

# Chapter 5

## Cellulose-Based Biodegradable Polymers: Synthesis, Properties, and Their Applications



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### 1 Introduction

Biopolymers have significantly drawn attention for creating numerous biodegradable products. These biopolymers are synthesized within the biological cells through enzymatic pathways. Among several polysaccharides such as starch, cellulose, xanthan, pullulan, hyaluronic acid, chitosan, etc., cellulose gains popularity as it contains large numbers of OH groups that can be modified according to the requirements. It shields plants, and fungi from external chemical, biological, and mechanical disturbances. The large supramolecular structure containing large amounts of OH moieties makes cellulose hydrophilic as they are susceptible to water attack. About 40–50 weight percent of the woody biomass in plants is made up of cellulose, the most common polymer, and a considerable amount of this biomass is found in crystalline forms [1]. Chemically, cellulose is a long linear chain of anhydro-D-glucopyranosyl units joined by  $\beta$ -(1,4)-glycosidic linkages (Fig. 1a) [2]. The cyclic hemiacetal groups contain equatorial OH groups (in the C<sub>1</sub> position) which defines that the cellulose structure exists in  $\beta$ -form (or  $\beta$  isomer), whereas, in the case of  $\alpha$  isomer, the OH group remains in the axial position. The different isomers differentiate cellulose from other polysaccharides in terms of its biodegradation through hydrolysis reaction [3].

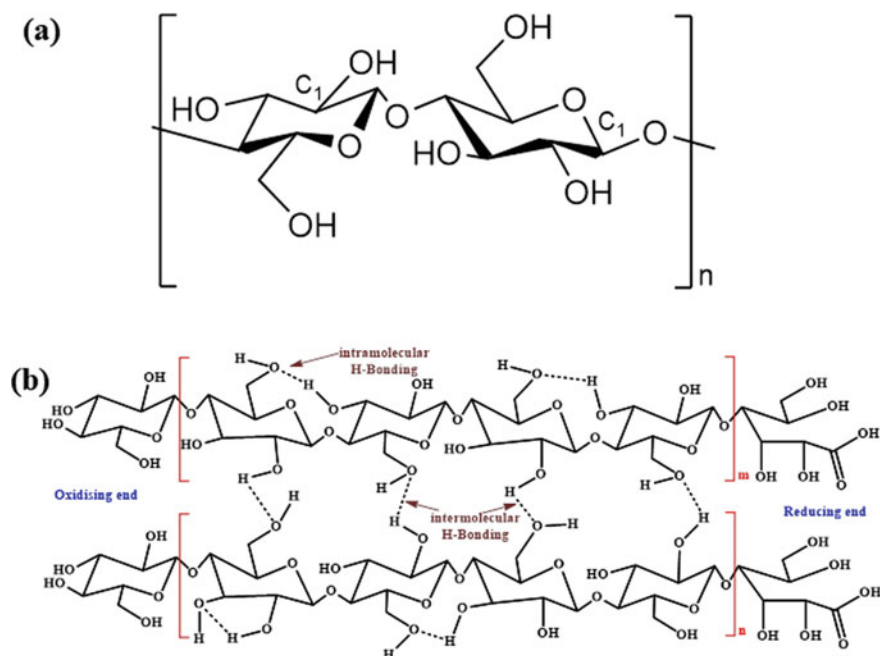
X-ray diffraction, <sup>13</sup>C CP/MAS, and solid-state NMR reveal two crystalline forms of cellulose structure, viz. I <sub>$\alpha$</sub>  and I <sub>$\beta$</sub> . These varieties completely depend upon sources.

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**Fig. 1** **a** Structure of cellulose; **b** Cellulose structure featuring inter and intramolecular H-bonding in the repeating unit and reducing (right) and non-reducing (left) end groups. Redrawn from the Ref. [4]

*Valonia*, bacterial cellulose, and the cell wall of *Glaucozystis* are rich in  $I_{\alpha}$ , whereas tunicate (*Halocynthia roretzi*) or animal cellulose are rich in  $I_{\beta}$  [5, 6]. The OH groups in the cellulose network control the chemical and physical characteristics of the cellulose chains. The intramolecular H-bonds between OH-groups of glucose units of the same cellulose units provide rigidity and thermostability of the chains, whereas, intermolecular H-bonds between two different cellulose chains are responsible for the development of the supramolecular structures as shown in Fig. 1b [4].

In addition to controlling their chemical reactivity, these OH groups can be modified by a variety of chemical reactions like esterification, acetylation, and nitration. Although cellulose is recognized to be the most plentiful and cost-effective biopolymer, its poor water solubility limits its commercial applications [7].

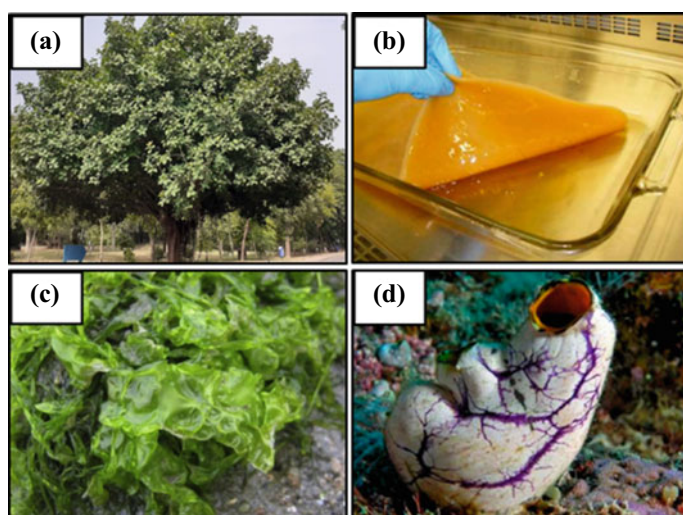
To address this problem, researchers have developed a variety of derivatives, including methylcellulose (MC), hydroxyethyl cellulose (HEC), carboxymethylcellulose (CMC), as well as cellulose acetate (CA) and hydroxypropyl methylcellulose (HPMC). The advantage of functionalization is to make a significant impact on the properties of cellulose by breaking H-bonding [2, 8]. Due to their simplicity of processing, these cellulose derivatives are also employed extensively in the

cosmetic and pharmaceutical industries. Mostly, industries employ them commercially in a variety of forms, including tablet and capsule coating materials, stabilizers, thickening agents, bioadhesives, mucoadhesives, pressure-sensitive adhesives, binders, gelling agents, flavor maskers, fillers, free-flowing agents, and hemostatic compounds [9, 10].

