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Review

Recent Developments of Pineapple Leaf Fiber (PALF) Utilization in the Polymer Composites—A Review

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Abstract: Plant fibers' wide availability and accessibility are the main causes of the growing interest in sustainable technologies. The two primary factors to consider while concentrating on composite materials are their low weight and highly specific features, as well as their environmental friendliness. Pineapple leaf fiber (PALF) stands out among natural fibers due to its rich cellulose content, cost-effectiveness, eco-friendliness, and good fiber strength. This review provides an intensive assessment of the surface treatment, extraction, characterization, modifications and progress, mechanical properties, and potential applications of PALF-based polymer composites. Classification of natural fibers, synthetic fibers, chemical composition, micro cellulose, nanocellulose, and cellulose-based polymer composite applications have been extensively reviewed and reported. Besides, the reviewed PALF can be extracted into natural fiber cellulose and lignin can be used as reinforcement for the development of polymer biocomposites with desirable properties. Furthermore, this review article is keen to study the biodegradation of natural fibers, lignocellulosic biopolymers, and biocomposites in soil and ocean environments. Through an evaluation of the existing literature, this review provides a detailed summary of PALF-based polymer composite material as suitable for various industrial applications, including energy generation, storage, conversion, and mulching films.

Keywords: pineapple leaf fiber; cellulose nanofibers; composites properties; biodegradation; lignocellulosic fiber

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1. Introduction

In the last few years, there has been significant emphasis on researching and prioritizing environmental security and preservation [1]. Petroleum-based primary raw materials are used in the development and synthesis of plastics. These resources, derived from organic remains buried underground for millions of years, are conventional but are depleting rapidly [2]. Their depletion not only poses environmental concerns such as pollution, climate change, and the greenhouse effect, but also health risks when they are burned [3]. The growing awareness of environmental pollution and climate change has spurred research into alternatives to these conventional resources. Natural materials such as plant waste, biomass, and cellulosic fibers are being explored as substitutes for petroleum-based products [4]. Various initiatives have been launched to raise environmental alertness, including the hosting of World Environment Day celebrations by the United Nations Environment Programme in 2018, focusing on issues like plastic pollution and promoting "Green Good Deeds". These efforts aim to educate the public about the harmful effects of single-use plastics and encourage activities like beach cleanups and waste reduction [5]. This has led to increased interest in utilizing natural sources based on biodegradable and renewable materials like plant waste and cellulosic fibers. To address environmental concerns, there is a growing interest in reinforcing plastics with fillers to enhance their strength and elasticity [6]. Natural fiber-reinforced composites, used extensively in the automotive, aerospace, and construction, industries, among others, are revolutionizing material science [7]. The use of locally available, biodegradable materials for value addition not only mitigates the environmental impact but also creates employment opportunities, particularly for marginalized communities [8]. Cellulose, both in the form of macro- and nanofibers, have garnered attention for their high strength, stiffness, biodegradability, and renewability. They are often used to reinforce composite materials, enhancing their mechanical, thermal, and biodegradation properties [9]. Using biomass to produce composites offers renewable and eco-friendly alternatives, with applications in various industries including healthcare, pharmaceuticals, aerospace, and medicine [10]. Depending on where they came from, cellulose fibers can be divided into leaf, bast, fruit, grass, stalk, and seed. In composite applications, the most utilized forms of hard fibers are bast and leaf (the hard fibrous portion covering the endosperm) [11,12]. Several plant fibers have been used as raw materials in various industries. Despite being a waste product of the agriculture industry, PALF is widely grown throughout Africa, including South Africa. The tropical fruits in the world include citrus, bananas, and pineapple [13]. Pineapple fruits are very valuable in the commercial sector, and the leaves are regarded as waste material that may be utilized to make natural fibers. However, because people are ignorant about the advantages and dangers of certain pineapple fruit pieces, the remaining portions of the fruit are still considered trash. Because fruit has a high biological and chemical oxygen demand, disposing of it contributes to environmental harm. Pineapple fibers are inexpensive, biodegradable, and renewable when derived from the leaves of pineapples. An illustration of pineapple leaves is shown in Figure 1.

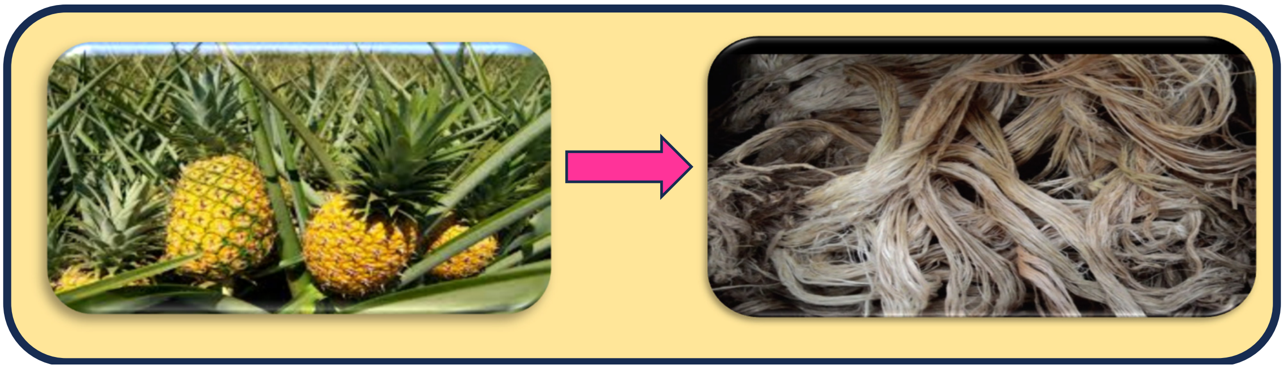

Figure 1. Image of *Ananas comosus* plants and pineapple leaf fibers.

Pineapple leaf fiber (PALF), an undervalued byproduct of pineapple cultivation, has gained attention for its renewable, cost-effective, and biodegradable properties. Despite being a waste material, PALF has potential as a reinforcement agent in composite materials, particularly in replacing pure resin to improve mechanical properties. PALF has diverse applications in textiles, cosmetics, medicine, and biopolymer coatings, and further research could unlock its potential for use in other products. However, challenges such as improving PALFs' compatibility with polymer matrices remain to be addressed. One of the main drawbacks of PALF is that, particularly at high temperatures, its hydrophilic nature makes it challenging to bond with hydrophobic materials [14]. Chemical treatments such as sodium hydroxide treatment onto dewaxed PALF improve the adhesion between PALF and polymer [15]. One example of a green composite is a bioplastic reinforced with natural fibers that is readily broken down by bacteria and enzymes [16]. Highly useful composites with superior strength are described in the current investigation. To enhance the mechanical qualities, PALF is employed as a reinforcing ingredient in a polypropylene matrix rather than pure resin. This review provides a basic overview of how PALF and other fibers function. Additionally, these fibers have a major economic advantage. In many applications, lightweight, less expensive natural fibers like pineapple leaf are presently replacing a considerable portion of synthetic fibers. In addition, PALF has potential uses in the fields of medicine, cosmetics, and biopolymers covering chemicals. Although PALF is already utilized extensively in the textile industry and in materials used in daily life, we believe that more research will improve its applicability in several other goods that are now on the market. Additionally, an economic perspective for lignocellulosic composites as well as the primary difficulties associated with the characterization of lignocellulosic fibers and PALF cellulose nanofibers and biodegradation of nature fibers, biopolymer and biocomposites in environmental conditions are discussed. The chemical composition, mechanical, and physical properties of PALF are shown in Table 1.

Table 1. Chemical, physical, and mechanical properties of PALF.

Properties	PALF
Ash content (%)	1.1%
Lignin content (%)	5–12%
Cellulose content (%)	70–82%
Density (g/cm^3)	1.53
Tensile modulus (GPa)	5.83
Tensile strength (MPa)	290.61

2. Pineapple Production in South Africa

The pineapple is part of the family *Ananas* and *Pseudoananas*, subclass Monocotyledons, with the species name Ananas comosus. Usually called the "Queen of Fruit Crops", this member of the Bromeliaceae family is regarded as one of the world's tropical fruits, comparable to bananas. Originating in Brazil, pineapples have spread to different tropical regions of the world, with major producers in Asia, South Central America, and Africa.

Pineapples are perennial herbaceous plants, reaching heights and widths of 1–2 m [17]. Cultivated mainly in coastal and tropical regions for their fruits, pineapples have a characteristic outer shell with hexagonal sections. Pineapple production generates significant waste: approximately 384,673 metric tons annually in South Africa [18]. Pineapple production in South Africa relies on dry-land farming, as irrigation is currently deemed uneconomical. The Eastern Cape focuses on processing, while Kwa-Zulu Natal supplies fresh produce markets. Unlike other agricultural sectors, there is no centralized association for pineapple growers in South Africa.

2.1. Pineapple Waste: An Environmental Risk

Disposal of pineapple fruits involves handling the remaining part of the pulp, peel, leaves, and stems left from processing. PALFs mainly consist of cellulose, lignin, and ash. Due to the fine, silky fibers in pineapple leaves, research has focused on utilizing this natural bio-adsorbent in polymer composites. The quantity of waste generated by pineapple parts is significant, comprising roughly 22,000–26,000 tons per acre annually. This waste is primarily due to inadequate management of fresh fruit, limited transportation options, or inadequate storage facilities. The pineapple industry's wastewater has high levels of biochemical oxygen demand (BOD) and chemical oxygen demand (COD), and contains Suspended Solids, which can harm aquatic life and the environment. Pineapple processing requires substantial water usage, resulting in substantial solid and liquid waste [19]. Unprocessed pineapple effluent contains high levels of carbohydrates, posing environmental risks when discharged into rivers. Despite the annual production of tons of pineapple leaf fibers, only a small portion is utilized in raw materials and energy production. Expanding the use of biocomposites might reduce waste and provide a market for agricultural products.

2.2. Classification of Natural and Synthetic Fibers

Cellulose fibers, which are composed of cellulose microfibrils wound helically and connected by an amorphous lignin matrix, make up plant fibers. Water is retained in the fibers by lignin, which also acts as a stiffener to give the stem resistance to wind and gravity pressures and defense against bacterial assault. It is thought that hemicellulose, which is present in natural fibers, acts as a compatibilizer between lignin and cellulose. Rong et al. [20] report that a fiber's cell wall is not a uniform membrane, as shown in Figure 2. In the fiber's complex, layered structure, a secondary wall is encircled by a thin primary wall, which is the first layer deposited during cell development. The thick intermediate layer, which makes up the secondary wall, controls the mechanical characteristics of the fiber. The intermediate layer is composed of helically coiled cellular microfibrils, which are generated from long-chain cellulose molecules. The microfibrillar angle is the angle that exists between the fiber axis and the microfibrils. Every fiber has a different characteristic microfibrillar angle value. Typically measuring between 10 and 30 nm in diameter, these microfibrils are composed of 30 to 100 cellulose molecules arranged in an extended chain conformation, which gives the fiber its mechanical strength.

