



## Review Article

# A review on engineering biocomposites and natural fiber-reinforced materials

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## ABSTRACT

Fiber-reinforced polymer composites are well-studied and established products, and today they are being used in different industrial and non-industrial areas. However, the increased interest in recyclability and the concerns about climate change caused materials scientists to look for a non-petroleum-based alternative to synthetic fibers and polymers. Since the beginning of this century, natural fibers and biopolymers have attracted increasing interest each year for composite applications. Thanks to this interest, studies on natural fibers and biopolymers have increased significantly. Despite the high number of studies on natural fibers and natural fiber-reinforced polymers (NFRP), there are gaps in the literature. This work reviews studies on natural fibers, biopolymers, and biocomposites with their advantages, disadvantages, and limitations. Studies that focus on the ways to reduce or eliminate these disadvantages and limitations have also been looked at. Also, current challenges and future perspectives for natural fibers, biopolymers, and NFRPs have been discussed.

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## 1. INTRODUCTION

Fiber-reinforced polymers (FRP) have replaced conventional materials since the mid-last century. This replacement was mainly due to their superior mechanical properties compared to traditional materials, such as steel and aluminum. Composite materials have a more specific strength and higher fatigue resistance than steel and aluminum and are lighter than conventional materials [1].

Today, FRPs have many applications in various industrial areas, such as the automotive, aviation, and defense industries. While composite materials solved many prob-

lems for these industries, they created new ones for the environment. Better mechanical properties with composite materials alone are not enough to solve today's problems. Better specific strength, higher fatigue resistance, and weight reduction do not solve the environmental issues associated with FRPs.

In the present century, thermoset polymers reinforced with synthetic fiber composites are being questioned due to problems related to environmental issues [2]. Due to thermoset polymers' nature, synthetic fibers with thermosets render FRP impossible to recycle. This causes irreversible problems for the environment.

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However, the substitution of natural fiber-reinforced polymers (NFRP) for the traditional (FRP) is not due to only environmental reasons. Natural fiber-reinforced polymer composites present the same or comparable mechanical properties to fiber-reinforced polymer composites with lower density [3]. Natural fibers (hemp fiber 1.48 g/cm<sup>3</sup>, flax fiber 1.4 g/cm<sup>3</sup>, kenaf fiber 1.45 g/cm<sup>3</sup>, jute fiber 1.46 g/cm<sup>3</sup>, banana fiber 1.35 g/cm<sup>3</sup>) are lighter than their synthetic counterparts (glass fiber 2.54 g/cm<sup>3</sup>, carbon fiber 1.75–2.00 g/cm<sup>3</sup>), this aspect makes the NFRPs favorable against FRPs in areas where weight reduction is essential. Natural fibers with very low densities, such as hemp, can be used in non-critical areas where solid mechanical properties are not needed [4].

In addition to the low-weight characteristics of natural fibers, NFRPs can be produced at a lower cost when compared with polymer composites with synthetic fibers. The automotive industry, where lower cost and lower weight are critical, started using natural fibers at the beginning of this century [4]. Furthermore, the aviation industry, another area where cost and weight reduction are essential to compete, started using natural fibers as a reinforcing material for polymer matrix composites [5].

Even though natural fibers bring new characteristics to the table, such as environmental friendliness, it comes with some drawbacks too. As they are crop-based materials that grow naturally on the soil, the mechanical properties of natural fibers depend upon the harvesting region, the soil condition, harvesting time, and the intensity of sunrays and rain [5, 6].

The compatibility between natural fiber and matrix polymer of NFRPs is poor [7]. This causes lower mechanical properties than expected due to the non-uniform dispersion of fiber in the matrix and low stress transfer between the matrix and the fiber.

Most polymers, especially thermoplastic ones, which are widely used with natural fibers due to their recyclability, are hydrophobic, in contrast to natural fibers, which are hydrophilic [5]. The hydrophilic characteristic of a natural fiber creates another problem: water or moisture absorption. They also have poor fire resistance [8].

Although their use in composite materials has increased over time, the subject of natural fibers as reinforcement materials is still a new topic and developing field. Therefore, research on natural fibers should be reviewed and summarized to guide future studies. In this paper, the studies on natural fibers and biocomposites are thoroughly reviewed and classified according to their content and purpose. Reviewed studies are arranged to create an orderly explanation of the subject in question.

This study aims to review the pros and cons of natural fibers and the studies that have been done to mitigate these disadvantages. First, an introduction to biocomposites is presented, and the classification of natural fibers is

explained. After that, the natural fibers' sources and macrostructure are explained, and the effects of surface modification on natural fibers are reviewed. In addition, the mechanical performance of surface-modified and non-surface-modified natural fibers is also reviewed. The last part, before the conclusions, explains the topic of green composites and potential areas for utilizing them.

## 2. BIOCOMPOSITES

Composite materials must consist of at least two or more materials. Biocomposites, a subcategory of composites, are no exception. However, the requirement of combining at least two materials is not enough on its own for biocomposites. For a material to be called a biocomposite, at least one of the materials that form the composite must be a natural material [3]. Composite materials made from natural fiber-reinforcements (hemp, jute, rami, etc.) and petroleum-derived polymer matrices can be labeled as biocomposites. Composite materials that consist of synthetic fiber-reinforcements and natural-based polymer-biopolymer matrices like PLA are also called biocomposites. If both the reinforcement and the matrix materials are naturally based, the material is considered biocomposite too, but to emphasize the material's biodegradability, they are classified as green composites [9].

Natural and synthetic fibers can be utilized in a single matrix to increase the performance of the biocomposite. The composite materials composed of this combination are called hybrid composites [10].

There are various reasons for using natural fiber-reinforced polymers over synthetic fiber-reinforced polymers. Aside from the low-cost and low-density benefits of biocomposites, biodegradability will become an essential feature of these materials as global environmental issues worsen. Biopolymers or biocomposites may be a long-term solution to 21<sup>st</sup>-century environmental problems (such as climate crisis, waste plastic pollution, etc.) [9].

### 2.1. Green Composites

Green composites are a subcategory of biocomposites. Green composites can be defined as fully biodegradable composite materials. A combination of natural fibers with natural polymers or biopolymers can be called a green composite [11]. It is critical to distinguish biocomposites from green composites; every green composite can be labeled as a biocomposite, but the reverse is not always achievable. Although hemp-polypropylene natural fiber reinforced polymer is a biocomposite, it is not a green composite due to polypropylene's nondegradability. Due to the degradable hemp and PLA, Hemp-PLA is both a biocomposite and a green composite.

The most crucial advantage of green composite is its biodegradable nature. This characteristic only makes green composites unrivaled against other synthetic or natural fiber-re-