## 2 Sources

French scientist Anselme Payen first extracted cellulose from the cell walls of the plant in 1938 and, later, the chemical structure was determined by Hermann Staudinger in 1920. Finally, synthetic cellulose was first chemically synthesized in the laboratory by Kobayashi and Shoda in 1992 [11].

Wood is the primary resource of cellulose. Others are bacterial cellulose, algal cellulose, and tunicate cellulose [2, 12, 13]. Bacterial cellulose is very pure, highly crystalline, and contains a high DP (degree of polymerization). It has been investigated that, annually a tree can produce  $10^{11}$ – $10^{12}$  tonnes of cellulose during photosynthesis [14]. Figure 2a illustrates the different sources of cellulose.



**Fig. 2** Different sources of cellulose **a** tree, **b** bacterial culture, **c** algae, and **d** tunicate. Reproduced with permission from Courtenay et al. [13] and Hardouin et al. [15]

## 2.1 *Wood and Plant Cellulose*

The primary source of cellulose is wood and plants. The sources vary as corn, cotton, jute, pineapple leaves, flax, hemp, potato peel waste, cereal straws, oil palm biomass, cotton, etc. [2, 16]. The cellulose content in the plant varies with the location. In the soft woody part, it ranges from 30 to 75%, while, in the case of the hard woody part, it is around 40 to 50%. Fibers extracted from both these parts exhibit similarities in composition and structure. They are made of carbohydrates, (such as cellulose, hemicellulose, and lignin) having a complex structure. Hemicelluloses are found in terrestrial plants and algae. Xylan-type polysaccharides are mostly characterized by hemicellulose, whereas lignin is the highly branched 3D complex network having several functional groups. The nature of cellulose highly varies with the amount of lignin, hemicellulose, and fibers.

## 2.2 *Bacterial Cellulose (BC)*

Bacterial cellulose (BC) is often very pure (no lignin or hemicelluloses). It is highly crystalline having a high degree of polymerization. It becomes more significant that numerous bacteria belonging to the genera *Acetobacter*, *Agrobacterium*, *Sarcina*, and *Rhizobium* produce cellulose [17]. Agro-industrial wastes are commonly used as carbon sources to manufacture BC, and the yield of bacterial culture can reach 40%. Most often, BC is acquired in pure form having a purity of more than 90%. BC is capable of being transformed into a variety of morphological forms, including spheres, films, fleeces, and hollow particles. Pure cellulose face masks also frequently have a higher biodegradability than commercial counterparts, which is an additional benefit. A fibrous network of cellulose chains gives them a porous structure, and they are typically wide ribbon-shaped fibrils (diameter ~100 nm), which contain numerous nanofibrils (diameter ~8 nm). As a result, distinctive nanomorphology has a wide surface area, high water holding capacity (~99% water), high wet strength, strong elasticity, and conformability.

## 2.3 *Algal Cellulose*

Another source of cellulose is algae. It is highly crystalline. Algae cellulose is specifically produced from brown species, red species (*Gelidium elegans*), and green species (*Cladophora*). Algal cellulose grows more quickly than plant cellulose, giving it a competitive edge in industrial applications. Oceans, lakes, ponds, and wastewaters are just a few of the environments where algae can develop, and they can produce cellulose [15]. Algal cellulose is not produced in its purest form because hemicellulose, protein, and lignin are highly associated with them. Red algae and green algae

are good sources of cellulose. The main types of carbohydrates found in red algae, or *Gelidium elegans*, are cellulose and agar. Green algal cellulosic cellulose exhibits outstanding qualities such as large specific surface area, high porosity (mesopores), and high crystallinity (~70%) among others.

## 2.4 Tunicate Cellulose

The only known animal source of cellulose is tunicates. They are marine invertebrate animals, and cellulose is extracted from the outer tissue of these animals. It contains a large amount of cellulose (~60%) and a small amount of nitrogenous compound (~27%). In tunicates, cellulose is also present as a nano fibrillar form arranged in a multi-layered structure at the surface of its epidermis. The shape and size of these nanofibril bundles vary with different species. Commonly, the length and width of nanofibrils are in the range of 100 nm–2  $\mu$ m and 10 nm–30 nm, respectively. It also has a high specific surface area (150–170 m<sup>2</sup> g<sup>-1</sup>). The extraction of cellulose from various sources is one of the important processes and most of the commercially used cellulose is primarily extracted from plant-based sources. In the following section, we are going to discuss the extraction process of cellulose from plant-based sources (Table 1).

**Table 1** Various sources of cellulose, cellulose content, properties, and their applications

Various sources	Cellulose content	Properties	Applications	References
Wood and plant	30–95%	Strength and properties of cellulose fibers vary with different varieties of wood and plants	The application also varies with the source, but the major application areas are paper, textile, and pharmaceutical industries	[2]
Bacterial	Approx. 90%	High degree of crystallinity (80–90%), high degree of polymerization, high mechanical strength, and degradation rate of BC is slightly higher than that of wood and plant cellulose	Biomedical applications, tissue engineering, pharmaceutical industry, emulsion and hydrogel stabilizers, drug-delivery systems, and smart artificial skin	[18–20]
Tunicate	Approx. 60%	Highly crystalline, high aspect ratio fibrils, high specific surface area	The excellent properties of tunicate cellulose is a good choice for various chemical and mechanical applications	[21, 22]

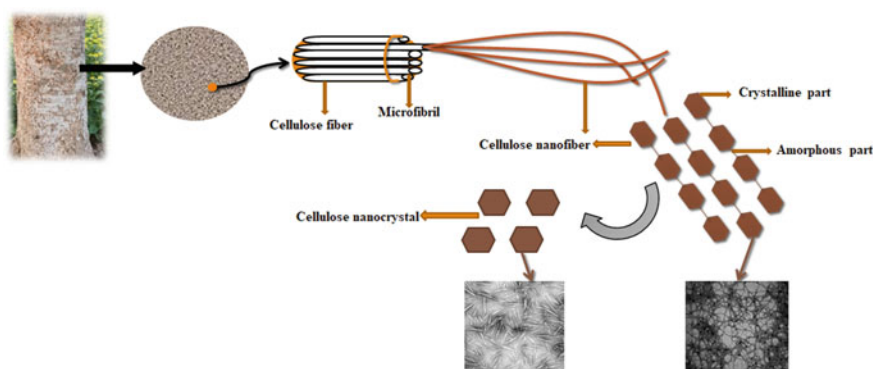
(continued)