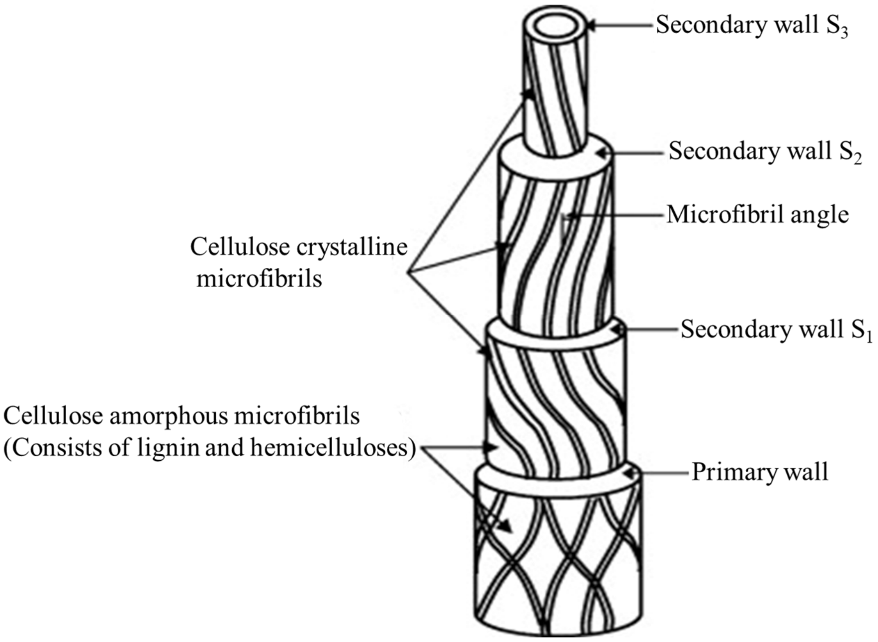

Figure 2. The natural fiber cell structural composition.

A diagram presenting a classification of the various fibers is shown in Figure 3. Numerous factors, including climate, variety, maturity, harvest degree of retting, disintegration, decortications, fiber modification, and technical procedures, influence the qualities of cellulose fibers [21]. The chemical, physical, and mechanical characteristics of natural fibers must be understood to comprehend the qualities of natural fiber composite materials. Plant fibers' mechanical, chemical, and physical qualities are greatly impacted by harvest, as well as by soil characteristics, climate, location, and weather. During fiber processing operations, including retting, scotching, bleaching, and spinning, these qualities are also impacted. The size, maturity, and the processing techniques used to remove the fibers are some of the variables that affect the quality and attributes of fibers. The internal structure and chemical makeup of fibers are linked to properties such as density, ultimate tensile strength, and initial modulus [22]. Fibers with modulus and strong tensile strength, good moldability, high durability, and recyclability possess desirable qualities.

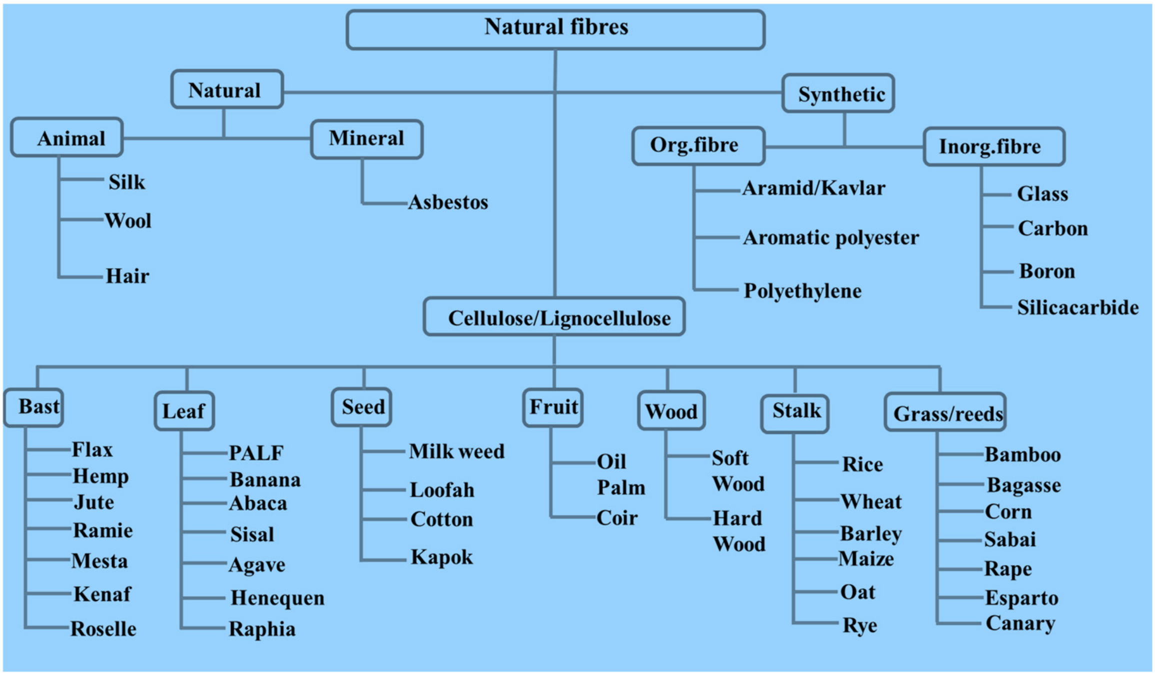

Figure 3. Classification of natural and synthetic fibers (Gurunathan et al. [21]).

The production process for the composites is optimized in terms of melding time, pressure, and temperature. Preheating the natural fibers to lower the moisture content is frequently required before the polymer composites can be processed. Elevated temperatures cause cellulose degradation, which hurts the composites' mechanical characteristics. Tensile strength is reduced by fiber agglomeration, which is brought on by ineffective fiber dispersion in the matrix. Many earlier studies on natural fiber composites concentrated on thermoset and thermoplastic matrices as well as reinforcements such as sisal, flax, hemp, jute, and pineapple leaf. To create completely biodegradable and biocomposites, some of these composites have been constructed using a matrix composed of cellulose, starch, and lactic acid derivatives [23]. In several applications in the packaging, construction, automotive, and transportation industries, natural fiber-reinforced polymer composites (NFRPCs) have already been shown to be a feasible substitute for synthetic fiber-reinforced polymer composites, or SFRPCs. Varieties of natural fibers have been found via ongoing research to increase the mechanical strength of polymer composites. Composite materials made with natural fibers are lighter than SFRPCs while maintaining the same mechanical strength. The manufacturing of natural fibers is linked to fewer emissions than that of synthetic fibers, and they are biodegradable.

Glass fiber polyester composites (GFPCs), pineapple leaf epoxy composites (PLECs), and pineapple leaf fiber polyester composites (PLPCs) were tested for mechanical qualities. The outcomes demonstrated that PLECs outperformed GFPCs and PLPCs in terms of mechanical qualities, especially at 40% fiber loading. Compared to fiberglass and pineapple fiber-reinforced polyester composites, pineapple fiber-reinforced epoxy composites exhibited superior flexural and tensile capabilities [24]. The commercially available composite made of natural fibers outperforms glass-reinforced composites in a variety of applications. Table 2 presents a comparison of several metrics between glass and natural fibers. While glass fibers in synthetic composites are very expensive and have a high density, flax fibers are a natural fiber with a relatively low density and a lower cost [25].

Table 2. Comparison between glass fiber and natural fibers.

Properties	Natural Fiber	Glass Fiber
Separation	Easy	Difficult
Availability	Very high	Low
Density	Low	Double
Cost	Low	High
CO_2 emissions	Low	High
CO emissions	Low	High
SOx emissions	Low	High
NOx emissions	Low	High
Phosphates to water	High	Low
Nitrates to water	High	Low
BOD to water	Low	High
COD to water	Low	High
Energy consumption	Low	High
Renewability	Yes	No
Recyclability	Yes	No
Energy consumption	Low	High
Distribution	Wide	Wide
Health risk when inhaled	No	Yes
Degradable	Biodegradable	Non-biodegradable
Distribution	Wide	Wide
End of life	Recovered energy	Waste
Pollution	Reduce	Less to natural Fibers

3. Classifications of Cellulose and Structure

Cellulose stands out as an abundant form of global biomass and holds diverse applications across modern industries. Functioning as a natural polymer, it comprises long chains formed by the linking of small molecules, with β-linked D-glucose and sugar as the constituents. While cotton fiber contains cellulose in an almost pure form, in materials like plant leaves, wood, and stalks, it is found alongside other substances such as lignin and hemicelluloses [26]. The hydroxyl equatorial positions in the cellulose chain promote hydrogen bonding, which results in the production of highly ordered structural formations. Despite variations in crystalline regions, interchain hydrogen bonds contribute to the fiber's good strength and insolubility in most solvents.

Cellulose, constituting the primary structural component of taller plants' cell walls, is prevalent in various natural sources including cotton (95%), pineapple leaf fiber (82%), jute (70%), wood (50%), and flax (80%). Agricultural byproducts such as corn stalks, sugarcane, and various straws offer sources for cellulose pulps. Comprising β-1,4-glycosidic linked D-glucose units, cellulose serves as the foundation for nanocellulose, a material that is gaining traction in research due to its favorable physicochemical properties like biocompatibility, availability, non-toxicity, hydrophilicity, low cost, and eco-friendliness. These nanofibers hold great potential for use as reinforcement in the development of nanocomposites. Several results have focused on isolating and characterizing nanocellulose fibers from many sources, with extraction methods ranging from simple mechanical processes to combined chemical and mechanical methods [27]. Nanocellulose finds application across several domains including automotive, structural components, and packaging, owing to its versatile characteristics. Nanocellulose is typically classified into three types based on its structure and properties, as shown in Figure 4.

