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# Production of Sustainable Bioplastic Derived from Renewable Lignocellulosic Agricultural Biomass: A Comprehensive Review

Muhammad Asif<sup>1,\*</sup>, Mohammad Siddique<sup>2</sup>, Azhar Abass<sup>3</sup>, Abdulhalim Musa Abubakar<sup>4</sup>, Musa Askira Abubakar<sup>4</sup>, Gaurav Kumar Pandit<sup>5</sup>, Minza Igunda Selele<sup>6</sup>

- <sup>5</sup> Department of Botany, Patna University, Patna, India
- <sup>6</sup> Department of Environmental Science and Technology, School of Engineering and Environmental Studies, Ardhi University, P.O. Box 35176 Dar es Salaam, Tanzania

ARTICLE INFO	ABSTRACT
<b>Article history:</b> Received 3 July 2024 Received in revised form 17 July 2024 Accepted 8 August 2024 Available online 30 September 2024	Environmental pollution is increasing due to plastic materials which may find their way into our food. Now, lignocellulose materials offer enormous potential for use in the production of eco-friendly bioplastics and decreasing the environmental impact of fossil fuels. These raw materials can be used to separate lignin and cellulose. Research on bioplastics, derived from biological sources is currently gaining attention for greener environment. Through surface alterations and other chemical derivatizations, several materials are readily adaptable to the production of diverse bioplastics. Polyhydroxyalkanoates, bio-polyethylene, polyurethanes, and nano cellulosic bioplastics are examples of common bio-based polymers produced from lignin or cellulose. The current review covers lignocellulose compositions, bioplastic
<i>Keywords:</i> Lignocellulose; Bioplastics; Nanocellulose; Biodegradable polymers; Cellulose; Biomaterials	manufacturing methods, and applications in a range of industries. Bioplastics made from lignocellulose will become a useful material in a variety of industries in no distant time. On the hand, bio composites from food sources are currently being recognized and researched.

### 1. Introduction

A significant amount of toxins have been produced by the extensive use of plastic products and are typically disposed of in landfills when environmental deterioration is ongoing and adversely affects natural resources as well as human health [1]. Due to the damage that plastics made from petroleum cause to the environment and the depletion of fossil fuel supplies, there is an increasing

\* Corresponding author.

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<sup>&</sup>lt;sup>1</sup> Department of Energy and Environment Engineering, Faculty of Agricultural Engineering, Sindh Agriculture University, Tando Jam, Hyderabad, Sindh, Pakistan

<sup>&</sup>lt;sup>2</sup> Department of Chemical Engineering, Balochistan University of Information Technology Engineering & Management Sciences (BUITEMS), Quetta, Balochistan, Pakistan

<sup>&</sup>lt;sup>3</sup> Department of Chemical Engineering, Muhammad Nawaz Sharif (MNS) University of Engineering and Technology (UET), Multan, Punjab, Pakistan

<sup>&</sup>lt;sup>4</sup> Department of Chemical Engineering, Faculty of Engineering, Modibbo Adama University, PMB 2076, Yola, Adamawa State, Nigeria

E-mail address: engr.asif86@yahoo.com (Muhammad Asif)

interest in encouraging the development of bioplastics. In comparison to plastics made from petroleum, bioplastics have a number of benefits, such as lower greenhouse gas emissions, quicker biodegradation, and the use of sustainable materials [2]. These kinds of materials are desirable inventions for environmental sustainability since they can be utilized to conserve natural resources. They are created utilizing renewable biomass feedstock, like lignocellulose, proteins, and biopolymers; vegetable oil; maize and pea starch; and so on. Lignocellulose feedstock is the most prevalent and non-edible biomass among them. Large amounts of lignocellulosic waste are produced daily by farming, food processing, the alcohol and timber industries, and the manufacturing of sugar [3]. They are often disposed of in an unregulated way. Numerous issues with safety, health, aesthetics, and the environment are brought on by their accumulation. Reusing these lignocellulosic wastes is the best and most practical way to address such problems [4]. These agricultural wastes can be used for a variety of purposes, including increasing soil fertility, producing paper, and supplying conventional fuels. Only a small part of the wastes is used for these functions, though. It is preferable to utilize this lignocellulosic biomass to create sustainable products using environmentally friendly practices.

