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REVIEW

Advances and Applications of Cellulose Bio-Composites in Biodegradable Materials

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Abstract

Cellulose is a natural polymer that has a lot of potentials. Cellulose gained more interest owing to its renewability, non-toxicity, economic value, biodegradability, high mechanical properties, high surface area, and biocompatibility. New sources, new isolation processes, and new treatments are currently under development to satisfy the increasing demand for producing new types of bio-based materials on an industrial scale. This article discusses the fundamentals and latest breakthroughs in cellulose biopolymer materials used in the fabrication of composite films owing to the cellulose forming films. Bio-polymers are finding wide applications due to their intrinsic properties such as low density, low thermal conductivity, corrosion resistance, and ease of manufacturing complex shapes. Cellulose possesses a highly crystallized structure, hence it is insoluble in typical organic solvents. Environmental restrictions are increasingly stringent, which is a key element leading to the growth of studies on this subject. These hydrocolloids have been modified by taking advantage of their valuable features; the mechanical strength and water resistance of cellulose make it being used as a thickener for large-scale applications such as cellulose composite films can extend the shelf life of a product while maintaining its biodegradability. New materials with high values are a hot topic for future research with commercial interest. These composite film potentials are contributing to the bio-economy. Here, the emphasis on the potential application of bio-composites of cellulose in various industries has been discussed.

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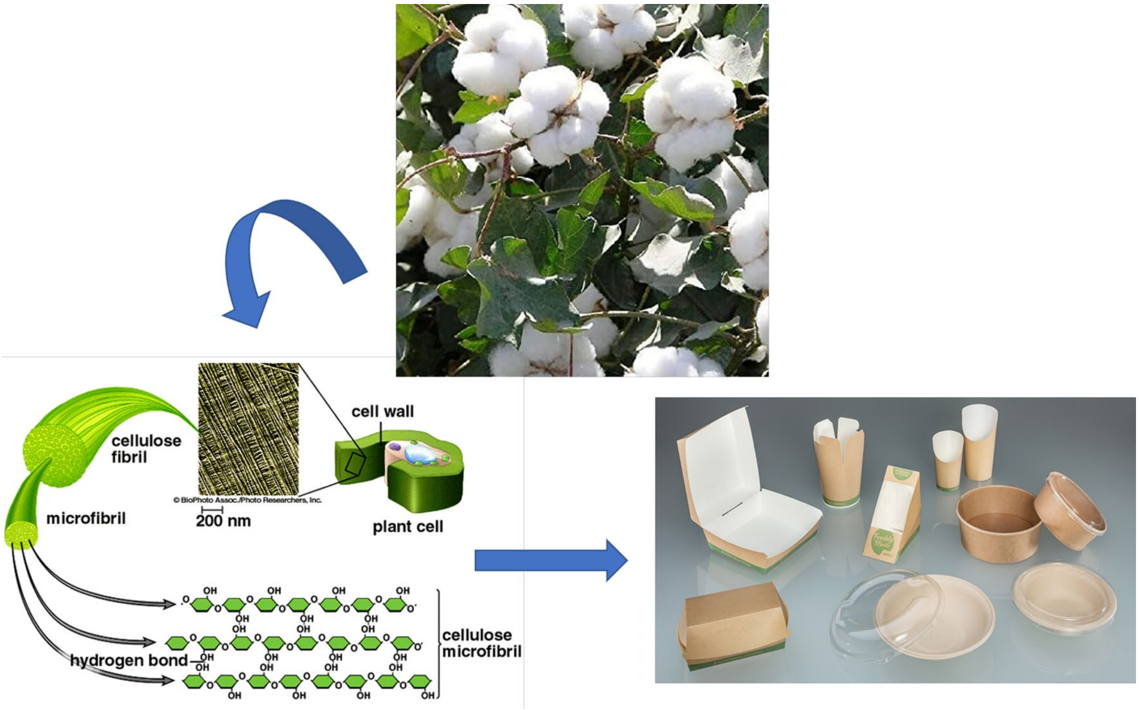
Graphical Abstract



