlighted the potential for using newly developed CAP/DABC dressing materials in wound treatment [211].

Cellulose-modified organic nanocomposite exhibits high performance as a wound dressing. A Three-dimensional (3D) network of collagen I (Col-I), hydroxypropyl trimethyl ammonium chloride chitosan (HACC), and BC was prepared via membrane–liquid interface (MLI) method [212]. HACC offered antibacterial activity during wound healing [212]. Inerpan (a polymer of L-leucine and methyl L-glutamate) and Procel-Super (SOD) accelerated the healing of burn wounds for BC-based wound dressing by 17.0 and 5.5%, respectively [213].

Cellulose nanocomposites are widely reported for wound dressing. A membrane of BC and chitosan (BC–CTS) was fabricated via the immersion of BC into a solution of CTS followed by freeze-drying [214]. The procedure can be applied for large-scale production. The presence of antibacterial agents such as CTS offered significant inhibition for the growth of bacteria cells, e.g., *E. coli* and *S. aureus* [214]. BC-CTs membrane exhibited higher antibacterial activity compared to pure BC membrane. BC and BC-CTS membrane offered antibacterial

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inhibition of 49.2% and 99.9% for *E. coli* and 30.4%, 99.9% for *S. aureus*, respectively. The BC-CTS membrane showed high epithelialization and regeneration of wound healing compared to wound treated with BC only or a commercial dressing such as Tegaderm^{TM} [214].

A sponge of carboxylated brown algae cellulose nanofibers (BACNFs) was prepared to contain organic rectorite (OREC) via freeze-drying [215]. OREC was organized via intercalation of chitosan (positive charge) into negative silicate layers *via* ion exchange. The sponge of BACNFs\OREC exhibits high antibacterial activity against *E. coli* and *S. aureus* without significant toxicity toward mouse fibroblast (L929). It also showed effective wound closure (100%) after 12 days compared to commercial gauze. It also exhibited high prevention for bacterial infections without significant inflammatory response [215]. It is essential to mention that BACNFs showed no antibacterial activity [215].

Cellulose was conjugated with several inorganic-based antimicrobial agents, such as silver nanoparticles [216], graphene oxide (GO)[217], and ZnO [218]. Silver-based drugs such as silver sulfadiazine (SSD) are widely used as antibacterial agents. BC/SSD was prepared via the impregnation of SSD into BC membrane via ultrasonication [201]. The BC/SSD membrane showed significant antibacterial activities against several bacteria such as *P. aeruginosa*, *E. coli,* and *S. aureus* [201]. The membrane exhibited high biocompatibility [201].

Cellulose offers several advantages for wound dressing (**Table 2**). It requires low cost. It can be fabricated into applicable forms such as membranes using cheap sources such as wood [41]. Cellulose-based membrane exhibits high performance for wound dressing than a commercial porous regenerating membrane [41]. The epithelialization of the wood-based dressing such as NFC showed faster healing compared to Suprathel® (commercial lactocapromer-based wound dressing)[43]. BC dressings are cheaper than conventional synthetic fiber dressings [219].

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The surface properties of cellulose-based wound materials can be modified via several methods, such as cross-linking with silane-based reagents [220]. The material was investigated for wounds for femoral artery and liver injury models. The chemical modification with organosilane enabled a hydrophobic layer that stoped the blood penetration (blood loss < 50%) and accelerated the process of blood clotting. It offered a short time for hemostasis for both models [220]. The high surface charge of cellulose nanomaterials enhanced the protein adsorption and could promote cell adhesion [162].

The dressing prepared from cellulose is usually transparent, allowing the evaluation of the wound treatment without removing or exchanging the dressing [41]. Cellulose-based membrane offers good adhesion to the moist wound surfaces due to the abundant hydroxyl groups present in cellulose structure without significant observation of allergic or inflammatory responses[41]. Cellulose-based dressing offers faster self-detachment than commercially available wound healing dressing [43]. They can be used for infected wounds [221]. Thymol enriched BC hydrogel can be used to treat injury healing of third-degree burns [222].

Among several cellulose types, BC-based membranes are used for healing wounds. However, the extraction process offers a low yield and requires a high cost. Nanocellulose extracted from wood pulp fibers can be an alternative to BC. Cellulose-based dressing suffers from swelling behavior due to the high affinity toward water molecules. The surface properties should be improved to meet the requirements for wound healing. BC shows no antimicrobial activity. Thus, antimicrobial agents are usually required.