Waste lignocellulosic materials hold great promise for the production of a number of useful goods [5]. Due to its chemical and morphological characteristics, as well as the fact that it is renewable, researchers are interested in employing it as an alternative feedstock for producing a range of important products. Due to its distinctive qualities, including biodegradability, biocompatibility, environmental friendliness, and low cost, biomaterials have recently attracted a lot of attention in a variety of disciplines. However, investigation from 2015-2024 in Figure 1 shows that research on the conversion of lignocellulose to bioplastic as few, as evidenced in Google Scholar.



Bioplastics have been developed as a result of several scientific investigations into the potential uses of lignocellulose waste conducted throughout the world [6,7]. Though the ecological and financially viable use of lignocellulose wastes for bioplastics have been confirmed. This review hence, provides an overview of the lignocellulose composition, sources, and numerous methods for producing bioplastics, as well as its prospects for the future.

### 2. Methodology

Cellulose and lignocellulose are crucial materials in the realm of bioplastic production, each presenting distinct advantages and challenges. Cellulose in Figure 2, a linear polysaccharide found abundantly in plant cell walls, offers good mechanical strength, high crystallinity, and biodegradability, making it a favorable candidate for bioplastics. It can be sourced from a variety of renewable biomass such as wood pulp, cotton, and agricultural residues. However, to enhance its processability and mechanical properties for bioplastic applications, cellulose often requires chemical modification, such as esterification. In contrast, lignocellulose encompasses a more complex structure comprising cellulose, hemicellulose, and lignin. While lignocellulose provides additional toughness and resilience due to the presence of lignin, it necessitates more intricate processing to separate and utilize its components effectively.



Fig. 2. Cellulose structure

This process involves breaking down the complex structure of woody biomass or agricultural residues into cellulose, hemicellulose, and lignin, with cellulose then typically undergoing further modification for bioplastic production. Despite these challenges, lignocellulose offers the advantage of utilizing abundant agricultural and forestry residues that would otherwise go to waste, contributing to the sustainability of bioplastic manufacturing. Both cellulose and lignocellulose play crucial roles in advancing bioplastic technologies, offering unique properties and contributing to the goal of developing environmentally friendly alternatives to conventional plastics. Extracting lignocellulose involves various methods depending on the source material and desired outcomes, as shown in Table 1.

#### Table 1

Method of extracting Lignocellulose					
S/No.	Method	Description			
1.	Steam Explosion	Biomass is treated with high-pressure steam and then rapidly decompressed, disrupting lignocellulosic structure and facilitating enzymatic hydrolysis.			
2.	Acid Hydrolysis	Biomass is treated with acids (e.g., sulfuric acid) to hydrolyze hemicellulose into sugars, leaving cellulose and lignin intact.			
3.	Alkaline Hydrolysis	Biomass is treated with alkaline solutions (e.g., sodium hydroxide) to remove lignin and hemicellulose, leaving purified cellulose.			
4.	Organosolv Fractionation	Biomass is treated with organic solvents (e.g., ethanol, acetone) under acidic or alkaline conditions to dissolve lignin and hemicellulose, leaving cellulose as a solid residue.			
5.	lonic Liquid Pretreatment	Biomass is treated with ionic liquids (molten salts) to dissolve lignin and disrupt the lignocellulosic structure, facilitating subsequent processing.			
6.	Mechanical Methods	Biomass is mechanically processed (e.g., milling, grinding) to reduce particle size and facilitate chemical or enzymatic treatment for lignocellulose extraction.			

These methods are often combined or tailored to specific biomass types and desired end products, such as cellulose for bioplastic production or sugars for biofuel production.

### 3. Availability of Lignocellulose Feedstock for Bioplastics Production

Bioplastics are beginning to replace fossil fuel-based plastics in order to decrease their detrimental effects on both human health and the environment. Innovative processes have been developed to transform lignocellulose-derived biomass into industrial-scale goods for a number of industries, including bioplastics, fuels, lubricants, drugs, solvents, surfactants, cosmetics, nutraceuticals and animal feed [8]. The environmental problems connected to the current petroleum-based polymers have significantly enhanced the need to develop bio-based and environmentally acceptable polymer systems over the past several years [9]. It is important because the long-term sustainability of the bio-based sector depends on the development of highperformance bio-based and renewable materials. Conventional biotechnological approaches to converting biomass to produce industrially significant polymers have only partially succeeded. This shows that complex networks must function together in order for biomass conversions to be successful. According to a research by Jung et al. (2020), cone stover and other organic wastes can greatly reduce the cost of making bioplastics from lignocellulosic feedstocks. Due to difficulties like high production costs, employing lignocellulosic waste to manufacture bioplastics emphasizes the need for technological advancements and innovations in the industry. It may lead to the development of novel strategies for redesigning biosynthetic pathways for biomass conversion synergistic behavior, and ultimately, inexpensive and effective ways to convert biomass into usable products like biopolymers. This is due to the fact that it is an interdisciplinary topic of research that is an excellent combination of engineering and living science [11].

