# Properties and Applications of Three Renewable/Sustainable Polymers: Cellulose, Lignin and Poly (Butylene Succinate)

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**Abstract.** Since its invention in the last century, plastic has been widely used in various fields and has become the most used material at present. While it is convenient for people's life, the massive use of plastics has also brought about serious environmental pollution problems. Therefore, the research on degradable polymer materials and their related applications has become a research direction for more and more scholars. This paper introduces several environmentally friendly polymer materials, including two natural polymers, cellulose and lignin, and a synthetic polymer, poly (butylene succinate) (PBS), they all have excellent biodegradability. This paper also discusses their structures, properties, and applications in related fields. Cellulose has good resistance to hydrolysis and can be used in food packaging and tableware; lignin can be extracted from wide range of sources and is suitable for medical applications; PBS has good overall performance and can be used in many fields after copolymerization or blending modification with other substances. The study of renewable/sustainable polymers is of great significance to environmental protection and to solve the environmental pollution problem, but the study of degradation properties and applications of many polymers is still at the initial stage, more research results are expected in the future.

**Keywords:** Degradable polymers; Applications; Cellulose; Lignin; PBS.

## 1. Introduction

Plastics, as one of the most important inventions of the 20th century, have been widely used in various fields because of their light weight, resistance to low temperature, chemical resistance, good insulation, and good processing properties. It can be seen from people's daily necessities, to industry, agriculture, medical and health fields, and even on the cutting-edge technology of national defense. The production of plastics has surpassed that of steel by volume as the most used material, and the production is increasing year by year. From 2010 to 2018, the production of plastics increased from 270 million tons to 359 million tons [1]. However, at the same time, plastics have high molecular weight and stable properties, which are difficult to be degraded under natural environmental conditions; and due to the defects of all current disposal methods for waste plastics, the massive use of plastics has brought about serious environmental pollution issues, including air and water contamination, and climate change [2].

At present, there are three main methods for the disposal of waste plastics, each of which has corresponding limitations. The disposal of waste plastics by landfill method will occupy the land for a long time due to the nature of plastics that are difficult to be degraded. In addition, the process of plastic degradation may produce leachate and contaminate groundwater. Disposing of waste plastics by incineration generates large amounts of toxic gases, including carbon dioxide, dioxins, furans, mercury, and polychlorinated biphenyls (PCBs), which not only pollute the air, damage vegetation, and harm to human and animal health, but also contribute to global warming. Disposal of waste plastics through recycling is the least environmentally impactful treatment method, but recycling of waste plastics is difficult and costly due to the variety of plastics and their stable properties, and is rarely used as a treatment method for waste plastics.

Therefore, developing biodegradable and environmentally friendly polymers to replace the current plastic is a very good solution to address the issue of environmental contamination brought on by improperly disposed waste plastics [3, 4]. This paper will focus on several common renewable/sustainable polymers, including bio-derived natural polymers as well as synthetic

polymers, and discuss their structures, degradation properties, modifications, and applications in related fields. All these polymers are of interest to many researchers because of their good degradation properties and potential for a wide range of applications.

## 2. Natural Polymers

Natural polymers are large organic compounds that are not artificially synthesized and are formed in nature (including microorganisms, animals as well as plants) and minerals through biochemical action or photosynthesis. Common natural polymers include starch, cellulose, natural rubber, proteins, and nucleic acids. Since the 19th century, people have been using natural polymers. In a time of environmental pollution caused by the massive use of plastics, natural polymers have excellent biodegradation properties because they are produced in living organisms or in minerals, and therefore their related properties and potential applications have been the focus of scholarly research.

In this paper, two natural polymers, cellulose and lignin, will be introduced and their structures, extraction methods, properties and applications will be discussed.

### 2.1. Cellulose

#### 2.1.1. Structure

Cellulose is a type of polysaccharide and is a linear polymer. It consists of multiple D-glucose units through β-1, 4-glycosidic bonds with the chemical formula $(C_6H_{10}O_5)_n$.

#### 2.1.2. Extraction Methodology

Cellulose is widely distributed throughout nature, and more than half of the carbon in the plant kingdom can be found in cellulose. As the primary structural component of cell walls of green plants and various forms of algae, cellulose is mostly present in the leaves and stems of plants. In addition to plants, extraction sources of cellulose also include tuna and bacteria [5]. In industrial production, the raw material for cellulose extraction is mainly derived from by-products of agriculture and forestry, including cotton, corn, pineapple, and rice straw [6]. These by-products contain large amounts of cellulose, and at the same time, the use of by-products for cellulose extraction can effectively reduce production costs, therefore, cellulose extraction from plants is still one of the most common ways to extract cellulose. In addition to cellulose extraction from plants, chemical acid hydrolysis is also used industrially to obtain a special type of cellulose, nanocellulose [7]. Compared to cellulose, nanocellulose has smaller scale (nm level), lighter mass, lower density and higher strength, and is increasingly used as an environmentally friendly material in various fields.