**Table 1** (continued)

Various sources	Cellulose content	Properties	Applications	References
Algae	Approx. 70%	High crystallinity (90%), low moisture adsorption capacity, and high porosity in the mesoporous range	Algal nanocellulose has excellent potential for biomedical applications such as tissue engineering because of its nontoxicity, and facile chemical modification	[23–25]

### 3 Extraction of Cellulose

Different types of structural polymers like polysaccharides and polyphenolic compounds make up the constituent of a typical plant. The cell wall of a plant comprises of cellulose, pectin, and hemicellulose. Cellulose is present in the form of fibrillar and is the major part of the biomass [16]. Conventionally, cellulose is isolated from plants starting with the pulping process. This process removes the extractable materials like lignin, and hemicellulose without degrading the fibrillar structure of cellulose. Bleaching is carried out as the next process which utilizes oxygen, ozone, and hydrogen peroxide. The final product after bleaching mainly contains alpha-cellulose and hemicelluloses in residual amounts. This process extracts 40% cellulose, 10–11% as secondary products like furfural, xylose, and acetic acid, and the remaining as waste material [26]. Salimi et al. elaborate on the stepwise extraction of cellulose [27]. In addition to fibrillated cellulose, cellulose nanoparticles can also be produced and a scheme to obtain the same is shown in Fig. 3, a detailed discussion of which is provided in the next section [28,29]

**Fig. 3** Scheme of extraction process of cellulose. Modified from Gopakumar et al. [28]

### **3.1 Cellulose Particles**

Based on the size and morphology, the cellulose particles have been broadly characterized into cellulose crystals and cellulose fibers. These differ in size, shape, crystallinity, aspect ratio, and physiochemical properties [2, 15, 20].

### **3.2 Cellulose Fibers**

Naturally found cellulose is present in the form of microfibrils which assemble and organize themselves in the form of cellulosic fibers. Three types of cellulose fibers are known viz. pulp fibers whose length is found in the range of 1–10 mm, staple fibers with a length of approximately 60 mm, and strand fibers with a length range of 20–100 cm. Strand fibers contain multiple cells, whereas staple fibers contain a single cell. Cotton fibers are an example of staple fibers with a length range of 25–45 mm. These two types of fibers are obtained from wild plants and crops via a pulping process. The fibers obtained are further disintegrated into micro and nanofibrils by a mechanical process that includes homogenization and microfluidization. The length of cellulose nanofibrils obtained is generally 1  $\mu\text{m}$  and the width ranges from 2 to 100 nm. The fiber size is mainly depending on the pretreatment, fibrillation process, and source. Cellulose nanofibrils have superior mechanical strength with a tensile strength of  $\sim 1$  GPa and an elastic modulus range of  $\sim 14$ – $36$  GPa [30].

### **3.3 Cellulose Crystals**

Enzymatic, mechanical, and chemical treatments of cellulose microfibrils lead to the formation of cellulose crystals. During treatment, the cellulose microfibrils get fragmented into microcrystalline cellulose and cellulose nanocrystals. Microcrystals have a high degree of crystallinity and form rod-like stiff particles. Microcrystalline cellulose (MCC) was first introduced as an ingredient for direct tableting. The common source for pharmaceutical MCC is wood cellulose. Cellulose nanocrystals (CNCs) particles are harnessed from the cellulose microfibrils' crystalline regions. CNCs are synthesized via sulfuric hydrolysis of cellulose, which leads to the disintegration of highly crystalline CNCs particles which are eventually extracted. The final characteristic of CNCs depends on the cellulose source and extraction parameters like temperature of hydrolysis, controlled time, and further modification like dialysis and neutralization. CNCs display exceptional mechanical and thermal properties like high TS, modulus, low density, and high thermal properties [31]. Due to the superior properties of CNCs, various potential applications have been explored [32].

## 4 Properties of Cellulose

### 4.1 Solubility

As we have seen that plants contain hemicellulose and cellulose known as holocellulose, lignin, and inorganic materials (such as ash). Cellulose can be simply hydrolyzed by acids to form water-soluble sugars and is resistant to strong alkalis. Cellulose is proportionally resistant to oxidizing agents. Hemicellulose forms a supportive matrix for the cellulose microfibrils which are hydrophilic in nature. This hydrophilicity helps its easy dissolution in alkali and easy hydrolysis in acids. The harder part (lignin) does not get hydrolyzed in acids but is soluble in hot alkali, gets condensed in the presence of phenol, and quickly oxidized. A mixture of dimethyl sulfoxide (DMSO) and 10–20% (w/v) tetra butyl ammonium fluoride trihydrate can dissolve cellulose without any pretreatment at room temperature. Cellulose is insoluble in water and other organic solvents which limits its applications. The OH groups present in the cellulose form H bonding restricting the entry of solvent molecules [33]. This is the main reason for the insolubility of cellulose. It can only be dissolved when this interaction is broken. This can be induced by the addition of functional groups in the cellulose backbone which break the intermolecular H-bonding and in turn solvate the chains. Due to the compact structure of cellulose, a complex solvent system that required minimum energy of dissolution is used in dissolving cellulose. Initially, Copper complexes (Cuoxam) were used to dissolve the cellulose [34]. However, other metal complexes based on Co, Ni, Cd, and Zn are also being used for dissolving cellulose. Thus cellulose can be easily dissolved in phosphoric acid and trifluoroacetic acids by disrupting the Hydrogen bonds. Various ionic liquids are found to be suitable agents for dissolving cellulose since the solvents are eco-friendly, less toxic, having good thermal stability and recyclability. However, the high cost and energy-intensive nature of ionic liquids limit their usage.