Keywords Composites · Cellulose · Properties · Environmental · Polymer

Introduction

Polymers are generated from renewable (polysaccharides) and edible (proteins) materials. It attracted vast attention, especially cellulose polymer used as a reinforcing agent in polymer matrices of composites and most importantly, they can bio-degrade easier and faster than non-renewable polymers [1–5]. Their bio-degradable potentials, preservation characteristics, cost-effectiveness, bio-compatibility, and environmentally friendly behavior have encouraged their consumption as food packaging materials, films, coatings, and wrappings. The polysaccharide is a promising edible polymer [6–8]. Cellulose and starch are the natural renewable resources of polysaccharides, which are widely used in agricultural products as edible film because they can extend the shelf life of fresh fruits and preserve flavor [9, 10]. Thus, the hydrocolloids have been modified to take advantage of their valuable features. Cellulose is a biopolymer demonstrating mechanical and water-repelling properties and has been evaluated as a biodegradable material [11–13], while inorganic materials pose a great threat to the environment and leave persistent pollutants such as dyes and heavy metals ions in water bodies [14, 15]. Thus, bio-polymer cellulose attracted attention in food, biosensors, and drug applications. [16, 17] It also shares similarities with other polysaccharide materials, which are abundantly available and commercially affordable [18]. In a proper solvent environment, cellulose can produce hydrocolloids, which can be used in the preparation of excellent film-building material. Additionally, cellulose films demonstrated good resistance to water and heat generated by microwave appliances [19, 20]. Mixing cellulose biopolymer with other hydrocolloids has been found to expand its application range [21]. It has been observed that the incorporation of original cellulose and its derivatives into a polymeric matrix contributes to the improvement in film's strength of tensile and stiffness [22–24]. In general, edible films are created from a single type of natural film-forming polymer that adds to the positive and negative characteristics [25, 26]. To improve the properties of edible films, an alternative strategy is combining biopolymers with bio-composite materials [27, 28]. Despite the development of new technologies and organizational measures up to the implementation of relevant laws, environmental pollution attracts severe concern worldwide [29, 30]. Certain attempts have been initiated to address the concerns. The creation of cellulosic materials with long operating life with an ideal covering of the operational unit is working for an entire period [31, 32]. The utilization of such cellulosic materials as a fuel for new industries following their regeneration. In the natural environment, the modified cellulose degrades rapidly [33]. Several technologies have been put into operation under the influence of sunlight, water, air, oxygen, bacteria, and other natural forces and degrade the cellulosic material's relatively innocuous compounds [34]. Nowadays, technological advancement triggers researchers to invent synthetic biopolymers with improved chemical, mechanical, morphological, and barrier properties, which not only overcome the drawbacks of natural polymers but also include other properties that help to enhance food safety, quality, and shelf-life. Moreover, due to environmental sustainability issues, a good hand of research work has Bio-composites reinforced with renewable bioplastic (cellulose plastic) has been seen in the use of bio-composites over the past few decades. There are a great number of achievements in eco-friendly technology in the field of materials science through the preparation of bio-composites like in automotive and decking markets, but the applications of these composites in other sectors have been seen as limited. However, with suitable developmental techniques and utilizing knowledge of science, the potential exists for bio-composites to be used in new markets.

This article emphasizes in detail of cellulose-based edible composite films' fundamental understanding, structures, and characteristics. It also further explains their compatibility with the environment and economic aspects.

Biodegradable Materials

Biodegradable materials are made from renewable resources. They are popular for their unique and fascinating properties: non-toxicity, biocompatibility, and biodegradability. Biodegradable materials decompose over time in natural circumstance without generating dangerous substances. Such materials are ideal alternatives to petroleum-based materials and contribute to environmental protection, for they can cut carbon dioxide emissions and reduce the use of fossil-based raw resources. These materials decomposing faster than traditional materials, are gaining popularity. Bio-surfactants and biopolymers are the most common biodegradable compounds utilized in the sample preparation process [35].

Bio-Surfactants

Bio-surfactants were found as extracellular amphiphilic molecules in hydrocarbon fermentation in the late 1960s. They were considered to be green materials due to their natural source and biodegradability. Bio-surfactants are biological products made of hydrophobic and hydrophilic moieties with varying polarity and a variety of functional groups with unique structural qualities and mostly isolated from microorganisms, fungi, and yeast. They are considered the most important and diverse families of biodegradable materials utilized in various types of applications such as agriculture and industry [36]. These materials demonstrate a strong sorption capacity, biological activity, and great tolerance to severe environments. Glycolipids, fatty acids, and polymeric bio-surfactants are the four types of microbial surfactants classified following their molecular structures [37, 38]. Bio-surfactants are molecules that are either low or high molecular weight materials that can reduce interfacial tension. It also acts as an effective stabilizing agent and attained the potential to be employed in liquid-phase micro-extraction to aid in the extraction of organic molecules from environmental samples [39].