### 4. Lignocellulosic Material

Lignin and holocellulose are components of lignocellulosic biomass. Holocellulose is the term for cellulose and hemicellulose together, such as small amounts of protein pectin as well as extractives. Lignocellulose consists of cellulose, hemicellulose, and lignin, providing structural integrity to plant cell walls and requiring complex processing for bioplastic production. Hemicellulose, a branched polysaccharide with diverse sugar units, offers potential as a source of sugars or as a component in bioplastic formulations due to its solubility and adhesive properties. The majority of lignocellulosic biomass is made up of the carbohydrate polymers cellulose (40-50%), hemicellulose (20-30%), and lignin (10–25%) contents. In Figure 3, the chemical composition of lignocellulosic biomass is shown schematically [12]. Due to cellulose crystallinity, lignin hydrophobicity, and lignin hemicellulose matrix encapsulations of cellulose, lignocellulose has evolved to be resistant to breakdown. There are several connections between cellulose, lignin, and hemicelluloses. The primary non-carbohydrate component of lignocellulose is the three-dimensional polymer of phenyl propanoid units known as lignin ( $C_9H_{10}O_2(OCH_3)n$ ) [13]. One of the many useful polyphenolic amorphous polymers is lignin, which is composed of three p-hydroxyphenyl groups that are o-methoxylated on propanoid bases (such as p-coumaryl, sinapyl alcohol, and coniferyl). P-hydroxyphenyl, guaiacol, and syringyl are the names of the similar monomeric phenylpropanoid units in the lignin polymer [14]. The primary noncarbohydrate component of lignocellulose is lignin, a phenyl propanoic-based polymer in three dimensions. Lignin, which is made up of three p-hydroxyphenyl groups that are o-methoxylated on propanoid bases, including p-coumaric, sinapyl alcohol, and coniferyl, is one of many important polyphenolic amorphous polymers. The units of p-hydroxyphenyl, guaiacyl, and syringyl, respectively, are designated as similar monomeric phenylpropanoid units in the lignin polymer [15].



Fig. 3. Primary chemical makeup and structure of lignocellulose biomass [16]

Carbon-carbon bonds and aryl ether bonds bind different monomer units together. Due to the abundance of polar groups and hydroxyls, which create strong hydrogen bonds, lignin is insoluble in the majority of solvents. It is a glassy transfer polymer that is amorphous. Hemicellulose  $(C_5H_8O_5)n$  is the second most common polymer. Xylan, galactomannan, glucuronoxylan, arabinoxylan, xyloglucan, and glucomannan are among the polysaccharides combined in this [4]. Hemicellulose lacks a crystalline structure, has a branching structure, is poor in polymerization, and contains the acetyl group, making it easier to degrade in the presence of heat and chemicals. There are numerous hemicellulose subclasses, including arabinoxylans, galactoglucomannans, linear mannans, glucuronoxylans, galactomannans, glucomannans, xyloglucans, and h-glucans. The content of hemicellulose varies by plant species. Hemicellulose can dissolve in both alkali and acidic solutions [17]. The cellulose-hemicellulose-lignin network becomes stiffer as a result of hemicellulose's role as a connection between lignin and cellulose. It serves as a physical barrier that restricts cellulase's ability to access cellulose. As a result, the synthesis of cellulose units was made simple by the removal of hemicellulose utilizing various pretreatments. Yang et al., [18] claim that hemicellulose produces acetyl groups through ester bonds, which function as a catalyst for the breakdown of lignocellulose. This process causes the hemicellulose to be completely removed, hastening the conversion. Cellulose  $(C_6H_{10}O_6)n$ , a linear syndiotactical glucose polymer coupled by  $\beta$ -l,4-glycosidic linkages, is the main component of lignocellulosic feedstock [19]. Due to its stereoregularity, hydrophobicity [20], and biocompatibility, it is the most frequently used polymer on the planet. Its distinctive polymer chains are strong and forceful, with a crystalline structure. Each cellulose glucosylic ring contains two hydroxyl secondary groups and one primary hydroxyl unit [21]. As a result, cellulose is capable of a variety of hydroxyl-based chemical reactions. The ability of these hydroxyl groups to establish hydrogen bonds with other molecules has a significant impact on the structure and reactivity of cellulose. Interchain hydrogen bonds in cellulose microfibrils regulate the crystalline or amorphous nature of the cellulose framework, while intrachain hydrogen bonds in cellulose micro fibrils preserve the chain's straightness. In most common solvents, including water, cellulose fiber does not dissolve