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Table 2 Cellulose-based materials for wound dressing.

Materials	Fabrication	Cellulose Source	Form	Study	Closure (%)	Time (days)	Bacteria	Efficiency	Ref.
BC–CTS	Immersing BC in chitosan followed by freeze-drying	*Acetobacter xylinum*	Membrane	In-vivo	85	8	*E.coli, S. aureus*	99.9%	[214]
Cellulose nanofibrils	Filtration technique	Birch pulp fibers	Membrane	In-vitro In-vivo Clinical studies		8-9	*S. aureus, P. aeruginosa*	None	[43]
Carboxylated CNF	1. Autoclaved using NaOH 2. TEMPO-mediated oxidation	*Pinus radiata* bleached kraft pulp fibers	Gels	In-vitro In-vivo		24h	*P. aeruginosa*, *S. aureus*	60%	[77]
SSD/BC	Impregnation of SSD into BC via ultrasonication	Commercial membrane	Membrane	In-vitro			*P. aeruginosa, E. coli, S. aureus*	6.5 mm	[201]
BC	Cultured the bacteria in Hestrin and Schramm (HS)	*Acetobacter xylinum*	Film	In-vitro In-vivo	90	24h			[207]
T-GNF	1.Alkali treatment 2.TEMPO-mediated oxidation	Ginger fibers	Hydrogels	In-vitro	67	72h	*E. coli, S. aureus*	0	[208]
BACNF/QCR	1. Cation exchange 2. Freeze-drying	Brown algae	Sponge	In-vitro In-vivo	100	12 d	*E. coli, S. aureus*	6 mm	[215]

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7. Drug and Gene Delivery using Cellulose-based Materials

Cellulose-based materials advanced drug delivery [51,223–227]. They can be conjugated with nanomaterials such as magnetic nanoparticles (MNPs) to offer multifunctional applications [228]. They can be used for drug encapsulation [229]. The functional groups of carboxymethyl cellulose (CMC) enabled the modification with folate for selective release of an anticancer agent such as 2,4- dihydroxy-5-fluorpyrimidin (5-FU) [230]. Cellulose's surface modification with folic acid ensures selective cell uptake and binding via folate receptor-mediated cellular mechanism [231,232]. Cellulose severs as effective carriers for the drug delivery of hydrophobic drugs such as docetaxel, paclitaxel (PTX), and etoposide [233].

Hydroxypropyl methylcellulose improved the cellular uptake of curcumin (CUR) to treat prostate cancer cells [234]. CUR-conjugated cellulose exhibited significant changes in apoptosis compared to CUR alone. Cellulose also showed the highest cellular uptake compared to other carriers such as β-cyclodextrin (CD), poly(lactic-co-glycolic acid) (PLGA), MNPs, and dendrimer [234]. TOCNF and MOFs such as zeolitic imidazolate frameworks (ZIF-8) were used for drug delivery of CUR (**Figure 8**) [235]. The composite of TOCNF/ZIF-8 offered the material processing into a 3D network via 3D printing [235]. The materials can release the CUR drug under physiological pH (5.5) [235].