Biodegradable polymer Film

A film of biodegradable polymer is a homogenous layer made of a single or combined biopolymer (polysaccharides, proteins, and lipids) [40]. Polysaccharide is the most promising polymer among edible and degradable polymers, which is affordable, widely available, biocompatible, and ecologically benign [41]. The cellulose, starch, and chitosan are naturally renewable polysaccharides that have been used to make edible films because of their hydro-colloidal properties as shown in Figure 1. In the world of food packaging, natural & biodegradable, and bioavailable polymers are gaining appeal owing to the hard film formed by synthetic material, which is hard to be broken [42, 43]. The current research is concentrated on generating biodegradable polymer films [44]. Microorganisms in the natural environment can disintegrate biodegradable polymers and eventually catabolize them to carbon dioxide and water. Polysaccharide is a natural biodegradable polymer that has been widely studied [45]. Apart from chemical and photochemical degradation mechanisms, the micro-organisms present in air, water, and soil may easily decompose cellulose and its derivatives [46, 47]. Cellulase enzyme can degrade cellulose into glucose, and glucose can be used to prepare bioethanol. The rate of cellulose biodegradation is determined by its crystallinity [48]. Low crystallinity cellulose degrades faster than high crystallinity cellulose. In comparison to the dissolved pulps, micro/nanoscale cellulose, and cellulose derivatives are found less crystalline [49]. Thus, cellulose derivatives have substantially attained better biodegradability than other types of celluloses. Due to its uniform dispersion and hydrophobicity, cellulose has substantially extended the shelf life of films [50].

Biodegradable Cellulosic Polymer as a Reinforcement Component

Cellulose can form films [51, 52] because of the crystalline structure and hydrogen bonding. It cannot be melted or dissolved in water or typical organic solvents. It is unable to form a gel or film in a normal state. Instead, it is usually modified to water-soluble compounds known as cellulose

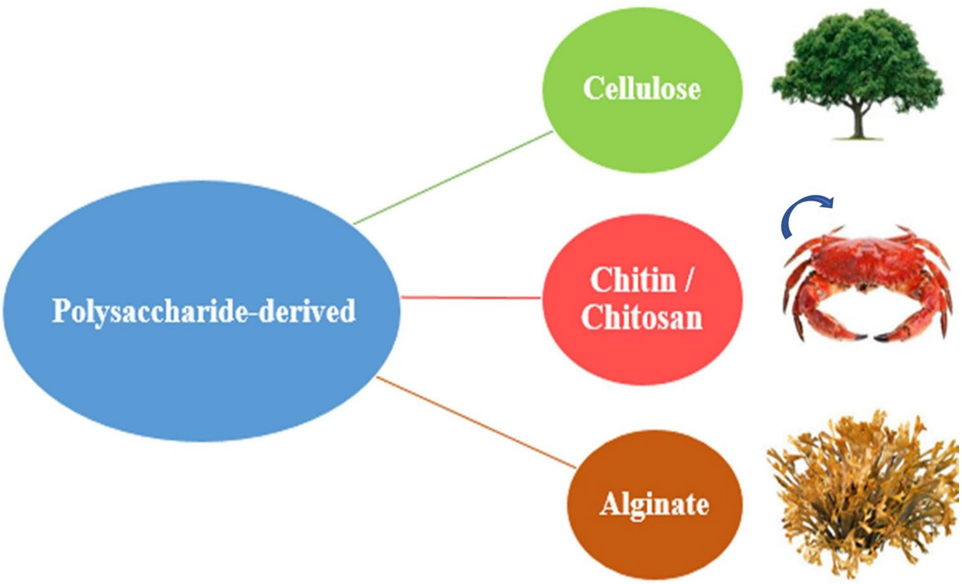


Fig. 1 Cellulose, Chitosan, and Alginate. (Copyright Permission Elsevier-2022)