due to strong contact forces [22]. In addition to these three primary components, lignocellulose also contains protein, trace amounts of pectin [23], and extractives such as waxes made from nonstructural sugars and chlorophyll. The properties of fermentation and enzymatic saccharification will be partially inhibited by colors. There are also trace amounts of many metals, alkaloids, and polyphenols [24].

# 5. Lignocellulose Sources

The term "lignocellulose feedstock" refers to the dry plant material that makes up a sizable component of the biomass present on the surface of the earth. The three main types of lignocellulose biomass are waste biomass, energy crops, and virgin biomass. Wood is a major source of lignocellulose. Among the sources of lignocellulose biomass are the waste products produced by natural forests, sawdust from sawmills, mulch, dead tree limbs, and swiftly expanding, transient forests. Forests contain two different types of wood: hardwood and softwood. Softwoods are made from gymnosperms. These trees have a faster rate of growth than hardwoods and have a lower density. Angiosperms are woody plants [25]. This feedstock is preferable to employing seasonal plants since it can be grown all year round, which allays concerns about long-term storage and ash production. When compared to herbaceous biomass as a feedstock, this provides a number of advantages in terms of production, processing, storage, and transportation. Non-wood sources are a good option since they are readily available, develop quickly, are inexpensive, are simpler to process, and require little time for growth [25]. Agricultural wastes, non-wood, and native plant fibers are the three categories under which popular lignocellulose non-wood biomass is grouped. Herbaceous crops are a term used to describe native plants. Comparing perennial grasses to other annual crops, perennial grasses are simpler to cultivate, harvest, and process. They have also garnered a lot of interest as active biomass feedstock since they produce more lignocellulose than typical tree species. A substantial source of biomass made of lignocellulose other than wood is agricultural waste [26]. Given that agricultural wastes are primarily eliminated during plant processing, using them to make bioethanol would be beneficial in any discussion regarding the use of agricultural land for both food production and energy production.

Non-wood biomass containing lignocellulose is extensively utilized as fillers, absorbents, and in the paper industry. Waste products created by civilization pose a range of risks to people's health, safety, and environment. However, lignocellulose found in municipal and commercial solid waste can be converted into biofuel. Food and kitchen waste, garden waste, paper and cardboard garbage, and other unrelated leftovers make up the majority of municipal solid waste [27]. They contain considerable amounts of biomass from plants, including lignocellulose, in the organic part, which has the potential to be turned into useful goods and can mitigate the negative impacts it causes. As a more affordable and plentiful raw substrate for lignocellulose biomass, marine algal sources can be used. There are several opportunities for genetically altering various algae to create new bio-based goods [28]. Algal biomass is not particularly suited to food production, unlike maize and sugarcane, and it doesn't need agricultural land or freshwater irrigation. Additionally, the huge amounts of CO<sub>2</sub> that marine algae absorb throughout their growth help to slow down global warming. This algae-based feedstock might be a practical and reachable source of third-generation energy. In addition, it can be used as a raw material to make crude oils, bioplastics, and aviation fuel [29]. Table 2 [29,30] displays the main sources and compositions of lignocellulose feedstock.