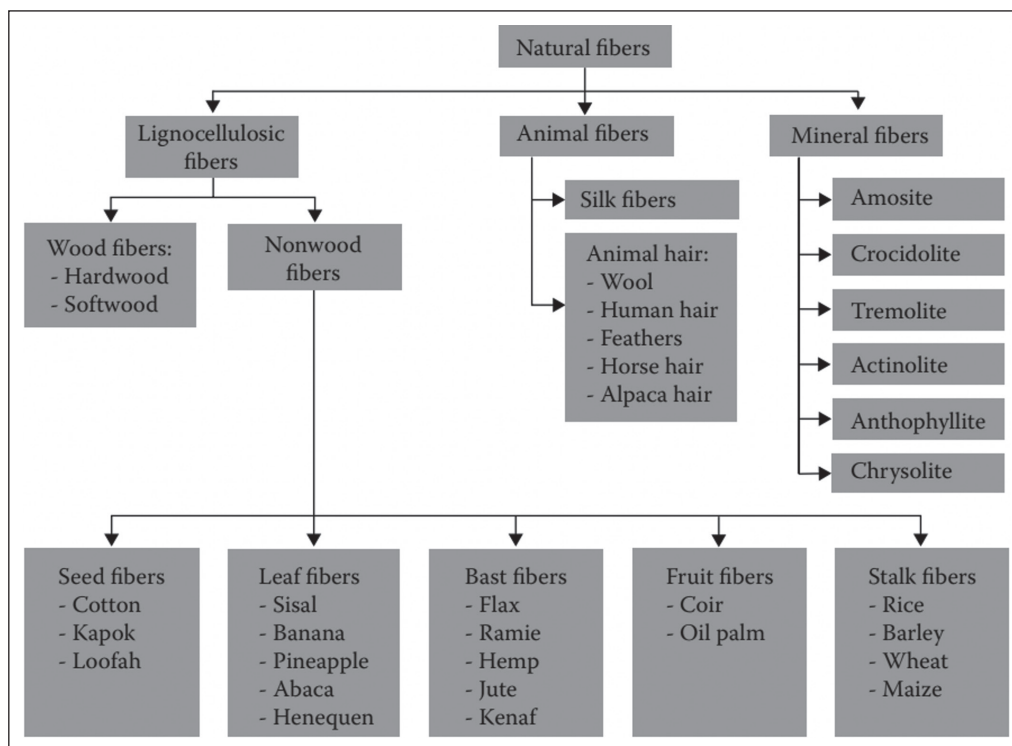


Figure 1. Classification of natural fibers [14].

inforced petroleum-derived polymers as an environmentally friendly alternative. In addition to this aspect, producing biodegradable polymers from renewable resources like animals, plants, and microbes through biochemical reactions offers lower reliance on petroleum-derived polymers, thus lower petroleum usage [11]. Increased use of green composites will help the struggle against environmental issues like climate crisis and plastic waste due to excessive usage of non-biodegradable and non-recyclable polymers [12].

### 2.2. Natural Fibers

Fibers can be divided into two categories: synthetic and natural fibers. Natural fibers can also be subdivided based on their origins, like animal, mineral, and plant (cellulosic/lignocellulosic). Most natural fibers are planted (lignocellulosic), like cotton, jute, flax, hemp, ramie, etc. These fibers have mainly lignified secondary cell walls, which give mechanical stability to the plant body [13]. The classification of natural fibers is shown in Figure 1. Some of the most popular plant (lignocellulosic) fibers are shown in Figure 2.

There can be significant differences between the different types of natural fibers. For example, their density, length, diameter, and mechanical properties can vary greatly. Although this difference might seem a disadvantage, it allows the engineers and designers to choose different materials with different physical and mechanical properties suitable for the design requirements. The physical and mechanical properties of various natural fibers are shown in Table 1.

#### 2.2.1. Sources of Natural Fibers

As shown in Figure 1, natural fibers are classified based on their origin, the part of the plant, animal, and mineral from which they are derived. Animal and mineral fibers are obtained from animals and minerals, respectively.

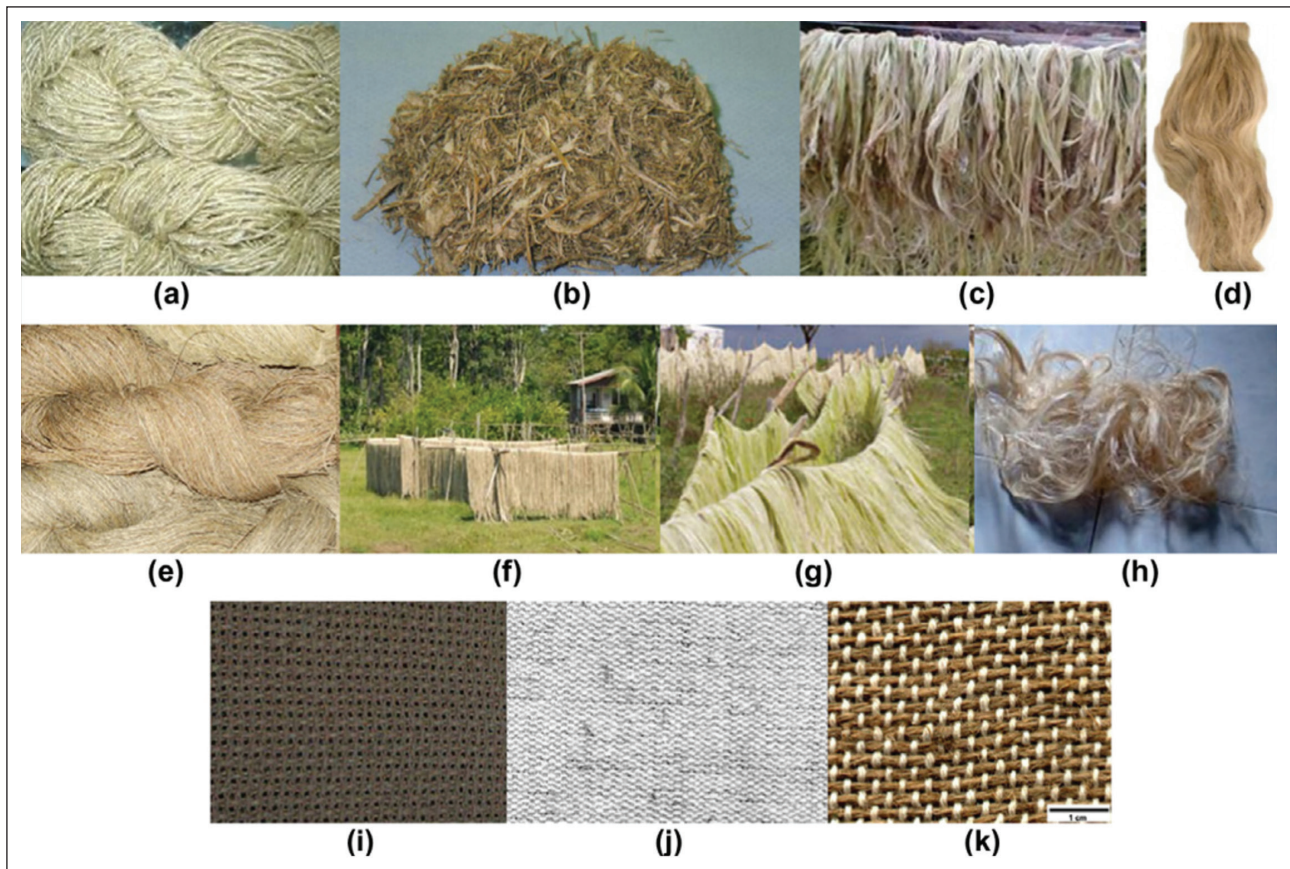
There are two types of wood fibers, hardwood, and softwood. Wood fibers are extracted from wood using various methods, but for natural fiber-reinforced polymer applications, wood flour is more widely used than wood fiber due to its low price and ease of processing with conventional polymers.

Seed fibers, cotton, kapok, loofah, etc., are obtained from the seeds of these plants. Leaves of monocotyledonous plants are used to obtain leaf fibers, such as sisal, banana, pineapple, abaca, henequen, etc. Bast fibers, which include flax, ramie, hemp, jute, kenaf, etc., are collected from the stems' inner bark, called phloem, of the dicotyledonous plants. Fruit fibers are the fruits of plants that bear the name. Stalk fibers of straw fibers, rice, barley, wheat, maize, etc., are the stalks of their plants [13]. The production of some plant (lignocellulosic) fibers and the largest producer countries is shown in Table 2.

Plant (lignocellulosic) fibers are more popular than animal and mineral-based natural fibers among all these natural fibers. Cotton (seed fiber), flax, jute, ramie, and kenaf (bast fibers) are plant fibers' most popular and most researched fibers.