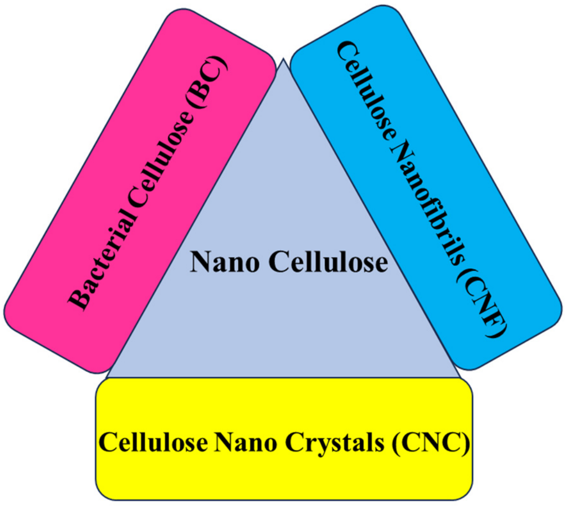

Figure 4. Nanocellulose is typically classified into three types based on its structure and properties.

3.1. Classifications of Nanocellulose

The reduction in the size of cellulose chains, either in diameter, length, or both, can be achieved through several methods such as mechanical, enzymatic, and chemical treatments. According to Klemm et al., for a material to be classified as nanocellulose as shown in Table 3, it must have at least one dimension within the nano range. Thus, nanocrystalline cellulose (CNC), bacterial cellulose (BC), and nanofibrillated cellulose (NFC) are recognized as the three main types of nanocellulose. The cellulose nanofibrils typically consist of bundles of fibers with diameters in the nanometer range and lengths in micrometers. Unlike CNC, cellulose nanofibrils can exist in both crystalline and amorphous forms, and they can be produced through both mechanical and chemical treatments. In the past, there has not been uniformity in the nomenclature of nanocellulose materials. Klemm et al. used terms like MFC (microfibrillated cellulose), NCC (nanocrystalline cellulose), and BNC (Bacterial Nanocellulose) [28,29]. They suggested that while "microfibrillated cellulose" has been widely adopted in scientific and commercial literature, terms like NCC and BNC are simple and descriptive. Over time, it is possible that the term "nanofibrillated cellulose" will become more prevalent, leading to more consistent terminology for nanocellulose.

Table 3. Types of nanocelluloses, production methods thereof, their building block dimensions, and mechanical strength.

Types of Nanocelluloses	**Typical Sources**	**Method of Production**	**Average Dimensions**	**Young's Modulus (GPa)**	
Cellulose nanocrystals (CNC)	Wood, cotton, tunicate, Ramie, bacterial cellulose, bamboo	Acid Hydrolysis	Diameter 3–50 nm Length: 30 nm to 300 nm (wood and cotton based) 100 nm to several microns (tunicate and bacterial cellulose based)	143 ± 0.9 GPa	[30]
Bacterial Cellulose (BC)	Acetobacter/Glucobacter xylinum	Biosynthesis of glucose and alcohol	Diameter 10–100 nm Length: Mostly several tens of micrometers up to a mm.	78 ± 17 GPa	[31]
Nanofibrillated Cellulose (NFC)	Wood, cotton, potato hemp, flax	High-pressure homogenization, microfluidization, grinding, cryocrushing	Diameter 5–60 nm Length: several microns regardless of the cellulose source	17.2 ± 1.2 GPa	[32]

Chemical Composition

According to TAPPI (Technical Association of Pulp and Paper Industry) standards from 1991, the chemical elements and extractives of PALF were analyzed across different fiber sources, ages, and climatic conditions. The extraction process may influence the variation in the chemical composition of PALF, as noted by Khalil et al. [33]. Furthermore, transmission electron microscopy reveals distinct layers within PALF cell walls, including primary, secondary, and tertiary layers. Chemical analysis reveals various constituents in PALF, such as α-cellulose, lignin and wax, pectin, ash content, and the crystallinity of α-cellulose, as reported by Asim et al. [34]. PALF is characterized by a high α-cellulose content (82%), with relatively low levels of hemicelluloses (12%), pectins (1.1%), and lignin (3%), as stated by Rahman et.al. Compared to several natural fibers like oil palm fronds and banana stem fibers, PALF exhibits a higher cellulose content, which correlates with the higher fruit weight [35]. The chemical composition of fibers directly influences their performance, as emphasized by Wirawan et al. [36]. Furthermore, Anon et al. reported that the T204 cm-88 and T264 cm-88 procedures were used to determine the extractive content. For 6 h, T204 cm-97 (Anon et. al) was the protocol for determining the solubility of ethanol in benzoene. The method of Le Wise et al. was used to quantify holocellulose, whereas T203 cm-74 was used to determine the α-cellulose base. The T222 cm-88 technique was used to measure the lignin content. The residue that remains after organic matter burns at a temperature of 52.5 ± 25 °C is referred to as the inorganic ingredient of lignocellulosic material or its ash content. The process described in the T211 cm-93 technique was used to determine the ash content [37–39].

4. Extraction of PALF

Various extraction methods exist for obtaining cellulose nanofibers, including mechanical techniques like grinding, cryo-crushing with liquid nitrogen, and high-pressure homogenization. Chemical processes such as alkali and enzymatic hydrolysis can also be employed before mechanical methods to enhance accessibility. Despite the excellent mechanical strength of pineapple fibers, their full potential remains underutilized due to a lack of understanding. These fibers hold promise for diverse applications including artificial fibers, sound absorption, thermal insulation, and drug delivery. The high tensile strength of plant fibers is attributed to cellulose nanofibers within the cell wall. Extracting these nanofibers from the cell wall is crucial for measuring their properties, though this process may risk chemical or mechanical damage. Once isolated, the reinforcing potential of these nanofibers in biomedical applications can be evaluated. Various methods are available for extracting PALF from pineapple leaves. One method involves the use of a scrapping machine, and the components of the machine are described by Kannojiya et al. [40]. The feed roller feeds the leaf into the machine, where it passes through the leaf scratching roller, which removes the upper waxy layer. The densely connected blade serrated roller processes the leaves last, crushing them and creating gaps that allow retting microorganisms to enter. It cuts the top layer of the leaf and eliminates the waxy layer as shown in Figure 5. PALF exhibits strength and stiffness but is hydrophilic due to its high cellulose content. The extraction methods for PALF include mechanical and retting methods, resulting in a yield of approximately 2% to 3% fibers from fresh leaves. PALF comprises various chemical constituents, including polysaccharides, lignin, wax, color pigments, and other substances. Given its unique properties, PALF presents an excellent alternative raw material for reinforcing composite matrices in various industries [41].

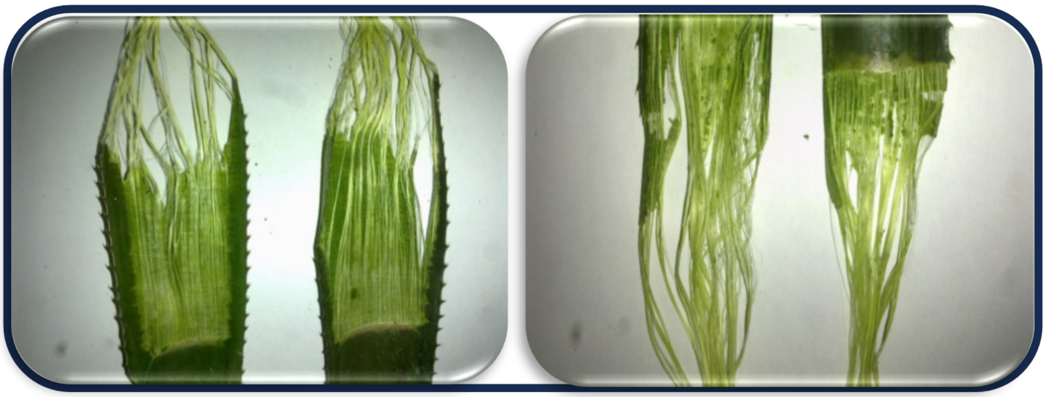

Figure 5. Shows the view of the wet fiber obtained after the extraction process.

4.1. Retting of PALF

During the retting process, small packages of scratched PALF are immersed in water containing a solution ratio of 1:20, along with 0.5% urea to expedite retting reactions. Regular checks are conducted by hand to ensure the fibers are loosened, facilitating the extraction of chemical constituents such as pentosans (glucose) monomers, lignin, fats, wax, and pectin from the materials in the water tank. Following the retting process, fibers are mechanically segregated by washing in water, and removed fibers are hung to air-dry [42]. To extract PALF from finely chopped fresh pineapple leaves, ball and disk mills can be used. While these procedures are more difficult than standard approaches, they also provide smaller fiber sizes and higher fiber production [43–45]. PALF has been demonstrated to be a viable substitute for synthetic fibers due to its economic and renewable source. The strength of natural fibers contributes to enhancing the physical and mechanical properties of the polymer composite.

4.2. Acid Hydrolysis

Among all the chemical techniques, acid hydrolysis is the most preferable method for producing nanocellulose [46]. Acid hydrolysis is a surface phenomenon that targets the amorphous parts of cellulose, increasing the crystallinity of the structure. Various acids such as sulfuric acid (H_2SO_4), formic acid (CH_2O_2), hydrochloric acid (HCl), and phosphoric acid (H_3PO_4), as well as organic acids, can be used for hydrolysis [47]. It is essential to control the reaction time and temperature, since the characteristics of nanocellulose are determined by the variation of these parameters. Therefore, the extraction of the final yield of nanocellulose is 76%. Figure 6 illustrates the effects of these controlling factors on the hydrolysis of cellulose. Sulfuric acid is generally preferred because it imparts negative charges on particles in solution, resulting in a more stable suspension, despite its thermostability.

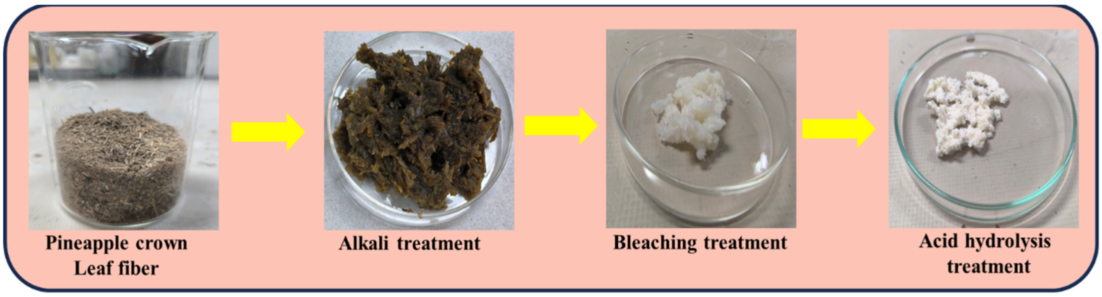

Figure 6. Illustration showing how the nanocrystalline cellulose is separated from pineapple crown leaf.