derivatives [53–56]. Due to the intrinsic hydrophilic nature of polysaccharides, the cellulose derivative films offer strong oxygen and aroma barrier but poor water vapor barrier and low mechanical properties [57]. One strategy is to demonstrate a moisture barrier by incorporating hydrophobic compounds. The fatty acids and essential oils with cellulose derivative matrix make a composite film [58]. It is very difficult to produce a homogeneous composite film containing both hydrophobic and hydrophilic chemicals [59]. A chemical modification of cellulose to water-soluble derivatives via cross-linking with citric acid has been found effective to enhance the moisture barrier characteristics, which excelled the thickness, molecular weight, and mechanical properties [60]. However, as the plasticizer content increases, the mechanical characteristics of fabricated films decreases, except for elongation at break. The latest research has discovered that adding a reinforcing substance such as nanomaterials (i.e. nano-clay/particles) to cellulose-derivative films can effectively improve their strength. Aside from employing the chemically modified cellulose as a film precursor, but its derivatives have sparked interest as reinforcement materials in biodegradable polymers both in academia and industry [61]. Cellulose reinforcement is primarily used to strengthen the polymer matrix structure, but it also creates a new composite material with a variety of unique physical properties and fascinating attributes [62]. When compared to non-biodegradable polymers manufactured from petroleum. The biodegradable polymer films typically have displayed poor mechanical, thermal, and barrier qualities [63, 64]. The inclusion of cellulose derivatives into a degradable polymer film is well known for reducing such limitations and the resulting materials are mixed with desirable functionalities and qualities [65]. Due to the chemical similarities of the hydrocolloids, which create positive interactions to considerably improve the film values. Biodegradable composite films are made by utilizing a mixture of hydrocolloids widely explored [66, 67]. Previous research has also demonstrated that cellulose combined with other polysaccharides can make a uniform bio-composite film [68, 69]. Recently, cellulosic materials have been employed as reinforcing elements in polysaccharide-based films [70]. Due to its excellent mechanical and water resistance, the micro/nanoscale cellulose has sparked a lot of interest in its use as a bio-composite material reinforcement [71, 72].