### 4.2 Mechanical Property

Cellulose shows high crystallinity and high intermolecular interaction which manifests itself in the form of superior mechanical properties, viz. tensile strength and modulus. The rigid cellulose nanocomposites have a modulus similar to that of Kevlar and Steel. Bacterial cellulose (BC) is softer and finds application in the replacement of collagen networks. Looking at the impressive mechanical properties of cellulose leads to potential applications in wound healing, tissue engineering, and drug delivery where a strong and stable building block is required [35].



### 4.3 *Hygroscopic Property*

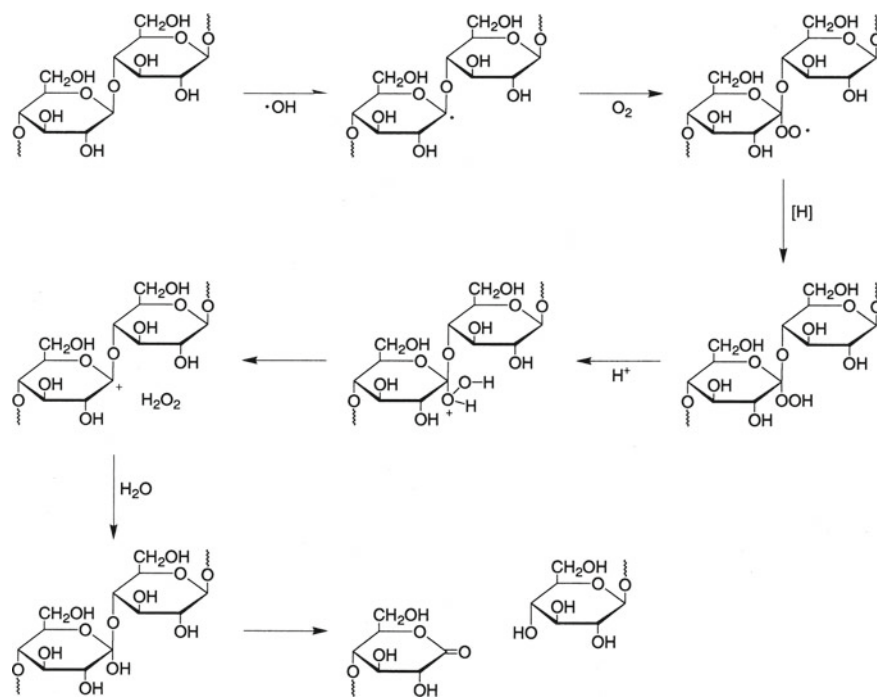
Cellulose is highly hygroscopic and attracts water, this is by the virtue of hydrogen bonds present in the structure. Cellulose absorbs water almost 30% of its biomass and gets swollen, the water permeates into the structure via the amorphous region (disordered domains) [36, 37]. This swelling of cellulose assists in the aqueous processing of cellulose and its composites. The hygroscopic nature of cellulose leads to lower wet strength and which might pose limiting parameters for various applications.

### 4.4 *Structure and Degradability*

The backbone of cellulose is structured on  $\beta$ -1,4-glycoside-linked glucose units. The rigid structure of cellulose can be attributed to the intermolecular hydrogen bonds leading it to good mechanical properties making it useful as a building material in plants. These superstructures produced are stable till significantly a higher temperature. The degradation of cellulose happens below the theoretical melting point of 260–270 °C. The melting point can be reduced by breaking the H-bonding in the structure, this can be effectively done by derivatizing the hydroxyl group of cellulose. Some of the most commonly used cellulose derivatives which are used industrially are cellulose acetate, cellulose acetobutyrate, ethyl cellulose, and benzyl cellulose. Thermoplasticity in these derivatives is achieved by complete derivatization of hydroxyl group. The average degree of substitution (DS) value is the number of substituted OH groups per anhydroglucose unit. The substitution level is inversely proportional to the biodegradability of cellulose derivatives as enzyme attack requires a free glucose unit [38].

### 4.5 *Mechanism of Degradation*

One of the well-recognized degradation routes for cellulose is by the brown rot fungi which produce  $H_2O_2$ , which in turn assists in the production of hydroxyl radical ( $\cdot OH$ ). The attack by  $\cdot OH$  leads to the cleavage of the cellulose chains generating lactone as shown in Fig. 4. The  $\cdot OH$  radical is produced via the Fenton reaction ( $H_2O_2 + Fe^{2+} \rightarrow H_2O + Fe^{3+} + OH$ ) in which iron is used from the wood itself. The hydrogen abstraction by  $\cdot OH$  is a fast reaction that leaves behind a carbon-centered radical which rapidly reacts with oxygen from the environment, which subsequently eliminates  $\cdot OOH$  [39].



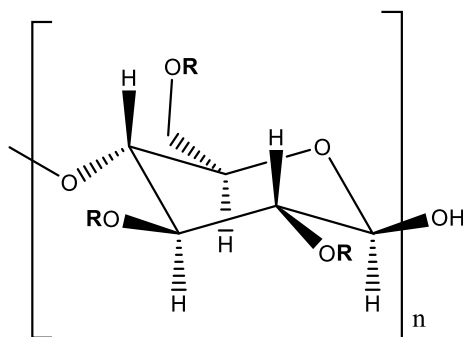
**Fig. 4** Plausible mechanism of degradation of cellulose by Brown rot fungi, reproduced with permission from Hammel et al. [39]

## 5 Functional Derivatives of Cellulose—Synthesis, and Biodegradability

As we have depicted in the previous section that crystallinity and hydrogen bonding in the cellulose structure leads to difficulty in the processability of cellulose; hence, it is essential to functionalize the cellulose. Esterification, etherification, nucleophilic substitution, oxidation, and copolymerization are the most common routes to achieve functionalization in cellulose. Functionalization has a two-fold advantage, viz. feasible processability, and better plastic properties. The functionalization helps in breaking the crystalline structure and assists in solubilization which further assists in processing. The amount of substitution has an impact on biodegradability. As the size and degree of substituent groups increase, the biodegradability of the cellulose decreases [40].

The OH groups in cellulose being susceptible to reactions, is utilized for the functionalization to undergo various kinds of reactions. Figure 5 shows the chemical structure of cellulose after functionalization. The degree of substitution affects the final properties of the polymer obtained. To design a new functionalized cellulose-based polymer, it is essential to understand the changes in properties achieved with

**Fig. 5** Cellulose derivatives, redrawn from [2]



the substitution. The various cellulose derivatives are discussed in detail below (Table 2).