4.3. Pre-Treatments of PALF

Hot water and an alkali pre-treatment were applied to the crushed pineapple leaves before the fibers were separated.

4.3.1. Hot Water Treatment

To perform pre-treatments, pineapple leaves were boiled for several durations of time (15, 30, and 45 min) at temperatures of 50, 70, and 90 °C. The fibers were then decorticated after the PALF was oven-dried for 24 h at 60 °C. After that, the physical characteristics of the isolated fibers were assessed.

4.3.2. Alkali Treatment

Three different concentrations (2, 4, and 6% w/w) and treatment times (2, 4, and 6 h) of alkali (NaOH) solutions were applied to whole pineapple leaves. After a 24 h oven-drying process at 60 °C, the fibers were extracted from the leaves (final yield of cellulose 87%). Subsequently, these fibers' physical characteristics were evaluated [48–51].

5. Characterization of PALF Cellulose Nanofibers Confirmed by FTIR Spectroscopy and FESEM, TEM Microscopy

FTIR spectroscopy is a valuable technique used to observe functional groups in natural fibers, including hydroxyl, carbonyl, vinyl, and ketone groups, and others. It helps functional groups change the chemical composition of natural fibers after and before chemical treatments. Table 4 presents the typical FTIR spectra of various untreated natural fibers. The characteristic hydroxyl group is common to all fibers, typically appearing with intensity between 3336 and 3442 cm^{-1}. Additionally, untreated fibers exhibit familiar peaks corresponding to C–H stretching and C–O stretching at 2924 and 1741 cm^{-1}, respectively. FTIR spectroscopy aids in identifying these characteristic peaks, allowing researchers to analyze the chemical composition of natural fibers and monitor any changes resulting from treatments or processing. Furthermore, the size and morphology of CNFs isolated from PALF were evaluated by FESEM and TEM. Cellulose nanofiber has a large amount of hydroxyl groups. This tends to cause accumulation to occur as the dark-colored networks in Figures 7 and 8, as reported by Klinthoopthamrong et al. and Ravindran et al. [52,53].

Table 4. The typical FTIR spectra of various untreated natural fibers.

Bond/Stretching	**–OH**	**C–H**	**C=O**	**C=C**	**C–H**	**C–H**
PALF (cm^{-1})	3349.9	2903.8	1737.4	1608.3	1374.2	-
Abaca (cm^{-1})	3446	2915	1740	-	1310	-
Hemp (cm^{-1})	3448	2920.5	-	1654	1384.1	-
Sisal (cm^{-1})	3447.2	2924.2	1736.5	1653.9	1384.1	1259.9
Jute (cm^{-1})	3447.9	2918.8	1737.2	1653.8	1384.1	1255.6
Kapok (cm^{-1})	3419.7	2918.1	1741.1	1596.1	1383.6	1245.5
Kenaf (cm^{-1})	3338	2899	1736	-	-	-
Oil palm fiber (cm^{-1})	3450	2850	1735	1606	-	-
Flax (cm^{-1})	3325	2900	1720	1615	1335	1228
Cotton (cm^{-1})	3330	2896	-	1622	1365	-
Banana (cm^{-1})	3379	2919	-	1602	1374	1238
Coir (cm^{-1})	3338	2845	-	1600	-	1252
Henequen (cm^{-1})	3431	2925	1741	-	-	1254
Bamboo palp (cm^{-1})	3400	2900	1750	1600	-	-
Elephant grass (cm^{-1})	3332	2888	1720	-	1319	1287
Rice straw (cm^{-1})	3446	2922	-	1642	1375	-
Sugar palp (cm^{-1})	3400	2931	1720	1600	-	-
Wheat straw (cm^{-1})	3422	2927	-	-	1388	1251
Ramie (cm^{-1})	3400	2900	1740	1630	-	1270

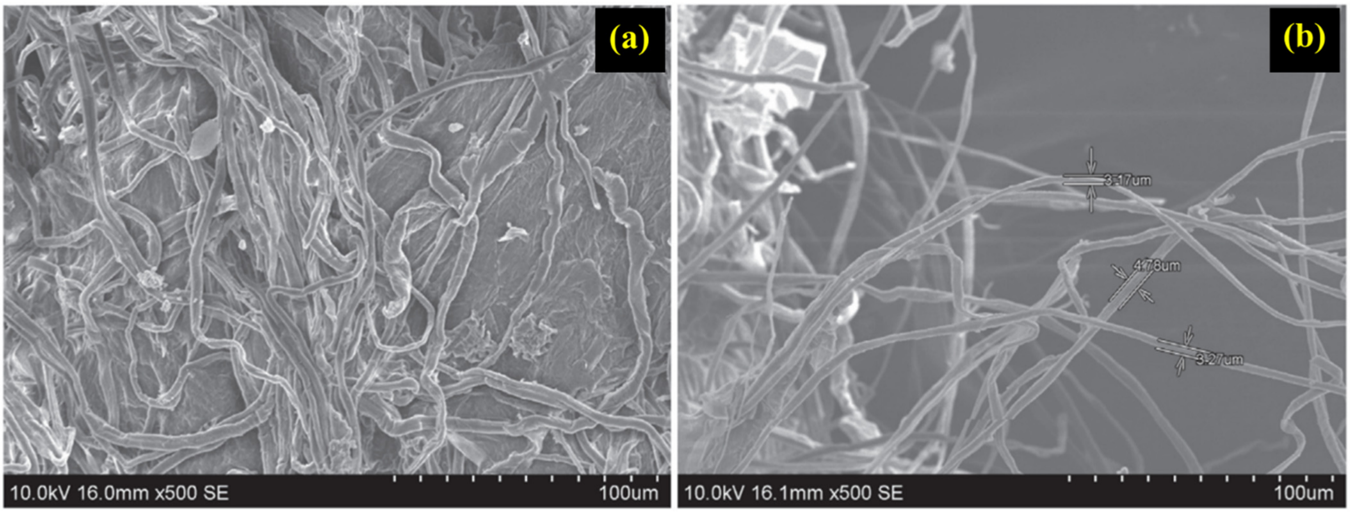

Figure 7. FESEM of different treatments of pineapple leaves ((**a**) Bleached pineapple leaves (raw), (**b**) hydrolyzed pineapple leaves) (Ravindran et al. [53]).

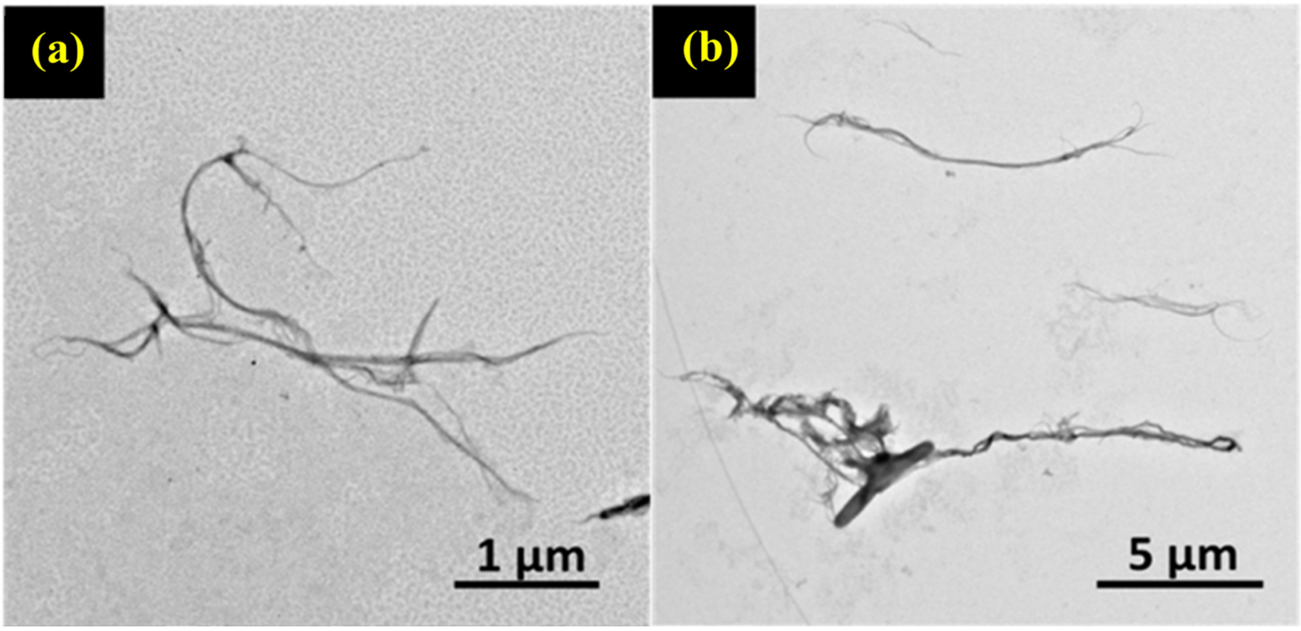

Figure 8. (**a**,**b**) TEM image of cellulose nanofiber (Klinthoopthamrong et al. [52]).