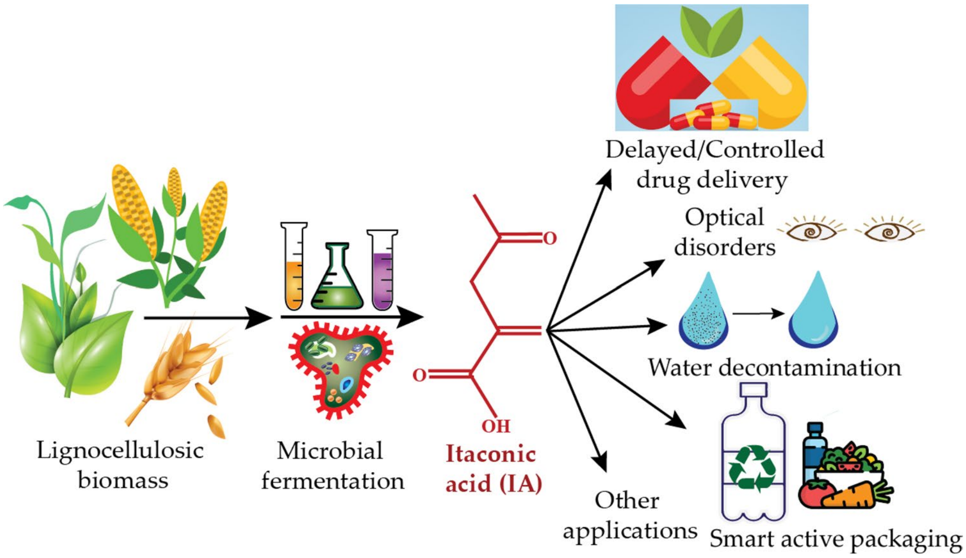
Polymers From Biomass

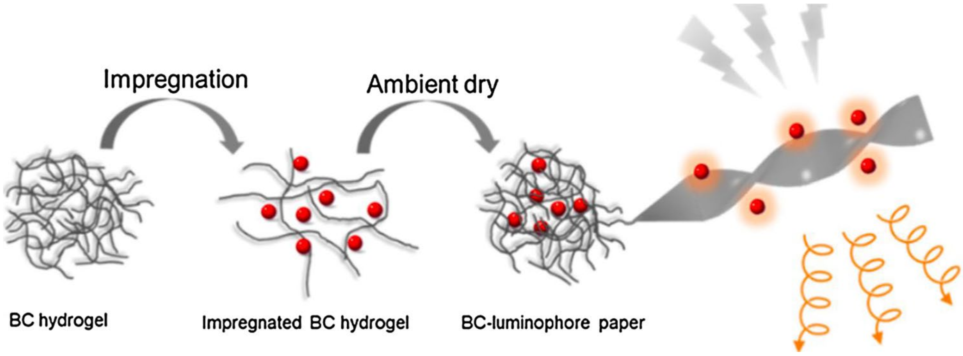
Polymers are most typically found in marine and agricultural environments, such as polysaccharides, cellulose, protein, and lipids. These are the basic materials being used to make polysaccharide films. Starch application in the synthesis of bio-plastic is a main focus. Cellulose is a plant-based biopolymer integrated by lysozyme into cellulose acetate (CA) films. The CA films are used in antimicrobial packaging materials. The film with the maximum release rate and antibacterial activity was made of 5% CA solution with 1.5% lysozyme. The increase in CA content has lowered the porosity of the film. It also lowered the release rate with the maximum release of lysozyme activities. The tensile strength of the films and the immobilized lysozyme activity were both boosted. The addition of lysozyme did not reduce the tensile strength of the films except for 15% CA-containing films. Asymmetric CA films have been found to possess a higher potential for achieving controlled release in antimicrobial packaging [73, 74]. Using N, N-dimethylacetamide/lithium chloride as a common solvent was effective in the fabrication of all-cellulose nanocomposite (ACNC) films from sugarcane bagasse nanofibers. A disk grinding technique was used for reducing the diameter. X-ray diffraction (XRD) revealed that crystal size was reduced as the duration of dissolution time has been increased such as a dissolution time of 10 min has led to the tensile strength of fiber sheet, nanofiber, and ACNC as 8, 101, and 140 MPa, respectively. The ACNC film water-vapor permeability increased as the dissolving time was increased. The ACNC has a great potential for use in cellulose-based food packaging owing to its positive characteristics [75]. During the production of biomass-based poly(l-lactic acid) PLA. Various epoxy-functional reactive oligomers have been developed and incorporated. The degraded fragments of chain extenders minimize the effects of hydrolytic degradation and maintain the acceptable viscosity of PLA. The molecular weight of PLA grew as the reactive oligomers' functionality has been increased. This is due to the carboxylic acid preferred reaction with the epoxy groups vs. the hydroxyl groups. The minimal reaction with the epoxy groups at the deteriorated PLA chains. The two ends have also been demonstrated in instances where PLA chains are severely degraded. Higher functionality and concentration of reactive oligomers are necessary to provide a substantial increase in molecular weight and enhance hydrolytic stability [76]. Biomass, the only source of renewable organic carbon on Earth, offers an efficient substrate for bio-based organic acid production as an alternative to the leading petrochemical industry based on non-renewable resources. Itaconic acid (IA) is one of the most important organic acids that can be obtained from lignocellulose biomass. IA, a 5-C dicarboxylic acid, is a promising platform chemical with extensive applications. IA production can take place through fermentation with fungi like *Aspergillus terreus* and *Ustilago maydis* strains or with metabolically engineered bacteria like *Escherichia coli* and *Corynebacterium glutamicum*. Bio-based IA represents a feasible substitute for petrochemically produced acrylic acid, paints, varnishes, biodegradable polymers, and other different organic compounds. IA and its derivatives, due to their trifunctional structure, support the synthesis of a wide range of innovative polymers through crosslinking, with applications in special hydrogels for water decontamination, smart nanohydrogels in food applications, coatings, and elastomers as shown in Figure 2 [77].

Bacterial cellulose

Aerobic bacteria synthesized cellulose is known as microbial cellulose or bio-cellulose (i.e., Acetobacter xylinum) [78, 79]. Bacterial Cellulose (BC) is a polymer alone, which does not require any chemical treatments for cellulose isolation. It's a unique and intriguing substance compared to green plant cellulose offering superior mechanical strength and degradability [80]. BC also features ribbon-shaped fibrils that are 20–100 nm in diameter and are made up of considerably smaller 2–4 nm nanofibrils and known as bacterial nanocellulose (BNC). Furthermore, unlike the methods used for obtaining nanocellulose through mechanical processes. BNC is mostly created by bacteria through the biosynthesis process as shown in Scheme 1 [81]. The elastic modulus of these microfibril bundles is 78 GPa and has a high crystallinity (84–89%). These microfibrils hold a larger moisture-holding capacity, a higher polymerization degree, and a finer web-like network. The BC's outstanding characteristics have widespread applications from food to functional materials like diaphragms in speakers, electronic gadgets, paper additives, membrane filters, and cosmetics [82–84]