#### 2.1.3. Properties

Cellulose has good biodegradability and resistance to hydrolysis, these properties make cellulose products environmentally friendly and can withstand harsh conditions, with potential applications in various fields. At the same time, cellulose is abundant in nature and has low extraction costs, making it suitable for mass production applications [7].

(1) Biodegradability

Since most cellulose is synthesized in plants, it is of good biodegradability. Cellulose can be degraded by microorganisms in nature into water and carbon dioxide. The degradation of cellulose occurs mainly through the hydrolysis of the glycosidic bonds between glucose by cellulase enzymes. Therefore, cellulose products can also be degraded by culturing microorganisms to obtain cellulase, which is a faster and more targeted degradation method than natural degradation.

(2) Hydrolysis resistance

Cellulose is also highly resistant to hydrolysis due to its structure. Because of its crystalline and non-crystalline topology, cellulose has a relatively stable structure and is resistant to attack by hydrolytic enzymes. According to studies, the topology of cellulose prevents enzymes from snapping it into the substrate site and, therefore, cellulose cannot be hydrolyzed by a single enzyme alone.

### 2.1.4. Applications

(1) Nanocellulose reinforced composites for food packaging and tableware applications.

Plastics are commonly applied in tableware and food packaging fields, such as cling film and disposable tableware. The reason is that plastics have low weight and high thermal deformation temperature. Most of these items are discarded after one use, which brings a large amount of waste plastic pollution. In order to solve this problem, environmentally friendly materials can be found to replace plastics in food packaging and tableware applications.

According to research, thermoplastic starch (TPS), polylactic acid (PLA), and PBS can be used as environmentally friendly polymeric materials to replace plastics in food packaging and tableware [7]. All three have good food contact properties and biodegradability, but at the same time, all three have certain drawbacks; TPS has significantly reduced mechanical properties when exposed to water and cannot be used alone as a material for food packaging and tableware; PLA and PBS have good mechanical properties but poor oxygen barrier, making them difficult to adapt to food packaging and tableware functions. Therefore, when they are used, they need to be modified according to the performance needs of the final product. Nanocellulose is able to be added to TPS, PLA and PBS as a reinforcing agent to co-blend and modify the three to improve the barrier and mechanical qualities of the materials [8]. According to the experiments, the results reveal that TPS, PLA, and PBS reinforced with nanocellulose have enhanced mechanical characteristics, oxygen barriers, and thermal shape change temperatures, which are suitable for food packaging and tableware applications. After the addition of nanocellulose, TPS and PBS have significant increases in the tensile strength, with increases in the range of 40-70% and 120% respectively; nanocellulose reinforced PLA, with increases in modulus of 58% and strength of 210% [7]. In addition, nanocellulose-reinforced composites have improved processing properties and can be crushed to very small sizes for various forms of processing.

(2) Plant fibers for tableware applications.

In addition to the use of nanocellulose-reinforced composites, tableware can also be manufactured using plant fibers as raw materials. Plant fibers are filaments produced by combining cellulose with various nutrients in plants and are mainly extracted from fiber crops. For the production of plant fiber tableware, fruit pomace, wheat straw and rice and wheat husk from agricultural crops are mainly used as the main sources of plant fiber. The production process can be divided into dry and wet methods [9]. The dry process is to crush wheat straw and other plant fiber sources, add water and binder, and then mix the three together and mold them by cold pressing. In contrast, the wet process involves the addition of catalyst, binder and curing agent to the crushed plant fiber source, and the final molding by hot pressing. Compared to the wet process, the dry process is shorter and cheaper, and is more often chosen for the production of plant fiber tableware in high volume production.

Plant fiber tableware also has very good biodegradability and food contact properties. Compared to nanocellulose-reinforced composites, the raw materials for plant fiber tableware are more readily available and the production process is simpler. The raw materials used in plant fiber tableware, such as pomace and wheat straw, are by-products of agricultural and forestry crop processing, which are easy to obtain and low cost. Additionally, the issue of an excess of these by-products can be efficiently resolved by using plant fiber tableware. Concerning the production process, plant fiber tableware is simpler, requiring only the crushing of raw materials and the addition of binders, followed by cold pressing, whereas the production of nanocellulose-reinforced composites requires more complex steps, including the extraction of nanocellulose from raw materials by chemical acid hydrolysis and the modification of TPS, PLA and PBS with nanocellulose in blends.