6. Micro- and Nanocellulose-Based Polymer Composites

The development of materials that are eco-friendly, sustainable, and easily accessible has attracted a lot of interest recently because of environmental issues and the depletion of petroleum products. Exploring alternatives to petroleum-based polymers by extracting cellulose from natural sources such as animals, plants, and microorganisms offers an excellent opportunity to protect the environment [54]. However, natural cellulosic polymers have certain limitations in their properties. Enhancing these properties broadens the potential applications of natural fibers. Typically, improving mechanical properties also enhances chemical and thermal properties [55]. This is because modifications to natural fibers improve particle homogeneity, leading to stronger chemical bonding. Important mechanical tests to assess the enhancements from these modifications include tensile strength, flexural strength, elongation, stiffness, and tensile modulus [56]. Investigating the films with different PNC concentrations (1, 2, 4, and 8 wt%) was the goal of Sukwijit et al. Their findings showed that whereas PNC concentrations decreased light transmission, they enhanced the composite films' water vapor permeability (WVP) and biodegradability. When compared to PLA films, films with 4PNC, specifically at 4 wt% (PLA/4PNC-4), showed an 11.18% improvement in elongation at break. Additionally, these films had increased barrier qualities, indicating better soil moisture retention, and lower light penetration, which correlates with decreased weed growth and WVP. Furthermore, PLA sheets containing 4PNC showed better stability against thermal deterioration compared to those containing unmodified PNC, indicating increased heat resistance [57]. Pornbencha, K. and Moreno et al. examined the mechanical/thermal, gas permeability, and UV absorption characteristics of PLA nanocomposite films that were made using a solution-casting technique. Additionally, the dispersion of fillers on the PLA matrix was significantly enhanced by the functionalization of cinnamonate on CNCs. The PLA sheets with three wt% Cin-CNCs showed excellent transparency and UV absorption in the visible spectrum. However, PLA films containing pure CNCs showed no UV-shielding characteristics. In comparison to plain PLA, the mechanical characteristics showed that adding 3 wt% Cin-CNCs to PLA boosted its tensile strength and Young's modulus by 70% and 37%, respectively [58,59]. Fitriani et al. studied the improved film formulation and discovered improved mechanical characteristics with a low thickness value and high elongation. The results demonstrated the potential for good model validation and effective use in food packaging applications using an improved bionanocomposite film formulation based on polyvinyl alcohol with glycerol and cellulose nanocrystals from pineapple crown leaves [60]. Consequently, many researchers have investigated (as Table 5 shows) the mechanical performance of biocomposites made from different natural fibers and biopolymer resins such as polylactic acid (PLA), thermoplastic starch (TPS), Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), polybutylene succinate (PBS), and Polyhydroxy Acids (PHAs). Since cotton fibers have high cellulose content, they offer good reinforcement to starch biocomposite to improve its mechanical performance and reduce water absorption compared to other natural fibers biocomposites. Cellulose fibers' and nanofibers' physical–chemical properties have roles in various polymer matrixes.

Table 5. Illustration of the effect of micro- and nanocellulose on biopolymer composites.

Source of Nanocrystalline Cellulose	Source of Biopolymer	The Effect of the Reinforcement	Ref.
PALF	PLA	The PLA film with 4PNC exhibited increased thermal degradation stability, indicating better heat resistance suitable for mulch film usage.	[57]
PALF	PLA	The mechanical properties revealed that adding 3 wt% Cin-CNCs to PLA increased its tensile strength and Young's modulus by 70% and 37%, respectively	[58]
PALF	PLA	The ultimate tensile strain values	[59]
PALF	PVA	Improved bionanocomposite film compositions have greater characteristics and properties that can be utilized as packaging in the food industry	[60]
PALF	TPS	Barrier properties were enhanced.	[61]
Microcrystalline cellulose	PLA	Acetylation can improve the performance of the composite by enabling linkages between carbonyl groups, helping to establish a good stress transfer between the fiber and the matrix.	[62]
Microcrystalline cellulose	PLA	The impact and the elongation at break increased from 0.864 to 2.64 kJ, and 22 from 11% to 106.0%, respectively.	[63]
Microcrystalline cellulose	PLA	The modified PLA nanocomposite is considered a practical candidate for hard tissue engineering applications according to cytotoxicity results.	[64]

Table 5. *Cont.*

Source of Nanocrystalline Cellulose	Source of Biopolymer	The Effect of the Reinforcement	Ref.
Microcrystalline cellulose	PBS/PLA	Thermal stability, storage modulus, glass translation temperature of nanocomposites increased.	[65]
Maize starch	PLA	Provided better filler dispersion and interaction with the matrix.	[66]
Banana waste	PLA	Dynamic mechanical thermal analysis (DMTA) exhibited a 30 to 50% reduction in storage modulus (stiffness) when compared to neat PLA.	[67]
Bamboo pulp	PHAs	The elastic and crystallinity properties of the nanocomposites improved with the increase in NCC loadings.	[68]
Kenaf	PHAs	The conductivity of the polymer nanocomposites improved.	[69]
Cotton	starch	Water absorption of the coated paper composite decreased by 50% at 5 wt% NCC concentration.	[70]
Kenaf fibers	starch	The water absorption by the biocomposite and the water sensitivity decreased.	[71]
Sugar palm	starch	Good compatibility between the nanofibrillated cellulose and the sugar palm fiber; the composition created intermolecular hydrogen bonds between them.	[72]
Cellulose nanocrystals	PHAs	Kraft paper was used to reinforce polyhydroxyalkanoate (PHAs). There was an increase in the crystallinity and stiffness of the nanocomposites	[73]
Cellulose nanocrystals	PHBV	Limited reinforcement was observed despite enhanced dispersion relative to the neat PHBV matrix due to the hydrophobization surface of NCC (plants).	[74]
Nanocrystalline cellulose	PBS	Nanocomposites from biodegradable poly (butylene succinate) (PBS), blended with nanofillers chitin whiskers. The tensile strength increased from 23.2 MPa to 32.9 MPa	[75]
Microcrystalline cellulose	PLA	NCC-reinforced PLA exhibited improvements in its thermal, mechanical, and UV barrier properties.	[76]

6.1. Applications of the Extracted PALF Cellulose Nanofibers

6.1.1. Drug Delivery

Drug delivery involves administering a drug compound to achieve a therapeutic effect [76]. Cellulose has a well-documented history of successful use in approved drugs in the U.S. For example, cellulose acetate (CA) has been successfully used in several HIV drugs, antibiotics, pain relievers, and flavonoids, among others. The application of cellulose nanofibers (CNFs) in the biomedical industry, particularly in drug delivery, is considered highly promising. Effective drug delivery systems possess key features such as targeting, improved solubility, controlled drug release, drug stability, and therapeutic effect. Some efforts have been focused on utilizing CNFs as suitable pharmaceutical excipients and carriers due to their colloidal stability, and negative surface charge. These properties allow for the loading of charged/neutral drugs, control of active compound release, and delivery of genes to target cells [77].

6.1.2. PALF Uses and Prospects for the Future

The diversified applications of PALF have expanded over the years, presenting promising prospects as depicted in Figure 9. Traditionally, PALF has been utilized in making threads for textile fabrics, but its potential applications now span various industries. Currently, PALF finds applications in textile manufacturing, sports equipment, automobiles, mats, and more. The surface-modified PALF is seeing increasing use in machinery parts such as conveyor belt cords, transmission cloth, and industrial cloths. Its suitability for carpet-making is notable due to its favorable chemical processing, dyeing, and esthetically pleasing fabric. In addition to its traditional uses, PALF is being explored for novel applications in the paper manufacturing industry [78]. Furthermore, its potential extends to industries such as cosmetics, medicine, and biopolymer coatings for chemicals. PALF stands out among natural fibers with its high cellulose content of 82%, comparable density to other natural fibers, and exceptional Young's modulus, resulting in high tensile strength. These properties make it suitable for applications in building and construction materials, automotive components, and furniture manufacturing. As research and development continue, PALF versatility and potential in various industries are expected to further unfold.

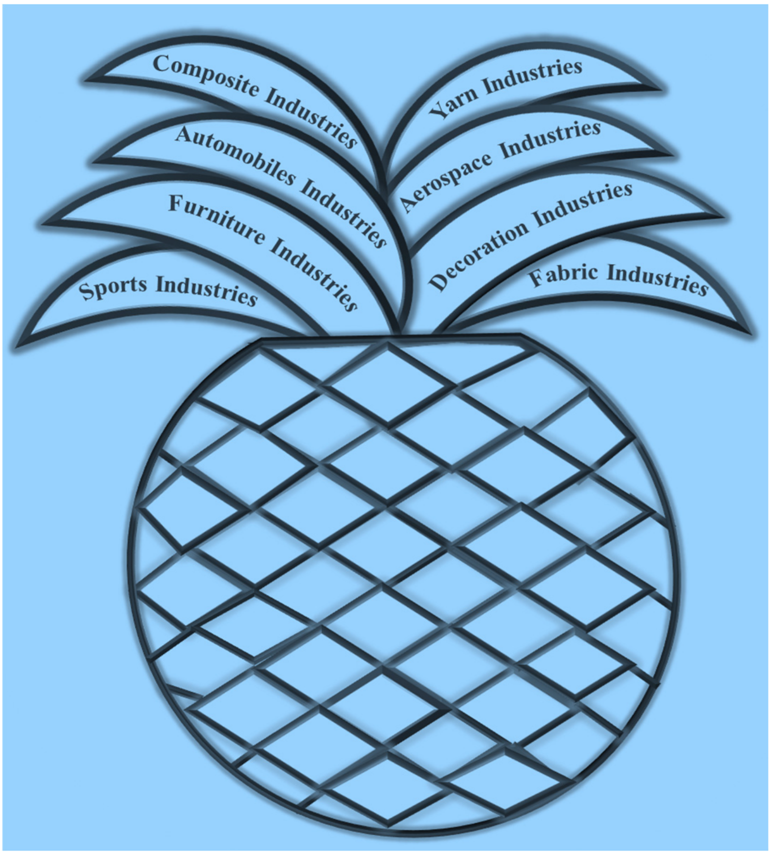

Figure 9. Several applications for pineapple leaves in the present and the future.

7. PALF-Based Biocomposites

The environmentally friendly characteristics of PALF-reinforced biodegradable composites make them suitable for usage in industrial applications. Non-biodegradable polymers like polypropylene (PP), polyethylene (PE), etc., are reinforced with PALFs of different lengths and fiber loadings. Using natural resource monomers to produce biodegradable polymers is a novel approach to the production of biodegradable polymers from renewable resources. Given that PLAF is derived from agricultural products and easily biodegrades, it is one of the most promising polymers in this area. Biodegradable polymers, include polylactic acid (PLA and polyhydroxy butyrate-co hydroxyvalerate (PHBV), etc.

1. Non-biodegradable biocomposites: PALF composite materials can be made into both petroleum-based plastics or renewable-based plastics, but if both polymers are conventional non-biodegradable polymers (PE, PP, and nylon) and biobased non-biodegradable polymers (Bio-PE, Bio-PP, and Bio-nylon), then the cellulose fiber composites are non-biodegradable biocomposites.
2. Biodegradable biocomposites: if PALF natural fiber composite is made from biodegradable polymer matrix incorporating petroleum-based biodegradable polymers (PBAT,

PCL) and/or renewable-based biodegradable polymers (PLA, starch, PHA), then these cellulose fiber composites are biodegradable.

PALF fiber composites can also be defined as biocomposites and whole green composites based on the origins of petroleum- and biobased polymers:

1. Biocomposites: PALF fiber-based composites are made from petrol-based polymers, but the biodegradability of these biocomposites depends on the polymeric matrix used. For example: cellulose fibers create a PP-based non-biodegradable composite, whereas cellulose results in BAT-based biodegradable composites.
2. Whole green composites: PALF-based composites are made from renewable resource-based polymers, but the biodegradability of these green composites depends on the polymeric matrix used. For example: cellulose—PLA-based biodegradable composites; cellulose—bio-nylon-based non-biodegradable composites, etc.