### Table 2

Туре	% Lignin	% Hemicellulose	% Cellulose	References
Cherry wood	18	29	46	[4]
Corncob	15	32	52	[5]
Corn Stover	14.4	30.7	51.2	[4]
Sunflower shell	17	34.6	48.4	[6]
Almond shell	27.5	27.9	48.6	[7]
Wheat straw	17.5	38.2	27.4	[4]
Beech wood	20.7	30.6	44.5	[4]
Spruce wood	26.7	20.2	48.2	[5]
Hazelnut shell	41.3	28	24.2	[8]
Wood bark	42.6	28	23	[9]
Olive husk	47	23	23	[10]
Walnut shell	51	22	25	[4]
Corn leaves	15	13	26	[4]
Wheat bran	8-11	34-38	12-16	[11]
Rice husk	24-30	17-20	36-44	[12]
Rice straw	16-18	22-24	28-33	[13]
Sunflower stalk	13	18	34	[4]
Willow	20	22	36	[4]

Various Lignocellulose biomass sources and their essential elements

### 6. Production of Bioplastics from Lignocellulose Feedstock

Although the markets for bioplastics are expanding steadily, they still only account for two percentage points of the total plastic market. It is crucial to combine biomass with other environmentally friendly elements to make profitable goods made from biomass technically, economically, and environmentally viable [20,32]. Lignocellulose biomasses are heterogeneous, complicated, and vary depending on the source, as was covered in earlier sections [33]. Bioplastics made from biomass materials are produced using a variety of techniques, including pretreatment, saccharification, liquid detoxification, fermentation, purification, and bio composite creation. Lignocellulose biomasses are heterogeneous, complicated, and vary depending on the source, as was covered in earlier sections. Bioplastics made from biomass materials are produced using a variety of techniques, including pretreatment, scarification, liquid detoxification, fermentation, purification, and bio composite creation [34]. Two separate conversion pathways, namely the lignin and cellulose pathways, are used in the creation of bioplastics from lignocellulose biomass. Figure 4 shows a schematic illustration of the primary two bioplastic manufacturing pathways. Three primary factors should be taken into account in order to produce bioplastics from lignocellulose materials in an economically viable and sustainable manner: namely, establishing a business model within a framework for the bio economy, addressing political and environmental challenges that affect the production of bioplastics, and following a novel approach. For lignin pretreatments, combining analytical chemistry, computer modelling, and genetic engineering will result in efficient lignin depolymerization and conversion [35].

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Fig. 4. Lignocellulose biomass-based materials [35]

# 6.1 Bioplastics

The names thermoplastics and thermosets refer to resins that, when heated, polymerize and create cross-linked chains to create thermosets that can be molded and plastic compounds that can be melt-molded (thermoplastics). Polyethylene terephthalate (PET), polycarbonate, and polyolefins (PE, PP) are a few thermoplastics made from petrochemical feedstock. Common thermosets include polyurethane (PUR) and phenol formaldehyde resin (PF) [36]. As discussed in the current review, biobased components can be used to upgrade or replace the aforementioned polymers. Some of the most common types of bio-based plastics are polylactic acid (PLA) [37], thermoplastic starch, cellulose (including cellulose acetate and cellulose xanthate/cellophane), and first-generation biodegradable bioplastics. Even though it is expanding, the market for bioplastics still only accounts for a small portion of the overall plastics business [38]. In 2014, the capacity for bioplastics reached over 215 million tonnes in 2012, thermoset plastic production exceeded 30 million tonnes [39]. Various production pathways can be used to describe the evolution of bioplastics. The most well-known ones for bioplastics made from forestry biomass are depicted in Figure 5.



Fig. 5. Primary methods of producing bioplastics based on forests [40]

# 6.2 Environmental Benefits of Some Bioplastics

Wood-based feedstock has a huge advantage over fossil-based feedstock because of their noticeably shorter carbon cycles, which lead to a balance between atmospheric carbon (carbon dioxide) and carbon bonded in organic material. Available cradle-to-grave studies in OECD (2013) show that, several bioplastics (e.g., PLA, BioPP, and BioPE) have lower greenhouse gas (GHG) emissions than polymers derived from fossil fuels [42]. The cradle-to-grave results did however, indicate that using bio-based products could reduce GHG emissions, although the findings were not conclusive [41]. Utilizing renewable resources is one way to lessen a material's environmental impact. In polyurethane polymers, for instance, polyols generated from plant oils are used commercially to replace polyols made from fossil fuels. A further problem is that bio based polymers (such as polyols formed from vegetable oils) may change how land is used, which could jeopardize the safety of food production [43]. Given that wood may be cultivated in locations that are unsuitable for growing food, the usage of polymers from wood, such as lignin-based polyols, can be a bio-based substitute that does not compete with food production. The type of use for the bioplastic influences the choice of waste treatment. Bio-based products may be used in durable applications, such as building supplies [44]. Consequently, the lifespan of plastics may be advantageous if the application's main goal is to reduce GHG emissions and the end-of-life option is energy recovery during burning or recycling. Biodegradable polymers, including PLA and PHA, may be appropriate for use in short-term applications like packaging and food disposal. This assertion must be taken with caution, though, as certain research has indicated that this is not always the case [45]. An environmentally friendly way to treat garbage is by biodegradation. The degradation process carried out by naturally occurring microorganisms in nature is referred to as "biodegradation." Microorganisms produce CO2 and water when the environment is aerobic, whereas methane is created when the environment is anaerobic. Unluckily, the word "biodegradable" is utilized indiscriminately when referring to bioplastics Figure 6, and not just in the contexts where it would be appropriate [46]. Remains of plastic, such as those with a high surface area, may pollute the water supply and other elements of the ecosystem.