According to Townsend [17], world fiber production in 2018 was approximately 110 million, including 32 million





**Figure 2.** Plant-based natural fiber-reinforcements, (a) Banana; (b) sugarcane bagasse; (c) curaua; (d) flax; (e) hemp; (f) jute; (g) sisal; (h) kenaf. The typical pattern of reinforcements used in the hybrid LC-based biodegradable composite synthesis. (i) Jute fabric; (j) ramie-cotton fabric; (k) jute-cotton fabric [15].

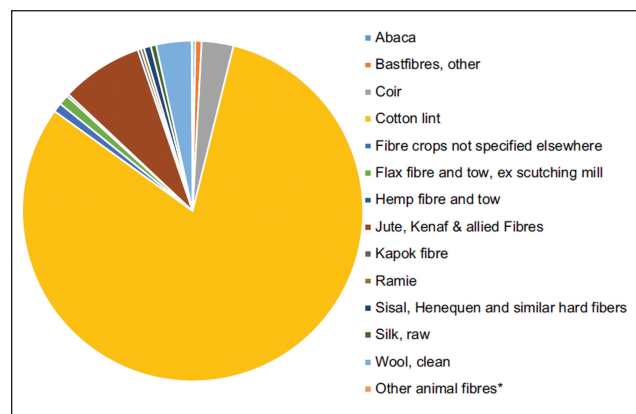
tons of natural fibers (Fig. 3). Cotton accounted for 80% of natural fiber production by weight, and jute production was approximately 3 million tons in 2018, with wool and coir each accounting for about 1 million tons.

Unlike synthetic fibers, natural fibers do not get manufactured at a fiber production plant. Instead, they come from "natural" sources, as the name implies. This brings biodegradability and new factors that can affect the fiber properties that synthetic fibers do not have. These factors are shown in Table 3.

One of the most critical factors in Table 3 is the harvesting time because the harvesting time of the plant would significantly affect the structure of fibers and their physical and chemical composition. Because of this, the best harvesting time for the desired natural fiber mechanical and chemical composition, which are the essential aspects in defining the fibers' overall qualities, must be determined [6, 14].

### 2.2.2. Chemistry of Natural Fibers

A single natural fiber is between 1 and 50 mm in length, and the diameter of a fiber is around 10–50  $\mu\text{m}$ . The fiber cell wall is composed of two main parts: the primary wall and the second wall. The primary wall's main purpose is to



**Figure 3.** World natural fiber production in 2018 [17].

control the fibers' growth direction and rate. The secondary wall provides mechanical strength to the fiber, composed of three main layers, S1, S2, and S3 [13]. The three-dimensional structure of fiber and a fiber's typical cross-section is shown in Figure 4.

The major constituents of natural plant fibers are cellulose, hemicellulose, and lignin. Natural fibers also include

**Table 1.** Physical and mechanical properties of some natural fibers [16]

Fiber type	Density [g/cm <sup>3</sup> ]	Length [mm]	Diameter [μm]	Tensile strength [MPa]	Tensile modulus [GPa]	Specific modulus (Approx.) ( $E/\rho$ ) [GPa.cm <sup>3</sup> .gr <sup>-1</sup> ]	Elongation [%]
Abaca	1.5	–	–	400–980	6.2–20	9	1.0–10
Alfa	0.89	–	–	35	22	25	5.8
Begasse	1.25	10–300	10–34	222–290	17–27.1	18	1.1
Bamboo	0.6–1.1	1.5–4	25–40	140–800	11–32	25	2.5–3.7
Banana	1.35	300–900	12–30	500	12	9	1.5–9
Coir	1.15–1.46	20–150	10–460	95–230	2.8–6	4	15–51.4
Cotton	1.5–1.6	10–60	10–45	287–800	5.5–12.6	6	3–10
Curaua	1.4	35	7–10	87–1150	11.8–96	39	1.3–4.9
Flax	1.4–1.5	5–900	12–600	343–2000	27.6–103	45	1.2–3.3
Hemp	1.4–1.5	5–55	25–500	270–900	23.5–90	40	1–3.5
Henequen	1.2	–	–	430–570	10.1–16.3	11	3.7–5.9
Isora	1.2–1.3	–	–	500–600	18–20	14	5–6
Jute	1.3–1.49	1.5–120	20–200	320–800	8–78	30	1–1.8
Kenaf	1.4	–	–	223–930	14.5–53	24	1.5–2.7
Nettle	1.4–1.55	–	–	650	38	25	1.7
Oil Palm	0.7–1.55	–	150–500	80–248	0.5–3.2	2	17–25
Piassava	1.4	–	–	134–143	1.07–4.59	2	7.8–21.9
PALF	0.8–1.6	900–1500	20–80	180–1627	1.44–82.5	35	1.6–14.5
Ramie	1.0–1.55	900–1200	20–80	400–1000	24.5–128	60	1.2–4.0
Sisal	1.33–1.5	900	8–200	363–700	9.0–38	17	2.0–7.0

**Table 2.** Production amount and producer countries of some plant fibers [13]

Plant (lignocellulosic) fibers	Type	Worldwide production amount (10 <sup>3</sup> tons)	Countries
Abaca	Leaf	70	Philippines, Ecuador, Costa Rica
Pineapple	Leaf	74	Philippines, Thailand, Indonesia
Sisal	Leaf	378	Tanzania, Brazil
Coir	Fruit	100	India, Sri Lanka
Cotton	Seed	25000	China, India, USA
Oil Palm	Fruit	40	Malaysia, Indonesia
Flax	Bast	830	Canada, France, Belgium
Hemp	Bast	214	China, France, Philippines
Jute	Bast	2300	India, China, Bangladesh
Kenaf	Bast	970	India, Bangladesh, USA
Ramie	Bast	100	China, Brazil, Philippines, India
Bagasse	Grass	75000	Brazil, India, China
Bamboo	Grass	30000	India, China, Indonesia

a small amount of pectin and wax. The amount of cellulose will vary depending on the type and age of the plant. The differences between the different natural plant fibers' cellulose, hemicellulose, and lignin values are shown in Table 4.

Cellulose microfibrils have a diameter between 10–30 nm. They act as the reinforcement material and remain

embedded in the hemicellulose/lignin matrix, which is responsible for providing mechanical strength to the fibers. These microfibrils are linked together to form the cellulose fibers, the main constituent of most plant-based natural fibers. The second most abundant natural fiber constituent in cell plant walls is hemicellulose. Lignin is another signifi-

**Table 3.** Factors related to the production of natural fibers affect fiber properties [14]

Stage	Factors affecting fiber properties
Plant growth	Plant species
	Crop cultivation
	Crop location
	Fiber location in plants
	Climate
Harvesting	Fiber ripeness, which affects;
	Cell Wall thickness
	iber coarseness
	Fiber-structure adhesion
Fiber extraction	Decortication process
	Type of retting method
Supply	Storage conditions
	Age of fibers

cant component of the plant cell wall that provides strength, rigidity, and protection against microbial pathogens of lignocellulosic-based natural fibers cell walls [13].