The focus on natural fibers as alternatives to glass fibers has intensified among researchers and industries due to the growing concerns about environmental issues. This trend is expected to accelerate the utilization of natural fibers in various applications in the coming decades. In recent years, PALF has emerged as an effective reinforcement material in polymer matrices to develop polymer composites with enhanced mechanical and physical properties. The remarkable mechanical and physical properties of individual PALFs translate into superior performance in the final composite products. Several studies have been conducted to reinforce PALF with various types of matrices, including thermoset, thermoplastic, and biodegradable plastics [79]. These efforts aim to leverage the unique properties of PALF to create composite materials that are not only environmentally friendly but also exhibit excellent mechanical properties suitable for a wide range of applications.

PALF-based thermoset, thermoplastic, and biodegradable polymer composites.

The utilization of PALF in polymer composites has been extensively studied with various types of polymer resins, each offering unique properties and applications. In comparison to other PALF-based composite materials, researchers have found that reinforcing PALF with thermoset, thermoplastic, and biodegradable polymers produce composites with exceptional stiffness and strength. The PALF-reinforced thermoset resins have been examined by several researchers (Table 6) After analyzing all resins, Sundeep et al.'s investigation into thermoset-based resins revealed that the configuration of composite layers significantly affected the materials' mechanical properties. The results of the experiment show that the alternate layer configuration of pineapple leaf fiber (P) and E-Glass fiber (G) had remarkable mechanical capabilities, especially in the areas of interlaminar shear strength, tensile strength, and flexural strength [80]. According to research on the tensile test by Venkatesan et al., increasing PALF improves the flexural modulus in the flexural test. In an impact test, the PALF rises but the impact strength decreases; in a compression test, when the PALF rises, the compressive strength increases at the same rate [81]. The tensile strength and flexural strength of the treated fiber-reinforced composite are, respectively, 132.12% and 109.58% greater than those of the untreated fiber-reinforced composite, according to research by Kumar et al. The incorporation of wire mesh improved the tensile strength by 11.22% but the bending strength decreased by 8% [82]. According to research published by Baigh et al., jute and PALF together increase tensile and flexural properties, exceeding those of pure PALF composites and falling short of pure jute composites. JFRP exhibited the maximum tensile strength of 35.16 MPa, whereas the hybrid composite had the highest tensile strength of 32.16 MPa. The hybrid composite 4P5J-2 has the highest tensile modulus (1.315 GPa) [83].

From Table 7, it was observed that the authors have studied the effect of strain rate, temperature, fiber orientation, fiber type, fiber loading, and chemical treatment on the PALF/thermoplastic resins. Studies have shown that PALF-reinforced polyethylene composites exhibit high performance, with techniques such as melt and solution mixing being used for their preparation. The solution-mixing technique has been found to yield better tensile strength compared to melt mixing. Factors such as fiber size, loading percentage, and orientation have been investigated, with a fiber length of 6 mm being deemed suitable for PALF-reinforced low-density polyethylene (LDPE) composites [84]. Additionally, longitudinal fiber orientation has been shown to offer better mechanical properties compared to random and transverse orientations. PALF-LDPE composites are eco-friendly and demonstrate superior performance compared to other cellulose-fiber-reinforced LDPE systems. The development of polyester composites reinforced with pineapple leaf fiber (PALF) at a weight fraction of 6 wt% was examined by Nurunnabi et al. [85] Utilizing synthesized graphitic carbon nitride (g-C3N4) as a filler, the mechanical and thermal characteristics of the resulting composites were enhanced at various weight fractions, including 0.25%, 0.50%, 0.75%, and 1.0% [86]. The materials with the highest tensile strength and elongation among the evaluated samples were found to be composed of 15% PP-PALF and 30% PLA-PALF. However, because of their greater resistance to deformation within the elastic range, 15% PP-PALF and 50% PLA-PALF were rated top in terms of elastic modulus. Longer PALF and PP provided a homogenous fiber-matrix distribution, which resulted in the maximum tensile strength, tensile modulus, bending strength, bending modulus, and impact strength, according to research by Zainudin AS and Rahman et al. [87–90]. Furthermore, Sundeep et al. observed that a composite with 16 mm of fiber length may achieve a maximum hardness of 77.8 ± 0.6 HV, as well as a higher coefficient of friction of 0.73 at 20 N at 1 m/s and enhanced wear resistance of 0.0065 mm^3/Nm at 10 N [91].

The environmentally friendly properties of PALF-reinforced biodegradable composites make them suitable for usage in non-structural applications. The reported investigation of biodegradable/PALF resins with appreciable improvements in mechanical characteristics is summarized in Table 8. GS Rao et al. The mechanical and thermal characteristics of unidirectional, biodegradable, environmentally friendly “green” composites composed of pineapple fibers and poly(hydroxybutyrate-co-valerate) (PHBV) resin were studied by Luo, Smitthipong et al. Both longitudinally and transversely, the tensile and flexural characteristics of the “green” composites with varying fiber contents were tested. The “green” composites’ tensile and flexural strengths are much higher in the longitudinal direction and significantly lower in the transverse direction when compared to those of virgin resin. TPS with PALF/PLA composite provides better mechanical properties and water resistance than the TPS/PLA blend, but it exhibits the same flow behavior as PLA alone [92–95]. The mechanical and morphological properties of a green composite reinforced with PALF are investigated in a study by Rao et al. in various environmental settings. A 20% fiber loading and a 3 mm length were used to create the PALF/PLA composite. Following production, the PALF/PLA composites were subjected to a range of environmental factors, such as water, soil, refrigeration, and room temperature. For four weeks, the effects of these various environments on the mechanical characteristics were examined scientifically [96]. The impact of incorporating dammar gum (DG), a renewable and biodegradable binder, on the mechanical characteristics of tapioca biopolymer (TBP) reinforced with short pineapple leaf fiber (PALF). By adopting an internal mixing technique and compression molding, samples with different TBP percentages were produced with DG concentrations (10%, 20%, 30%, and 40% by weight) and a constant 30% PALF composition. Biodegradable polymers can be classified based on various criteria such as their properties and synthesis methodology the classification of biodegradable polymers depends on the synthesis as shown in Figure 10. The findings demonstrated that PALF-TBP with 10% DG had the best mechanical characteristics, as studied by Rahman et al. and Zainudin AS [97,98] Furthermore, many researchers have investigated the mechanical characteristics of PALF based on polymer resins such as PALF-epoxy, Polyester (PS), Polypropylene (PP), Polyhydroxybutyrate-co-hydroxy valerate (PHBV), polylactic acid (PLA), and thermoplastic starch (TPS) as shown in Tables 6–8.

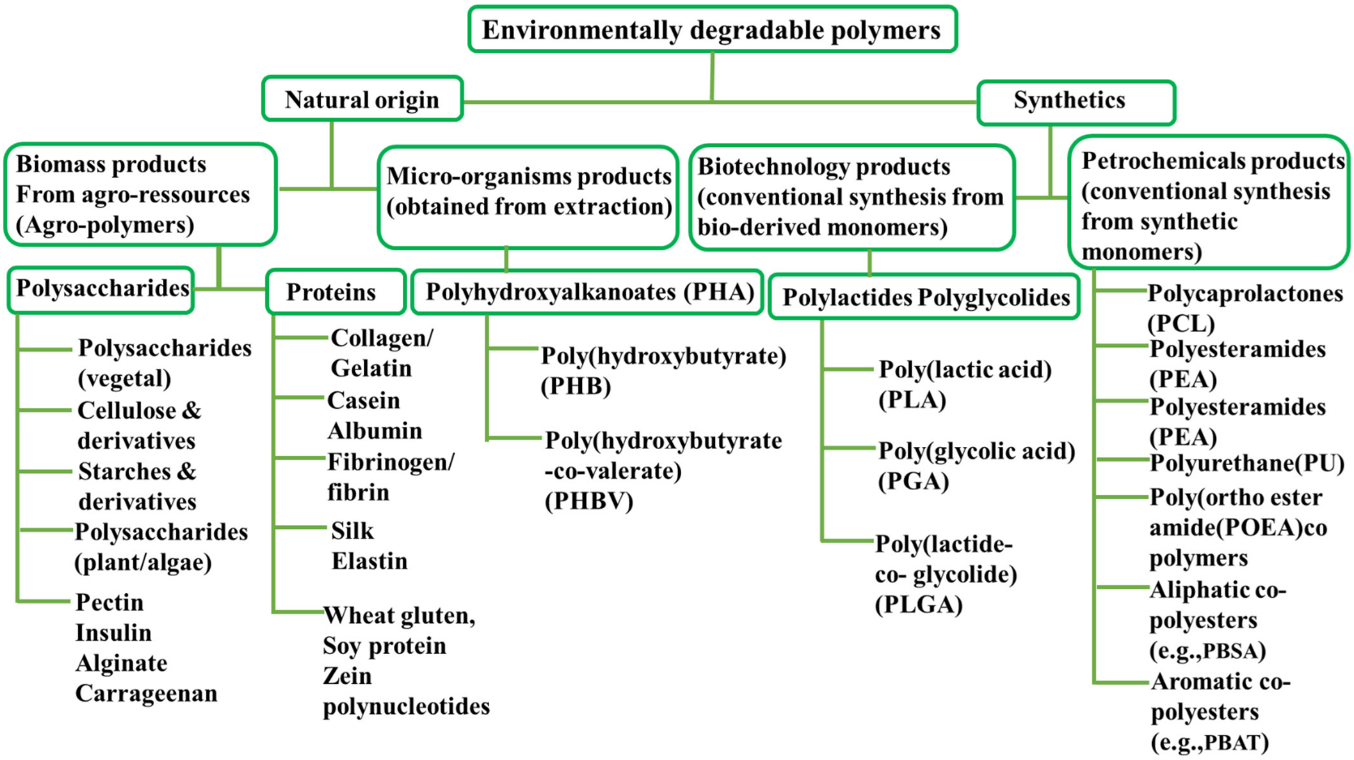

Figure 10. Classifications of the different biodegradable polymers.

Table 6. Impact of factors and specifications on the mechanical characteristics of thermoset polymer/PALF composites.