Significant volumes of plastic waste pollute marine areas, which is a huge environmental problem [47].



Fig. 6. The distinction between biodegradation and degradation/fragmentation [47]

### 6.3 Major Bioplastics Made of Monomers

PHA, BioPE, and PLA are the three monomer-based bioplastics that are now offered for sale (which are all made, for example, from maize by NatureWorks<sup>™</sup>). The raw materials derived from forests can be used to make the monomers needed to make PLA, PE, and PHA Pretreatment is necessary to separate the polysaccharides from other wood constituents when using wood-based lignocellulose in manufacturing processes that call for monomeric sugar; however, maize and sugarcane have an advantage over it because their sugars are easier to extract from plants and have a lower degree of polymerization [48]. There are application and prospects for the bioplastics and biopolymer shown in Figure 7. In addition, as previously mentioned, conversion procedures are needed to break down the polysaccharides into monomeric sugar. It is difficult to carry out the degradation without also generating substances that have a deterrent impact on the microbes utilized in later process steps [49].



Fig. 7. Applications of biopolymer and bioplastics [19]

### 6.4 Biomass Treatments for Bioplastics Production

Due to their complex and diverse structure and content, lignocellulose raw materials are difficult to use to manufacture biochemical and bioplastics. Therefore, in order to assure the technological, economic, and environmental viability of a given product [4], it is essential to combine the production of that product from biomass with that of other components [50]. The technology and methods used in bio refineries for the extraction and conversion of various products, the separation and purification of the streams of intermediate compounds and end products, and their full integration into the process as a whole are relevant to the topic at hand. These issues will be covered in the sections that follow [51].

### 6.5 Separation Process of Forest Biomass

Depending on the sort of plant, forest biomass are softwoods, hardwoods, and/or gramineous species. Lignocellulose materials can have a variety of compositions. Fortunately, the primary elements are about 10-25% lignin, which is a complex volatile structure high in energy that is resistant to biochemical transformation, 35-50% cellulose, which is a polymer of sugar, the most common form of carbon on Earth, and 20-35% hemicelluloses, which are primarily composed of xylose, the second-most abundant sugar in the environment [52]. Similar to hardwoods, agricultural residues' lignocellulosic materials have a cellulose content of 41-52%, a hemicellulose content of 25-27%, a lignin content of 18-25%, and an additional 0.5-10% of extractives and inorganics [53].

# 7. Conclusion and Future Outlooks

Lignocellulosic raw materials are a viable resource, notably in the creation of bioplastics, due to their vast availability, range of surface changes, and usable mechanical characteristics of derived materials. The origin and methods used for isolating certain biomaterials, such as lactate, alkanoates, acids, or cellulose, define the precise properties and functionality of the finished goods. This review gives readers a better understanding of the numerous lignocellulose raw material sources, production processes, and forms of bioplastics. The optimal technique to generate a certain bioplastic depends on the kinds of biomass feedstock that are accessible, the yield costs of the raw materials, the recovery costs, and the infrastructure that is in place, making it difficult to pinpoint a precise manufacturing path. These factors may also influence which specific bioplastic should be developed. Green chemistry makes an effort to develop toxic-free industrial processes with lessen reaction conditions, boosted yields, and a means to salvage enzymes used. Lignocellulose has the potential to be a biomass that is utilized in large quantities to produce a number of valuable goods with a variety of uses.

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