However, at the same time, plant fiber tableware still has deficiencies in mechanical properties. Since the research on plant fiber tableware is still in the development stage, no relevant studies on the modification of plant fiber to enhance its mechanical properties have been seen. More research results are expected in the future, so that plant fiber tableware can be applied on a large scale.

## 2.2. Lignin

### 2.2.1. Structure

Lignin is a complex, 3D polymer derived from the polymerization of three main precursor lignans: p-hydroxybenzoate alcohol, coniferyl alcohol, and sinapyl alcohol. Depending on the plant species, tissue type, and extraction method, lignin can have a variety of configurations [10]. This structural diversity gives lignin great potential for applications in a variety of fields.

### 2.2.2. Extraction Methodology

Lignin is a fundamental structural element of cell walls and is mainly found in plants. Especially in bark or wood, the presence of lignin allows them to be extremely stiff in order to support the weight of the whole plant. In addition to its structural function, lignin is critical for the transport of water and nutrients throughout plants and aids in their defense against pathogens and pests.

There are various methods of lignin extraction, including kraft pulping, sulfite pulping, organosolv process, ionic liquid treatment, deep eutectic solvent treatment, steam explosion process, dilute acid process, ammonic fiber explosion process and pyrolysis [11].

(1) Kraft pulping

Among these extraction methods, kraft pulping is the most commonly used extraction method in industry, and it accounts for 85% of the total global lignin production. Kraft pulping first uses a high pH solution to dissolve most of the lignin (90%-95%) in the plant material, and subsequently acidifies the solution by adding sulfuric acid or carbon dioxide to the solution, allowing the lignin to precipitate and be processed further [11].

(2) Sulfite pulping

Sulfite pulping is another widely used method for lignin extraction, which is mainly carried out at pH values between 2 and 12, with different solution pH values according to positive-charged components of the pulping solution. The advantage of this extraction method is the ability to obtain lignosulfonates as product, which contain high amounts of sulfur because of the sulfonic acid groups in lignin and can be widely used in dispersants, concrete additives, and composites [12].

(3) Organosolv process

Compared to the previous two extraction methods, organosolv process has become a new trend in lignin extraction methods through reducing the potential contamination possibility without the presence of sulfur during extraction. The organosolv process mainly involves the extraction of lignin by fractionation of the components in the plant raw material through organic solvents, including acetone, methanol, ethanol, and ethylene glycol, at high temperatures, usually between 170-220 °C [13]. In addition to organosolv process, newly developed extraction methods including dilute acid process, steam explosion process, and ammonic fiber explosion process to remove the presence of sulfur in the extraction of lignin, reducing the possibility of contamination and can extract higher quality lignin.

### 2.2.3. Biodegradation

Lignin has good biodegradability. Depolymerization and mineralization are the two steps that often characterize its biodegradation. In depolymerization process, oxidoreductases, including lignin peroxidase, versatile peroxidase, and laccase generated by white-rot fungus can cause lignin to depolymerize into lignin phenoxy radicals. After this, the aromatics generated by depolymerization undergo a series of mineralization reactions conducted by bacteria and finally enter the tricarboxylic acid (TCA) cycle [14].

In addition, lignin is widely available and can be extracted from many plants. These two properties allow lignin to be well used as environmentally friendly polymer materials in various fields.

### 2.2.4. Applications

(1) Wound Dressings

In the medical industry, lignin is used for a variety of purposes, including the delivery of medications and the wound dressings. Recent studies have shown that one of the raw materials for

wound dressings: hydrogel, can be prepared from lignin. Wound dressings prepared from lignin-based hydrogels have good healing ability to wounds. In the study, the researchers applied regular and lignin-based hydrogel wound dressings to the back wounds of two groups of mice and observed the healing situation of the wounds. The results showed that lignin-based hydrogel wound dressings have a significant improvement in the healing ability compared to the regular wound dressings. The wound healing rate reached 48% on the tenth day with the regular wound dressings, while the wound healing rate reached 87% on the tenth day with the lignin-based hydrogel wound dressings [15]. Lignin, gantrez, and polyethylene glycol (PEG) are combined through an esterification reaction to create lignin-based hydrogels.