Parameter	Material	Fiber Length	Fabrication Method	Feed Rate	Observations	Ref.
Fiber loading	PALF/epoxy	160 mm	Hand layup	TS = 0.5 mm/min	Increase in longitudinal Young's modulus, transverse and longitudinal shear modulus observed	[99]
Fiber loading	PALF/polyester	-	Hand layup	TS = 100 mm/min	(1) TS and TM was comparatively high during NaOH treatment	[100]
Fiber loading	PALF/epoxy	-	Hand layup	FS = 5 mm/min	Linear increase in FS with the increase in volume fraction	[101]
Fiber loading	PALF/epoxy	35 mm	Hand layup	-	Average TS and FS found to increase with increasing fiber volume fraction	[102]
Fiber loading	PALF/Epoxy	2–5 mm	Hand layup	-	10% PALF loading delivered better mechanical properties.	[103]
Fiber loading	PALF/E-glass/epoxy	-	Hand-layup	-	TS and elasticity modulus: PALF/E-glass/epoxy > PALF composites	[80]
Fiber loading	PALF/Glass Fiber/epoxy	-	Hand-layup	-	15% PALF loading delivered better mechanical properties.	[81]

Table 6. *Cont.*

Parameter	Material	Fiber Length	Fabrication Method	Feed Rate	Observations	Ref.
Fiber loading	PALF/Steel wire mesh/epoxy	-	Hand-layup	-	TS: PALF/Steel wire mesh/epoxy > PALF/epoxy	[82]
Fiber loading	PALF/jute fiber/epoxy	-	Hand-layup	-	TS: PALF/jute fiber/epoxy > PALF/jute fiber	[83]
Fiber loading	PALF/sisal fiber/epoxy	100 µM	Hand-layup	50 mm /10 min	PALF/sisal fiber loading delivered better mechanical properties	[104]
Fiber loading	PALF/banana /sisal/epoxy	-	Hand-layup	5 mm/min	The treated PALF/banana/sisal/epoxy improved the TM, and TS.	[105]

Table 7. The effect of factors and specifications on the mechanical characteristics of composites made of thermoplastic polymers and PALF.

Parameter	Material	Fiber Length	Fabrication Method	Feed Rate	Observations	Ref.
Strain rate	PALF/LDPE	6 mm	Solution-mixing technique	TS = 50 mm/min	TS and TM = 500 mm/min > 50 mm/min >5 mm/min and elongation at break goes on reducing	[106]
Fiber loading	PALF/PP	50 mm	Injection molding	-	50/50 fiber matrix loading exhibited highest TM, FS and FM	[107]
Fiber extraction process	PALF/PP, PALF Loading -5 and 30 wt%	5–8 mm, Longitudinal direction loading	Melt mixing, prepreg, compression molding	5 mm/min	PALF with 30 wt% > 5 wt% fiber loading	[108]
Fiber loading	Randomly oriented PALF/PP	100 mm	Compression molding machine	-	PALF with 30 wt% offered highest TS and hardness due to good adhesion between fiber matrix	[109]
Fiber type	PALF/PF/PP	Powder size—0.2 mm	Twin screw extrude	TS = 10 mm/min.	TM and elasticity modulus: PALF/PP > PF/PP composites	[110]
Fiber type	PALF/LDPE	17.5 mm	Heat press	TS = 10 mm/min	TS, and elasticity modulus: PALF/LDPE > LDPEcomposites	[85]
Fiber loading	PALF/GCN/polyester	500 nm	Hand-layup	15 mm/4 min	TS modulus: PALF/GCN/polyester > PALF/polyester composites	[86]
Fiber loading	PALF/PP/PLA	-	Molten mixing	50 mm/min	TS: PALF/PLA > PP	[87]
Fiber loading	PALF/PP	60 µM	Compression molding machine	2 mm/min	TS: PALF/PP > PALF	[88]
Fiber loading	PALF/MA-g-PP	500 µM	Twin-screw extruder machine	50 mm/min	TS: PALF/MA-g-PP > PALF/PP	[89]
Fiber loading	PALF/Ramie Fabric-PP	500 µM	Hand-layup	2 mm/min	TS and flexural: PALF/Ramie Fabric-PP > Ramie Fabric	[90]
Fiber loading	PALF/polyester	10 µM	Hand-layup	1 m/s	16 mm length PALF/polyester composites have good wear resistance	[91]
Fiber loading	Paddy straw/PALF/polyester	-	Compression molding	-	TS and flexural: Paddy straw/PALF/polyester > polyester	[111]
Fiber loading	PALF/kenaf fiber/vinyl ester	-	Compression molding technique	-	Increasing fiber loading by 20% improved the TM and TS of the Composite	[112]
Fiber loading	PALF/Palm/PP	500 µM	Molten mixing	-	Increasing fiber loading by 15% improved the blending strength of the composite	[113]

Table 8. Parameter and specification effects on the mechanical characteristics of biodegradable reinforced polymer composites made of PALF.

Parameter	Material	Fiber Length	Fabrication Method	Feed Rate	Observations	Ref.
Fiber loading	Unidirectional PALF/PHBV	-	Hot compression molding, with alternate layers of PALF and PHBV in 0/90/0 orientation	TS- strain rate = 0.04/min, FS = 0.043 mm/min	Tensile and flexural properties high PALF/PHBV composites	[92]
Fiber varieties	PALF/PLA, PALF/PP	-	Melt mixing, compression molding	-	TS and FS: PALF/PP > PALF/PLA	[93]
Fiber length	PALF/PLA	short-30 mm, Long-100 mm	Short PALF and melt mixing, long PALF and hand lamination	TS, FS = 2 mm/min	(1) Long PALF composites delivered the highest TS and FS compared to the short PALF	[94]
Fiber loading	PALF/TPS	1 mm	and single-screw extruder Compression molding machine	100 mm/min	Increasing fiber loading improved the TM and TS of the Composite	[95]
Fiber loading	PALF/PLA	50µM	Injection molding process	1 mm/s	TS and elasticity modulus: PALF/PLA > PLA composites	[96]
Fiber loading	PALF/TBP/DG	200 µM	Brabender Palatograph EC internal mixing	1.28/min	TM, TS: PALF-TBP with 10% DG > DG	[97]

8. Lignocellulosic Fibers

Lignocellulosic fibers have gained increasing attention as fillers and reinforcing agents in polymer composites due to their renewability, cheapness, and rigidity. These fibers offer attractive natural materials for various applications in the plastic industry. The demand for materials suitable for effectual applications has driven manufacturers to adopt technological advances, and lignocellulosic fibers provide a sustainable solution in this regard. Additionally, the use of lignocellulosic fibers helps address concerns related to rising oil prices, depletion of natural resources, and environmental issues by utilizing annually renewable materials. The adoption of lignocellulosic fibers in polymer composites benefits not only the plastic industry but also the agricultural community, as many of these fibers are obtained from agriculture byproducts. Natural resources, such as wood, also play a significant role in the utilization of lignocellulosic fibers in polymer composite applications. Lignocellulosic fibers can be classified as primary and secondary fibers based on their usage and utilization. Primary fibers are fibers from purpose-grown plants, including flax, jute, hemp, kenaf, etc. Secondary fibers are fibers obtained as byproducts of plants, including agro-residues, coir fibers, and pineapple fibers. There are several types of lignocellulosic fibers which are as follows: bast fibers (flax and kenaf, etc.), leaf fibers (sisal and PALF, etc.), seed fibers (cotton and coir, etc.), and other types (wood) [114,115]. Lignocellulosic fibers can be classified into primary and secondary fibers based on their usage and source, as shown in Table 9. The development of lignocellulosic polymer composites has evolved beyond simply mixing plastics and fibers; today, biobased plastics are seeing increasing use in the design of these composites, reflecting a growing market for such materials. Moreover, bioplastics made of reinforced lignocellulosic fibers are an excellent example of a green composite, since they break down quickly in the presence of enzymes and bacteria [116]. The biodegradation of lignocellulosic and plastic composites is a topic of attention for both academia and industry, reflecting the growing importance of sustainable materials and waste management practices.

Table 9. Chemical composition and structural parameters of common lignocellulosic fibers.

Fiber	Cellulose (wt%)	Hemicellulose (wt%)	Lignin (wt%)	Pectin (wt%)	Wax (wt%)	Water Soluble (wt%)	Microfibrillar Angle (°)
Hemp	70.2–74.4	17.9–22.4	3.7–5.7	0.9	0.8	2.10	2–6.2
Jute	61–71.5	13.6–20.4	12–13	0.2	0.5	1.2	8
Flax	71–78	18.6–20.6	2.2	2.3	1.7	2.3	5–10
Kenaf	45–57	21.5	8–13	3–5	-	-	-
Leaf fiber Sisal	67–78	10–14	8–11	10	2	1.3	10–22
Curaua	73.6	9.9	7.5	-	-	-	-
Abaca	56–63	21.7	12–13	1.0	0.2	1.6	-
Henequen	77.6	4–8	13.1	-	-	-	-
Fruit/Seed fiber Cotton	85–90	5.70	0.7–1.6	0–1	0.6	1.0	20–30
Coir	36–43	0.15–0.25	41–45	3–4	-	5.2–16.0	30–49
Wood Hardwood	43–47	25–35	16–24	-	-	-	-
Banana	63–64	10	5	-	-	-	11
Pineapple	80–83	15–20	8–12	2–4	4–7	1–3	8–15

8.1. Lignocellulosic Materials and Their Composites

Lignocellulosic materials, comprising both lignin and cellulose, are commonly derived from sources such as wood, grasses, plants, and agricultural residues, all obtained as a result of photosynthesis. Despite their inherent variability in properties and composition, lignocellulosic materials suggest numerous advantages in the development of polymer biocomposites [117,118]. These materials are renewable, possess low abrasiveness, exhibit strong load-bearing capacity, and are biodegradable, making them attractive for various engineering applications. Polymer composites incorporating lignocellulosic fillers can be low cost and find use in diverse sectors, including the manufacture of strong and biodegradable products, as shown in Figure 11.