### 2.2.3. Polymers and Biopolymers

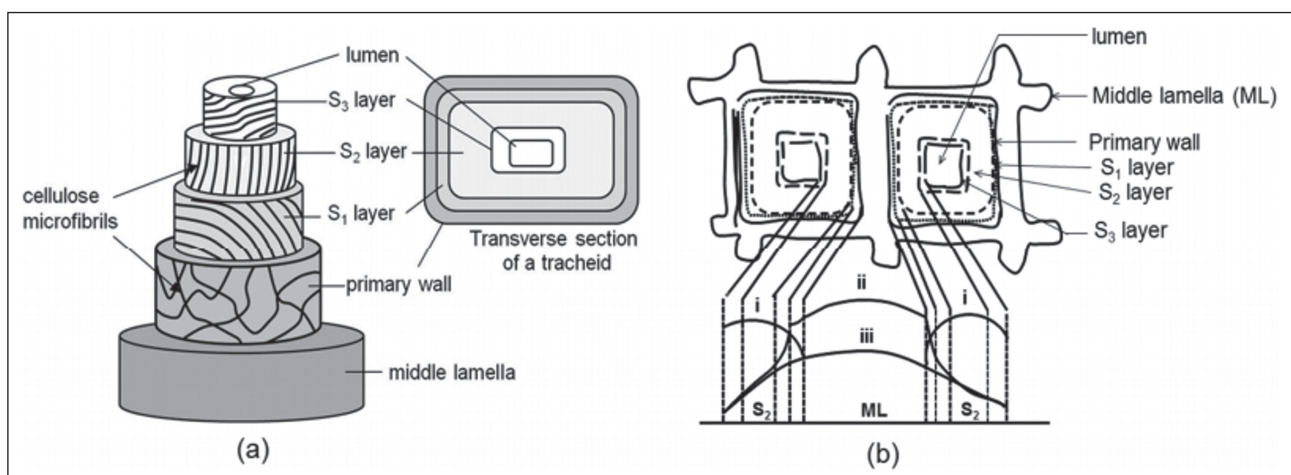
Thermosetting polymers (thermosets) are widely used with natural fibers to produce biocomposites. Thermosets commonly used as matrix material in biocomposites are polyester resin, epoxy resin, and vinyl ester resin [18]. Due to their chemical composition, thermosets are non-recyclable and non-biodegradable materials. Biocomposites are preferred because of their environmentally favorable properties like recyclability and biodegradability, but thermosets render them unusable as the environmental problems due to the excessive use of non-recyclable polymers increase, the use of thermoset biocomposites decreases. However, to

address these environmental problems, there are efforts for thermoset biopolymers from vegetable oils, e.g., castor oil, soybean oil, rapeseed oil, etc. [13].

Linear chain molecules characterize thermoplastic polymers. The most crucial aspect of thermoplastic polymers is that they can be repeatedly melted or reprocessed. This characteristic makes the thermoplastic polymers favorable to use over the thermoset polymers as a matrix material in biocomposites. The degree of crystallinity of the thermoplastic is affected by the cool-down time because of its reusability. This is because the polymer chains need time to get organized in the orderly pattern of the crystalline state; too quickly of a cooling rate will not allow crystallization to occur [1].

On the other hand, too slow of a cooling rate may cause thermal degradation in the polymer. Thermoplastics with different cool-down times may show different mechanical behaviors under static or dynamic loads. This makes the temperature and cool-down time control very important during reprocessing. Commonly used thermoplastic polymers as matrix materials in biocomposites are polypropylene (PP), polyethylene (PE), low melt poly(ether-sulfone) (PES), and polyethylene terephthalate (PET).

For a biocomposite to be classified as a green composite material, it must consist of a biodegradable and biobased reinforcement material with a biodegradable and biobased matrix material. Conventional thermosets and thermoplastics are not enough for a composite material to be considered a green composite. In order to achieve complete biodegradability, biopolymers or biobased polymers must be used as matrix materials. Only biodegradable and biobased polymers can be defined as biopolymers. Due to the action of microorganisms over a determined time and in a specific environment, the material undergoes a degradation process; this material's ability is called biodegradability [13].



**Figure 4.** (a) Three-dimensional structure of the secondary cell wall of a xylem cell (b) the relative amounts of cellulose, hemicellulose, and lignin across a cross-section of two wood cells (i: cellulose, ii: lignin, iii: hemicellulose) [18].



**Table 4.** Composition of some natural fibers [16]

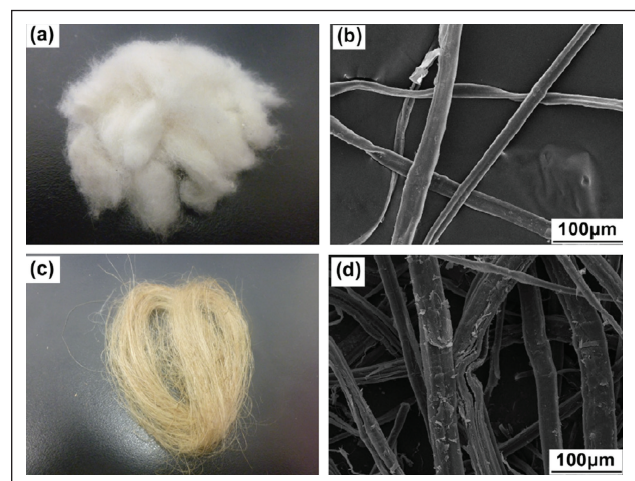
Fiber type	Cellulose [wt%]	Hemicellulose [wt%]	Lignin [wt%]	Pectin [wt%]	Waxes [wt%]	Micro-fibril angle [deg]	Moisture content [wt%]
Abaca	56–63	20–25	7–13	1	3	–	5–10
Alfa	45.4	38.5	14.9	–	2	–	–
Begasse	32–55.2	16.8	19–25.3	–	–	–	–
Bamboo	26–65	30	5–31	–	–	–	–
Banana	63–67.6	10–19	5	–	–	–	8.7–12
Coir	32–43.8	0.15–20	40–45	3–4	–	30–49	8
Cotton	82.7–90	5.7	<2	0–1	0.6	–	7.85–8.5
Curaua	70.7–73.6	9.9	7.5–11.1	–	–	–	–
Flax	62–72	18.6–20.6	2–5	2.3	1.5–1.7	5–10	8–12
Hemp	68–74.4	15–22.4	3.7–10	0.9	0.8	2–6.2	6.2–12
Henequen	60–77.6	4–28	8–13.1	–	0.5	–	–
Isora	74	–	23	–	1.09	–	–
Jute	59–71.5	13.6–20.4	11.8–13	0.2–0.4	0.5	8	12.5–13.7
Kenaf	31–72	20.3–21.5	8–19	3–5	–	–	–
Nettle	86	10	–	–	4	–	11–17
Oil Palm	60–65	–	11–29	–	–	42–46	–
Piassava	28.6	25.8	45	–	–	–	–
PALF	70–83	–	5–12.7	–	–	14	11.8
Ramie	68.6–85	13–16.7	0.5–0.7	1.9	0.3	7.5	7.5–17
Sisal	60–78	10.0–14.2	8.0–14	10	2	10–22	10–22

Biopolymers can be classified into two main categories. The first category is biopolymers made from natural raw materials (biobased) and biodegradable. The most popular biopolymer, PLA (Polylactide acid), is considered in this category along with other biopolymers, such as PHAs (polyhydroxyalkanoates), starch, and chitosan [19]. The second category belongs to biopolymers made from fossil resources but biodegradable, such as PCL (polycaprolactone), PBS (polybutylene succinate), and PBAT (Polybutylene adipate terephthalate) [13].

**2.2.4. Macrostructure of Natural Fibers**

Natural fibers used in NFRPs can be divided into three categories according to their fiber orientation: non-woven, woven fabric, and unidirectional (UD). Non-wovens are mainly produced with randomly oriented short fibers. Felts, a non-woven type, use randomly oriented short natural and polymer fibers as reinforcement and matrix materials. Felts have excellent sound absorption and thermal insulation properties due to their high thickness values. However, unlike other NFRP types, felts have lower strength than woven fabrics and UD.

The second type is woven fabrics. This type of fabric is generally used for composite applications where the composite is produced by combining natural fiber fabrics with polymer pellets or films. Depending on the needs, they can



**Figure 5.** Photos and micrographs of NHF (a, b) and SHF (c, d) [27].

be produced in bi-directional or multidirectional form. These fibers are easy to produce and are used to obtain high mechanical strength in multiple directions. However, this multidirectional fiber structure might create extra weight than felts and UD.

Unidirectional (UD) fibers' structure is similar to the woven ones with a difference: all-natural fibers run in a

single, parallel direction. UD s show the best mechanical performance under loads with the same direction as their fibers. In addition, UD s are lighter than their woven counterparts. These properties allow for more precise production with even lighter weights. However, UD s are not appropriate for parts where a great anisotropic strength property is required.