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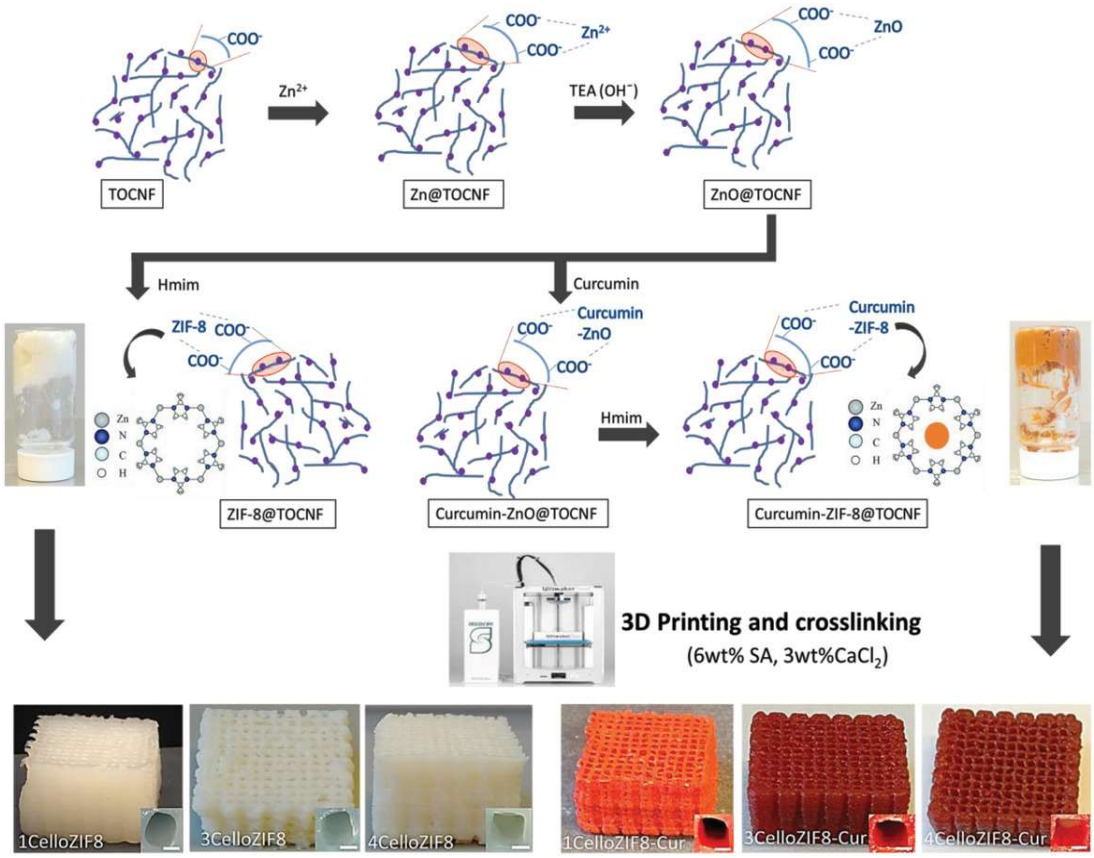

Figure 8 Synthesis procedure of cellulose-ZIF8 bioink and their processing into the 3D network via 3D printing. Figure reprinted with permission from Ref. [235]. Copyright belongs to John Wiley & Sons (2019).

Cellulose-based materials were also used for gene delivery of oligonucleotides such as siRNA [236,237]. CNCs were modified with poly(2-dimethylamino)ethyl methacrylate) (PDMAEMA) via atom transfer radical polymerization (ATRP) for gene delivery of pDNA. The polymerization occurs via forming a disulfide (SS) bond, and the product was denoted as CNC-SS-PDs [238]. The CNC-SS-PDs exhibited good transfection efficacy with low cytotoxicity [238]. Cellulose-based materials are promising non-viral vectors for gene delivery [239–245].

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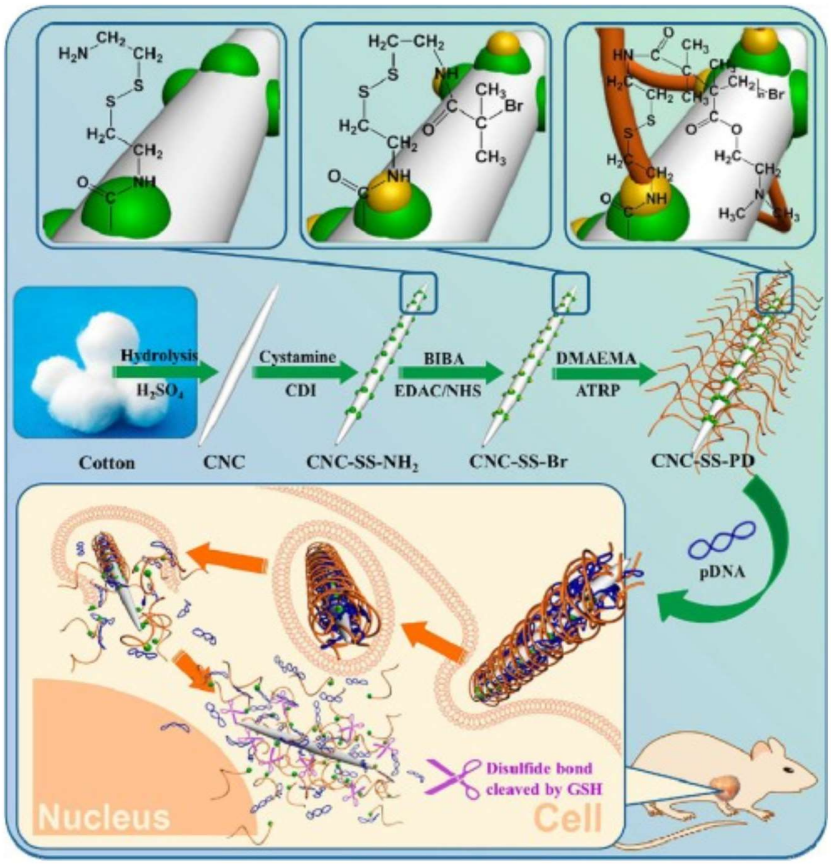