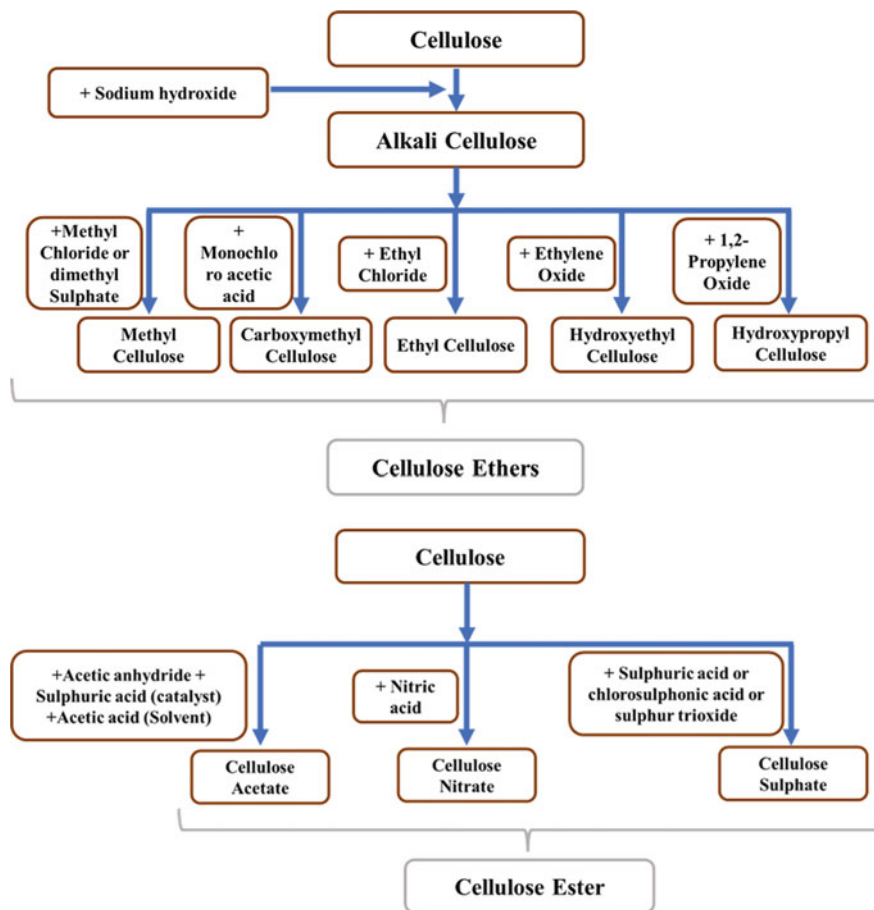
### 5.1 Cellulose Ethers

When OH in the anhydroglucose unit of cellulose is substituted with the alkyl group, cellulose ethers are obtained. Cellulose ethers are synthesized by the reaction of cellulose with NaOH and subsequently reacting with alkyl chloride as shown in the scheme below (Fig. 6). Cellulose ethers consist of methylcellulose, ethyl cellulose, hydroxyethyl cellulose, hydroxypropyl cellulose, and carboxymethylcellulose and their derivatives.

#### Methylcellulose (MC)

Methylcellulose is the simplest functionalized biodegradable cellulose ether. It has a vast application in the food industry, tissue engineering, coating, and preparation of mulch films. It comes with the advantage of easy processability, large availability, and low cost. The biodegradability however limits its application in high-end products. Hence, two methods which helps to modulate the biodegradability of the polymers with required properties are the incorporation of fillers with a high aspect ratio to increase the tortuous path, and secondly by incorporation of crosslinks such as glutaraldehyde (Glu) between the polymer chains. This will delay the hydrolysis and provide an enhanced property with the loss in biodegradability [41].

Methylcellulose is synthesized in an alkaline medium in the presence of methylating agents like methyl chloride or dimethyl sulfate [42]. Degree of substitution depends on the synthesis conditions such as reaction time and methylating agent. As the degree of substitution is increased from the range 1.4–2.0 to 2.4–2.8, the solubility of methylcellulose is enhanced in water as well as in some organic solvents [43].



**Fig. 6** The above scheme represents the summary of the synthesis of cellulose derivatives viz. esters and ether, redrawn from [2]

### Carboxymethylcellulose (CMC)

CMC is one of the most popular and commercially available cellulose ethers. It is biodegradable, non-toxic, water-soluble, and has better chemical stability. CMC has myriad applications by the virtue of its properties in the food industry, textile industry, paper industry, drugs and cosmetics, leather, films, filaments, paints, and lacquers [44]. CMC is an efficacious thickening agent, binder, and emulsion stabilizer, and has film formability. CMC is synthesized by a reaction of cellulose with monochloroacetic acid in which the hydroxyl group (predominantly C<sub>2</sub>) is substituted by carboxymethyl groups (–CH<sub>2</sub>–COOH).

The degree of substitution is typically between 0.6 and 1.25. The properties are dependent on the large molecular structure, degree of substitution, and molecular

**Table 2** Derivatives of cellulose

Functionalization	Derivatives	R
Cellulose ethers	Methylcellulose	H or CH <sub>3</sub>
	Carboxymethylcellulose	H or CH <sub>2</sub> COOH
	Ethyl cellulose	H or CH <sub>2</sub> CH <sub>3</sub>
	Hydroxyethyl cellulose	H or CH <sub>2</sub> CH <sub>2</sub> OH
	Hydroxypropyl cellulose	H or CH <sub>2</sub> CH(OH)CH <sub>3</sub>
Cellulose esters	Cellulose acetate	H or (C=O)CH <sub>3</sub>
	Cellulose sulfate	H or SO <sub>3</sub> H
	Cellulose nitrate	H or NO <sub>2</sub>

weight [45]. The sodium salt is soluble in water and hence CMC is usually used as its sodium salt.

### Ethyl Cellulose (EC)

Ethyl cellulose is another commercial biodegradable cellulose ether that is formed when the hydroxyl group is substituted by the ethyl group. Ethyl cellulose is synthesized by the reaction of cellulose alkali with ethyl chloride at 60 °C for several hours [46]. For commercial products, normally the degree of substitution lies in the range of 2–2.6 [47]. The properties of ethyl cellulose depend on the molecular weight, degree of etherification, and molecular uniformity.