Two of the most popular natural fibers are hemp and flax [20]. Therefore, the macrostructure of natural fibers is explained in this section with hemp and flax fibers studies.

Numerous studies focus on hemp fiber-reinforced polymers and flax-reinforced polymers. In the case of hemp fiber-reinforced non-wovens, most studies focused on their superior noise reduction properties [21–23]. The works of Nick et al. [21], Yilmaz et al. [22], and Oldham et al. [23] utilize non-wovens in their felt forms, where their sound absorption properties are most robust, but their mechanical strength values are lowest. Numerous studies exist for woven hemp fiber-reinforced polymer biocomposites [24–28]. Corbin et al. [24] have studied the effects of weave patterns and features on hemp fabrics and have concluded that high-performance woven hemp fabric composites made from low-twisted roving can be obtained. The study presented by Bonnafous et al. [25] compared the damage mechanisms in woven hemp fiber composites and glass fiber composites; they established that the damage development of these two composites is different. Hasan et al. [26] studied the mechanical performance of hemp/glass woven fabric hybrid composites treated with greenly synthesized silver nanoparticles. Berhanu et al. [27] have studied the sliding behavior of woven hemp fabric reinforced biocomposites and have established good compatibility between the woven hemp fabric and polypropylene. Baghei et al. [28] have studied the characterization of biocomposite reinforced with woven hemp fabrics and lyocell fabrics. They have used polylactic acid (PLA) as matrix material; PLA is a filament produced using natural resources. Thus the biocomposite in this study can be classified as a green composite. It should be noted that the performance of this green composite was lower than its lyocell reinforced counterpart.

However, the composites with woven fiber are mainly manufactured with polymer pellets and resins as thin plates [24–27]. This feature seriously limits the sound absorption capability of hemp fibers but increases their mechanical strength remarkably. Even the woven hemp reinforced biocomposites with polymer fiber matrix are investigated for superior mechanical properties and water absorption characteristics instead of poor noise reduction performance [28].

As mentioned, non-woven NFRPs are weaker than their woven counterparts, considering their mechanical performance. To use the felts in the areas where high mechanical strength is needed, they must be hot-pressed and formed as biocomposite plates. Hargitai et al. [29] have worked on hemp-PP and flax-PP non-wovens to find the optimal re-

inforcement to matrix ratio for best mechanical strength values. They have concluded that 50 % by weight is the optimal value for hemp fiber-reinforced PP felts. The studies of Shahzad [30] and Stelea et al. [31] have also focused on the characterization of hemp-PP felts by testing them as plates. Chen et al. [32] have compared the mechanical performance of hemp-PP non-woven to bagasse-PP, kenaf -PP, and ramie-PP non-wovens. They revealed that the hemp-PP composites, compared to other biocomposites, featured similar tensile and flexural modulus values with better thermal properties [32]. It should also be noted that there are also studies on the mechanical strength of unpressed hemp fiber-reinforced felts, but their mechanical properties are inferior compared to their hot-pressed counterparts [33].

For flax-reinforced biocomposites, the studies for non-woven flax composites also focus on sound absorption and thermal insulation properties [34–36]. Rasyid et al. [34], Velayutham et al. [35], and Muthukumar et al. [36] studied the sound absorption and acoustic performance of non-woven flax fiber fabrics. They have established that the sound absorption performance of flax fibers is good due to the macrostructure of the natural fibers [34, 35] and can be improved with the addition of low-melt PET [36]. However, since the flax fibers are mechanically stronger than hemp fibers, as established by Pil et al. [37], Shahria et al. [38], and Maity et al. [39], the emphasis is on the mechanical strength of non-woven flax fiber-reinforced polymers. Maity et al. [39], John et al. [40], Omrani et al. [41], Bachmann [42], and Alimuzzaman [43] have studied the mechanical performance of flax fiber reinforced biocomposites by using characterization tests and by employing different techniques and methods to increase its performance, such as surface modification. Compared with hemp felts, flax fiber-reinforced biocomposites show better mechanical performance in their felt forms but with high material deformation [41]. Due to the high mechanical strength of their fibers, non-woven flax fibers may even be used as structural building materials [44, 45]. Woven flax fibers have higher mechanical properties than non-woven flax fibers [46] and woven hemp fibers [47].

Unidirectional flax fibers, like all-UD fibers, perform best when the load is in the same direction as the fibers. Depending on the load, their mechanical performance may be better or worse than their woven counterparts [48]. There are numerous studies that focus on the polyesters [49], polylactic acid (PLA) [50], polypropylene (PP) [51, 52], bio-based resins [53], and geopolymers [54] that are UD flax fibers reinforced.

Hybrid biocomposites reinforced with two different types of fibers or reinforced with the different fiber orientations of the same fibers are also possible to produce. Due to their higher mechanical performance, flax fibers are mostly combined with glass fibers [55] or carbon fibers [56]. However, they can also be combined with other natural fibers, including hemp fibers [57, 58], to preserve their recyclabil-



ity and to be able to classify them as biocomposites [38]. However, hemp fibers are not as strong as flax fibers, and because of this, the most used synthetic fiber for hybrid hemp fiber composites is glass fibers [26, 59].

#### *Limitations of Natural Fibers*

Natural fibers have higher moisture absorption than synthetic fibers due to their hydrophilic nature. The long-term effect of moisture absorption is developing micro-cracks, thus lowering the mechanical properties of NFRP significantly [1, 5, 29]. However, there are some contradictory research results about the effect of moisture in the short term. For example, Hargitai et al. [29] proposed that wetting the hemp fiber-reinforced polypropylene fiber composite increases the impact strength but decreases the bending properties. On the other hand, Munoz et al. [60] have studied the water absorption of flax fiber-reinforced bio-epoxy composite and concluded that water absorption increases tensile strength but reduces the flexural properties, similar to those proposed by Hargitai et al. [29]. Even though the impact strength aspect has positive effects in the short term, moisture, and water absorption, wear down the mechanical properties of natural fibers by creating micro-cracks or fiber swelling and weaken the NFRP structurally [4].

Another problem with natural fibers is that they have limited thermal stability. The temperature of natural fibers should not exceed 200 °C. Beyond this temperature, the fibers will degrade, and the mechanical properties of the natural fibers will be significantly reduced. Several studies found that this thermal stability problem depends on the lignin rate of natural fiber [4]. Manfredi et al. [61] proposed that the decomposition of the lignin starts at 200 °C, which is the main reason for natural fiber's thermal problems. Kumar et al. and Sarkar et al. [62, 63] proposed that lower lignin content in the natural fiber causes degradation to begin at a higher temperature. This means that there are differences between different natural fibers in the aspect of thermal stability. Due to this characteristic, Manfredi et al. [61] proposed that flax fiber has the best thermal resistance since its lignin content is the lowest among natural fibers.

Regarding the dependence on harvesting time, Pickering et al. [6] have found a significant increase in the average tensile strength of hemp fiber during the growth period from 99 days to 114 days and reaches optimum average tensile strength at 114 days. Das et al. [64] have found that the optimum days of growth for jute fibers should be 120 days; beyond 120 days, tensile strength reduces significantly.

### **2.3. Surface Modification of Natural Fibers and Polymers**

The problems mentioned in the previous section are severe limitations to developing and applying natural fibers. However, especially in the last ten years, much progress has been made to reduce or, if possible, eliminate these prob-

lems. The most important limitations are dependence on harvesting region and time, poor compatibility with the polymer matrix, the tendency to moisture absorption, and thermal stability issues. Each of these limitations became the subject of different studies, and efforts have been made to find a solution to these problems.

The problems and the solutions and solution proposals to these problems have been thoroughly investigated, and these findings will be explained in a detailed manner with different studies to introduce the solutions for these disadvantages to make biocomposites more favorable.