Lignin is considered as a suitable material to produce hydrogels because of its good antioxidation and antibacterial properties. The experiment measured the number of free radicals consumed to investigate antioxidation properties of lignin. The test findings revealed that lignin solution at 0.2 mg/mL has good antioxidant qualities since it could consume 70% of free radicals [15]. Besides, the high mechanical strength of lignin also makes the wound dressings effective in protecting the wound site from further injury and contamination. The structure of the lignin-based hydrogel is presented as a 3D network of the polymer. Such a structure can absorb and retain large amounts of water, helping to remove undesirable metabolites released from the wound site [16].

(2) Blending modification of lignin-calcium carbonate composites with PLA

PLA has good biodegradable and mechanical properties and is a good alternative to plastics that can be commonly utilized as the environmental-benign material in various fields, such as in medical applications and films used in agriculture [17]. Despite the excellent properties, the high cost has been one of the reasons limiting the large-scale application of PLA in various fields. According to the study, lignin-calcium carbonate composites can be co-blended with PLA to improve the tensile strength and effectively reduce production cost [18]. In Ding et al.'s study, calcium hydroxide feedstock was first converted into calcium carbonate, then carbon dioxide carbonation method was used to combine calcium carbonate and lignin to produce lignin-calcium carbonate composites, and finally the composites and PLA underwent blending modification by using the melting method to obtain PLA/lignin-calcium carbonate composites. After testing, the new composites showed a significant increase in tensile strength compared to PLA. At a filling of 40 wt% of lignin-calcium carbonate, the increase in tensile strength was able to reach 214% [18]. By this method, industry can use less PLA to obtain composites with higher tensile strength, effectively reducing production and application costs.

## 3. Synthetic Polymer: PBS

Among the polymer materials, there are not only natural polymers which exist in nature, but also synthetic polymers which are synthesized artificially, both of which together constitute polymer materials. Synthetic polymers are polymers synthesized by chemical synthesis in industry, and these polymers are produced by different polymerization methods and conditions according to different performance requirements. Polypropylene (PP), nylon, and plastics are examples of common synthetic polymers. Compared to natural polymers, synthetic polymers are utilized in a variety of fields due to the designability and controllability of their products to meet the needs of the final product. In addition, many synthetic polymers are suitable for mass production and application due to their inexpensive raw materials, but they also cause serious environmental pollution due to their non-natural degradable nature. This paper focus on an environmentally friendly synthetic polymer material, PBS, which can be naturally degraded, and introduce its structure, synthesis methods, degradation properties and modification [19].

### 3.1. Structure

PBS is an aliphatic polyester with good biodegradability and is a white crystalline polymer. It is polymerized from butylene glycol succinate ($C_8H_{12}O_4$) as monomer, with the chemical formula H-[O $(CH_2)_4$OOC $(CH_2)_2$CO] $_n$-OH.

### 3.2. Synthesis

There are three main methods for the synthesis of PBS: bio fermentation, chemical condensation polymerization, and direct esterification with acid anhydride. Biological fermentation method for the synthesis of PBS is more expensive and difficult to prepare high molecular weight PBS, which is not suitable for use in industrial mass production. Chemical condensation polymerization method is able to design the molecular structure of PBS, and the synthesis cost is low, so it is the most commonly used synthesis method in industrial production. Chemical condensation polymerization method mainly including ester exchange method, acid anhydride reaction method, solution polycondensation method and chain expansion method. Ester exchange method is to obtain PBS by the ester exchange reaction between diol and dimethyl dibasic acid under vacuum, high temperature and catalyst conditions, and to remove methanol. The acid anhydride reaction method can obtain PBS by ring-opening polymerization of succinic anhydride and butanediol also under vacuum, high temperature and catalyst conditions. Solution polycondensation method obtains PBS by esterifying succinic acid and butanediol in different solutions, followed by higher temperature to complete the polycondensation reaction. This synthetic method requires relatively low temperature, but the reaction rate is slow, and it is not possible to prepare large molecular weight PBS. Chain expansion method is a method to increase the molecular mass of PBS. During the reaction of the condensation polymerization method, side reactions including thermal degradation and thermal oxidation occur, especially when the reaction proceeds to a later stage and after the temperature exceeds 200 °C. These side reactions affect the improvement of molecular mass of PBS, and molecular mass can continue to increase by adding chain extenders. Although the chain expansion reaction can improve the molecular mass of PBS, the reaction rate is fast and difficult to control, meanwhile, the commonly used chain expansion agents are toxic, such as diisocyanate. The direct esterification method is a synthetic method that has emerged in recent years and is still in the development stage. This method obtains PBS by direct condensation of succinic acid and butanediol. The direct esterification method consists two steps. Firstly, esterify dibasic acid with excess diol at a lower reaction temperature to produce end-hydroxy prepolymer, and then the excess diol is removed under vacuum, high temperature and catalyst conditions to obtain PBS.