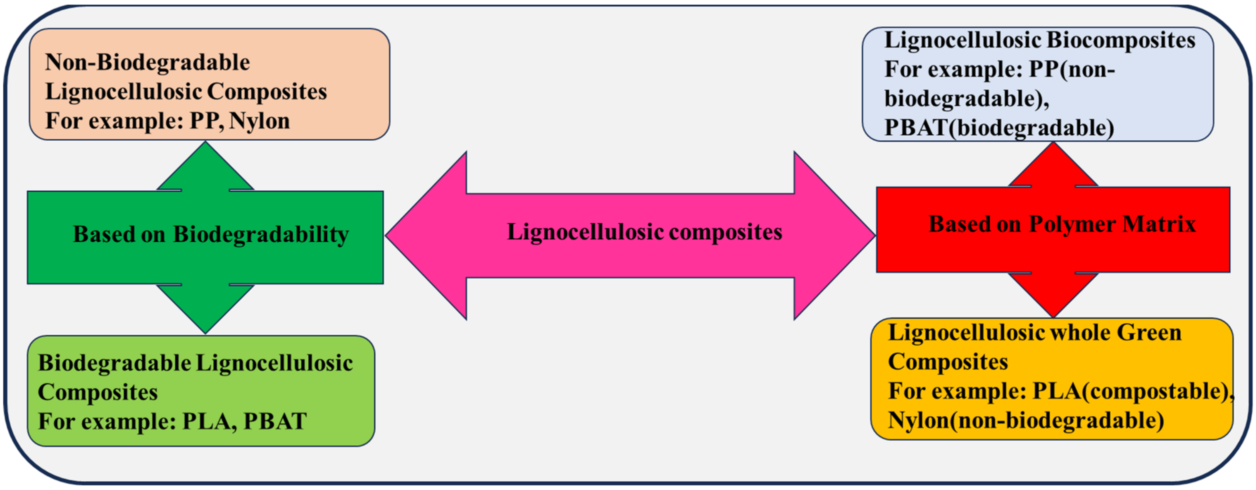

Figure 11. Classification of lignocellulosic composites.

8.2. Lignin-Based Polymer Composites

Lignin has been blended with various polymers to create materials for high-value applications. However, efficient blending can be challenging due to lignin's complex structure and reactivity. Lignin exhibits thermoplastic behavior due to its network of strong intra- and inter-molecular hydrogen bonds but acts as a thermoset material at high temperatures due to extensive cross-linking reactions [119]. Therefore, it is crucial to thoroughly investigate lignin's miscibility with other polymers, monitoring phase evolution, as their compatibility significantly affects the structure–property relationships of lignin-based blends [120,121]. Depending on the compatibility between lignin and different polymer matrices, the resulting blends can either be miscible or display phase separation. Lignin, a relatively polar polymer, requires bond cleavage to minimize condensation, which often limits its conversion into value-added products [122]. Table 10 provides examples of lignin-based polymer composites and their properties.

Table 10. Lignin-based polymer composites.

Source	Polymer Matrix	Type of Lignin	Ref.
Lignin	Thermoplastic polyolefins (TPO)	Industrial lignins (hardwood and softwood Kraft lignins, wheat straw soda lignin, hydrolysis lignin from wheat)	[123]
	Vinyl polymers	Softwood Kraft lignin; softwood lignosulfonate; hardwood lignosulfonate	[124]
	Polyamides	soda lignin from annual plants (sisal and abaca), hardwood	[125]
	Polyether	Softwood Kraft lignin	[126]
	Polyesters	soda lignin from wheat straw	[127]
	Thermoplastic elastomer (TPE)	Eucalyptus wood Kraft lignin, softwood, and hardwood Kraft lignins	[128]
	Polysaccharides	LignoBoost softwood Kraft lignin soda lignin from wheat straw and bagasse	[129]
	Polyethylene teripthalate (PET)	Lignin from pineapple	[130]

8.3. Biodegradation of Lignocellulosic Composites

Research on the biodegradability of lignocellulosic fiber-based biocomposites has been extensive, with investigations focusing on lignocellulosic fillers to modify the physicochemical properties of the polymer biocomposites. Nevertheless, there is still a lack of well-established research regarding their ultimate biodegradability. Biodegradation processes usually provide end products such as CO_2, H_2O, and cell biomass in aerobic environments. On the other hand, in anaerobic conditions, cell biomass, CH_4, CO_2, and H_2O are the end products of biodegradation. While studies have reported observations of weight loss and changes in mechanical properties during biodegradation in soil, these observations provide only a limited understanding of the intricate processes involved [131]. To address this, various standardized test methods established by organizations like ISO and ASTM have been developed to estimate the mineralization of polymeric materials. Improvements in testing equipment, like respirometers, which are fully closed-circuit devices coupled with CO_2 and CH_4 sensors, have helped these procedures. A more sensitive and precise evaluation of biodegradability is provided by respirometers, which automatically quantify the byproducts of biodegradation under both aerobic and anaerobic circumstances. Respirometers are widely used to test the biodegradability of biopolymers and biocomposites due to their suitability for this application. Their use enables researchers to obtain valuable insights into the biodegradation kinetics and mechanisms of these materials, facilitating informed decisions in the development and selection of environmentally sustainable materials.

9. Biodegradation of Nature Fibers, Biopolymers, and Biocomposites in Environmental Conditions

Sustainability and eco-friendliness in material development are being challenged by the growing concerns about environmental contamination caused by the disposal of plastic waste from past consumer products. Sustainable biopolymers and bio-based materials derived from bioresources for green manufacturing are of great scientific interest globally [132]. Biopolymer composites based on natural fibers are currently receiving much attention as eco-friendly alternatives to synthetic fibers in various industrial applications, such as the automotive industry, construction, and packaging [133]. This interest is mainly due to the favorable properties of natural fibers, such as low density, mechanical strength, low cost, environmental friendliness, renewability, and sustainability. Microbial biopolymers, primarily produced through microbial fermentation of renewable feedstock from biomass, represent a new generation of materials. Their renewability offers an intrinsically neutral carbon footprint and conserves primary resources [134].

Biopolymer composites are currently gaining popularity as very appealing materials for a variety of purposes, including biomedical applications like pharmaceutical capsules, bone prosthesis, and stiff packaging. In recent years, significant research has been conducted on developing biocomposites made from biopolymers and natural fibers, focusing on improving fiber/polymer interfacial adhesion and impact resistance [135]. Muniyasamy et al. incorporated flax into PHBV (Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)) composites, demonstrating biodegradation applications. Their report studied the effects of 2% alginic acid-treated flax fibers on the thermal–chemical properties and degradation behaviors of PHBV/flax composites under composting conditions, compared to untreated flax fibers of PHBV composites and neat PHBV [136]. Considering that textile fabrics biodegradation is affected by their structure and characteristics such as thickness, production process, weave, and yarn, systematic research on fiber biodegradation is needed. Brunsek et al. investigated the biodegradation properties of jute, hemp, and sisal fibers (lignocellulose fibers); viscose fibers (regenerated cellulose); and polylactide (PLA) (biopolymer) in farmland soil using a soil burial test [137].

Fibers that are designed to biodegrade in the soil after the end of their service life are being utilized in open environment applications with increasing frequency. A flexible alternative to natural fibers is biodegradable polymer fibers. The degradation behaviors of fibers composed of polylactic acid (PLA), a mix of polyhydroxyalkanoate (PHA) and PLA, and a bicomponent fiber (BICO) composed of PLA and polybutylene succinate (PBS) were examined by Brunsek, R. et al. [138]. Using the CO_2 evolution respirometric approach, Nomvuyo et al. examined the rate and processes of the aerobic biodegradation of biopolymer blends under controlled composting conditions. The quantity of carbon mineralized under home and commercial composting environments was assessed using melt extrusion tests on biopolymer blends of poly(butylene adipate terephthalate) (PBAT) blended with poly(lactic acid) (PLA) and PBAT blended with poly(butylene succinate) (PBS) [139]. The mineralization of poly(lactic acid) (PLA), poly(3-hydroxybutyrate-co-valerate) (PHBV), and blends of PLA and PHBV in compost and soil burial settings was investigated by Sudhakar et al. [140]. International standards and guidelines are followed in the discussion of various analytical techniques and standard test procedures for assessing the possible biodegradability and toxicity levels of polymeric materials in various situations [141]. Ramengmawii et al. studied the biodegradability, color, and texture qualities compared with PLA by conducting accelerated weathering and soil burial experiments on biocomposites of different ratios of coir (CF)/pineapple leaf fibers (PALF) with polylactic acid (PLA) [142]. The polymer matrix of a lignocellulosic composite plays an important part in determining its biodegradability, and the rate of biodegradation is affected by a variety of environmental conditions. It is interesting that the rate of biodegradability exhibited by natural fibers and biopolymer composite materials varies according to environmental conditions, as shown in Table 11.

Table 11. Overview of biodegradation results of nature fibers, biopolymer, and biocomposites in environmental condition.

Material	Biodegradation (%) in Days	Ref.
PHBV/flax	48.9–30 days	[136]
PHBV/flax/alginic acid	65.6–30 days	[136]
Hemp Fiber	6.73–13 days	[137]
Jute Fiber	10.82–13 days	[137]
Sisal Fiber	28.83–13 days	[137]
Viscose Fiber	3.12–13 days	[137]
PLA	1.5–13 days	[137]
PBS/PLA/BICO	65–30 days	[138]
PHA/PLA/BICO	55–30 days	[138]
PBAT/PLA	25–30 days	[139]
PBAT/PBS	20–30 days	[139]
PHBV	15–30 days	[140]
PLA/PHBV	25–30 days	[140]
PBAT	80–33 days	[141]
PCL	100–67 days	[141]
TPS	45–67 days	[141]
PALF	15.2–30 days	[142]
CF/PALF/PLA	18.6–30 days	[143]

10. Conclusions

The utilization of PALF in various applications has created new opportunities for both academia and industries to develop sustainable solutions for future use. PALF has found extensive use in composite and biomedical industries, contributing to socio-economic empowerment. The fabrication of PALF fiber-based polymer biocomposites using different matrices has led to the development of cost-effective and eco-friendly biomaterials, influencing market dynamics. A thorough investigation of the fundamental, mechanical, and physical properties of PALF is crucial for designing such polymer biocomposites. Despite the wide range of studies conducted globally on PALF fiber-based polymer biocomposites, the goal of fully utilizing PALF remains unrealized, particularly in South Africa. In contrast, countries like India and China have made significant progress in leveraging PALF for socio-economic development. Achieving a sustainable future for the PALF-based polymer biocomposites industry requires innovative approaches beyond traditional modes of utilization. Efficient characterization techniques for PALF and PALF-based polymer biocomposites need to be advanced to enhance analysis and testing capabilities. Although significant research has been conducted in this area, further innovation and exploration are necessary to overcome potential challenges. PALF, with its high cellulosic content and superior mechanical properties, holds promise for more applications. Lignocellulosic composites, in which lignocellulosic materials serve as major fillers, are categorized based on their biodegradability in soil and ocean environments. This classification helps us understand the environmental impact and disposal options for these materials, further contributing to sustainable practices in material design and engineering.

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