Various researchers proposed that surface modification must be used to increase poor compatibility with polymer matrix. Surface modification of NFRPs not only increases the poor compatibility problem of natural fibers but also solves other essential issues of biocomposites, such as decreasing water hydrophilicity and increasing thermal stability by removing lignin and hemicellulose. However, different views on surface modification should be applied to different natural fibers. Due to these significant advantages and differences in the application of surface modification, this topic will be investigated thoroughly.

Wu et al. [65] have proposed that applying magnesium hydroxide (MH) as the surface modification significantly increases the rupture and tensile strength of kenaf fiber. First, the surface of kenaf fibers has treated with a five wt% alkali solution, then modified with MH. According to Wu et al. [65], these mechanical properties improved interfacial compatibility between kenaf fibers and polyester resin after modifying the surface of kenaf fibers with MH. In addition to improved mechanical properties, kenaf fiber-reinforced polymer composites with MH modification showed excellent thickness swelling and water absorption with deficient water intake, thus improving another problem of NFRPs.

Suizu et al. [66] have suggested that mercerization of ramie fiber-reinforced biopolymer composites increases the fracture strain of ramie fibers, thus improving the toughness of the green composite significantly. In addition to the toughness, mercerized ramie fibers absorb almost twice as much impact energy as the green composite using untreated ramie fiber reinforcement. Other than mechanical properties, water absorption was also studied by Suizu et al. [66], and they found that the impact energy of the ramie fiber-reinforced biopolymer composite with mercerization increased as the water content increased. However, the article also indicates that this increase in impact properties is due to softening of the ramie fiber reduces the deformation resistance and the strength of the composites. Also, they have established that, even though low water addition (2 wt% to 5 wt%) increases the composites' impact energy. It can be anticipated that the impact energy and general mechanical properties of the composite would decrease with further increased water content. This finding on water absorption coincides with Hargitai et al. [29] and Munoz et al. [60].

Qiu et al. [67] have worked on surface modification of hemp fiber-reinforced unsaturated polyester composites to increase the composite's mechanical properties by improving the interfacial adhesion. They have proposed that treating hemp fibers with 1,6-diisocyanatohexane (DIH) and 2-hydroxyethyl acrylate (HEA) significantly increases the tensile strength, flexural modulus of rupture and flexural modulus of elasticity, water absorption resistance, and dramatically improves the interfacial adhesion between hemp fiber and UPE.

Lu et al. [68] have worked on the effect of 5 wt% alkalis and 5 wt% silane treatments on the thermal stability of hemp fiber. They have proposed that both treatments effectively removed pectin and wax, along with partial removal of lignin and hemicellulose, which increased the mechanical properties and adhesion with the polymer matrix. It is worth noting that while both treatments improved the thermal stability of the hemp fiber, alkali-treated fibers showed higher thermal stability compared with the silane treatment results.

Oh et al. [69] have modified the surface of hemp fibers using five wt% alkali solution and oils (soybean and corn) on hemp-PP composites. They have found that alkali treatment removed the non-cellulose parts, such as lignin and hemicellulose, thus reducing the fiber diameter of hemp fibers. Higher temperatures during the surface treatment process mean better removal of non-cellulose parts. However, when the fiber was treated with alkali solution at temperatures higher than 100 °C, the tensile strength was decreased due to the accelerated cellulose hydrolysis. Cellulose hydrolysis occurs at higher temperatures and causes the cellulose to be decomposed by alkali into aldehydes and acetone. This causes a reduction in tensile strength and poor mechanical performance overall. Alkali-treated hemp fibers treated with soybean and corn oil also have increased water resistance and interfacial performance. Even though both oils have increased the compatibility between hemp fiber-reinforcement and PP matrix, soybean has performed better than corn oil, thus making soybean a better choice. The paper concluded that 100 °C is the optimum temperature for maximum lignin and hemicellulose removal without damaging the cellulose part of the fiber to increase the overall mechanical performance of hemp fibers.

There are different results on the alkali treatment of hemp fibers, however. According to Islam et al. [70], alkali treatment removes lignin and hemicellulose, resulting in better-separated fibers with cleaner surface topography and better thermal stability. However, they have also found that the fiber's tensile strength was reduced due to the removal of lignin and degradation of the cellulose chains. It is worth noting that at the surface treatment stage of this paper, hemp fibers were treated with five wt% alkali solution at 120 °C for 60 minutes, which further proves the argument made by Oh et al. [69] that when fiber treatment

temperature is higher than 100 °C, alkali removes cellulose along with excessive amounts of lignin and hemicellulose. Väisänen et al. [71] also proposed that the alkali treatment of hemp fibers reduces tensile strength slightly. They treated the hemp fibers with ten wt% alkali solution at 95 °C for 60 minutes. They have established that the main reason for this reduction is the removal of lignin and degradation of cellulose chains, the same as Islam et al. [70]. However, Väisänen et al. [71] suggested that the main reason for this contradiction in the effect of alkali treatment on natural fibers is the longer treatment time and relatively high concentration of the alkaline solution (NaOH) used in the current study in terms of increasing or decreasing the tensile strength. It is also worth noting that previous research made by various researchers [65–70] has used five wt% surface treatment solution utmost, thus proving the point of Väisänen et al. [71] in using excessive rate (10 wt%) of alkali solution.

There are various types of surface modification or surface treatment methods mentioned above. According to surveys, one of the most effective surface treatment chemicals is maleic anhydride grafted polypropylene (MAPP). Unlike other surface treatments applied to natural fibers, MAPP is added to the polymer matrix in this study to polypropylene (PP). Sullins et al. [72] have worked on the effects of alkali treatment on hemp fiber and MAPP treatment on PP pellets. Three different composites have been used: only alkali treated, only MAPP treated, and both alkali and MAPP treated hemp-PP composites. Both surface treatments have increased hemp-PP composite's tensile and flexural properties by having excellent interfacial adhesion with no apparent gaps between the fiber and matrix. A critical aspect of this work is that only MAPP-treated hemp-PP composites showed better tensile and flexural properties than only alkali-treated and both MAPP and alkali-treated hemp-PP composites. This paper indicates that instead of treating the surface of hemp fiber reinforcement, better results can be achieved with the MAPP treatment of the PP matrix. In addition to the significant increase in mechanical properties and better interfacial adhesion, Bledzki et al. [73] and Schirp and Stender [74] have established that the application of MAPP reduced the water uptake of NFRPs significantly, thus strengthening the idea of using untreated fibers with thermoplastic polymers with maleic anhydride grafted polymer (MAH) or when used with polypropylene, MAPP addition.

Niu et al. [75] have worked on hemp fiber-polypropylene composites' mechanical properties and thermal stability with MAH addition. They also used maleic anhydride grafted styrene-(ethylene-co-butylene)-styrene copolymer (SEBS-MAH) and maleic anhydride grafted Poly (ethylene octane) (POE-MAH) as a compatibilizer to improve the fiber-matrix interactions. They found that all composites containing compatibilizers showed higher storage modulus and interfacial adhesion between hemp fiber and PP

matrix than total composites. The addition of PP-MAH increased the tensile flexural strengths of the composite. It is also worth noting that the addition of SEBS-MAH and POE-MAH elastomer remarkably increased the notched and unnotched impact strengths, thus proving itself a valuable compatibilizer for areas with strong impact resistance is needed.

Another study on PP-MAH or MAPP is the paper by Merotte et al. [76]. This study used four non-woven biocomposites; flax-PP, flax-MAPP, hemp-PP, and hemp-MAPP. A critical aspect of this work is that they used PP and MAPP fiber instead of the usual PP and MAPP pellet or powder. They used a fiber-matrix ratio of 50 wt% at all these four composites. As expected, hemp fibers exhibited poorer tensile properties but showed better interfacial shear strength with PP and MAPP than flax. Both hemp and flax fiber have shown better tensile strength and tensile modulus values with MAPP fiber matrix than pure PP fiber.