Figure 9 Schematic representation for the preparation of CNC-SS-PD and their use for gene delivery process. Figure reprinted with permission from Ref. [238]. Copyright belongs to ACS (2015).

Cellulose-based materials offer several advantages for drug delivery. They can be used to release water-soluble and insoluble drugs, ionizable, and hydrophobic drugs [233,246]. Thus, they can be used to co-deliver two drugs [247]. CNC hydrogels offered locally targeted drug release [248] with sustainable properties [249]. Cellulose can be fabricated as capsules without gelatin [250]. It can be used for oral drug delivery [251]. The drug delivery of cellulose-based hydrogels can be simulated under pH- and temperature-responsive.

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8. Scaffolds for skin and tissue engineering

Nanocellulose-based materials offer several advantages as scaffolds for tissue engineering (**Table 3**) [252]. They show high biocompatibility, good water absorption, high water retention, high optical transparency, and good mechanical properties. They can be performed using comprehensive methods, including solvent casting, electrospinning, freeze-drying, and 3D printing [252]. They can easily be custom-made for tissues engineering of damaged tissues or organs. They can be optimized to ensure the required hierarchical structure, pore size, surface functional groups, and mechanical properties. Thus, they can be used for blood vessels, skin, and organs engineering [252]. Scaffolds should offer several essential requirements, including high biocompatibility to mimic the natural extracellular matrix (ECM) of native tissue. They should also serve as carriers for cell growth, proliferation, and differentiation (**Table 3**).

Bacterial cellulose (BC) was used as tissue-engineered blood vessels (TEBV) [253]. BC-based TEBV promises caliber vascular grafts to reconstruct tissues associated with vascular diseases [253]. BC was used to fabricate artificial blood vessels that can be used for microsurgery [53]. The vessels were abbreviated as BASYC®, referring to **BA**cterial **SY**nthesized **C**ellulose. The prepared vessels exhibit high mechanical strength in wet form with high water retention. They also showed low roughness of the inner surface and can provide a complete 'vitalization' in the rat. BASYC® was proposed as an artificial blood vessel in microsurgery [53].

BC was reported as a temporary skin substitute [254] and as a scaffold for cartilage tissue engineering [255]. It was used for in-vitro seeding of cells such as Human adipose-derived stem cells (hASCs)[256]; Human urine-derived stem cells (hUSCs) [257]; Human keratinocytes (HaCats) [258]; Human umbilical vein endothelial cells (HUVECs)[259]; Equine-derived bone marrow mesenchymal stem cells (EqMSCs)[260]; Human embryonic kidney 293 cells (HEK) [261]; Bovine smooth muscle cells (SMCs)[259]; Endothelial cells

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(ECs)[262], Chondrocytes[255]; Epidermal cells[201]; Mouse leukaemic monocyte-macrophage cells (RAW 264.7, **Table 3**) [207]. It showed high biocompatibility and promoted the proliferation of cells such as SMC, leading to ingrowth of size > 40 µm after two weeks of culture on BC pellicle [253]. The proliferation of hASCs into BC film showed a plateau phase after 9 days, indicating a single layer on the film [256]. Thus, BC is promising for tissues engineering.

BC pellicle showed the formation of an exemplary network similar to a collagen network [253]. It can form porous scaffolds with different pores sizes using sterile paraffin particles of varying size ranges (90–150 µm, 150–300 µm, and 300–500 µm) [257]. Biomolecules such as alginate were used to prepare sponges for oral tissue regeneration [258]. It can also be shaped into tubes using tubular template materials such as PDMS (polydimethylsiloxane) [259].