Fig. 2 Biomass-Derived Production of Itaconic Acid as a Building Block in Specialty Polymers (Copyright Permission MDPI-2022)





Scheme 1 Bacterial cellulose luminophore paper prepared by impregnation and ambient drying, (Copyright Permission MDPI-2022)

attributed to its extreme crystalline structure, where dry cellulose is neither easily soluble in water nor organic solvents. Cellulose-based gels/films have limited applicability [85, 86]. Chemical alteration of the cellulose surface is necessary to produce soluble cellulose. Esterification is a typical method of converting dry cellulose to a water-soluble solution. This modified cellulose is an environmentally benign product that can be used in a variety of applications [87]. Methylcellulose (MC), hydroxypropyl methylcellulose (HPMC), and carboxymethyl cellulose (CMC) are water-insoluble materials produced by etherification with methyl chloride, propylene oxide, or mono-chloroacetate. Furthermore, cellulose acetate (CA), hydroxypropyl methylcellulose phthalate (HPMCP), etc. are popular derivatives employed in commercial items or pharmacological research [88]. Ionic liquids (ILs), which are sophisticated green solvents have also been widely used as the media in cellulose modification. In previous studies, it is discovered that cellulose may dissolve in ionic liquids, which encourages the development of novel cellulose solvent solutions. Organic salts consist of cations and anions that result in ionic liquids. During cellulose dissolution in ionic liquids, the OH group of cellulose forms compounds with the ionic liquid. The oxygen atoms in cellulose act as electron donors, while the hydrogen atoms act as electron acceptors [89]. During the reaction between cellulose-OH and the ionic liquid, the hydrogen bonding network of cellulose is disrupted, which results in the dissociation of hydrogen bonds between the molecular chains of cellulose. Among other applications, this novel cellulose material can be utilized to generate cellulose composites for thin films supporting reaction media, and cellulose-based ions gel in fuel cells [90]. Electrospinning of cellulosic fibers for biological applications, biosensors, and separating membranes [91]. The biosynthesis and [92] the formation of microfibril bundles have opposed to mechanical or chemo-mechanical approaches for obtaining nanocellulose [93, 94].

Future Development of Cellulose Composites

Throughout the review, cellulose composite offer several advantages, which are of great potential to food, medical and high-end industries [95]. However, the development of these composites is still in the preliminary stages due to their undetermined functions, quality, and cost [96]. To expand the application of cellulose bio-composites across various industries [97], several aspects must be considered for future developments and applications. Cellulose has been recognized as a good film-forming material [98]. However, several studies reported the utilization of its derivatives instead of its original form for biodegradable film production. For cellulose derivatives exhibit better transparency, mechanical and water vapor barrier properties [99]. Cellulose derivatives can satisfy electrical applications such as polymer electrolytes, wound dressings, scaffolds, hydrogel for cell and drug delivery, biomedical [100], as well as soft gel capsules as pharmaceutical devices. The production of these derivatives is environment friendly during the extraction or isolation process. Hence, the production of the edible film from the original cellulose should be focused and developed for future applications. Although the edible film produced from untreated cellulose is relatively simple, environment friendly, and cheaper than cellulose derivatives. But on the other side, cellulose in its original form has poor physicochemical properties, which limited its extensive applications. It is believed that the development of cellulose composites may attain equal or even better performance than the derivatives' films. In the food industry, cellulose derivatives are widely used as a gelling agent for processed foods. This polysaccharide also has significant potential for development as a source of biodegradable [101] or edible film in packaging applications. Nevertheless, cellulose derivatives exhibit some poor physical and thermal properties that are needed for the food packaging process [102, 103]. In this regard, cellulose especially in its nano-size has been proposed as a reinforcing agent in derivatives film owing to its high adhesive strength and tensile modulus [104]. Thus, the bio nano-composite-based film revealed remarkable improvement in its properties as compared to pure derivatives films. Nevertheless, this nanocomposite-based film has limited usage in some food packaging applications, wherein it cannot be classified as edible food wrapper due to cellulose being considered non-edible material, although cellulose is found in all plants and most prevalent carbohydrate on the planet. Humans are unable to consume it since no vertebrate has the enzymes necessary for its breakdown. Herbivores, on the other hand, have symbiotic bacteria in their intestines, which help in cellulose digestion. To increase the use of cellulose composite film in this food industry, the recent progress in research has effectively turned cellulose into an edible form of material to use in food and food wrappers that may be digested by the consumer [105]. This incredible accomplishment has paved the way for the future development of modified cellulose in degradable materials [106] as well as food packaging that can be consumed. The cellulose or its derivative may be blended with modified cellulose to obtain new exciting products [107], novel and multifunctional composite films, which may be utilized and developed for advanced applications.