Yan et al. [77] have compared two different types of hemp fibers: noil hemp fibers (NHF) and scutched hemp fibers (SHF). SHF is mechanically degummed raw material for textile hemp fiber production, and NHF is a by-product of textile hemp fiber production from SHF. It is the over-degummed hemp fiber with a much smaller diameter and length. These two types of hemp fibers are very different from each other visually, as shown in Figure 5. The NHF reinforced PP matrix composites and SHF reinforced PP matrix composites have their surface treated with MAPP, and the results have been compared with the NHF-PP and SHF-PP composites without surface treatment. They have found that most of the pectin, hemicellulose, and lignin were removed during the degumming process of noil hemp fibers. Due to this removal, NHF has performed better thermally when compared with SHF. They have also established that MAPP significantly improved the hemp fiber-PP adhesion, which leads to significantly better tensile strength, flexural strength, and impact strength by increasing the fiber-resin adhesion. The positive effects of MAPP coincide with the findings of Sullins et al., Bledzki et al., Schirp et al., Niu et al., and Merotte et al. [72–76].

Talla et al. [78] have worked on hemp-Polyethylene terephthalate (PET) composites. PET has been compounded with Polycaprolactone (PCL) to reduce the melting point of PET, and hemp fibers have been treated with an alkali solution to increase their thermal stability to be able to use PET and hemp fiber together. The results from pure hemp-PET composite have been compared with hemp-PET composite with additives such as clay grade Cloisite 30B, pyromellitic dianhydride (PMDA), and glycidyl methacrylate (GMA). The results have shown that melt-processed PET can be a suitable matrix for hemp-thermoplastic polymer composites. Hemp and PET, without any additives, can have a good bonding interface, thus making rearing coupling agents optional. With some trade-off between the mechanical and

structural properties, hemp-PET composites can be used without any added chemical and can be used instead of the widely popular hemp-PP. However, it should be noted that hemp fibers have been treated with an alkali solution to increase the fiber's thermal performance to match it with PET's thermal degradation ( $T_m > 200$  °C), so the hemp fiber-reinforced composite is not 100% chemical-free.

#### 2.4. Impact Performance of Biocomposites

Studies in the literature so far are mainly about improving tensile and flexural strength, with only a few studies on impact strength using surface modification. Although the tensile and flexural strengths are essential parameters to determine the mechanical characteristics of an NFRP, they are not sufficient alone. Impact behavior should always be considered, especially for composite materials, since composites react differently to impacts when compared with conventional materials, and most composite materials are used in areas where high impact strength is needed; in addition to that, impact performance is an essential indication of the overall strength of the composite materials [79]. Because of these requirements, papers and studies focusing on the impact properties of NFRP must be surveyed thoroughly too. For this reason, the focus of the following studies will be on the impact performance of natural fiber-reinforced biocomposites.

The Charpy impact behavior of gigantic bamboo fibers reinforced with epoxy composites was studied by Glória et al. [80]. They employed bamboo fibers with no surface modification as the matrix and DGEBA epoxy resin as the reinforcement material. Charpy impact testing was performed on 10%, 20%, and 30% bamboo fiber-reinforced composite biocomposites. According to the findings of this study, the Charpy toughness of biocomposites increases with increasing bamboo fiber density. This work shows that the Charpy toughness of biocomposites increases with increasing bamboo fiber density. However, they also note that this increase in Charpy toughness is relatively smaller than the other natural fibers due to defects introduced in the giant bamboo fiber during extraction by a manual cut of the hard culm with a razor blade, thus rendering the use of bamboo fibers ineffective at the areas where strong impact toughness is needed.

As in the previous study, Assis et al. [81] have worked on the Charpy impact behavior of natural fibers without surface modification as reinforcement and DGEBA epoxy resin as a matrix with triethylenetetramine (TETA) as a hardener. This study used banana fibers with 10%, 20%, and 30% in volume as natural fiber reinforcement instead of bamboo fibers. They have established that as banana fiber volume increased, the Charpy toughness of biocomposite increased significantly. This increase in toughness is apparently due to the low banana fiber/epoxy matrix interfacial shear stress. This results in higher absorbed energy because of a longitudinal propagation of the cracks throughout the



interface, which generates larger rupture areas than a transversal fracture. They have concluded that banana fibers have the best impact characteristics compared with other fibers. However, low banana fiber/epoxy matrix interfacial strength will be a significant problem and a limiting factor for other mechanical performance characteristics such as tensile, bending, and shear.

Pereria et al. [82] have worked on the Charpy impact behavior of epoxy matrix composites with jute fibers. They have used aligned jute fibers without surface modification as reinforcement material and DGBEA epoxy resin as a matrix material with TETA as a hardener. Similar to previous papers with giant bamboo fibers [29] and banana fibers [30], this paper also used three different biocomposites with 10%, 20%, and 30% in volume as a natural fiber matrix. They have found that jute fibers have performed well at the Charpy impact test. Similar to the previous results, impact toughness was measured as a function of fiber, so composites with a higher volume of jute fibers showed better impact characteristics than the lower volume of jute fiber composites. Pereria et al. also note that even though jute fibers have good impact properties, they underperform compared to banana fiber-reinforced biocomposites [80, 81].

In addition to these studies, previously surveyed papers from Suizu et al. [66] too, have focused on the impact properties of green composites with mercerized ramie fibers. Niu et al. [75] have worked on hemp-PP composites and proposed that with the addition of SEBS-MAH and POE-MAH, the impact strength of the composites increased significantly. Another study on hemp-PP composites that has tested the impact properties is the paper published by Yan et al. [77]. They have established that with the addition of MAPP, the impact strength of the composite increased. From the studies of Niu et al. and Yan et al. [75, 77], the conclusion can be drawn that with the correct chemical additions, impact strength enhanced-hemp fibers can compete against jute and banana fibers, which have higher impact resistance than pure hemp fibers [81, 82].

Thanks to the data and results from previous studies, recent studies on the impact performance of biocomposites and green composites are focused on the different areas with different natural fibers. Reddy et al. [83] studied the mechanical and wear performance of three different epoxy biocomposites reinforced with different natural fibers (Tapsi, Abolition Indicum, and Prosopis). They have concluded that Abolition Indicum has shown better impact properties among the biocomposites.

Al-Oqla et al. [84] have studied the flexural and impact performance of LDPE reinforced with green olive leaves. The increase in impact performance of the biocomposite with adding olive leaves indicates that olive leaves can be considered a low-cost and eco-friendly alternative reinforcement material.

The studies of Hassan et al. [85] and Liang et al. [86] focused on the impact performance of green composites. Hassan et al. [85] studied green composites' acoustic, mechanical, and thermal properties reinforced with three types of fiber wastes: cotton fly, coconut/coir husk, and sugarcane, with green epoxy resin as matrix material. They found an increase in impact performance with all natural fiber additions, with the cotton fiber reinforced green composite performing the highest. The study of Liang et al. [86] has worked on the impact performance of PLA reinforced with sisal fibers. However, it should be noted that sisal fiber was treated with an alkali solution before blending with PLA. They have also found increased impact performance with the addition of sisal fibers. They have also established that long-sisal fiber reinforced green composites have performed better than their short-sisal fiber reinforced counterparts.

### **2.5. Hybrid Biocomposites**

As mentioned before, for a material to be considered a composite material, it should consist of at least two different materials; this means that there is a possibility of manufacturing a composite material that consists of more than two different types of materials if it is feasible to use. These materials are called hybrid composites. Such examples may be suitable for this definition; a composite material with two reinforcement materials and a polymer matrix or one reinforcement material with two polymer matrices. However, even though there are unlimited possibilities that can be created by using this formula, there are few real-world applications with acceptable results.