Cellulose offers several advantages. The cellulose composite exhibits high cell compatibility, water uptake, and mechanical strength. Cellulose-based materials exhibit good stability in water and phosphate buffer saline (PBS) buffer. BC offers the growth of multipotent mesenchymal stem cells (MSCs) [260]. Thus, BC/MSCs hydrogel can be used to construct musculoskeletal tissue. The BC scaffolds improved the adhesion, proliferation, and differentiation of MSCs [260].

The proliferative rates of several cells such as HaCat and gingival fibroblasts (GF) using different supports were ordered in the following order; tissue culture plastic (TCP) > BC > BC-alginate (BCA) > alginate [258]. This character depends on the cell types. BC offered significantly higher chondrocyte growth levels than TCP and calcium alginate [255]. Unmodified BC supports the proliferation of chondrocytes at levels of 50% of the collagen type II substrate [255]. However, BC-based materials exhibited good mechanical properties [255].

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3D printing of bioink consisting of NFC and alginate was reported [263]. NFC ensures high shear-thinning properties, while alginate enables fast cross-linking ability. 3D printing of bioink can be used for cartilage structures, including a human ear and sheep meniscus (**Figure 10**). The method can be used for printing human chondrocytes nanocellulose-based bioink. Cellulose bioink showed 73% and 86% high cell viability after 1 and 7 days, respectively [263].

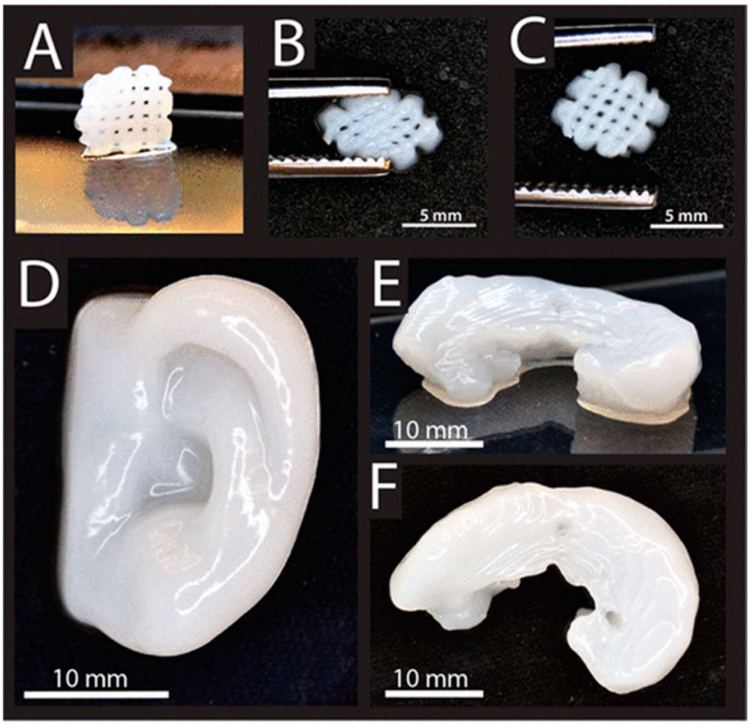

Figure 10 3D printing of NFC-alginate into A) small grids (7.2 × 7.2 mm^2), B) after squeezing, C) restored after squeezing, D-F) 3D printed human ear in different views. Figure reprinted with permission from Ref. [263]. Copyright belongs to ACS (2015).

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Table 3 Applications of cellulose-based materials for tissue engineering.

Cellulose	Source	Form	Fabrication method	Study Type	Cells	Time (d)	Evaluation Method	Comments	Ref.
BC	*Acetobacter xylinum*	films	Shaken in a culture flask	In-vitro In-vivo	hASCs	9	Optical density (OD)		[256]
BC	*Acetobacter xylinum*	Scaffold	Fermentation into sterile paraffin particles	In-vitro In-vivo	hUSC	7	Histology	No effect of pore size	[257]
BC/alginate (BCA)	*Acetobacter xylinum*	Sponge	Freeze-drying	In-vitro	HaCat	2	MTT assay	30% alginate	[258]
BC	*Acetobacter xylinum*	Scaffold	Culture on TCP	In-vitro	EqMSCs	14	OD	The cells seeded were metabolically active.	[260]
BC	*Acetobacter xylinum*	Tubes	Culture on PDMS tubes	In-vitro In-vivo	SMCs	7	OD	No signs of inflammation	[259]
BC-CMC	*Gluconacetobacter saccharivorans*	Gel	Agitation overnight at room temperature	In-vitro	HEK	1	Optical microscope		[264]
BC	*Acetobacter Xylinum*	Tubes	Fermentation in glass tubes using a silicone support	In-vitro	ECs	28	Fluorescence microscope		[262]
BC	*Acetobacter xylinum*	Scaffold	Freeze-drying	In-vitro In-vivo	Chondrocyte	8	Fluorescence microscope		[255]