The bio-composites derived from renewable resources have been the subject of attention. The abundant and cheap availability of petroleum-based materials restricted the earnest efforts for the development of eco-friendly materials. Presently, increasing environmental concerns and regulations have put a deliberate interest in this direction. The very important advantages of natural fibers as filler over traditional carbon and glass fibers are their eco-friendly nature. In most cases, unfortunately, the natural fiber composite does not reach the same strength level as glass fiber composites mainly because of incompatibility between generally hydrophobic host polymer matrix and hydrophilic natural fiber, combined with a lower thermal resistance of the cellulosic material. In the last couple of years, it has been observed that highly crystalline cellulose has some unique and outstanding potential to increase the composite material properties at lower filler concentration, in comparison to unfilled polymer matrix counterparts. Cellulose has to overcome many obstacles against industrial practices due to time-consuming preparation procedure with very low yield, highly hydrophilic surface, commercial unavailability, poor dispersion due to high agglomeration tendency, low thermal stability, and most importantly, in general, comparatively higher cost through the expensive source [108, 109].

Biodegradation of Cellulose

The primary objective to fill the polymer matrix with cellulose is to develop eco-friendly green composites with the potential of degradation in the biocycle by the action of different microbes, leaving behind unharmful residue biomass with the emission of carbon dioxide (CO_2) and water. Therefore, the evaluation of the environmental biodegradability of cellulose-based composites is a highly important factor in order to expand their applicability. Cellulose is not uniform in structure and there are some imperfections, mostly due to the various chain dislocations and ends [110, 111]. It is known to degrade by the action of exoglucanases initiated through the action from the end. There is limited research conducted in the area of biodegradation of cellulose-based composites. In a report on the biodegradation study of cellulose-reinforced rubber, the biodegradability of the sample was enhanced with the amount of filler, where the results indicated that crystallinity caused important effects in promoting the biodegradability of rubber. Similarly, the presence of bagasse whiskers resulted in an increase in moisture sorption of rubber films where the highest weight loss in soil was observed at 12.5% whisker content fueling the conclusion that the presence of cellulose whiskers increased the rate of degradation of rubber in soil. Poly(lactic acid) (PCL) reinforced with cellulose whiskers highly dispersed with poly(ethylene glycol) were examined for biodegradation in simulated body fluid, where an improvement in the water absorption and biodegradation of the nanocomposites was observed. Cellulose whiskers isolated from bagasse have been filled in polycaprolactone after modification with n-octadecyl isocyanate and nanocomposites were fabricated by a casting/evaporation technique. Bio-disintegration studies of the PCL/cellulose in soil were carried out and an increase in the bio disintegration was found after the addition of 7.5% modified whiskers. At higher loadings of modified cellulose whiskers, the weight loss tended to decrease, but it was still higher than that of neat PCL [112, 113]. The effect of compatibility on the biodegradation of cellulose reinforced composites has not been quantified till now in relation to the mechanical performances. However, the reports on the macro natural fiber-filled composites indicated a role of compatibilization in the degradation of resulting composites, and there was a significant effect of the method of preparation on the degradability of the composites. The composites prepared by direct reactive mixing were found more degradable. It has been proposed that compatibility may increase the properties and biodegradation of the host matrix [114, 115].

Conclusions

This review emphasizes cellulose film generation, which offers great compatibility, biodegradability, and potential contribution to the established economy. It is suitable for the production of biodegradable and cost-effective mix films for a variety of applications. The current state and future possibilities of the most promising natural polymer cellulose, and its derivatives, such as hydrogels, films, and composites are elaborated here in detail. Cellulose is the most abundant renewable material in the biosphere, for it is cost-effective, non-toxic, and biodegradable. Thus, it's worth looking into the newly synthesized composite materials for the preparation of goods that are sustainable, useful, and cost-effective. This incredible accomplishment has paved the way for the future development of modified cellulose in degradable materials in food packaging that can be consumed. The cellulose or its derivative blended with modified cellulose will be a new exciting product that can form novel and multifunctional composite films and should be investigated further for future development and advanced applications.

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Declarations

Conflict of interest There are no conflicts to declare.

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