EC has an excellent film-forming capacity with superior barrier properties and hydrophobicity making it a suitable candidate for biomedical applications, majorly in drug delivery. Ethyl cellulose is generally brittle and hence plasticizers are added in order to enhance its flexibility, thermal stability, and processability [48].

### Hydroxyethyl Cellulose (HEC)

Hydroxyethyl cellulose is synthesized by reacting alkali cellulose with ethylene dioxide to achieve hydroxy ethyl group (–CH<sub>2</sub>–CH<sub>2</sub>–OH) at 2, 4 and 6 positions of glycosyl unit of cellulose. This OH group acts as a reactive center which can be utilized for further modification. Hydroxyethyl cellulose is highly soluble in water and other organic solvents. Mixing cellulose with HEC allows easy processing of it, and diversifies its biomedical application [49].

### **Hydroxypropyl Cellulose (HPC)**

Hydroxypropyl cellulose is a biodegradable polymer in which the OH group is substituted by 2-hydroxypropyl. Like HEC, HPC also has a secondary alcohol group that can be utilized for further functionalization of the cellulose chains for biomedical applications such as tissue engineering. HPC forms a liquid crystal when dissolved in water depicting an interesting property of mechanochromism meaning it shows a change in color on the application of pressure. This property makes it a suitable candidate for application in sensing applications [50].

## **5.2 Cellulose Esters**

Cellulose esters offer the advantage of solubility in a common solvent and easy melt processability over cellulose. It also affords cellulose to be molded into 3D shapes, drawn into thin wires, and solution cast into films, sheets, or coating applications [51]. The modification can also be done over the surface of the cellulose substrate keeping the crystallinity intact to avail high mechanical properties [52]. Common cellulose esters are discussed below.

### **Cellulose Acetate (CA)**

Cellulose acetate is synthesized from the acetylation of cellulose. The DS defines the properties of the CA. DS of 2.5 is the most common level that provides optimum molecular weight and rheological/solution properties. These properties help CA to be used in various applications like textiles, thermoplastic, films, and cigarette filters [53].

The key mechanism of biodegradation of CA is chemical hydrolysis and acetyl esterases as the first step which subsequently leads to the degradation of backbone which mainly contains cellulose. Various studies have confirmed that CA is indeed biodegradable in natural environment. One interesting study is done by Komarek et al. in which acetyl carbon was labeled with  $^{14}\text{C}$  and  $\text{CO}_2$  evolution was monitored. The study compared CA with DS of 1.85, 2.07, and 2.57, and found a reduction in degradation rate by increasing the level of acetylation [54].

### **Cellulose Nitrate (CN)**

Cellulose nitrate also known as nitrocellulose, is synthesized by the substitution of an OH group with a nitrate group by treating cellulose with concentrated nitric acid. The typical DS is 2.2–2.8 which eventually decides its properties and application. A major application of CN is in explosives, plastics, coating, and ink industries [55].

CN is found to be 40% biodegradable only due to inhibition by various products formed during the degradation of main degrading enzymes [56].

### **Cellulose Sulfate (CS)**

Sulfonation of cellulose is carried out using sulfuric acid, sulfur trioxide, and chlorosulfonic acid. It is water soluble, antiviral, antibacterial, and has anticoagulant properties due to the presence of a sulfate group. CS shows excellent biocompatibility, film-forming ability, and biodegradability. These properties make it highly viable for application in tissue engineering and drug delivery [57]. In vitro studies showed favorable biodegradable properties of sodium cellulose sulfate (NaCS) and chitosan-based films. The DS also has an inverse effect on the biodegradability of CS [58].

## **6 Applications**

### **6.1 Packaging Industry**

Plastics are commercial materials derived mainly from synthetic polymers. These materials are cheap and remain intact throughout their packaging life. But synthetic polymer leads to some side effects on human beings, animals, and the environment. The usage of plastic bags cannot be reduced as they are easy to handle. That's why to get rid of the sustainability issues, it is important to introduce biodegradable polymers to reduce the solid waste in the environment. Among the well-known biopolymers, cellulose has been extensively used in the food packaging industry [59].

In order to convert cellulose into a polymeric substance, first cellulose has to be extracted from natural resources. The best methods to extract cellulose were described by Han and Rowell and later by Borella et al. [52, 53, 60]. Recently, partially biodegradable biocomposites and green biocomposites gained much attention in synthesizing biobased products, which can be degraded easily by microorganisms [3, 52, 53]. Biocomposites consist of biofiber and matrix polymer. Partially biodegradable biocomposites contain biodegradable biofiber and non-biodegradable polymer matrix whereas, biodegradable biocomposites consist of both biodegradable fiber and polymer. Also, there exist hybrid biocomposites which contain two or more biofiber in combination with a polymer [61]. In recent days, biodegradable polymers blended with inert polymer matrices are becoming more attractive. In this case, upon disposal of the plastic in the environment, the biodegradable part can be degraded by microorganisms and the residual polymer component loses its integrity and fades away. A cellulose ester known as cellulose acetate butyrate (CAB) can be added to other cellulose ester-based film formers as well as employed as a reactive polyol during curing of the coating. The regenerated cellulose fiber, i.e., cellophane is used as breathable packaging material for baked food. Apart from that, cellulose

esters and cellulose ethers are being used in the processing of film coatings, edible films, etc. [62].

## 6.2 *Pharmaceutical Industry*

Cellulose-based polymers are gaining much importance in the field of pharmaceuticals. Especially, cellulose ethers and cellulose esters are having unique physico-chemical and mechanochemical properties by which they can be used broadly as healthcare products in the form of tablets, gelling agents, bioadhesives, mucoadhesives, etc. Another well-known derivative of cellulose MCC is a multifunctional excipient such as silicified MCC (SMCC) which is being extensively used instead of only MCC. Recently, cellulose is converted to nanocomposites which are also used as drug-delivery agents and as ion-sensing materials [63–65].