Notes: 3-(4,5-dimethythiazolyl-2)-2,5-diphenyltetrazolium bromide, MTT) assay; *Acetobacter xylinum is the same as Gluconacetobacter xylinus.*

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Cellulose was used for bone regeneration. BC assisted the synthesis of hydroxyapatite (HAP) with calcium-deficient and nanocrystallites particle size [265,266]. BC was oxidized before the formation of HAP to offer high degradable materials. HAP/BC was used for bone regeneration using osseous tissue [265]. The material can be degraded under physiological conditions, i.e., pH and temperature of 7 and 37 ºC, respectively. Thus, it was proposed to stimulate bone colonization [265].

A nanocomposite material of BC networks and HAp was prepared via a wet chemical precipitation method using aqueous solutions of calcium nitrate and di-ammonium phosphate salts [261]. The dispersion can be improved by adding CMC (1% w/v). CMC increased the pore size of BC by 47.8%. The prepared composite, i.e., BC/HAP/CMC, supported the growth of HEK cells [261].

BC scaffolds were used for the growth of EqMSCs to apply bone and cartilage tissue engineering [260]. They can be fabricated via freeze-drying. They can be prepared with fiber diameters and pore sizes of 32.08 ± 10.85 nm and 254.16 ± 76.65 nm, respectively. They exhibited high cytocompatibility and can support the cellular adhesion and proliferation of the cells. They kept the differentiation of EqMSCs. EqMSCs/BC scaffolds are promising for bone and cartilage regeneration tissue engineering, similar to those reported using tissue culture-treated plastic (TCP)[260].

Cellulose-based materials offer several advantages for tissue engineering. They provided high biocompatibility [267], self-healing properties [268]. Cellulose materials can be conjugated with polymers to improve their properties. A rigid composite of tannic acid (TA)\CNC (TA@CNC) was incorporated into poly(vinyl alcohol) (PVA)-borax networks [269]. TA@CNC\PVA-borax hydrogels offered high toughness with self-healing properties [269].

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Outlook

CNCs were commercialized via several companies such as Bio Vision (Canada), CelluForce (Canada, the trade name is *NCC*™), and US Forest Service Forest Products Laboratory (USA). While CNFs were marketed by several European companies such as Centre Technique du Papier (France)), Borregaard ChemCell (Norway), Innventia AB (Sweden), Stora Enso (Finland), and UPM fibril cellulose (Finland). Plant-based cellulose was commercialized into several products such as cellophane (transparent films), Rayon or Tencel™ (synthetic textile fibers), Surgicel™, Interceed®. BC-based materials were commercialized into several products such as Bioprocess®, BASYC®, Biofill®, XCell, and Gengiflex®. Microbial-based cellulose is free of lignin and hemicelluloses compared to cellulose extracted from a plant source. However, the use of microbial-based cellulose requires high-security precautions to avoid the presence of microbial species inside the extracted cellulose. Cellulose can be produced from cheap sources and waste materials [270–272]. However, the current technologies for cellulose nanomaterial fabrication are still expensive, require tedious efforts, and lack large-scale production for industrial and biomedicine applications.

Cellulose-based antimicrobial agents are promising for the fabrication for membrane [40], antimicrobial textiles [273,273,274], and food packing [275,276]. They offer durable antibacterial activity with tunable properties such as hydrophobic [277]. They can be modified with different antibacterial agents with permanent antibacterial properties [278]. Nanoparticles exhibit undesirable toxicity. Thus, some precautions or post-synthetic treatments should be performed. For instance, sulfidation was proposed to transform Ag NPs into highly insoluble forms for minimal cytotoxicity [279].

Cellulose-based materials offer green cross-linkers [280] and flexible platforms [281] for tissue engineering. It can be modified via in-situ and ex-situ procedures [282]. They can

proceed into several forms using various methods. Most of the available techniques lack large-scale production and require other materials such as binders or modifiers. Further investigations to move cellulose materials into customized requirements with minimal cost are needed.

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