Petrucci et al. [87] have used various hybrid composite laminates based on basalt fibers combined with flax, hemp, and glass fibers. They have manufactured three different hybrid composites using EC360 epoxy as a matrix material: GFB, which consists of glass fiber, flax fiber, and basalt fiber; GHB, which contains glass fiber, hemp fiber, and basalt fiber; FHB, with flax fiber, hemp fiber, and basalt fiber, respectively. Their mechanical tests have shown that in terms of flexural performance, GFB has performed the best, followed by FHB and GHB, respectively. This order is also preserved in the case of post-impact flexural loading, suggesting that the addition of glass fiber offers a much better result in the presence of flax fibers than in the presence of hemp fibers. This formula also increases the impact toughness of composite by adding basalt fibers.

Another study on natural and glass fiber-reinforced polymer hybrid composites was carried out by Kong et al. [88]. They have used regenerated cellulose fiber with glass fiber as reinforcement material and epoxy resin as a matrix material. They have established that the natural fiber must be sandwiched between the glass fiber layers to maximize the toughness of hybrid composite material. Clark and Ansell [89], which is considered one of the first studies on a



hybrid glass fiber/natural fiber composite, found that sandwiched jute fibers between glass fibers maximize the toughness of hybrid composite, which is the source of the claim made by Kong et al. [88].

Sanjay and Yogesha [90] have established various advantages of hybrid composites against pure glass fiber or pure natural fiber composites, but the problem with hybrid composites is not due to their compatibility between synthetic and natural fibers; instead is due to using synthetic fibers in the composite.

Due to the general characteristics of synthetic fibers, hybrid composites with synthetic fibers are non-biodegradable, non-recyclable, and non-green, thus rendering compositely unusable in the areas where using biodegradable or recyclable materials is compulsory due to regulations [4]. No matter how good hybrid composites are, this problem prevents the wide use of synthetic fibers with natural fibers.

Due to some obligations to use biodegradable and recyclable materials in specific industries (especially in the automotive industry), natural fiber's use has increased significantly over the last 20 years. Thermoplastic polymers reinforced with natural fibers are only recyclable [4]. However, to manufacture a fully biodegradable composite material, matrix material should be biodegradable (biopolymer), just like the natural reinforcement fiber.

Various studies used a biodegradable matrix (biopolymer), as mentioned by Mitra [11]. Considerable studies have been made on green composites with natural fibers and biopolymers such as starch, polylactide (PLA), polycaprolactone (PCL), and Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV). Even though they have environmental advantages, Mitra [11] also noted that these biopolymers have tensile and flexural strengths lower than 100 MPa, limiting their usage in high-strength applications.

The application areas of green composites are extensive [91]. They can be used for environmental concerns and sustainability [92, 93] or as construction materials [94]. The production of green composites can also be achieved with significantly different materials, as long as they are recyclable and biodegradable [95, 96]. The study by Scaffaro et al. [97] shows a wide variety of areas for the use of green composites. They used PLA-based green composites reinforced with agricultural and marine wastes and studied the possibility of three-dimensional printability of these green composites. They have established that with a slight difference in molecular weight and filler aspect ratio, it is possible to 3D print the biocomposite. Another study by Leow et al. [98] utilized spent coffee grounds as reinforcement materials. They established that with the addition of spent coffee grounds and acetone, the performance of the composite material increased. Kamble et al. [99] have worked on green composites reinforced with waste cotton fibers. The increase in performance of an epoxy matrix with reinforcement

indicates that waste cotton fibers are an environmentally friendly and economical alternative to synthetic fiber reinforced composites.

The increased concern for the environment heightens the search for sustainable and environmentally friendly materials for various applications. When a wide variety of application areas and types of reinforcement materials are considered, green composites may be an answer to this search [100, 101].

## 2.6. Performance and Degradability of Green Composites

Various studies focused on using and performing natural fiber-reinforced and biopolymer matrix biocomposites, with their specific name, green composites. The study by Stapulionienė et al. [102] used short hemp fibers (20–30 mm in length) as reinforcement material with PLA fibers as matrix material. They have found that the direction of fibers at the stress direction may significantly influence the mechanical properties. However, this study's most important result is the possibility of successfully using hemp and PLA fibers together.

Song et al. [103] have also worked on hemp-PLA fiber green composites. The degummed and surface-treated hemp fibers are mixed with PLA fiber to create fiber pellets; then, fiber pellets are mixed with PLA pellets. Thus, composite pellets were created. Hemp-PLA fiber composite pellets have been manufactured from these composite pellets with the help of injection molding. After the mechanical tests, they concluded that this process increases the mechanical properties with the help of PLA fibers and silane treatment.

Mukherjee and Kao [104] proposed that both PLA and natural fibers are hydrophilic so that this property will facilitate better adhesion. However, they have found poor adhesion between natural fiber and PLA, probably due to debonding during mechanical testing or poor approximation during composite production. The hydrophilicity of PLA means that, as seen in natural hydrophilic fibers, PLA is also subjected to water or moisture absorption. The surface modification increases the interfacial adhesion and eliminates the natural fiber and PLA's water or moisture absorption problems. Therefore, it is essential to compete with synthetic and other natural fibers with synthetic polymers without compromising the biodegradability of these two materials.

It is also worth mentioning that degradability features are essential to compete with sustainability issues. For example, Shibata et al. [105] have worked on the degradability of biodegradable polyesters reinforced with abaca fibers and found from the soil-burial test that the composites containing untreated abaca fibers have shown the highest weight loss after being soil-buried for 24 weeks, thus indicating the highest biodegradability when compared with treated abaca fibers.

### 3. CONCLUSIONS, CHALLENGES, AND FUTURE PERSPECTIVES

In this work, the studies on natural fibers and natural fiber-reinforced polymers have been reviewed. The micro-and macrostructure of natural fibers has been explained. The advantages and the limitations of natural fibers have been discussed, and the methods used to eliminate these limitations have been examined.

It can be seen from this review that natural fibers can replace synthetic fibers as the primary reinforcement materials for composites. Despite their limitations, their advantages of recyclability and degradability make them a critical part of environmental issues. Also, it has been seen that with the help of correct surface modification, limitations of natural fibers, namely, moisture absorption, poor compatibility, and low thermal stability, can be reduced to acceptable levels or, if possible, wholly eliminated.

These aspects make natural fibers and biocomposites a critical material for composite materials and the environment. This importance will be increased with new natural fibers, biopolymers, and methods for producing these natural fibers and biopolymers.

However, there are still challenges for natural fiber-reinforced polymers to be utilized in areas with high safety standards, such as the cabin interior of commercial aircraft. Poor fire resistance of natural fibers is a big concern for this area. There is also the issue of the degradability of natural fibers and biopolymers. This might be considered an advantage for parts with a short life span or those used as consumables, but this aspect might be a disadvantage for long-life span parts. The degradability of the NFRP must be examined and correctly understood to predict a part's life limit appropriately, whether structural or not.

As an alternative to petroleum-based synthetic polymers, biopolymers can be enhanced. More natural materials can be introduced as resource materials to minimize reliance on petroleum and expand the range of biopolymer resources. These new natural resources can help achieve thermoplastic biopolymers' objectives with improved fire resistance and strength.

In addition, there is a gap in the literature on the aging of biocomposites subjected to different service environments. The studies must cover this area to increase the possible service areas of biocomposites. For example, a study that includes an aging test at cryogenic temperatures may enable biocomposites for deep-space applications.

For the implementation of large-scale production of NFRPs, different and more efficient production methods must be studied. Biocomposites can be produced with the same production methods as traditional synthetic composites. However, novel production methods must be introduced to NFRPs for mass production. Additive manufacturing of NFRPs can be an alternative to traditional composite production methods.

There are many areas for the potential use of biocomposites. More studies on NFRPs are needed to fill the literature gap about biocomposites and make them available for more areas and industries. The areas mentioned above and the challenges that come with them are the future of biocomposites and green composites.

#### DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

#### CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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#### PEER-REVIEW

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