## 6.3 *Electronic Industry*

In order to meet the sustainability issues, it is necessary to utilize renewable energy sources. Thus biodegradable materials such as cellulose derivatives are highly used in the field of the electro-technological field for their lightweight, low cost, transparency, portability, and flexibility in electronic and solar energy conversion [66, 67]. Cellulose fibers having a diameter of  $\sim 20 \mu\text{m}$ , would have been suitable for usage in solar cells, but their high surface roughness and porous structure render their effectiveness in coatings. Also, the fiber diameter is much larger than the wavelength of visible light. So, traditional papers made of cellulose are opaque which is difficult for using the materials for transparent devices. Chang et al. showed that *O*-(2,3-dihydroxypropyl) cellulose (DHPC) produces a smooth surface with excellent ductility and transparency and can be effectively used in a flexible solar cell. However, due to poor mechanical strength, this derivative of cellulose is further reinforced by rigid tunicate cellulose nanocrystals (TCNCs) [68].

## 6.4 *Metal Industry*

Water bodies are playing a major role as a carrier of heavy metals, non-metals, and organic dyes. Cellulose backbone contains a huge amount of OH groups which can interact with metal ions through electrostatic force and hydrogen bonding. Besides, these OH groups can be converted into several other functional groups (phosphonate, sulfonate, amine, carboxyl, ether, ester, etc.) which change the surface charge over the cellulose matrix. These modifications provide excellent interaction with metal ions. Hajeeth et al. employed cellulose extracted from sisal fiber in the form of a



cellulose-g-acrylic acid copolymer to adsorb Ni(II) and Cu(II) from aqueous solutions [69]. Cellulose was also successfully modified to produce cellulose-amine and cellulose-thio adsorbents. The obtained sample showed strong adsorption potential towards Hg(II) in aqueous solutions [59]. Cellulose can also be used as an adsorbent when combined with montmorillonite (NaMMT). It was applied to eliminate Cr(VI) from the aqueous solutions. Other modified cellulose were listed in Table 3.

## 6.5 Biomedical Industry

Cellulose contains abundant OH groups over its backbone. Hence, cellulose hydrogel can be easily produced by physical crosslinking [91]. There are some water-soluble cellulose derivatives, such as Methylcellulose, Hydroxypropyl Cellulose, Hydroxypropyl Methylcellulose, and Carboxymethylcellulose that are often used for the fabrication of cellulose-hydrogels. It was shown that, when OH moieties are partially substituted by methyl or hydroxypropyl groups, both hydrogen bonding and hydrophobic interaction play a key role in determining the gelation ability of the hydrogels [92]. In the case of chemically crosslinking structures, some di-functional reagents are employed to provide a three-dimensional shape to the cellulose hydrogel structure. Sannino et al. prepared super-hydro porous hydrogels based on crosslinking of CMC and HEC with a crosslinker such as DVS (divinyl sulfone) (Fig. 7). The main feature of these hydrogels is that they can change their sorption capability depending on the ionic strength and pH of the excipient. The super-adsorbent hydrogel has been efficiently applied for the treatment of edemas [93–95]. Apart from medication, cellulose-based hydrogels are often used in tissue engineering, sensors, agriculture, water purification, and chromatographic support which are discussed below [96–99].

In the biomedical field, hydrogels have never-ending applications. It has the ability to remove excess water from the body and is highly effective in the treatment of edema. Polyelectrolyte-based cellulosic hydrogels, such as sodium salt of carboxymethylcellulose and hydroxyethyl Cellulose are highly pH-dependent and get removed from the body with feces without affecting the function of other body parts. The water uptake capability varies with the changing ratio of NaCMC/HEC [101]. Besides, superabsorbent cellulose-based hydrogels are being extensively used in personal care products like diapers as these hydrogels absorb liquid and keep that

**Table 3** Several cellulose derivatives and their applications

Purpose	Cellulose components	Applications	References
Metal adsorption	Phosphorylated cellulose (nano paper)	Copper ions adsorption	[70]
	Tetraethoxysilane-cellulose acetate composite material	Cr(VI) adsorption	[71]

(continued)

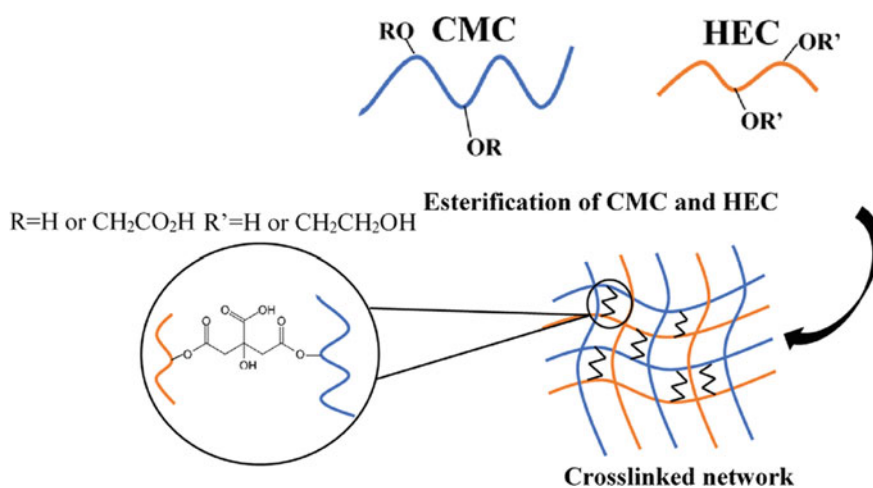
**Table 3** (continued)

Purpose	Cellulose components	Applications	References
	Fe(III) metal based aminofunctionalized poly(glycidylmethacrylate)-grafted TiO <sub>2</sub> -densified cellulose	As(V) adsorption	[72]
	Sulfonated wheat pulp nanocellulose	Pb(II) adsorption	[73]
	Amino-propyl-triethoxy silane modified micro fibrillated cellulose	Cu(II), Ni(II), Cd(II) adsorption	[74]
	Cellulose functionalized methyl-iodide and triethylenetetramine	Cr(VI) adsorption	[75]
	Cellulose nanofiber	Ag <sup>+</sup> adsorption	[76]
Packaging	Cellulose esters	Laminates, optical films, and laminates	[77]
	Cellulose acetate butyrate, and methylcellulose	Coatings and additives in film former	[78]
	Carboxymethylcellulose	Thickener, stabilizer, and suspension agents	[79]
	Cellophane	Breathable packaging for baked goods and textiles	[80]
	Methylcellulose with polyethylene glycol, and PEG400 plasticizers	Edible film	[62]
	Nanocellulose	Food packaging	[81]
Hydrogels	Si impregnated hydroxypropyl methylcellulose (Si-HPMC)	Biomedical applications	[82]
	Chitosan/methylcellulose/Na <sub>3</sub> PO <sub>4</sub> hydrogels	Tissue engineering	[83]
	Poly(N,N-dimethyl acrylamide)/cellulose hydrogels	Optical applications	[84]
Smart material	Cellulose electro-active paper (EAP)	Biosensors	[85]
	<i>O</i> -(2, 3-dihydroxypropyl) cellulose (DHPC) with tunicate (TCNCs) cellulose nanocrystals as reinforcing agents	Flexible solar cells	[68]
	Sodium cellulose sulfate	Biomaterials for microcarriers' designing	[58]