### 3.3. Properties

PBS has good combination properties, including high melting point, good mechanical and processing properties, and the ability to be biodegraded. These properties allow PBS to be used in a wide variety of applications while avoiding environmental pollution.

#### 3.3.1. High heat deflection temperature

As a crystalline polymer, PBS has a high melting point and can reach heat deflection temperatures close to 100 °C. Compared to other common aliphatic polyester compounds, PBS has good heat resistance. When blended or copolymerized with other substances, the heat deflection temperature of PBS composites can exceed 100 °C, which makes PBS very suitable for food packaging and tableware manufacturing.

#### 3.3.2. Excellent mechanical properties

PBS has excellent mechanical qualities, with tensile strength, elongation, flexural strength, and flexural modulus very close to those of the general-purpose plastics polyethylene (PE) and polypropylene (PP). This allows PBS to meet the requirements of applications in most fields.

### 3.3.3. Excellent processing properties

PBS also has excellent processing properties and is not easily degraded when melted at high temperatures in a liquid state. Therefore, industry is able to process PBS using different molding processes, including extrusion, injection molding and blister molding, to produce different forms of products to meet the needs of different fields of application.

### 3.3.4. Biodegradability

The presence of easily hydrolysable ester groups in the structure of PBS gives it excellent biodegradation properties. Under specific microbial exposure conditions such as composting, PBS can be hydrolyzed by a variety of microorganisms and enzymes produced in plants and animals, ultimately producing water and carbon dioxide, and avoiding environmental pollution. During biodegradation, the rate of degradation is also influenced by the type of enzyme, the molecular weight size of PBS, and the degree of crystallinity. To achieve more efficient degradation, specific enzymes derived from cultured microorganisms are used to target the degradation of PBS with different molecular weights and crystallinity to increase the degradation rate.

### 3.4. Modification

Although PBS has good overall performance, its rigidity is poor, and its production cost is still relatively high compared with general-purpose plastics. To enhance the functionality of PBS, reduce its production cost, and allow PBS to be used more widely, it is necessary that PBS should be modified.

The modification of PBS includes copolymerization modification and blending modification. Copolymerization modification is to add other different monomers in the polymerization stage, design from the perspective of molecular structure, and obtain PBS copolymer materials with different mechanical and degradation properties. The monomers used in PBS copolymerization modification can be mainly divided into two categories, one contains aliphatic components and the other contains aromatic components. Common monomers include adipic acid, hexanediol and terephthalic acid. Adding monomers containing aliphatic components can improve the brittleness of PBS and enhance its biodegradability, while adding monomers containing aromatic components can increase the rigidity and thermal deformation temperature of PBS. The blending modification of PBS is generally to mix PBS with other materials in a molten state to obtain products with better performance, and can greatly reduce the production cost of PBS. Common blend materials include starch and PLA.

## 4. Conclusion

This paper focuses on two natural polymers, cellulose and lignin, and one synthetic polymer, PBS, and discusses their structures, extraction or synthesis methods, properties and related applications as sustainable/renewable polymer materials. All three polymers have excellent biodegradation properties and can be degraded by microorganisms in nature. This has also led to the development of more efficient degradation methods, i.e., obtaining specific enzymes to degrade these polymers by artificial cultivation of microorganisms. Cellulose, mainly extracted from plants, is able to be used in tableware, and it’s another form, nanocellulose, can be used in food packaging and tableware by reinforcing other polymers; lignin can be extracted from plants in a variety of methods, and different structures are obtained depending on the extraction method. Lignin has a very good performance in the medical field due to its excellent antibacterial and mechanical properties, and at the same time, the composite material formed by lignin and calcium carbonate can effectively reduce the production cost of PLA; PBS is mainly synthesized by three methods, with good combination performance and can be copolymerized or blended with a variety of substances for modification. Although the applications in many fields are still in development, all three polymers offer great potential for utilization in various fields. It is expected that more research results in the future will enable these environmentally friendly polymers to be increasingly used in life, lessen the reliance on plastics and mitigating contamination of environment.

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