(continued)

**Table 3** (continued)

Purpose	Cellulose components	Applications	References
	Nitrocellulose	Explosives	[86]
Biofuel	Cellulose	Energy production	[87]
Others	Oxycellulose	Bioadhesives and cosmetics	[88]
	Hydroxyethyl cellulose	Agriculture, textile, and paper industries	[89]
	Hydroxypropyl cellulose	Lubricant in artificial eyes and food additives	[90]

**Fig. 7** Scheme of formation of HEC and CMC crosslinked network, redrawn with permission from Ayouch et al. [100]

place dry, and also prevent diaper rash and any other fungal attack. These products are also cheap and safe to use. Furthermore, these superabsorbents prevent leakage and reduce the risk of fecal contamination, and gastrointestinal problems [102, 103]. In 1966, Harmon and Herper separately patented superabsorbent materials [104].

The superabsorbent cellulosic system contains three main parts (1) an envelope of non-oven tissue, (2) a plastic cover material, and (3) an absorbent made of wood pulp cellulose mixed with some hydroporous superabsorbent polymer (SAP). The latter two components were usually non-biodegradable. Hence, for the recovery of cellulose materials and recycling of the absorbent, usage of cellulose-based hydrogels was suggested. Among them, NaCMC and HEC crosslinked with DVS show similar properties to SAP and produce higher retention ability and swelling ratio [105].

## 6.6 *Agricultural Industry*

In order to optimize the water resources in the agricultural field, especially for the cultivation in the desert area, a constant water supply is highly needed. So, the super-absorbent provides the required quantity of water throughout the time by releasing the water slowly through a diffusion-drive mechanism [106]. Cellulose-based hydrogels perform as an alternative to acrylate-based hydrogel which is not biodegradable. Some cellulose-based SAP hydrogels were suggested which have the absorption ability of almost one liter of water per gram of hydrogel [107, 108].

## 6.7 *Biofuel Industry*

In recent days biofuel supplies 86% of the energy of the world. With the increment in population, energy usage is also expected to surge in the coming days. High usage of petroleum-based fuels leads to an impact on environmental changes like global warming and the reduction of biomass. In that case, cellulose is the most active renewable resource used in the production of energy in the world (chemical energy stored in biomass is approximately 6–7 times that of total human energy consumption per year) [109]. Moreover, a human can not digest cellulose, so it is advantageous to use this polymer as it does not compete with food resources. Production of biofuel from cellulosic materials is generally carried out by the processes of gasification, pyrolysis, and hydrolysis. First, the woody biomass is disintegrated to produce isolated cellulose from other constituents of lignocellulosic materials. Next, cellulose is depolymerized (basically converted to glucose units), followed by conversion of glucose into biofuels via biological treatment, and finally purification of the biofuel. In the meantime, it is necessary to deoxygenate the biomass as it reduces the heat content thereby lessening the chance of blending with other fossil fuels [110]. The detailed procedure for the formation of biofuel was studied by Fatehi [87].

## 6.8 *Paper Industry*

The major ingredient of paper is cellulose fibers. The quality of paper highly depends on the size and quality of cellulose fiber. Higher the cellulose content in the paper pulp, the better the quality of the paper. In earlier times, the paper used to be made up of bamboo fiber, and silk. Nowadays, it has been replaced by cellulose-based, plant fiber, which has excellent writable properties and is also cheap. Cellulose-based papers are highly porous, rough, and hydrophilic. Three kinds of pulp were studied such as chemical pulp, semi-chemical pulp, and mechanical pulp during paper production. Industrially, the chemical pulp is being extensively used as paper produced from this becomes strong, stable, and easy to bleach, as compared to other pulps [111].

Recently, nano-fibrillated cellulose has become an industrially promising material for the formation of papers. It acts as a reinforcing material on the fiber surface. Also, the nano-fibrillated cellulose plays the role of filler and is incorporated in the voids and pores between the fibers [112].

## 7 Conclusion

Cellulose being a ubiquitous and easily biodegradable polymer has an immense potential to replace petroleum-based non-degradable polymers. Cellulose possesses fascinating mechanical and thermal properties making it comparable with that of conventional polymers. Cellulose is majorly extracted from the plant via multiple stages of chemical treatment. Other sources include bacteria and marine invertebrate animals called tunicates. Hydrogen bonding present in the Cellulose makes them crystalline, thereby enhancing their mechanical property, but reducing solubility in various solvents, thus affects its solution processability. To enhance the processability of cellulose and to provide better plastic properties, it has been functionalized via various routes viz. etherification and esterification. The free OH group in cellulose assists in their functionalization to produce cellulose derivatives. For example, CMC and EC are some of the commercially available Cellulose ether derivatives used in myriads of application. Cellulose acetate, cellulose nitrate and cellulose sulfate are some of the common cellulose esters discussed in this chapter. By a virtue of their property, cellulose has a great potential to be used in various areas, viz. packaging, pharmaceutical, electronic, biomedical, biofuel, and paper industry. With the development of nanotechnology, cellulose–inorganic hybrid hydrogels are found to have multifunctional applications. Besides, microbial cellulose has become popular in the generation of biomedical devices such as in wound healing, organ replacement etc. Cellulose has become the most studied biopolymer that can be easily processed and industrialized. Therefore, the development of cellulose derivatives along with other polymers as well as inorganic fillers has the potential to show extensive application in our daily life. Nonetheless, they suffer from challenges such as source variation, availability, lack of economically attractive extraction procedures, etc., which require to be overcome in the near future for their widespread applications.

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